Influence of catalyst packing configuration on the discharge characteristics of dielectric barrier discharge reactors - a numerical investigation

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A two-dimensional numerical fluid model is developed for studying the influence of packing configurations on dielectric barrier discharge (DBD) characteristics. Discharge current profiles, and time averaged electric field strength, electron number density and electron temperature distributions are compared for the three DBD configurations, plain DBD with no packing, partially packed DBD and fully packed DBD. The results show a strong change in discharge behaviour occurs when a DBD is fully packed as compared to partial packing or no packing. While the average electric field strength and electron temperature of a fully packed DBD are higher relative to the other DBD configurations, the average electron density is substantially lower and may impede the DBD reactor performance under certain operating conditions. Possible scenarios of the synergistic effect of the combination of plasma with catalysis are also discussed.

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I. INTRODUCTION

Packed bed dielectric barrier discharge (DBD) reactors that can operate at atmospheric pressure and ambient conditions are being increasingly used in remediation of a variety of gaseous pollutants such as SOx, NOx, VOCs, etc. When a catalyst is used as a packing material, the synergistic effect of the so-called ‘plasma-catalysis’ helps to activate the catalyst at relatively lower temperature and improve the selectivity towards desirable products. The catalysts can either be fully packed in the entire discharge gap or be placed either radially or axially covering only a fraction of the discharge gap. This second configuration known as partial packing, has been shown to avoid the typical disadvantages of the “packed bed effect”.

While many studies with DBD in a fully packed configuration have found enhanced performance in comparison to a plasma alone system, some studies have also reported a decrease in efficiency. Experimental studies comparing the nature of interactions between packing materials and plasma in fully and partially packed DBDs have observed a significantly strong filamentary discharge in the partially packed DBD as compared to the combination of weak filamentary discharge and surfaces discharge observed in fully packed DBD. It is postulated that suppression of filamentary discharges in a fully packed DBD may be the reason behind the negative performance observed in some cases. Packing typically induces effective polarisation and enhances the electric field strength at the contact points, however the discharge behaviour changes depending on the particular packing configuration. The changes in polarisation have a strong influence on the electron density and electron energy in the discharge gap, which play a major role in deciding the DBD performance.

However in experimental studies it is difficult to characterize the discharge parameters, and the packing itself adds an extra hindrance in visibility for discharge diagnostics. Computational modelling can be used as a complimentary tool to understand the discharge characteristics and optimize the system in a directed way, providing more quantitative process-parameter relationships. There have been some computational studies on packed bed DBD reactors in the past. Kang et al. developed a two dimensional (2D) model for ferroelectric packed bed barrier discharge reactor but did not include any plasma chemistry. Russ et al. used the so-called ‘donor cell’ method and developed 2D
However this study presented very short one-directional discharge (of a few 10s of nanoseconds) with limited results of spatial electric field and electron density distributions. Van Laer and Bogaerts developed a fluid model for a packed bed DBD with pure helium gas. The 3D packing was represented using two complementary axisymmetric 2D geometries. They highlighted that addition of packing led to a higher electron field strength and electron temperature at the contact points. Van Laer and Bogaerts also studied the effect of discharge gap size and dielectric constant of the packing material on the discharge characteristics using a 2D axisymmetric fluid model. In our previous work, we have developed a 2D fluid model for a partially packed DBD and showed the increase in electric field strength, electron energy and electron density as compared to a DBD with no packing.

However at present there is no numerical study comparing the discharge characteristics of DBDs in partially packed and fully packed configurations. In this work, for the first time, we have developed a 2D fluid model for helium DBD in three configurations, no packing, partial packing and full packing, to study how inclusion of packing material in different configurations affects the discharge characteristics and the reactor performance.

II. MODEL DESCRIPTION

In this work, the 2-D fluid model is applied to a cylindrical DBD reactor with two co-axial metal cylinders as electrodes. The dimensions of the reactor, plasma chemistry and the governing equations used in the fluid model have been described in our previous study. The fully packed DBD is represented using a similar approach as described by Van Laer and Bogaerts. However instead of using two separate 2D geometries, we have used one geometry, as shown in Figure 1. This geometry provides a comparative representation of the three dimensional packing. While the the packing pellets are not touching each other or the walls, the gap in between the beads and in between the walls and the pellets is optimized using electrostatic models to represent a similar electric field strength as observed when the contact points are included. This simplification not only saves a large computational expense, it also resolves the problem of using two separate geometries to represent the 3D problem. The proposed geometry also confirms with the so-called ‘channel of voids’ approach recommended by Van Laer and Bogaerts. The geometries used for representing the plain
FIG. 1. 2D geometry used in the model for representing a fully packed DBD.

DBD with no packing and the DBD with partial packing have been described in our previous study\(^ {24}\).

III. RESULTS & DISCUSSION

Figure 2 shows the discharge current profiles for the three DBD configurations. The current profile for DBD with no packing exhibits a single distinct pulse in both the positive and negative half cycle of applied potential, which is characteristic of atmospheric pressure glow discharge (APGD), typically obtained for helium DBD\(^ {25}\). Inclusion of packing, both partial and full, alters the discharge behaviour of the DBD, which is reflected in the change in discharge current profiles. However, while partial packing leads to a very small change (exhibiting two current peaks in the positive half cycle as opposed to the single peak observed in DBD without packing), the fully packed DBD shows a much dramatic transition in the discharge current signal and amplitude.

For the fully packed DBD, both the current signal profile and amplitude undergo a major transition suggesting a large alteration of the plasma discharge. The current profile (figure 2C) shows multiple current peaks (4-5) of varying amplitude in both the positive and the negative half cycle of the applied potential. Such a transition in discharge current profile has been observed previously in both experiments\(^ {5,13}\) and numerical simulations\(^ {18}\) of fully packed DBDs. It is postulated that this occurs due to the multiple breakdowns across the different points in the discharge gap as the gap voltage crosses the breakdown voltage multiple times.
FIG. 2. Discharge current profiles (red solid lines) in an atmospheric DBD in helium with (A) no packing, (B) partial packing, and (C) full packing, during one cycle of the applied potential of 3 kV peak-to-peak (black dashed lines) at a frequency of 20 kHz.

... during one period of applied potential\textsuperscript{13,18}. It should also be noted that the amplitude of the discharge current also decreases significantly, which is in accordance to that observed in experimental studies\textsuperscript{13}. One reason for such striking change in discharge current profile signal and amplitude is the ‘packed-bed effect’, which occurs due to the significant decrease in discharge volume that reduces the distance a typical microdischarge can travel in the discharge gap\textsuperscript{5,13,26}.

Based on the packing configurations, the void fraction of the DBD also changes. For the fully packed configuration, packed with spherical pellets, the void fraction if estimated to be about 39\%\textsuperscript{27}. Such dramatic change in void fraction has been shown to cause a significant change in discharge behaviour\textsuperscript{5}. Discharge of molecular gases in a fully packed DBD is modified from the typical filamentary mode of discharge to a prevalent surface discharge on the packing surface and spatially limited microdischarges\textsuperscript{5,28,29}. Comparatively, the void
Fig. 3. Time averaged logarithm to the base 10 of electric field strength (V/m) over one period of applied potential 4.0 kV peak-to-peak for (A) plain DBD with no packing, (B) partially packed DBD and (C) fully packed DBD.

The fraction of partially packed DBD is 99.5%, almost same as the DBD with no packing, which is also reflected in the similar discharge current profiles obtained for the two discharges (figure 2 A and B).

More information on the exact nature of the discharge mode can be obtained by observing the distributions of electric field, electron density and electron temperature (energy) in the discharge gap. Figures 3, 4, and 5 show the time averaged electric field strength, electron number density, and electron temperature distributions over one period of applied potential for the three DBD configurations. The time averaged electric field distributions for the three DBD configurations are quite different from each other (figure 3). As expected there is an
electric field enhancement at the contact points between pellet-pellet and pellet-dielectric barrier. Such an enhancement is typically attributed to increased charge deposition due to more effective polarisation at the contact points\textsuperscript{13,18}.

The electric field strength for partially packed and fully packed DBD are in the similar range i.e. between 4.5 to 7. Comparatively, the same value for an empty DBD is far less, reaching to a maximum of only 4.9. The packing in both the configurations, distorts and enhances the electric field strength, mainly at places where there is close contact between packing pellets or between packing and dielectric barrier. The distortion of the electric field distribution in the discharge gap depends on the shape of the packing material. For spherical objects, intensification of the electric field occurs at the poles of the solid object with a local minima observed at the equatorial plane\textsuperscript{30,31}. For the particle packing arrangements described in this work, this enhancement in electric field occurs at the vertical poles (top surface of the particles), which can be clearly seen in case of fully packed DBD from the time averaged distribution plot shown in figure 3C. Enhanced electric field can be observed at the vertical poles and in the contact region between the two pellets. However there is also a local minima in the equatorial plane, which falls along the mid section of all pellets parallel to the dielectric barrier. Thus we obtain regions of low electric field strength between the two pellets along these equatorial planes. Similarly for the partially packed DBD, we obtain electric field strength enhancement at the contact points and at the top surface of the packing, and the local minima in the region between the two pellets along the equatorial plane. The partially packed DBD is discussed in more detail in our previous study\textsuperscript{24}. These results on electric field strength enhancement at the poles of packing material and at the contact points between packing and dielectric layer are in accordance with that reported previously in experimental and numerical studies on packed bed DBDs\textsuperscript{13,16,18,22}.

The time averaged electric field strength of fully packed DBD is of similar magnitude to that of the partially packed DBD. However it should be noted that there are more regions of high electric field strength in the discharge gap (i.e. values > 6) for fully packed DBD compared to partially packed DBD. This factor needs to be seen in the context of discharge volume and the high energy electron density distribution in that volume. As discussed in the previous study\textsuperscript{24}, partial packing does not lead to any major reduction in discharge volume or the total electron count compared to a plain DBD with no packing. On the other fully packed configuration leads to a drastic reduction in discharge volume\textsuperscript{12}. To reflect on this
more, we need to study the time averaged electron density distributions, which are shown in figure 4.

As can be seen from figure 4, the time averaged electron density for plain DBD and DBD with partial packing are of similar magnitude (\( \sim 10^{10} \)), which falls within the typical range for APGD. For the fully packed DBD however, the time averaged electron density is three orders of magnitude smaller (\( \sim 10^7 \)), and this value is characteristic of atmospheric pressure Townsend discharge (APTD)\(^{32} \). Based on the magnitude of electron density, it can be inferred that inclusion of full packing inside the discharge gap of a helium DBD operating at applied potential 4.0 kV peak-to-peak and 20 kHz, leads to a transition of discharge mode from APGD to APTD. This also explains the significantly different discharge current signal and amplitude, as observed in figure 2 and is in accordance with the experimental observations of a significant transition in discharge behaviour reported previously for fully packed DBDs\(^{5,13} \).

The electron density distribution shows that for the DBD with no packing, the maximum electrons are located in the vertical central plane (parallel to the dielectric barrier), whereas for DBD with partial packing, the maximum electron density is concentrated along the horizontal central plane (perpendicular to the dielectric barrier). While the electron density of the fully packed DBD is significantly lower compared to the other two DBD configurations, the distribution is more spread out across the discharge gap in between the pellets (figure 4C).

Electron temperature is another important parameter of a DBD. This is directly correlated with the decomposition efficiency of the reactor, as higher the electron temperature, more will be the energy to break down chemical bonds of molecular pollutants. As can be seen from figure 5, the maximum electron temperature observed for DBD with no packing (\( \sim 3 \) eV) is substantially less than that obtained for DBDs with packing.

Comparing between partially and fully packed DBDs, the time averaged electron temperature is higher for fully packed DBD with a maximum of \( \sim 18-19 \) eV, while the same for partially packed DBD reaching only up to \( \sim 14 \) eV. It should also be noted that there are more regions of high electron temperature intensity in the discharge gap of the fully packed DBD, resembling the same points which showed high electric field strength, as seen in figure 3. Whereas for partially packed DBD, the high electron temperature is restricted to the contact points between pellets and dielectric barrier. Looking only at the electron
FIG. 4. Time averaged electron density (cm$^{-3}$) distribution over one period of applied potential 4.0 kV peak-to-peak for (A) plain DBD with no packing, (B) partially packed DBD and (C) fully packed DBD.

temperature distribution, one can conclude that the decomposition efficiency of the fully packed DBD would be higher than that compared to both partially packed DBD and DBD with no packing. However we also need to account for the significant reduction in electron density and the dramatic change in discharge behaviour caused by a reduction in discharge volume in fully packed DBDs. It should also be noted that this work only accounts for the change in discharge behaviour based on the electrical properties of the packing material, whereas the influence of the chemical and physical characteristics of the packing and how they would change under the influence of discharge have not been considered. It has been observed through experimental studies that the catalytic activity of a packing material can be enhanced under exposure of plasma, and this factor would also play an important role in
FIG. 5. Time averaged electron temperature (eV) distribution over one period of applied potential 4.0 kV peak-to-peak for (A) plain DBD with no packing, (B) partially packed DBD and (C) fully packed DBD.

calculating the performance efficiency of the DBD. We have shown here numerically that a helium DBD discharge behaviour would undergo a substantial change in fully packed configuration as compared to DBD with no packing or partial packing. This is in accordance with previous experimental studies that have also shown a significant change in discharge behaviour of fully packed DBDs when compared to DBDs with no packing\textsuperscript{5,13}.

Another important parameter that governs the performance of a DBD reactor is the dissipated power density. Table I shows the spatially and time averaged dissipated power density, electron density and electron energy for one voltage cycle, at applied potential 4.0 kV peak-to-peak for the three DBD configurations.

As we can see from table I, as the packing inside the discharge gap of DBD increases,
TABLE I. Spatially and time averaged dissipated power density \( (W m^{-3}) \), electron density \( (cm^{-3}) \) and electron temperature \( (eV) \) over one period of applied potential 4.0 kV peak-to-peak with applied frequency 20 kHz for different DBD configurations

<table>
<thead>
<tr>
<th>DBD configuration</th>
<th>Power density ( (W m^{-3}) )</th>
<th>Electron density ( (cm^{-3}) )</th>
<th>Electron energy ( (eV) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Packing</td>
<td>4.00x10^5</td>
<td>2.3x10^10</td>
<td>2.19</td>
</tr>
<tr>
<td>Partial Packing</td>
<td>5.64x10^5</td>
<td>2.20x10^10</td>
<td>3.21</td>
</tr>
<tr>
<td>Full Packing</td>
<td>7.13x10^5</td>
<td>5.8x10^6</td>
<td>7.33</td>
</tr>
</tbody>
</table>

the power density also increases. Thus while the DBD with no packing has a power density of 4.00x10^5, inclusion of partial packing increases it \( \sim 40 \% \) to 5.64x10^5, and the full packing increases the power density by \( \sim 80 \% \) to 7.13x10^5. Similarly we see an increase in average electron temperature as the volume fraction of packing material increases. However the increase in electron temperature is much more pronounced for a fully packed DBD, which shows \( \sim 230 \% \) increase when compared to DBD without any packing. These values contribute to the higher decomposition efficiency of full packed DBDs as observed in several experimental studies\(^2,6,7,8,9\). However the dramatic change in discharge behaviour in a fully packed DBD also decreases the electron density in the discharge, which shows a drop of \( \sim 100 \% \) when compared to DBD with no packing. On the other hand, the partially packed DBD does not show any significant drop in electron density (given negligible change in discharge volume and discharge behaviour), while showing a \( \sim 40 \% \) increase in power density and \( \sim 46 \% \) increase in electron temperature. This explains the relatively higher performance efficiency of partially packed DBD as compared to plain DBDs with no packing as observed in some experimental studies\(^5,33,34,35\).

The discharge characteristics in a DBD are closely linked to the available void volume in the discharge gap. Compared to the DBD with no packing, the void volume of partially packed and fully packed DBD are 99.5% and \( \sim 39 \% \) respectively. The smaller void fraction of fully packed DBD reduces the electron density, however the close packing also intensifies the electric field strength near the several contact points between the pellets. The higher electric field strength increases the electron energy and the power density as the amount of packing is increased.
The overall performance of the DBD in a fully packed configuration would depend on the trade-off between the reduction in efficiency due to the packed bed effect (leading to change in discharge mode and drastic reduction in electron number density), and the enhancement in electric field strength leading to the increase in electron temperature and the possible improvement in the catalyst activity of the packing under exposure of plasma discharge. If it is assumed there is no influence of plasma on the catalytic activity of the packing material, the major factor that can reduce the efficiency of fully packed DBD compared to plain DBD would be the reduction in electron density. This factor is related to the chemical composition and the residence time of the pollutant gases in the DBD. If the high energy electron count is proportionate enough to disintegrate the pollutants in the given operating conditions, the fully packed DBD would show a better decomposition efficiency compared to other configurations. However if the high energy electron count is inadequate, the fully packed DBD may show no change or even reduction in efficiency compared to DBD with no packing.

IV. CONCLUSION

To summarize, we have used a 2D fluid model to understand the influence of different packing configurations on the discharge characteristics of a helium DBD. For the operating parameters used in this study, we have found that there is a complete change in discharge mode in the fully packed DBD when compared to either partially packed DBD or DBD without any packing. However at a given applied potential, a fully packed DBD shows more effective polarisation, leading to enhanced electric field strength and higher average electron temperatures compared to the other DBD configurations. The interactions between plasma and packing are very complex and instead of a general rule, the synergistic effect of plasma-packing interaction needs to be understood individually for each specific case. For packing material that also acts as a catalyst, it would be essential to evaluate both the chemical and physical changes that packing undergoes due to plasma discharge and the change in electrical discharge behaviour induced by the packing. Influence of plasma on the catalytic activity can be studied using atomic scale simulations based on molecular dynamics or density functional theory\textsuperscript{36}. 

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