Modelling of a packed bed dielectric barrier discharge plasma reactor

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Abstract: A packed bed dielectric barrier discharge plasma reactor was computationally studied with a fluid model. Two different complementary 2D geometries were used to mimic the intrinsic 3D problem. It was found that the contact points between the pellets are of direct importance to initiate the plasma. When a high enough potential is applied, the plasma will be able to travel through the gaps in between the pellets from wall to wall.

Keywords: plasma, modelling, fluid, helium, DBD, packing, dielectrics

1. Introduction

The dielectric barrier discharge (DBD), as a source of non-thermal plasma, has been of interest for environmental applications for quite some time. Indeed, plasma can be an interesting alternative for conventional thermal methods, because the input energy solely goes to heating up the electrons, while the rest of the plasma particles (i.e., radicals, ions, neutrals) stay at room temperature. However, the feasible energy efficiency appeared to be on the low side. In order to overcome this, a dielectric packing was introduced in the gas gap of the reactor, forming a so-called packed bed plasma reactor (PBPR). A PBPR is built by packing dielectric pellets inside a dielectric barrier discharge (DBD) reactor. Some commonly used dielectrics are glass (dielectric constant ~4-6), quartz (dielectric constant ~4-7), aluminium oxide (dielectric constant ~10), zirconium oxide (dielectric constant ~10s), titanium oxide (dielectric constant ~100s), ceramic (dielectric constant ~10s-10000s), and ferroelectric materials (dielectric constant ~100s-10000s) [1]. This type of reactor has been widely experimentally investigated in the last few years in applications like ozone generation [2, 3], gaseous pollutant removal [4, 5] and H_2 production [6, 7] resulting in a significant improvement in energy efficiency, up to a factor 12. The improvement is attributed to the increased electric field at the various contact points of the packing pellets with each other and with the walls, due to the polarization of these materials. This strong field induces a locally higher electron temperature and thus higher reaction rates, but also a lower plasma density [8]. However, the underlying mechanisms are still poorly understood. In order to improve the applications, a better understanding of the packing effect is needed. Plasma diagnostics, e.g., by probes, optical emission or laser diagnostics, is not straightforward in a PBPR, as the optical and physical access is limited due to the presence of the pellets. Therefore, a computational approach is very much of interest to gain more insight.

In the past, only a few numerical studies have been performed for packed bed DBD reactors [9-12]. Chang and Takaki *et al.* [9, 10] developed a simplified 1D parallel plate N_2 plasma model, focusing on the determination of the electron density, electron energy and electric field strength, based on the applied voltage, the distance between the plates, the free volume and the dielectric constants of the pellets and the gas. In the 1D model, however, the void between the pellets was assumed to be spherical, which is not the case in reality. Kang *et al.* [11] developed a 2D model to study the impact of the introduction of dielectric pellets in a parallel plate DBD reactor on the evolution and characteristics of the typical micro discharges. However, the arrangement of the packing pellets was limited to two pellets on top of each other and furthermore no plasma chemical reactions were included in this model. Finally, Russ *et al.* [12] used a 2D hydrodynamic model, to study transient microdischarges in a packed bed DBD reactor filled with dry exhaust gas (80% N_2, 20% O_2 and 500 ppm NO). This model did include an extensive chemical reaction set with 23 different species reacting in 120 different plasma reactions, but it only simulated a short one-directional discharge (of a few 10s of nanoseconds) via a constant applied potential difference, and the presence of a void channel true the packing from one wall to the other (see below) was lacking in their model.

In principle, a PBPR can only be studied in three dimensions to take into account the packing geometry as it is in real life. However, the duration of such 3D simulations is predicted to be well over a few months, for just a couple of periods of the applied potential. Due to these computational limitations, a 2D model is needed to gain the first insights in the mechanism of a plasma discharge in a PBPR. It is of course key in such model to simplify the 3D geometry without compromising its authenticity.

2. Model description

Using COMSOL's built-in plasma module, a two-dimensional axisymmetric fluid model is built with semi kinetic treatment of the electrons. The model is based on solving a set of coupled differential equations that express the conservation of mass, momentum and energy, for the different plasma species. These equations contain production and loss terms for the different species, based on the chemical reaction set. For the electrons and positive ions, the flux is based on the drift-diffusion approximation. The Poisson equation will also be solved to self-consistently calculate the electric field distribution, using the densities of the different plasma species as input. Further details on the model scheme can be found elsewhere [13].

The 3D unit cell contains two important geometrical properties, which need to be taken into account in the 2D adaptation. The contact points between the dielectric materials (i.e., pellets, walls) are the first property. The contact points will strongly change the electric field distribution in the gas gap and will therefore have a big influence on plasma generation and distribution throughout the gap. On the other hand, these contact points also cause the voltage-driven electrode to be in direct contact with the grounded electrode, through a "channel of dielectric material", lowering the electric field strength over the entire gap. Studies with simple electrostatic models (i.e., not including any plasma reactions) in 2D and 3D, however, indicated that its influence was only minor. Therefore, we did not necessarily focus on including this phenomenon. The second property is the existence of a so-called "channel of voids". Indeed, just like the fact that all the dielectric materials are linked, all the voids in between the pellets are also connected to each other, resulting in a direct channel from the dielectric layer on top of the voltage-driven electrode to the grounded electrode. This channel must be present because the plasma must be able to travel from one side of the discharge gap to the other. However, it is not possible to make a single axisymmetric 2D adaptation of the 3D unit cell with both the "channel of voids" present and all the packing pellets in direct contact. Therefore two different geometries are studied, each focussing on one of the properties. In both geometries the rotational axis is located on the left side. The first geometry shows two packing pellets with a dielectric constant of 25 (zirconia), with diameters of 2.25 mm, on top of each other making direct contact with each other and with the walls, which are 4.5 mm apart. A 2.5 mm thick layer of alumina (dielectric constant of 9) is covering the voltage driven electrode. To overcome computational difficulties, the contact points are slightly enlarged and rounded. This geometry will be called the "contact point model". The second geometry shows three packing pellets, i.e., two pellets on the left, now with a diameter of 2.00 mm and spaced apart so that they both are in contact with the opposite wall, leaving a gap of 0.5 mm in between. On the right side, the third packing pellet is added to recreate the "channel of voids", therefore we will call this the "channel of voids model". It must be stated that this packing pellet after rotation around the axis will not be a sphere but a torus. We are aware that both geometries are not a true copy of the real life geometry, but we are convinced that they can give us insights to understand the real life plasma better.

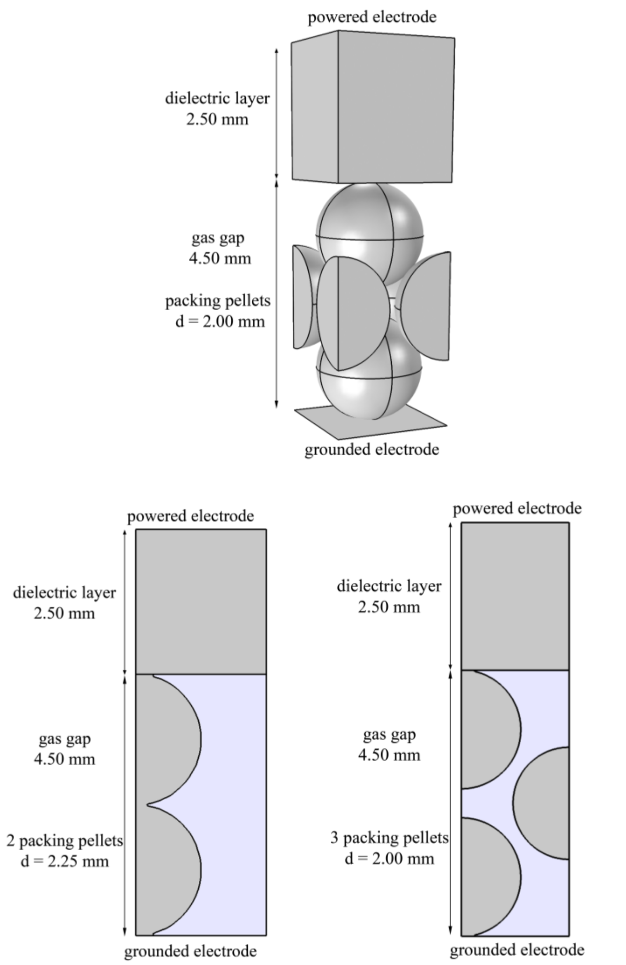

Fig. 1. 3D unit cell of the packed bed plasma reactor (PBPR) and its 2D representations used in the model.

Two different 2D axisymmetric geometries, based on a 3D unit cell of a close-packed PBPR are used to best represent the 3D problem (Fig. 1.). For two reasons we have chosen helium as discharge gas. First of all, the reaction chemistry of this noble gas is quite simple. Second, it has the ability of forming a homogeneous instead of a filamentary discharge in a DBD reactor, and this is what we will be simulating by using a fluid model to describe the plasma. The model considers six different species, i.e., electrons (e), neutral helium atoms (He), positive helium ions (He^+), positive helium molecular ions (He_2^+), metastable helium atoms $He(2^1S)$ and $He(2^3S)$ combined into one effective level He* and helium dimers (He_2^*). The different species interact with each other by 23 elementary reactions. "Bolsig+", a software programme that solves the Boltzmann equation for the electrons, uses the input collision cross-sections form the LXcat database, to generate five electron impact reaction rates as a function of the mean electron energy [14, 15]. It also calculates the transport coefficients of the electrons as a function of mean energy. The reaction rate coefficients of the other 18 reactions, namely recombination reactions with electrons and heavy particle reactions between ions, atoms and exited species, are taken from literature, if possible as a function of electron temperature [16, 17]. Some reaction rate expressions even have the gas temperature as a variable, but we kept it constant at 300 K. The mobilities of the ions are taken from literature [18], and then used to calculate their diffusion coefficients using the Einstein relation. For the neutral particles, the diffusion coefficients are calculated with the Chapman-Enskog equation. Furthermore also four surface reactions are included, namely the quenching of helium atomic and molecular metastables, and the electron-ion recombination of He^+ and He_2^+ ions to ground state helium atoms with a 0.05 probability to send out a secondary electron of 5 eV.

A simple 1D model for a reactor without packing, with a similar geometry, was also simulated to benchmark the reaction set to existing results of Golubovskii *et al.* [19], and excellent agreement was reached. The results will also make it possible to, in a later stage, compare a PBPR with an empty reactor, to clearly indicate the influence of the introduction of a packing in a reactor.

3. Results and discussion

We first present the results for the "contact point model" with an applied sinusoidal voltage of 3.5 kV peak-to-peak with a frequency of 23.5 kHz.

Fig. 2. shows the time averaged electric displacement field and electron temperature over one period of applied potential. Due to the polarization of the packing pellets as a result of the applied potential over these materials, the electric field at the contact points will be enhanced. The electrons in this region will receive more energy than in the rest of the reactor and therefore, this will lead to a breakdown. In other words, the plasma will be initiated near the contact points. If the applied potential is on the low side, i.e., 3.5 kV peak-to-peak, the plasma will stay in this region and pretty quickly die out because of recombination at the opposite pellet wall. A second breakdown in the same half period of the applied potential can occur when the breakdown voltage in this region is met again. When a higher potential of 10 kV peak-to-peak is applied, the discharge can receive more energy and spread out beyond the region in between the pellets towards the bulk. The reason for this is that the electric field over the full height of the gap will be enhanced, and eventually be strong enough to cause a breakdown. Fig. 3. illustrates this difference by showing a plot of the electron density at the moment of discharge breakdown, with a low and a high applied potential (left and right, respectively).

At low applied potential, the "channel of voids model" shows some similar results. Since the pellets are not in contact in this model, the plasma will not be directly initiated at this location as in the "contact point model", but still similar "rules" are followed. The plasma will be initiated in the region with a strong electric displacement field, namely in between the two packing pellets on the left. When the sinusoidal applied potential increases towards one of its maxima, the electric field at the region above and below the third pellet on the right will also be strong enough to initiate a discharge. For a relatively low applied power, i.e., 4 kV peak-to-peak, the plasma stays in these respective regions, as seen in the time averaged plot of the electron density in Fig. 4 on the left. On the other hand, when the applied potential is increased to 10 kV peak-to-peak, displacement of the discharge will occur through the so-called "channel of voids" from one

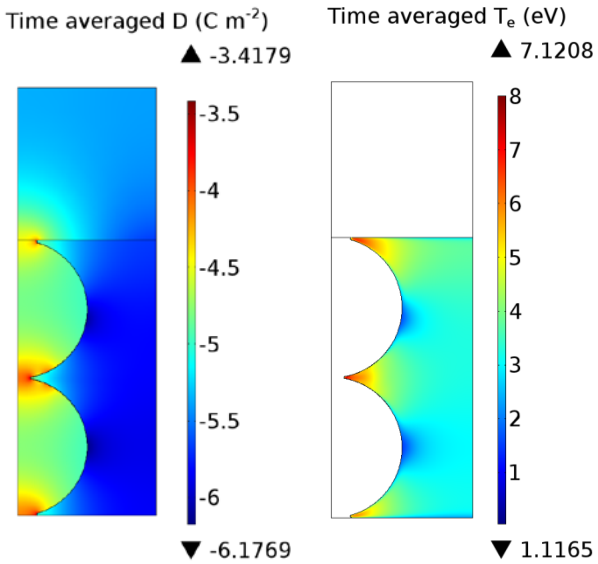

Fig. 2. Time averaged electric displacement field D (C m^{-2}) and electron temperature T_e (eV) over one period of applied potential. The scale on the left is logarithmic.

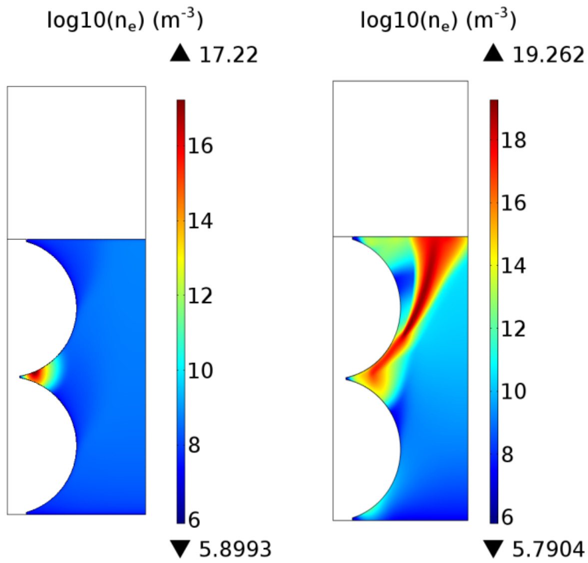

Fig. 3. Electron density n_e (m^{-3}) around the time of gas breakdown for an applied voltage of 3.5 kV peak-to-peak (left) and 10.0 kV peak-to-peak (right), both in logarithmic scale.

void to the other, all the way towards the opposite wall. This result implies that the inclusion of a "channel of voids" is of key importance when studying a packed bed DBD reactor.

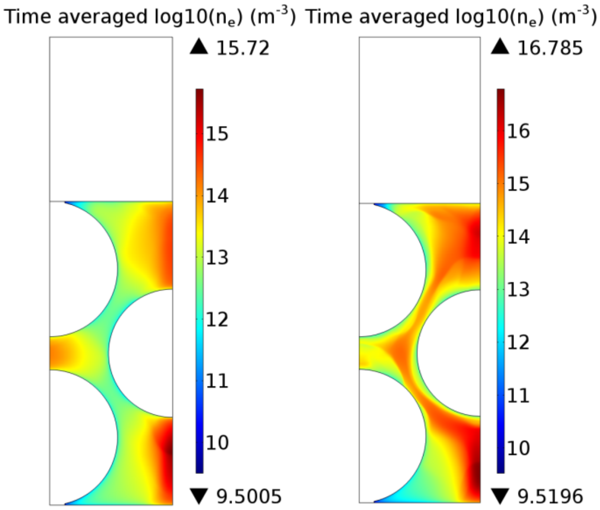

Fig. 4. Time averaged electron density n_e (m^{-3}) for an applied voltage of 4.0 kV peak-to-peak (left) and 10.0 kV peak-to-peak (right), again both in logarithmic scale.

4. Conclusions

The results from the "contact point model" teach us that the plasma in a PBPR is initiated at the contact points, since this is the place with the highest electric field strength and thus the highest electron energy. When a low potential is applied, the plasma stays in this region. A higher applied potential will cause the discharge to further spread out into the bulk of the reactor towards the walls. The "channel of voids model" shows some similar results. In this model, the plasma also initiates at the position with high electric field strength, and stays in this region at lower applied potentials. Yet again, when a higher potential is applied, the plasma can travel away from these regions from one void to the other and eventually towards the walls. It is therefore of uttermost importance to include this "channel of voids" in a packed bed model, since the plasma will not stay localized when the applied potential meets a certain value.

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6. References

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