Postdoc Fellowships for non-EU researchers

Final Report

Name	Ya Zhang
Selection	2013
Host institution	University of Antwerp
Supervisor	Annemie Bogaerts
Period covered by this report	from 01/12/2013 to 13/9/2015 (incl. maternity leave)
Title	Comprehensive Simulations of microdischarges with a Particle-In-Cell/Monte Carlo/Direct Simulation Monte Carlo model

1. Objectives of the Fellowship (1/2 page)

"Microdischarges (MDs)" or "microplasmas" are gas discharge plasmas generated in small dimensions, typically 100's of µm, and operating at gas pressures up to 1 atm [1]. The most remarkable advantage of MDs is that the discharge configurations can still be stable at atmospheric pressure and high discharge power densities (100 kW/cm^3). Indeed, it is hard or even impossible to maintain a non-equilibrium high pressure plasma in other configurations for any length of time, because small fluctuations tend to be unstable and trigger a rapid rise in the gas temperature (i.e., transition to a thermal plasma arc). Because of the important use of MDs, as they can indeed produce a stable non-equilibrium plasma at atmospheric pressure, a good insight in the MD behavior is desirable. During the research fellowship of Dr. Ya Zhang, she has obtained this insight by computer simulations.

Specifically, a comprehensive model based on the Particle-In-Cell/Monte Carlo/Direct Simulation Monte Carlo (PIC/MC/DSMC) method was developed and applied to simulate the behavior of an atmospheric pressure MD with both rf and dc frequencies for different gap distances, as well as an rf driven atmospheric pressure dielectric barrier glow discharge in argon at steady state.

Moreover, a packed bed dielectric barrier discharge was simulated by a two-dimensional particle-in-cell/Monte Carlo collision method developed within the VSim simulation package [1]. This type of discharge is of special interest in the field of gas pollutants and hydrocarbon conversion.

[1] Nieter C and Cary J R 2004 Journal of Computational Physics 196 448–473.

2. Methodology in a nutshell (1/2/ page)

MDs are generally at non-equilibrium and the electron velocity distribution is typically non-Maxwellian. It is therefore most accurate to use a kinetic model, like a PIC/MC model. On the other hand, the plasma density in MDs can be as high as 10^{23} m^{-3}, and the electron temperature is about 1 eV, which will lead to a small Debye length (in the order of 10^{-4}-10^{-5} cm) and a high electron oscillation frequency (around 10^{14}-10^{15} Hz). As is well known, an explicit PIC model requires to resolve the Debye length and the electron oscillation frequency, which necessitates the use of several thousands of cells and billions of time steps. This will lead to a high computational cost. Moreover, the high pressure makes the situation even worse, because the simulations require a much longer time to reach steady state. The alternative approach is to adopt an implicit PIC method [2], which allows us to use much larger space and time steps, while keeping the same accuracy. Here, we thus use such a direct implicit method.

In order to reach the objectives mentioned above, the three models that constitute the PIC/MC/DSMC method, i.e. Particle-In-Cell, Monte Carlo and Direct Simulation Monte Carlo, have been coupled and incorporated into a unified framework.

The PIC model is used to describe the motion of the charged particles and the electromagnetic field. The charged particles included in the model are singly charged positive or negative ions, as well as the electrons. The MC model handles the collision processes between the charged particles and the background gas. Finally, the DSMC model deals with the collisions and chemical reactions among the neutral species.

The particle motion can be decomposed into the movement and the collisions. In the so-called "motion stage", the charged particles move in the self-consistent electromagnetic field generated by the other charged particles, and both velocities and coordinates will change; the neutral particles are not affected by the electromagnetic field, but also move as they have some velocity. In the "collision stage", only the velocity of the particles will change, but the coordinates remain the same, due to the relatively short interaction time.

[2] G. Lapenta, J. U. Brackbill, and P. Ricci, Phys. Plasmas 13, 055904 (2006).

3. Results (6-8 pages)

During these 18 months, I have studied several types of MDs:

(1) Study of radio-frequency (rf) driven MD

We applied a direct implicit PIC-MC method to study a radio-frequency (rf) argon MD at steady state in the glow discharge limit, in which the microdischarge is sustained by secondary electron emission (SEE) from the electrodes. The plasma density, electron energy distribution function (EEDF) and electron temperature were calculated in a wide range of operating conditions, including driving voltage, MD gap and pressure. Also the effect of gap size scaling (in the range of 50-1000 µm) on the plasma sustaining voltage and the peak electron density at atmospheric pressure were examined, which has not been explored before. In our simulations, three different EEDFs, i.e., a so-called three temperature hybrid mode, a two temperature α mode and a two temperature γ mode distribution, are identified at different gaps and voltages. The maximum sustaining voltage to avoid a transition from the glow mode to an arc is predicted, as well as the minimum sustaining voltage for a steady glow discharge. Our calculations elucidate that secondary electrons play an essential role in sustaining the discharge, and as a result the relationship between breakdown voltage and gap spacing is far away from the Paschen law at atmospheric pressure. The effect of rf-voltage on the electron and ion densities at atmospheric pressure is shown in Fig. 1. In all cases, the electron and ion densities are more or less equal to each other as expected, except in the sheaths, where the ion density is slightly higher, creating a net positive space charge. The densities rise with applied voltage in both discharge gaps, as expected. Furthermore, the maximum plasma density in the 100 µm gap is significantly larger than in the 500 µm gap for the same rf-voltage (i.e., a difference of one order of magnitude at 100 V, as is clear from Fig. 1). Again, it is important to realize that, despite the high collision frequency at atmospheric pressure, the ions and electrons do not necessarily exhibit a local behavior in a MD. When the discharge is in a so-called hybrid mode (i.e., sustained by SEE), a "local discharge" means that the SEE electrons cannot reach the bulk region and most of them will lose their energy in the sheath. A "nonlocal discharge", on the other hand, means that the SEE electrons can reach or pass through the bulk region and induce ionization in the bulk plasma.

This work has been published in JOURNAL OF APPLIED PHYSICS: Y. Zhang, W. Jiang, Q. Z. Zhang and A. Bogaerts, Computational study of plasma sustainability in radio frequency microdischarges, J. Appl. Phys. 115, 193301 (2014).

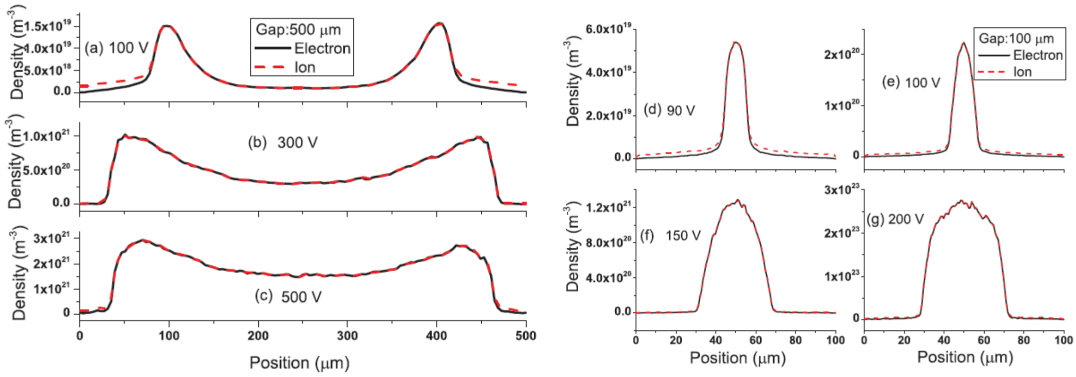

FIG. 1: Electron (solid lines) and ion (dashed lines) density profiles for two different gap sizes, i.e., 500 μm (left panel, with rf-voltages of 100 V (a) , 300 V (b) and 500 V (c)) , and 100 μm (right panel, with rf-voltages of 90 V (d) , 100 V (e), 150 V (f) and 200 V (g)), at atmospheric pressure.

(2) Study of direct current (dc) driven MD

Second, we applied the PIC-MC model to simulate the plasma kinetic properties at steady state in a parallel-plate direct current argon glow MD under various operating conditions, such as driving voltage (30-1000 V) and gap size (10−1000 μm) at atmospheric pressure. Also the effect of gap size scaling (in the range of 10 − 1000 μm) on the breakdown voltage, peak electron density and peak electron current density at breakdown voltage is examined. The breakdown voltage is lower than 150 V in all gaps considered, as shown in Fig. 2.

This work has been published in JOURNAL OF PHYSICS D: APPLIED PHYSICS: Y. Zhang, W. Jiang and A. Bogaerts, Kinetic simulation of direct-current driven microdischarges in argon at atmospheric pressure, J. Phys. D: Appl. Phys. 47 (2014) 435201.

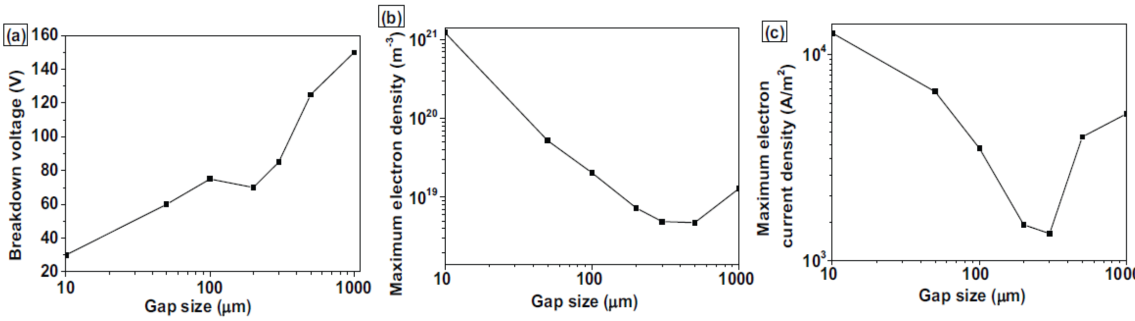

FIG. 2: Breakdown voltage (a), peak electron density (b) and peak electron current density (c) at this breakdown voltage, as a function of gap size.

(3) Study of local electrical breakdown in sub-micrometer metallized film capacitors (MFCs)

We applied the PIC-MC model to study the local electrical breakdown in sub-micrometer (0.2 μm) metallized film capacitors as a direct-current MD with 0.2 μm gap in a range of different voltages and pressures. The discharge process is significantly different from a conventional high pressure discharge. Indeed, the high electric field due to the small gap makes that the discharge is sustained by field emission. At low applied voltage (15 V), only the electrons are generated by field emission, while both electrons and ions are generated as a stable glow discharge at medium applied voltage (50 V). At still higher applied voltage (100 V), the number of electrons and ions is rapidly multiplying, the electric field reverses and the discharge changes from glow to arc regime, as shown in Fig. 3. It is easy to see that at high voltage (100 V), the electric field is reversed, and a positive electric field (of 1.3 × 108 V/m) is observed in the bulk around 0.06 μm, forming a potential barrier. This potential barrier prevents the electrons from being lost, so the bulk electrons are rapidly multiplied, and at 1 ps, an avalanche occurs, forming a quasi-neutral density region. Moreover, this leads to two potential wells at both sides of the barrier. A deep potential well near the cathode (with amplitude of -7×10^9 V/m) results in significant heating of the electrons up to 45 eV.

This work has been published in NEW JOURNAL OF PHYSICS: W. Jiang , Y. Zhang and A. Bogaerts, Numerical characterization of local electrical breakdown in sub-micrometer metallized film capacitors, New J. Phys. 16 113036 (2014).

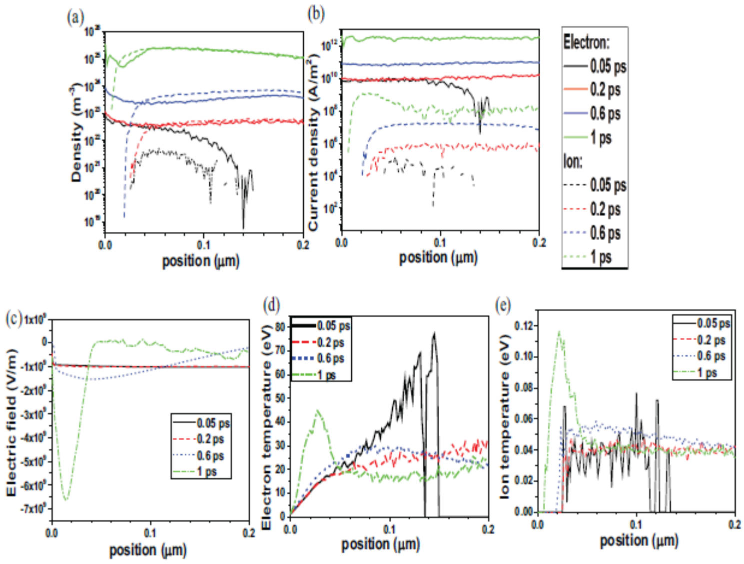

FIG. 3: Space-time evolution of (a) the electron and ion densities, (b) the electron and ion current densities, (c) the electric field, (d) the electron temperature and (e) the ion temperature, at10 atm and 100 V applied voltage.

(4) Study of packed-bed dielectric barrier discharge (DBD) with 2D PIC/MC method

We used a two-dimensional particle-in-cell/Monte Carlo method to simulate a packed-bed DBD in air at atmospheric pressure. The plasma behaviour in a parallel-plate DBD is simulated, comparing for the first time an unpacked (empty) DBD with a packed bed DBD, i.e., a DBD filled with dielectric spheres in the gas gap. The geometry used in the model is shown in figure 4(a-b). The calculations are performed in air, at atmospheric pressure and with applied voltage amplitude of −20 kV. When comparing the packed and unpacked DBD reactors with the same dielectric barriers, it is clear that the presence of the dielectric packing leads to a transition in discharge behaviour from a combination of negative streamers and unlimited surface streamers on the bottom dielectric surface to a combination of predominant positive streamers and limited surface discharges on the dielectric surfaces of the beads and plates. Furthermore, in the packed bed DBD, the electric field is locally enhanced inside the dielectric material, near the contact points between the beads and the plates, and therefore also in the plasma between the packing beads and between a bead and the dielectric wall, leading to values of 4×10^8 V/m, which is much higher than the electric field in the empty DBD reactor, i.e., in the order of 2×10^7 V/m, thus resulting in stronger and faster development of the plasma, and also in a higher electron density. The locally enhanced electric field in Fig. 5 and the electron density in Fig. 6, in the case of a packed bed DBD are also examined for different times with dielectric constant $\epsilon_r = 22$ (ZrO_2). It is clear that the presence of the dielectric beads in the discharge greatly enhances the electric field inside and near the dielectrics, both between the packing beads, and between the beads and the dielectric surfaces. This is especially true inside and at the surface of the bottom dielectric plate and around the contact points between the bottom dielectric surface and the beads in the bottom row, due to the increased charge deposition on the bottom dielectric surface and between the packing beads in the bottom row, as shown in Fig 5 (d). Indeed, the electrons are accumulated and trapped in the gap between the packing beads in the bottom row and the bottom dielectric plate, which is clearly seen in Fig. 6 (c-d). The maximum electric field and electron density are also shown in tables 1 and 2 respectively, for three different dielectric constants, i.e., $\epsilon_r = 22$ (ZrO_2), $\epsilon_r = 9$ (Al_2O_3) and $\epsilon_r = 4$ (SiO_2). The enhanced electric field is stronger and the electron density is higher for a larger dielectric constant, as shown in tables 1 and 2, respectively, because the dielectric material is more effectively polarized. These simulations are very important, because of the increasing interest in packed bed DBDs for environmental applications.

This work is accepted in NEW JOURNAL OF PHYSICS: Y. Zhang, H. Wang, W. Jiang and A. Bogaerts, Two-dimensional particle-in cell/Monte Carlo simulations of a packed-bed dielectric barrier discharge in air at atmospheric pressure, New J. Phys, in press (2015).

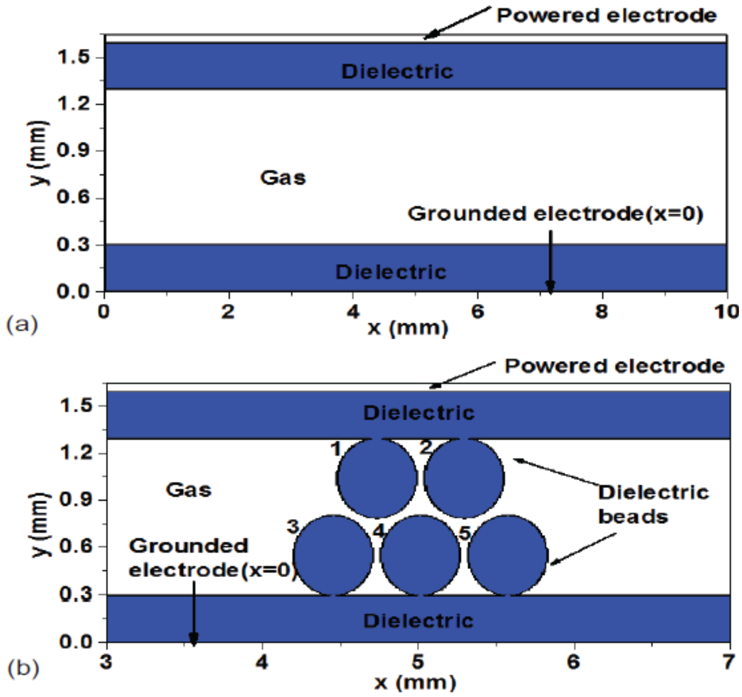

FIG. 4: Geometry used in the model with a domain of 10 mm*1.65 mm, for the unpacked DBD (a) and packed bed DBD (b). The 1 mm discharge gap is bounded by 0.3 mm thick dielectric plates at both the top and the bottom, and the top dielectric plate is covered by 0.05 mm thick metal electrodes. The bottom electrode is grounded (x=0). In (b) dielectric spheres with radius of 0.25 mm are inserted in the gap.

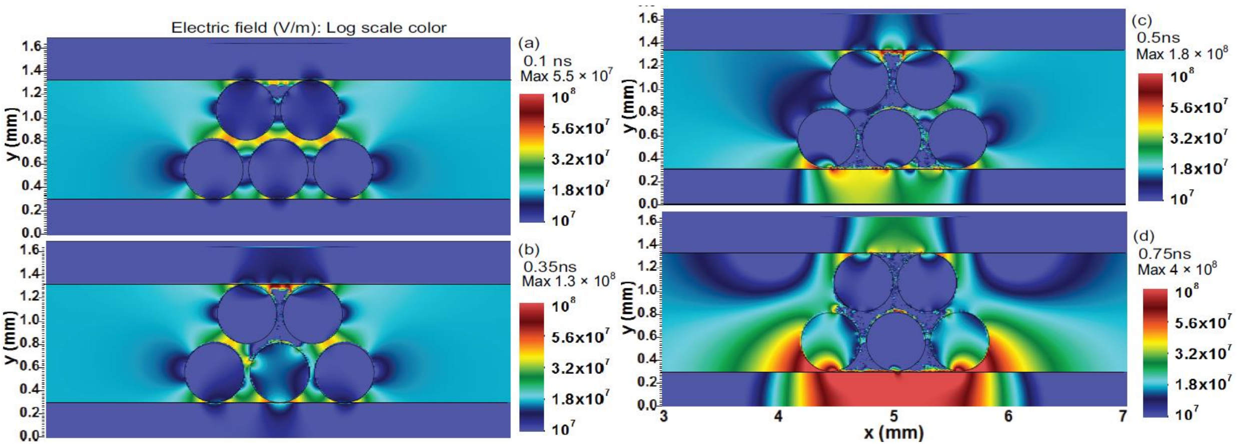

FIG. 5: Electric field amplitude |E| at different times, in the packed bed DBD case, with $\epsilon_r = 22$ (in all dielectrics).

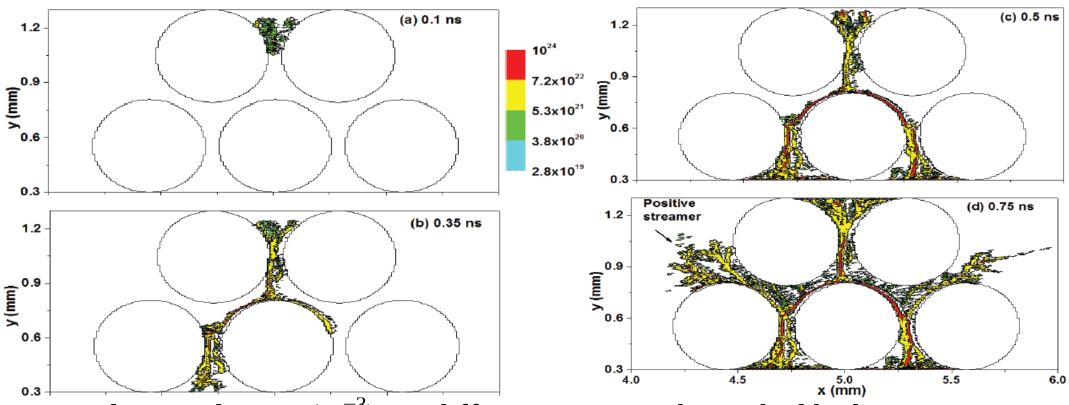

FIG. 6: Electron density (m^{-3}) at different times, in the packed bed DBD case, with ϵ_r = 22 (in all dielectrics).

Electric field (V/m)	0.1 ns	0.35 ns	0.5 ns	0.75 ns
ε_r=22	5.5×10^7	1.3×10^8	1.8×10^8	4.0×10^8
ε_r=9	3.1×10^7	6.0×10^7	1.4×10^8	3.4×10^8
ε_r=4	1.9×10^7	4.9×10^7	6.0×10^7	1.5×10^8

Table 1. Maximum value of the electric field at different times, for three different dielectric constants in all dielectrics (plates and beads), i.e., ϵ_r = 22, ϵ_r = 9 and ϵ_r = 4.

Maximum Electron density (m^{-3})	0.1 ns	0.35 ns	0.5 ns	0.75 ns
ε_r=22	3.8×10^{22}	2.6×10^{23}	1.4×10^{24}	1.1×10^{24}
ε_r=9	1.4×10^{22}	1.1×10^{23}	5.2×10^{23}	5.6×10^{23}
ε_r=4	1.1×10^{22}	8.5×10^{22}	2.2×10^{23}	2.5×10^{23}

Table 2. Maximum electron density at different times, for three different dielectric constants in all dielectrics (plates and beads), i.e., ϵ_r = 22, ϵ_r = 9 and ϵ_r = 4.

(5) Study of rf driven DBD

We investigated an rf driven atmospheric pressure dielectric barrier glow discharge (DBGD) in argon at steady state with a one-dimensional PIC/MC/DSMC method, where the dielectric barriers are self-consistently included in the simulation region and coupled with the gas gap region. The dielectric barrier glow mode at steady state is sustained even at high driving rf voltage, indicating that the glow discharge is more stable with dielectric barriers compared to a bare electrode case. The discharge is in the α mode even for large driving voltage, as shown in Fig. 7, with most of the electrons having low energies trapped in the bulk plasma region due to negative charge accumulations on the dielectric barrier surfaces. As shown in Fig. 8, two sharp peaks appear in the sheaths. In detail, the temperature presents the α mode distribution, with a wide peak in the bulk region and subpeaks in the sheaths. The EEDFs transit from a three-temperature distribution to a Druyvesteyn distribution, with increasing rf-voltages. Negative charges are deposited on the dielectric surfaces, inducing an effective negative well in which the majority of the electrons with low energy are trapped in the bulk plasma region.

This work will soon be submitted to APPLIED PHYSICS LETTERS: Y. Zhang, W. Jiang and A. Bogaerts, Kinetic modelling of radio-frequency driven dielectric barrier glow discharges in argon at atmospheric pressure (2015).

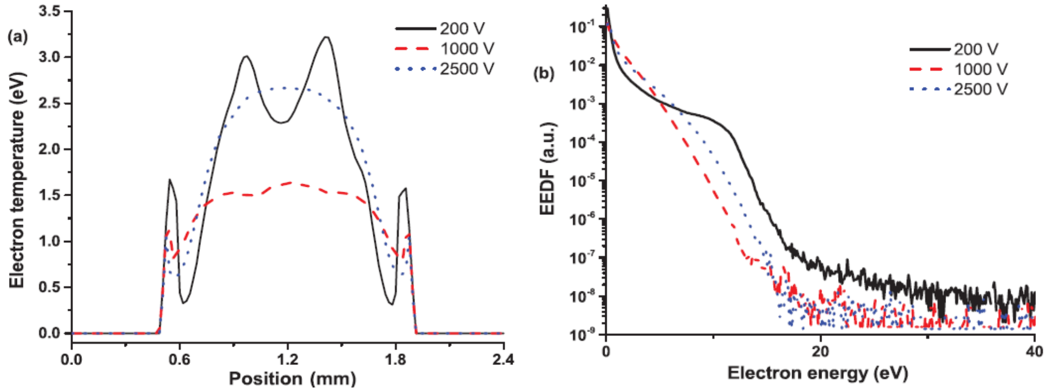

Fig 7. Electron temperature profiles (a) and space-time-averaged EEDFs (b) for the 1.4 mm gap DBD with two parallel dielectric barriers of thickness 0.5 mm, for three different rf-voltages.

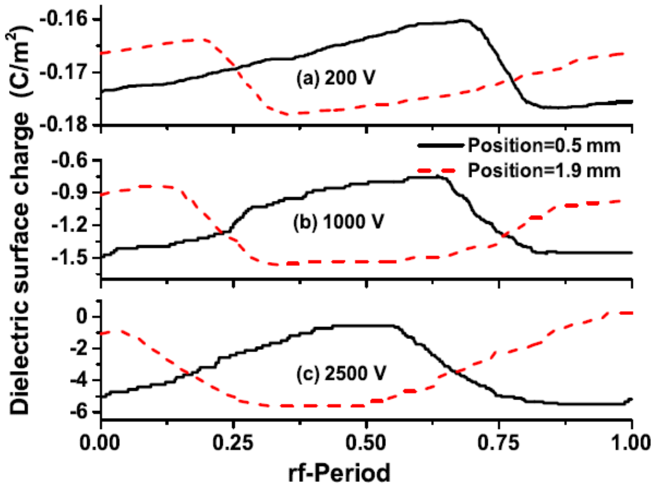

Fig 8. Dielectric surface charge at position of 0.5 mm (solid line) and 1.9 mm (dashed line), for the 1.4 mm gap DBD with two parallel dielectric barriers of thickness 0.5 mm, for different rf-voltages.

4. Perspectives for future collaboration between the units (1 page)

This postdoctoral fellowship was very useful, as it yielded interesting results, but also because it forms the starting point for continued collaboration between the host group and the postdoctoral researcher. More specifically, in our future collaboration:

(1) We will improve the 1D PIC/MC/DSMC code and extend it to a 2D model with considering photo-ionization effects that may have important features on the filamentary and streamer behavior.

(2) We will modify the 2D PIC/MC collision method developed within the VSim simulation package, to a 3D model that is more realistic, and easier to compare with experiments.

(3) For the packed bed DBDs, we plan to consider radical formation and its transport to the surface, as well as the surface conditions (such as conductivity, porosity, and the effect of adsorbed gas molecules), by coupling the PIC model with a fluid model. Indeed, the PIC method alone is too time consuming to consider these processes.

(4) We will try to compare the simulation results for the packed bed DBDs with experimental data, as soon as they become available in literature. However, it is not straightforward to perform plasma diagnostic measurements in a packed bed DBD.

5. Valorisation/Diffusion (including Publications, Conferences, Seminars, Missions abroad...

[1] Y. Zhang, W. Jiang, Q. Z. Zhang, and A. Bogaerts, Journal of Applied Physics 115, 193301 (2014).
[2] Y. Zhang, W. Jiang and A. Bogaerts. Journal of Physics D: Applied Physics 47, 435201(2014).
[3] W. Jiang,Y. Zhang and A. Bogaerts. New Journal of Physics 16 113036, (2014).
[4] Y. Zhang, H. Wang, W. Jiang, and A. Bogaerts, accepted by New Journal of Physics (2015).

6. Skills/Added value transferred to home institution abroad (1/2 page)

(1) The PIC/MC/DSMC model and code that has been developed in this project, is a general-purpose tool, that cannot only be applied to MDs, but also to many other types of low temperature plasmas, such as low and high pressure capacitively coupled plasmas, inductively coupled plasmas, etc. So it will significantly strengthen the simulation capabilities, both in the host lab and the home institution abroad.

(2) Various configurations of MDs and DBDs have been studied in detail. Especially, the electron and ion densities, the EEDFs, and the electron temperature profiles are presented in a wide range of operating conditions to obtain a better insight in the general plasma behavior. Furthermore, the results illustrate that (atmospheric pressure or lower pressure) rf and dc MDs in the glow regime can be operated in an energy efficient manner, which is of crucial importance because of energy considerations. These results will be a good reference for the ongoing experimental and theoretical research both in the host and home institute, as well as for the design of some practical industrial reactors.