

# A mechanism for anomalously high voltages in high-pressure dc microdischarge mixtures of He, Ne and Xe.

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Electrical characteristics in the microdischarge experiments of Postel and Cappelli (*J. Appl. Phys.* **89**, 4719, 2001), show that voltages are higher in mixtures of helium and xenon than in pure xenon in the abnormal glow discharge regime. While originally attributed to the possible formation of heterodimer ions which have lower secondary emission coefficients, we show here that we can reproduce the general trends seen experimentally with a one-dimensional fluid model by taking into account the charge exchange process,  $\text{He}^+ + \text{Xe} \rightarrow \text{He} + \text{Xe}^+$ . The reaction rate coefficient used ( $10^{-9} \text{ cm}^3 \cdot \text{s}^{-1}$ ) corresponds to that for ions with energy of around 1 eV, which is not an uncommon energy for ions in the cathode sheath of strongly collisional microdischarges. Experimental results are also presented for mixtures of He, Xe, and Ne, at 50 Torr and 250 Torr.

In a plasma display panel (PDP) cell, as in some fluorescent lamps, ultraviolet (VUV) photons are produced by glow discharges in mixtures of xenon with other inert gases such as neon and/or helium<sup>1-3</sup>. The composition and proportions of each gas used for such a discharge is an important factor affecting the emission efficiency and the life-limiting sputtering of the cathode due to ion bombardment<sup>4-7</sup>.

Despite the importance of mixture chemistry in the performance of these light sources, researchers still lack a detailed understanding of the reactions which lead to the production of excited (radiating) states and ions, and to the establishment of the electrical characteristics of the discharge (e.g., sustain voltage in PDP cells).

Our research aims at isolating these reactions from affects associated with complex geometries and/or non-steady fields by carrying out experiments in direct-current microdischarges of planar geometries. In recent papers<sup>8,9</sup>, we have presented results on the vacuum ultraviolet emission and the electrical characteristics of these planar dc microdischarges. We noted that for the range of current density studied (abnormal glow regime) the discharge voltages in mixtures of helium and xenon were surprisingly higher than in either pure xenon or pure helium, when the xenon fraction exceeded about 10%. While we originally attributed this to the possible formation of  $\text{HeXe}^+$  ions that have lower secondary emission coefficients, Veronis and Inan<sup>10</sup> have modeled our specific experiment, and concluded that the required rate coefficient for the formation of this species would have to be unacceptably high. In this letter, we have revisited this problem, extending our experiments to mixtures of Xe and Ne, and to a wider range of current densities and pressures, and we show here that we can reproduce the general trends seen experimentally with a one-dimensional fluid model by taking into account charge exchange process. As shown below, the rate coefficients needed for the charge exchange reactions are consistent with those available in the literature for the ion energies expected.

Our experiments are performed in dc discharges between plane parallel stainless steel electrodes under conditions in which the pressure-electrode spacing product ( $pd$ ) is 5 Torr ·

cm, typical of conditions in PDP cells<sup>11</sup>. The diameter of each electrode is 2.3 mm and the electrodes are confined by ceramic sleeves to maintain predominantly one-dimensional current flow. Mixtures of either He/Xe or He/Ne are reported here, although we have also studied more complex tertiary mixtures, which we will report on in a future paper. It is noteworthy that the reproducibility of a given voltage measurement after a complete polishing of the electrodes was within 20 volts.

The central frame of Fig. 1 shows the measured discharge characteristics for different mixtures of helium and xenon. The voltage and current density are scaled by  $pd$  and  $p^2$  respectively, to directly compare conditions in which pressure is varied<sup>12</sup>. A striking feature in the experiments is that the minimum in the discharge voltage is about the same for all mixed gas compositions studied, and equal to that measured for the pure Xe case. It is also clear that for scaled current densities above  $10^{-2} \text{ mA} \cdot \text{cm}^{-2} \cdot \text{Torr}^{-2}$  the discharge voltages are higher than for pure xenon or pure helium, as we had seen previously<sup>8,9</sup>. We would expect the discharge voltage for He/Xe mixtures to be in between those of the pure Xe and pure He cases. We show later that indeed this is the case for Ne/Xe mixtures (e.g., left panel of Fig. 2).

We propose that charge exchange between  $\text{He}^+$  ions and Xe atoms ( $\text{He}^+ + \text{Xe} \rightarrow \text{He} + \text{Xe}^+$ ) may explain the relative positions of the voltage-current density ( $V$ - $j$ ) curves in these He/Xe discharges. The relative concentrations of  $\text{He}^+$  ions and  $\text{Xe}^+$  ions affects the discharge voltage because the secondary emission coefficient for  $\text{Xe}^+$  is much lower than that for  $\text{He}^+$ <sup>13,14</sup>. To our surprise, many simulations of PDP discharges that contain Xe and He do not include this charge exchange process<sup>15-17</sup>.

In order to test the importance of this charge exchange reaction in determining the discharge voltage, we have simulated our experimental conditions using the commercially available one-dimensional SIGLO model<sup>18</sup>. Although these fluid models, which are routinely used for simulating PDP discharges, do not capture all the complexity of the non-local effects of electrons or ions<sup>12,19,20</sup> they are well suited for testing the effects of charge exchange reactions on the trends expected

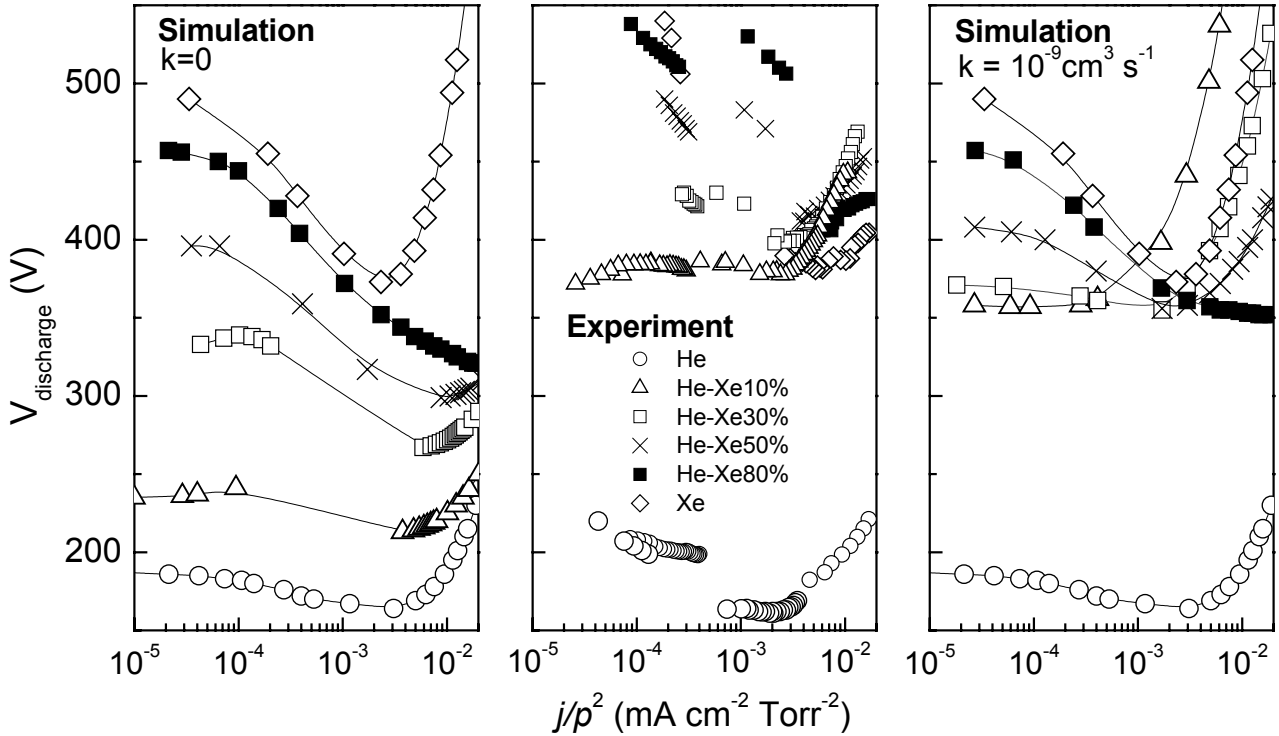


Fig. 1: Current-voltage characteristics for various He-Xe mixtures simulated without the charge exchange reaction:  $\text{He}^+ + \text{Xe} \rightarrow \text{He} + \text{Xe}^+$  (left), measured (middle) and simulated with charge exchange (right). Simulation conditions:  $\gamma_{\text{He}^+}=0.17$ ,  $\gamma_{\text{Xe}^+}=5 \cdot 10^{-3}$ , 5 Torr-cm,  $k$  represents the charge exchange rate coefficient.

in the discharge characteristics. The fluid model required as input, the mobility for the dominant ionic species (which is found to also control the discharge voltage). For the calculations presented here, we have used commonly employed mixture rules<sup>21</sup> in combination with ion mobilities in pure gases determined experimentally<sup>22,23</sup> and from Monte Carlo simulations<sup>24</sup>. It is noteworthy that the predicted voltages in the abnormal glow regime are found to be sensitive to the ion mobility, and so a direct quantitative comparison between predictions and measurements should be viewed with caution. Panels on either side of the experimental  $V$ - $j$  curves in Fig. 1, show the simulated results including (right) and excluding (left) the charge exchange reaction discussed above. The secondary emission coefficients for  $\text{He}^+$  and  $\text{Xe}^+$  are chosen in order to accurately reproduce the measured characteristics in the abnormal glow regime (beyond the minimum in the curve) in pure He and pure Xe respectively ( $\gamma_{\text{He}^+}=0.17$ ,  $\gamma_{\text{Xe}^+}=5 \times 10^{-3}$ ).

A charge-exchange rate coefficient of  $10^{-9} \text{ cm}^3 \text{ s}^{-1}$  was used in the results presented in Fig. 1. It is apparent that this rate leads to good qualitative agreement with experimental measurements. In particular, we see that in the abnormal glow regime, the simulations predict that for cases of 10-30% xenon in helium, the discharge voltage exceeds that of pure xenon, as is seen experimentally. It is also apparent that without this charge exchange process, the simulations fail to reproduce even the qualitative trends seen experimentally. Experimental measurements of this rate indicate a sensitivity to the relative energy between the colliding ions and neutral particles, and is found to vary between  $7 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$  at room temperature to  $10^{-9} \text{ cm}^3 \text{ s}^{-1}$  at 1eV<sup>25,26</sup>. In our discharge conditions, most of

the ions, which are produced at the boundary between the plasma and the cathode sheath edge, will gain an energy of a few eV's based on our estimated mobilities and a reduced electric field of  $E/p \sim 600 \text{ V} \cdot \text{Torr}^{-1} \text{ cm}^{-1}$  at the cathode surface. This should be seen as an upper limit on the mean ion energy, and so it is quite reasonable to use the value of  $10^{-9} \text{ cm}^3 \text{ s}^{-1}$  for the rate coefficient in our simulations. The use of this value leads to  $\text{Xe}^+$  as the dominant ion in all conditions shown, which is why the reduced voltage curves all have minimums of about the same value.

At this point, a discussion is warranted about why the predicted voltages including charge exchange for the gas mixtures of 10% - 30% Xe are close to or exceed those of the pure xenon case. The efficient charge exchange leads to a condition that limits the discharge voltage to at least that which is seen for the pure xenon case. In addition, however, our calculations indicate that for these conditions, the sheath thickness (largely controlled by the electron transport through the sheath, dominated by electron-He momentum transfer collisions) is significantly larger than the case of a pure Xe discharge, even for as low a He dilution of 20%. The discharge voltage is expected to scale as the square of the sheath thickness divided by the ion drift velocity (at constant current density)<sup>12,27</sup>. These two effects combined – the efficient charge exchange and the larger sheath thickness – gives the high discharge voltages seen in the abnormal glow regime, for the Xe-He discharge mixtures.

The measured discharge characteristics for Ne-Xe mixtures are compared to those predicted by the simulations in Fig. 2. The results of the simulations shown do not include the process

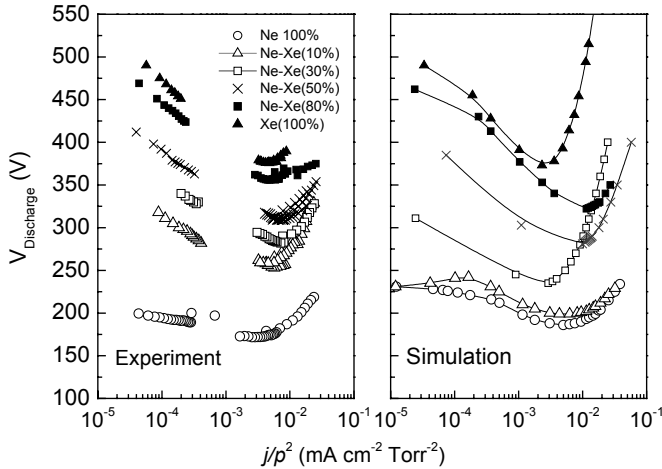


Fig. 2: Measurements (left) and simulated (right) current-voltage characteristics for various Ne-Xe mixtures.  $p = 50$  Torr,  $d = 1$  mm,  $\gamma_{Ne^+} = 0.17$ ,  $\gamma_{Xe^+} = 5.10^{-3}$ .

of charge exchange between  $Ne^+$  ions and Xe ( $Ne^+ + Xe \rightarrow Ne + Xe^+$ ). The rate coefficient for this reaction is found to be (and expected to be<sup>28</sup>) several orders of magnitude lower than that for the  $He^+ - Xe$  case<sup>26,28,29</sup>. It is apparent that there is good agreement between the simulations (excluding charge exchange) and measurements, and including charge exchange only has a minor effect on the predicted characteristics.

As mentioned in our previous studies, another possible explanation for the ordering of  $V$ - $j$  curves in the abnormal glow regime is the formation of molecular ions (such as  $HeXe^+$ ), which have a lower secondary emission coefficient. The formation of these heterodimer ions requires three-body collisions that are favored at higher pressures. Figure 3 depicts the results of measurements taken at two widely varying pressures, 50 Torr and 250 Torr. Focusing largely on the abnormal regime ( $j/p^2 > 10^{-2}$  mA·cm<sup>-2</sup>·Torr<sup>-2</sup>), it is apparent

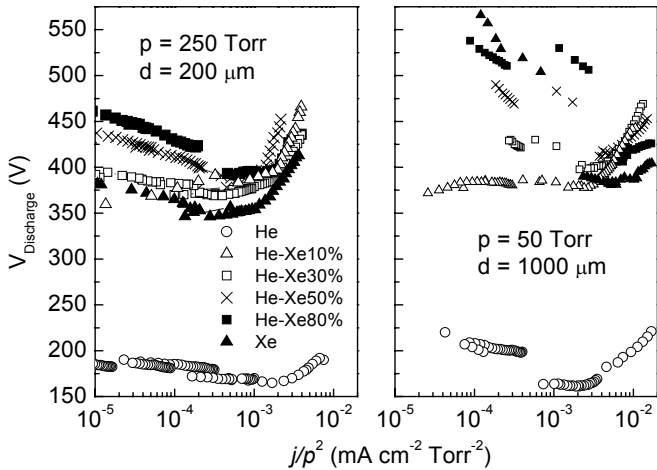


Fig. 3: Measurements of current-voltage characteristics for various He-Xe mixtures (left): 250 torr / 200  $\mu$ m, (right): 50 torr / 1000  $\mu$ m.

that the unusual ordering of the  $V$ - $j$  curves persists (indeed even further exaggerated) in the lower pressure case, despite the fact that the frequency of production of these molecular ions should be reduced by a factor of about 25. This provides further evidence that the formation of these heterodimer ions is not responsible for these unusual voltages.

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