1-1-1990

Ion-Ion Recombination In A Gas-Discharge

I. M. Littlewood

University of Central Florida

R. E. Pyle

University of Central Florida

Find similar works at: <https://stars.library.ucf.edu/facultybib1990>

University of Central Florida Libraries <http://library.ucf.edu>

This Article is brought to you for free and open access by the Faculty Bibliography at STARS. It has been accepted for inclusion in Faculty Bibliography 1990s by an authorized administrator of STARS. For more information, please contact STARS@ucf.edu.

Recommended Citation

Littlewood, I. M. and Pyle, R. E., "Ion-Ion Recombination In A Gas-Discharge" (1990). *Faculty Bibliography 1990s*. 101.

<https://stars.library.ucf.edu/facultybib1990/101>

Ion-ion recombination in a gas discharge

I. M. Littlewood and R. E. Pyle

Department of Physics, University of Central Florida, Orlando, Florida 32816

(Received 13 July 1989)

We present results of a Monte Carlo simulation of the recombination of positive methane ions and negative sulfur hexafluoride ions in an electric discharge at atmospheric pressure. The results of the simulation are directly compared with recent experimental measurements of the ion-ion recombination rate in discharges through gas mixtures of methane and sulfur hexafluoride.

Although the theory of ion-ion recombination has a long history,¹⁻⁶ comparison between theoretical and experimental results has been hampered by a lack of experimental data. This has been particularly true for measurements of the ion-ion recombination rate in discharge plasmas with an applied electric field. However, a recent experimental study of the recombination of ions in gas discharges in mixtures of methane and sulfur hexafluoride⁷ makes such a comparison possible.

In an electric discharge in a pure gas it is expected that the rate of recombination of the ions would depend on both the gas pressure and the applied electric field. In the case of recombination in discharges through gas mixtures, it is also to be expected that the recombination rate will depend on the relative proportions of the components making up the gas mixture. Cornell and Littlewood have shown empirically⁷ that the dependence of the recombination rate on each of these parameters can be described by a single parameter, an effective ion temperature, defined by

$$\frac{3}{2}kT^* = \frac{3}{2}kT + \frac{1}{2}m_+v_+^2 + \frac{1}{2}m_-v_-^2. \quad (1)$$

In Eq. (1) T is the ambient gas temperature, $m_{\pm}$ are the ion masses, and $v_{\pm}$ are the ion drift velocities given by

$$v_{\pm} = K_{\pm} E, \quad (2)$$

in which $K_{\pm}$ are the ion mobilities, and E is the applied field. In terms of the effective ion temperature, the recombination rate was found to follow an empirical power law

$$\gamma = \gamma_0 (T/T_0)^{-n} \quad (3)$$

over a wide range of discharge conditions. Only one pair of parameters, γ_0 and n , were needed to fit the measured rates for all pressures and compositions studied. We have used the Monte Carlo method to calculate the recombination rate for these ions in gas discharges under conditions corresponding to those used in the experimental study.

The Monte Carlo method has been described in detail elsewhere.⁴⁻⁶ We have used the same model for our calculations, with modifications to simulate the recombination of ions in a mixture of gases, and with an applied electric field.

For a given set of discharge conditions (gas pressure,

gas composition, and applied field strength) the trajectories of a large number of ion pairs were followed from an initial separation r_0 , chosen to be larger than the mean interion distance, but large enough that the distribution of ionic velocities is close to Maxwellian. For each ion pair the initial velocities at separation r_0 are selected from the Maxwellian distribution using three computer generated random numbers.

As the ions approach each other and then separate they suffer elastic collisions with gas neutrals. A fourth random number, selected from an exponential distribution, is used to determine the time of each collision between an ion and a neutral, such that the mean time between collisions is τ . For each collision, in the center-of-mass frame of the colliding ion and neutral, the speed of the ion is unchanged, although the ion may be scattered into a different direction. The direction of the scattered ion was determined from random numbers, chosen such that the scattering cross section was isotropic.⁶ It is then a simple procedure to determine the scattered velocity of the ion in the laboratory frame.

Since the recombination is being simulated in a gas mixture containing both methane and sulfur hexafluoride molecules, it is necessary, for each collision, to determine with which of these two molecules the ion has collided. For each species a mean time between collisions is determined from

$$\tau_i^{-1} = \nu_i = \frac{2\pi e N_i}{(4\pi\epsilon_0\alpha_i)^{1/2}}, \quad (4)$$

in which N_i and α_i are the number density and polarizability of species i . The overall collision time is then given by

$$\frac{1}{\tau} = \frac{1}{\tau_1} + \frac{1}{\tau_2}. \quad (5)$$

The probability of colliding with species 1 (methane) is then given by

$$P_1 = \tau/\tau_1. \quad (6)$$

If the next random number generated by the computer was less than P_1 , the ion was taken to have collided with a methane molecule (species 1), otherwise the ion was assumed to have collided with a sulfur hexafluoride molecule.

After each collision with a neutral molecule the energy of the ion pair was calculated. If the energy dropped below a preselected critical energy the ions were assumed to have recombined and the simulation for that ion pair was terminated. Otherwise, the trajectories of the ions were followed through subsequent collisions with neutral molecules. After each collision the ion energies were again compared with the critical energy to test for recombination. If the ions again separated to a radius r_0 , the ions were taken to have not recombined, and the simulation for that ion pair was terminated. After a large number of ion pairs had been simulated, the recombination probability was defined as the ratio of the number of ion pairs recombining to the total number of ions in the simulation. The probability was found to be insensitive to the value chosen for the critical energy, providing the critical energy was less than $-5kT^*$.

Once the recombination probability is known it is a simple procedure to calculate the recombination rate. The rate is given by⁴

$$\gamma = \pi r_0^2 \langle v \rangle P n_0 / n_\infty. \quad (7)$$

In Eq. (7) P is the recombination probability, n_0 is the ion density at r_0 , n_∞ is the macroscopic ion density, and $\langle v \rangle$ is the mean velocity of the ions. Since the applied electric field gives the ions a directed motion toward each other, the mean velocity is given by

$$\langle v \rangle = \left[\frac{8kT}{\pi\mu} + v_d^2 \right]^{1/2}, \quad (8)$$

in which μ is the reduced mass of the ions and the drift velocity toward each other, v_d , is given by

$$v_d = (K_+ + K_-)E. \quad (9)$$

The ratio of the ion densities, n_0/n_∞ , was calculated from diffusion theory.⁶

The predicted recombination rates are shown in Fig. 1 as a function of the applied electric field and gas composition at a total pressure of 250 Torr. Similar results were obtained for a gas pressure of 500 Torr, except the results

FIG. 1. Calculated rates for the recombination of methane and sulfur hexafluoride ions in a gas discharge as a function of the applied field strength.

FIG. 2. Recombination rates in a gas discharge as a function of the effective ion temperature of the ions.

at the higher pressure were consistently 10–15 % higher than those at the lower pressure. At low to moderate field strengths, where the recombination rate is high, good statistical accuracy can be obtained with 2500 ion pairs in each computer run. At the highest field strengths, for which the rate has fallen by two orders of magnitude, maintaining the statistical accuracy required an increase in the number of ion pairs in each simulation, eventually leading to a prohibitive increase in computer time. Results are shown for three gas mixtures containing 50%, 80%, and 95% methane, respectively. Comparison of the predicted rates with the published experimental results⁷ for the same gas mixtures shows good agreement.

Figure 2 shows the same predicted rates, this time plotted as a function of the effective ion temperature. The observed dependence of the rate on the concentration of methane, which was evident in Fig. 1, has been removed by using T^* as the abscissa. As a function of the effective ion temperature, the predicted rate follows the same power law as that observed experimentally by Cornell and Littlewood. At the highest field strengths the data do show a decrease in the slope of the curve. However, as already noted, the statistical accuracy of the results at

FIG. 3. Comparison of the measured and predicted rates of the recombination of ions in a mixture containing 95% methane and 5% sulfur hexafluoride.

the high field strengths could only have been maintained by prohibitively large increases in computer time, and the data in this regime may not be reliable.

The experimental data and results of the Monte Carlo simulation are directly compared in Fig. 3. For clarity, results for a single gas composition (95% methane) have been included in the figure. However, when plotted against the effective ion temperature, there is no dependence of either the experimental recombination rate or the Monte Carlo predictions on the percentage of

methane in the gas mixture. Hence a similar agreement between experiment and theory can be obtained for the other gas mixtures.

Cornell and Littlewood introduced the effective ion temperature as an appropriate empirical parameter for displaying the measured recombination rates. The agreement between the predicted and experimental results shows that the effective ion temperature can also be a useful parameter for numerical simulation of the recombination of ions in a gas discharge.

¹P. Langevin, *Ann. Chem. Phys.* **48**, 433 (1903).

²J. J. Thomson, *Philos. Mag.* **47**, 337 (1924).

³G. L. Natanson, *Zh. Tekh. Fiz.* **29**, 1373 (1959) [*Sov. Phys.—Tech. Phys.* **4**, 1263 (1959)].

⁴W. L. Morgan, B. L. Whitten, and J. N. Bardsley, *Phys. Rev. Lett.* **45**, 2021 (1980).

⁵W. L. Morgan, J. N. Bardsley, J. Lin, and B. L. Whitten, *Phys. Rev. A* **26**, 1696 (1982).

⁶J. N. Bardsley and J. M. Wadhera, *Chem. Phys. Lett.* **72**, 477 (1980).

⁷M. C. Cornell and I. M. Littlewood, *J. Phys. D* **20**, 616 (1987).