Using COMSOL to Build a Transient Model of a Drift Tube Ion Mobility Spectrometer with Improved Performance

J. Langejuergen[*], P. Cochems, M. Allers and S. Zimmermann
Leibniz University Hannover, Institute of Electrical Engineering and Measurement
Technology, Department of Sensors and Measurement Technology
*Corresponding author: Leibniz University Hannover, Appelstr. 9a, 30167 Hannover,
Langejuergen@geml.uni-hannover.de

Abstract: A drift tube Ion Mobility Spectrometer (IMS) is a device to analyze different components of a gas mixture. One particular feature of this technology is its ultra-low detection limit. However, due to a limited resolution required to differ between different components an improved IMS field geometry would help to increase resolution and to analyze even complex mixtures of multiple gases. Furthermore - especially for novel, non-radioactive ionization sources [1] - the influence of Coulomb repulsion needs to be taken into account. In this work a transient and parametric model of a drift tube IMS based on COMSOL 4.1 is presented in good agreement with the experimental results. The exact shape of the electrical field, the loss of ions at metallic surfaces and the timing of ion generation and injection is considered. Furthermore, coulomb repulsion caused by the transient charge distribution is taken into account.

Keywords: Drift Tube Ion Mobility Spectrometer, Transient Simulation of Ions under Atmospheric Pressure Conditions, Ion Drift Tube, Coulomb Repulsion.

1. Introduction

A drift tube Ion Mobility Spectrometer (IMS) is a device to analyze different components of a gas mixture. Therefore, the gas sample is first ionized and then transported in an electrical field $\mathbf{E}$ towards a detector (fig. 1). A detailed description of IMS and the wide field of applications can be found in [2].

The IMS is usually operated at atmospheric pressure conditions. Therefore, the ionized molecules collide with neutral molecules within the drift tube resulting in a constant ion drift velocity $\mathbf{v_D} = K \cdot \mathbf{E}$. Due to their specific mobilities K the ions of different components are separated within the drift tube and arrive the detector at different times leading to a transient detector current (fig. 2).

The two major advantages of an IMS are the comparably small size and the excellent sensitivity caused by atmospheric pressure

ionization leading to ultra-low detection limits [3] for most trace gases. Nevertheless, the analytical power of an IMS is limited by a comparably low selectivity making it difficult to separate ions with similar K or to identify individual components in a complex matrix of multiple substances. This limited separation power is mainly caused by inhomogeneities of the electrical field within the IMS, the diffusion of ions, coulomb repulsion and the extent of the volume in which the molecules are initially ionized.

In this work we present a transient model of a drift tube IMS which is considering these effects to design an optimized geometry for an IMS with improved resolution.

2. Experimental Setup

In fig. 1 the experimental setup is shown. The model is based on this setup. The sample gas is ionized by an electron emitter. The ionization depth can be calculated by the kinetic energy of emitted electrons. The ions are injected into the drift tube by an injection voltage ($U_{source} = U_{injection}$) at $t = t_0$. The duration of the ion injection is $t_{injection}$.
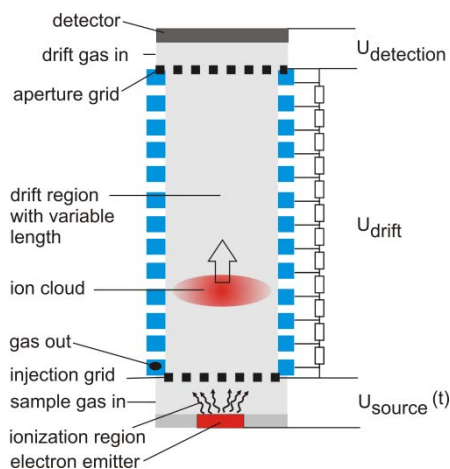

Figure 1: Experimental setup of a drift tube IMS.

Along the drift tube a drift voltage (U_{drift}) is applied which causes the ions move towards

the detector grid. The drift tube consists of multiple ring electrodes with an inner radius of 7.6 mm, an injection grid and a detector grid in order to obtain a homogenous electrical field. The length of the drift tube can be varied by changing the number of ring electrodes. A default length is about 75 mm and a default voltage is U_{drift} = 2000 V. Between the detector grid and the detector plate (Faraday cup) a voltage ($U_{detection}$) is applied. When the ions hit the metallic detector at $t = t_{time\ of\ flight}$ they cause a transient current of some pA which is amplified and recorded. Depending on the ion mobility the resulting time of flight at the mentioned parameters is about 5 ms to 20 ms.

3. IMS resolution

As an example, in fig. 2 the detector current for two substances (A and B) with different mobilities ($K_A > K_B$) is shown. The shape of the current peaks is supposed to be Gaussian. One major parameter describing the analytical power of an IMS is the resolution R. It is defined as quotient $R = t_{time\ of\ flight}$ / FWHM (Full Width at Maximum Height). The higher the resolution the better the separation of the ions of different mobilities within the drift tube, see fig. 2.
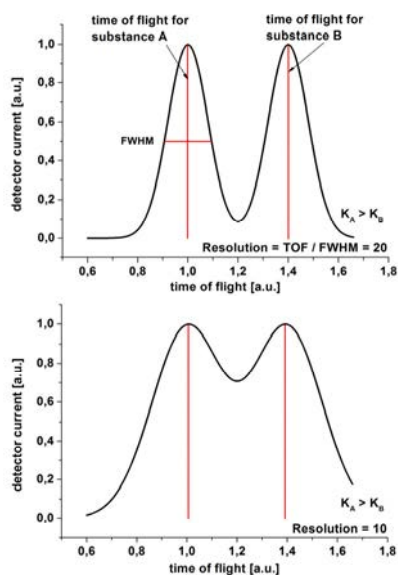

Figure 2: Ion mobility spectrum of two different substances (A and B) at different IMS resolutions.

4. Use of COMSOL Multiphysics

In [4] Hill et al. derive an analytical expression to rapidly predict the IMS resolution R considering diffusion, applied

voltages and the length of the drift tube. However, due to field inhomogeneities and coulomb repulsion the measured resolution can significantly differ from the estimated value.

In this work we present a model using the COMSOL Chemical Reaction Engineering Module to simulate the movement of the ions within the electric field $\mathbf{v_D}=K \cdot \mathbf{E}$. As electrical boundary conditions the electric potentials of the different electrodes are defined. The potential of the injection grid is a function of time and changes after $t_{injection}$.

The model is using cylindrical symmetry which also applies for the real experimental setup except for the two grids. In the model the grids have the same thickness, web thickness and diameter of holes as the real grids. This leads to a slightly increased optical transmission of the simulated grids which has to be taken into account when comparing experimental with simulated results.

The initial concentration of ions within the IMS is defined to be zero except for the ionization region (fig. 4) where it is defined to be c = c_{init}. The default ion mobility is defined to K = 2.3 cm²/(Vs). The diffusion coefficient D is derived from K according to the Einstein-Nernst relation ($D = k_B \cdot T \cdot K/q = 5.8\ 10^{-6}$ m²/s) where T is the temperature in Kelvin and q is the charge of one ion (here equal to the elementary charge). The concentration at metallic surfaces is set to be zero.
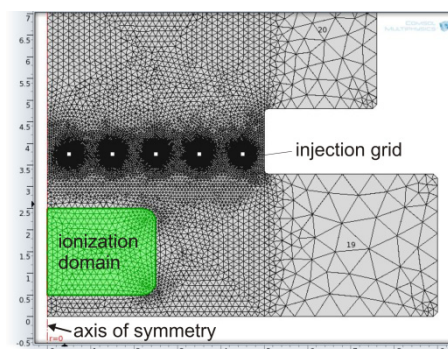

Figure 3: Meshing of the ionization domain and the injection grid.

To calculate the effect of high ionization densities the ion concentration c is coupled to a space charge density ρ leading to Coulomb repulsion.

The generation of ions can be described by defining a reaction rate coefficient $dc/dt = R_C$. The ion-ion recombination due to the generation of ions of different polarities (c_+ for positive ions, c_- for negative ions) can be described by defining a concentration-

dependent recombination coefficient $dc/dt = -R_{ionion} \cdot c_+ \cdot c_-$ within the entire ionization source. All other parameters like pressure and temperature are set to standard values (p = 1013 mbar, T = 293 K).

A moving mesh with approx. 400k elements is used to obtain sufficient results (fig. 3). Especially the injection grid and the detector grid need to be highly resolved. Furthermore, the time steps taken by the solver need to be adjusted. By doing so, no more artificial diffusion is needed to eliminate computational artifacts like negative concentrations.

5. Results

In fig. 4 (left) a detailed view of the simulated ion distribution is shown in the ionization chamber prior to injection into the drift tube. During injection (center and right) the ion cloud is deformed due to field inhomogeneities close to the injection grid (white dots). About 50 percent of the ions are lost due to collision with the metallic injection grid (fig. 5). This value is simulated for a symmetric grid. The grid used in the experimental setup has an optical transmission of only 90 % of the value for a symmetric grid resulting in an ion loss of approx. 55 percent. Due to field inhomogeneities this value is expected to be even higher than the ratio of optical transmissions. However, no significant amount of ions is lost within the drift tube or at the detector grid.
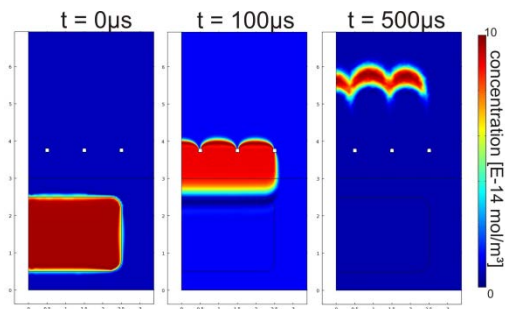

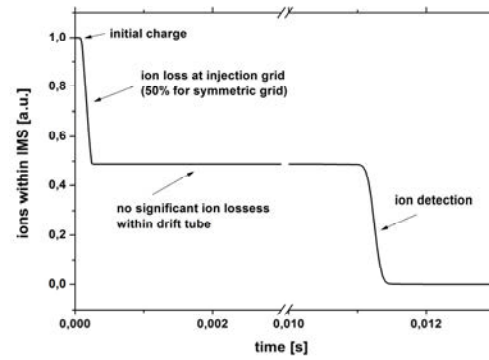**Figure 4:** Ion distribution at different times of IMS operation.

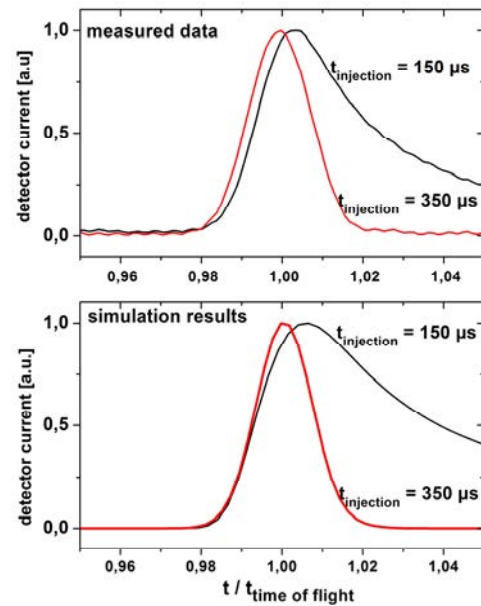**Figure 5:** Ion loss at the injection grid and the detector plate.

Figure 6: Simulated and measured detector currents for different values of $t_{injection}$.

In fig. 6 the measured (top) and the simulated (bottom) detector currents are shown for two different IMS operating parameters. For an injection time of 350 µs (red) the ion cloud is transferred into the drift tube leading to an almost symmetric detector signal and an optimal resolution. For an injection time of only 150 µs only part of the generated ions are directly transferred into the drift tube before the voltage at the grid is changed. This causes an asymmetric detector signal with a reduced resolution. This effect is clearly visible in the simulated and measured data. A good match of the simulated and the measured detector currents is given in both cases.
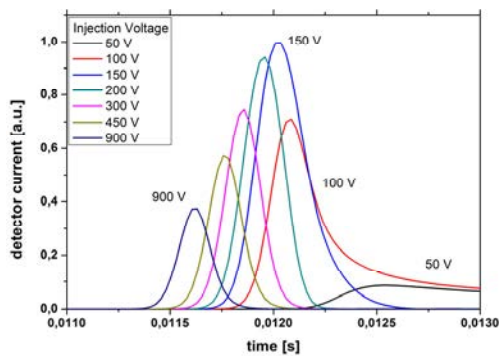
Figure 7: Detector current for different values of $U_{injection}$.

In fig. 7 the influence of the injection voltage on the detector current is shown. For voltages lower than 150 V the ion cloud is injected too slowly into the drift tube. This again results in an asymmetric peak shape and a reduced resolution. For voltages above 150 V a decreasing time of flight with increasing injection voltage is visible. This is due to a decrease in the time necessary to transfer the ions out of the ionization source. Furthermore a decrease of the detector current is visible. This is caused by an increase of the ion loss at the injection grid at higher voltages. Hence, an optimum injection voltage is 150 V.
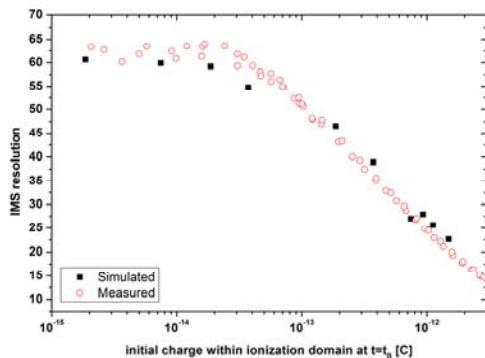

Figure 8: Measured and simulated influence Coulomb repulsion on the IMS resolution.

In fig. 8 the influence of Coulomb repulsion on the IMS resolution is shown. The measurements (red circle) are performed with an adjustable electron gun. In the simulation (black squares) the electric field caused by the charge density of the ion cloud is recalculated for every time step. Simulated resolutions show a very good match with the experimental results.

For an initial ionization of less than $5 \cdot 10^{-14}$ C the influence of Coulomb repulsion can be neglected while diffusion and field inhomogeneities are limiting the IMS resolution. For an increasing amount of initial ions the influence of Coulomb repulsion increases causing the resolution to decrease.

6. Conclusions

In this work a model of a drift tube IMS is presented in good agreement with the experimental results. The ion distribution within the IMS can be calculated at any time of IMS operation. This allows simulating the influence of static values like the geometry of the injection grid or the voltage applied along the drift tube. Furthermore, transient parameters like the injection time, ion generation and Coulomb repulsion due to the moving charge distribution can be calculated.

Besides Coulomb repulsion and diffusion the deformation of the ion cloud at the injection grid could be identified as a major factor affecting IMS resolution.

Since the loss of ions at metallic surfaces is considered, the model gives a good prediction of ion losses for different field geometries. An optimum set of parameters for the injection voltage and injection time giving maximum resolution and detector signals were computed.

7. References

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