Characterization of an Integrated Low Pressure Gas Dynamics and Ion Migration Simulation Method within the SPARTA-DSMC Framework

Robin Hillen; Walter Wissdorf, Hendrik Kersten, Thorsten Benter

Physical & Theoretical Chemistry

Wuppertal, Germany

Institute for Pure and Applied Mass Spectrometry

Introduction

Direct Simulation Monte Carlo (DSMC) is a well-established method to investigate the dynamics in low density gases with a particle based numerical approach. For the simulation of neutral gas flows, SPARTA (Stochastic PArallel Rarefied-gas Time-accurate Analyzer [1]), is a popular open-source DSMC code.

SPARTA allows basic chemical reactions and arbitrary surface geometry in the simulation domain. However, albeit charged particles (charged droplets, molecular ions) are basically implemented in SPARTA, their interaction with external electric fields is currently not.

To widen the applicability of the code, we present a new module in SPARTA, which allows the simultaneous determination of the electric field force and the interaction between charged and neutral particles by the soft-sphere collision model.

To evaluate the simulations the ion mobility is calculated and compared with literature data.

Methods

Simulations:

- Modified SPARTA open source DSMC Code (based on version from jan20) [1]

Data Analysis:

- Python 3 with numpy, pandas, matplotlib and scipy libraries
- Paraview 5.7.0 [2]

Machine:

- Workstation Computer (Dell Precisions T5700) Xeon E5530, 24 GiB RAM, Ubuntu 18.04 LTS

Benchmark Setup

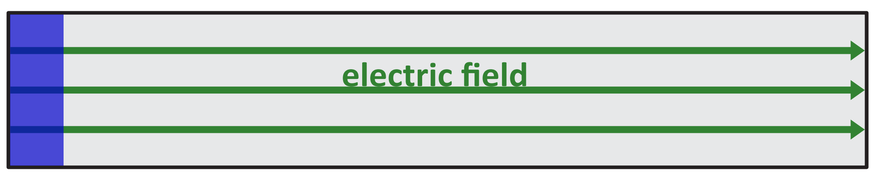

Fig 1: Generic high kinetic energy IMS; applied electric field is parallel to the x axis

A generic high kinetic energy IMS (cf. **Fig 1**) is used as benchmark model. The two dimensional simulation consists of a 0.16 m long and 0.03 m wide IMS.

The pressure of the background gas within is approx. 20 mbar and reduced field strengths from 20 up to 400 Td are applied. The electric field orientation is parallel to the x axis of the simulation domain.

The particles are generated in the blue area at the left. Approximately 100 charge particles are introduced to each simulation. For collisions between the charged and neutral particles the soft sphere collisions model is used [3].

Goals

- Observe the acceleration of charged particles
- Determine equilibrium drift velocities
- Calculate reduced electrical ion mobilities for different reduced field strengths
- Compare ion mobility with literature

Results

Application of an electric field in the simulation domain leads to electric acceleration of the charged particles. The ions reach an equilibrium and a static drift velocity due to the collisional interaction with the background gas.

The charged particles start in the blue area in the left end of the IMS (cf. **Fig. 1**) at thee beginning of each simulation. The initial velocity of these particles is zero.

In **Fig. 2** an acceleration curve is shown: The ions have reached the steady state drift velocity after 25 ns.

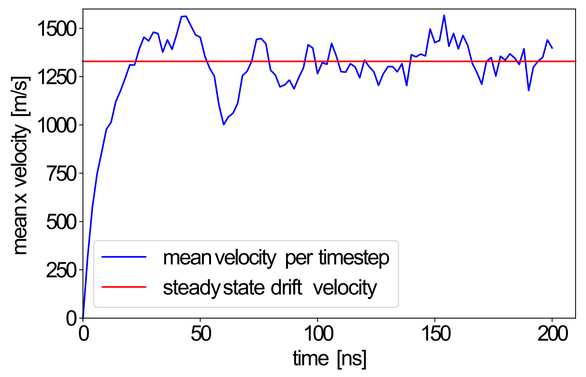

Fig. 2: Acceleration and steady state drift velocity of 84 simulated ions

$$K = \frac{v_d}{E} \quad \text{(Eq. 1)}$$

$$K_0 = K \frac{N}{N_0} \quad \text{(Eq. 2)}$$

To compare the simulation results with literature data, the electrical ion mobility **K** (cf. **Eq. 1**) of charged particles is calculated from the drift velocity v_d and the electric field E.

To be able to compare different ion mobility experiments, a normalized reduced ion mobility $\mathbf{K_0}$ is calculated by correcting the ion mobility with the background gas density (N) (cf. **Eq. 2**) [4].

The difference between the calculated values of K_0 and the literature data is shown in and **Fig. 3** and **Tab. 1**. The difference decreases with increasing reduce field strength. The simulated values are a factor of 3.2 up to 5.2 too high.

Tab. 1: Comparison between simulated and literature values of K_0 for different reduced field strength [5]

Species	E/N [Td]	K_0(sim) [cm²/Vs]	K_0(lit) [cm²/Vs]
N_2^+ in O_2	20	10.10	1.83
	40	7.96	1.75
	60	6.98	1.63
O_2^+ in CO_2	100	6.08	1.38
	200	5.77	1.39
	400	4.85	1.50
	600	4.41	1.36

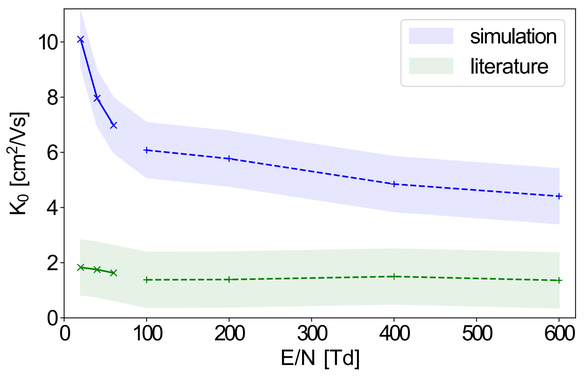

Fig. 3: Simulated K_0 for N_2^+ in O_2 (unbroken line) and O_2^+ in CO_2 (dashed line) in comparison with literature data [5]

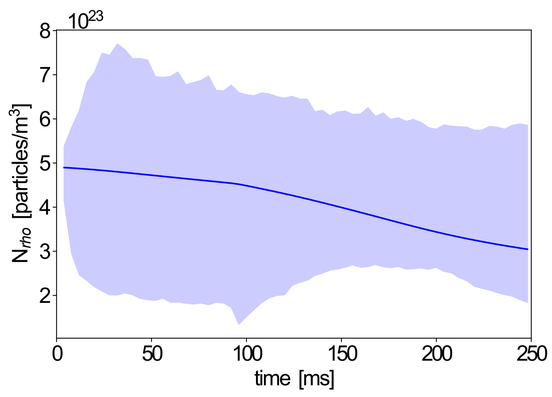

Fig. 4: Trend of number density per volume in one simulation run with the min/max range covered by the blue area

There are a few possible reasons why the calculated values of the normalized reduced ion mobilities are too high:

The background gas density decreases constantly with the simulation time as shown in **Fig. 4**. Therefore the number of collisions of the charged particles with the background gas decreases too. This leads to higher drift velocities and thus to higher ion mobilities.

The decreasing of the number density of the background gas has several potential causes: The ratio between simulated neutral and charged particles could be too small resulting in significant impulse transfer to the neutral gas. This potentially leads to ions “pushing” the neutral gas out of the simulation domain. Alternatively, some simulation parameters are not chosen appropriately. The time step size is particularly critical: A too large step size can lead to a wrong collision frequency of the charged particles.

Similarly, parameters like the collision cross sections between the simulated particles also affect the collision frequency.

Conclusion / Outlook

Conclusion:

- The electric field accelerates the simulated charged particles
- The ions reach a steady state drift velocity after a short acceleration period
- The calculated ion mobilities are in the right order of magnitude but substantially too high

Outlook:

- Decrease the impact of the ions on the background gas
- Implement more complex and temporally changing electric fields
- Enable the possibility of resonant charge transfer (e.g. N_2^+ in N_2) via SPARTA reaction module

Literature

[1] Plimpton, S.J., Moore, S.G., Borner, A., Stagg, A.K., Koehler, T.P., Torczynski, J.R., Gallis, M.A.: Direct simulation Monte Carlo on petaflop super computers and beyond. *Physics of Fluids*. *31*, 086101 (**2019**).

[2] Ayachit, U.: The ParaView guide: updated for ParaView version 4.3. Kitware, Los Alamos (**2015**)

[3] Bird, G.A.: Molecular gas dynamics and the direct simulation of gas flows. Clarendon Press ; Oxford University Press, Oxford : New York (**1994**)

[4] Eiceman, G.A., Karpas, Z., Hill, H.H.: Ion mobility spectrometry. (**2016**)

[5] Viehland, L.A., Mason, E.A.: Transport Properties of Gaseous Ions over a Wide Energy Range, IV. *Atomic Data and Nuclear Data Tables. 60*, 37–95 (**1995**).

Acknowledgement

Support is gratefully acknowledged:

- Deutsche Gesellschaft für Massenspektrometrie (DGMS), Halle , Germany
- German Research Foundation (DFG), BE2124/8-1