# Multiphysical simulation of a multicomponent trace gas electric swing adsorption

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## Introduction

Gas cleaning of technical gas mixtures is a complex task. There are technical challenges such as finding the cost optimum or treating a time dependent gas composition. For instance, current methods for the adsorptive gas cleaning utilize multiple adsorbent layers. Therefore, in some cases desorption times of different species differ significantly.

Thus, within this work a (pre-) adsorber is modeled to treat heavy components e.g. tars. As a result, one can adjust adsorption und desorption times of the pre- and mainadsorber independently.

For desorption an electric field heating the conductive adsorbent is modeled. In literature [1], this novel method is considered as customizable and energy-efficient.

In the following chapters, the multiphysical and multiscale kinetic model is shown and discussed. Furthermore, a new method to implement multicomponent adsorption data using lookup tables is demonstrated. Finally, a model validation and results are shown.

## Theory

Adsorbers for gas cleaning are vessels filled with an adsorbent or multiple adsorbent layers. There are two different adsorber types: pressure swing adsorbers and temperature swing adsorbers.

For a pressure swing adsorption the gas mixture is compressed during the adsorption cycle. The high pressure shifts the adsorption equilibrium towards the adsorbed form of the adsorbate. Throughout the adsorption cycle the so-called loading of the vessel increases. Eventually, the vessel is filled and the desorption cycle begins.

During the desorption the pressure in the vessel is decreased. This causes the equilibrium to shift and ultimately desorbs the loading. Usually the direction of flow changes during the adsorption and desorption cycles.

For a temperature swing adsorption the gas mixtures is not compressed during the adsorption cycle. To shift the equilibrium for desorption the temperature is increased. In general, a hot gas stream is introduced for desorption. In contrast, the electric swing adsorption uses resistive heating to generate the required heat.

## Multiscale kinetic model and lookup tables for multicomponent adsorption data

Within this chapter, the multiscale model including a macro and a micro scale is shown and discussed. In the COMSOL modeling environment the Transport of Diluted Species in Porous Media physic interface is used in combination with the reactive pellet bed feature to build the multiscale model, see figure 1.

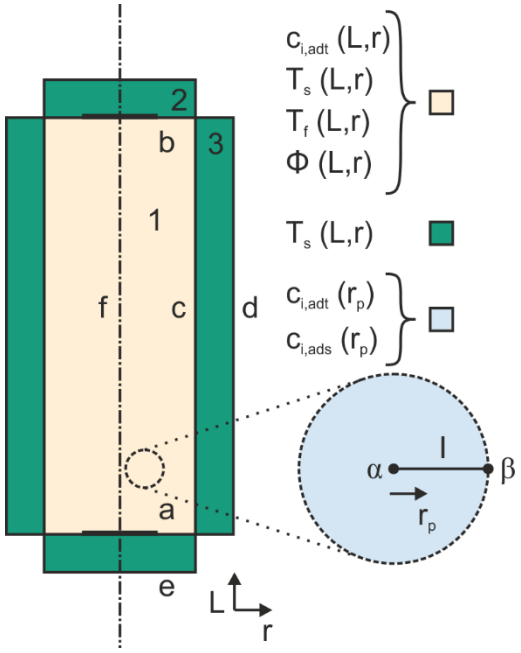


**Figure 1. Two-dimensional macroscopic model of the apparatus and one-dimensional microscopic model of the pellets. 1: vessel/gas bulk, 2: flanges, 3: walls, I: pellet volume, a: inlet and electrode, b: outlet and electrode, c: contact gas bulk and wall, d; contact wall and environment, e: contact flanges and environment, f: symmetry axis, α: symmetry, β: contact gas bulk and microscale. The dependent variables are shown.**

The Chemistry physic interface is applied to model the adsorbate formation as an equilibrium reaction inside the micro scale. The temperature and concentration dependent equilibrium coefficients are calculated based on the adsorption isotherms.

Dealing with a technical gas mixture requires information about the multicomponent adsorption behavior of all adsorbing species – but literature data is usually short. Therefore, one can use the ideal adsorbed solution theory (IAST) to predict the multicomponent adsorption based on the single component isotherms. A more rigorous approach is the real adsorbed solution theory (RAST) which considers the real behavior of the mixture.

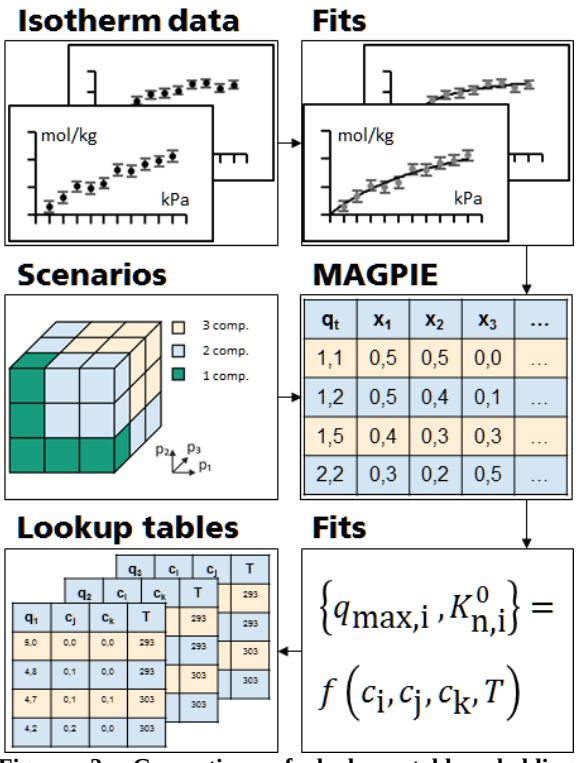


**Figure 2. Generation of lookup tables holding multicomponent adsorption isotherms.**

Direct implementation of the IAST or the RAST is complex and requires an external procedure within COMSOL. Therefore, this work suggests an indirect implementation featuring lookup tables. One can calculate the desired multicomponent isotherms beforehand using any software tool – in this case MAGPIE [2]. Thus, it is necessary to predict all occurring scenarios (ranges of concentrations and temperature) beforehand to avoid extrapolation.

Afterwards, the multicomponent isotherm data is processed by a fit to compress the amount of data and to allocate the isotherms to the according species. As a result, one receives the temperature and concentration dependent adsorption isotherms for each species. The generation of the look up tables is shown in figure 2.

The lookup tables are implemented in COMSOL using the interpolation function. The arguments of the interpolation are the concentrations and the temperature. The interpolated data points are the parameters to describe the isotherms.

The multiscale model also uses the Electric Currents, the Heat Transfer in Solids and the Heat Transfer in Fluids interfaces to account for the current and energy conservation. The Multiphysics feature is set up to calculate the Local Thermal Non-Equilibrium connecting the heat transfer in solids and fluids. Furthermore, the Multiphysics feature is used to consider the Electromagnetic Heat Source of the current conservation.

Finally, a combination of the Global ODEs and DAEs and the Events interface is used to implement the time dependent behavior of the direction of flow and the resistive heating.

This setup of the model allows one to simulate the dynamic behavior of an electric swing adsorber. There are three phases in one adsorption/desorption cycle:

1. Adsorption
2. Desorption and resistive heating
3. Desorption

During the adsorption phase concentration fronts of the adsorbates move through the adsorber. During the first step of the desorption phase the direction of the flow is changed and the resistive heating begins. Therefore, the temperature of the adsorbent rises decreasing the loading. During the second step of the desorption phase the resistive heating is switched off to allow the adsorbent to cool down. Afterwards, the next adsorption/desorption cycle begins.

The setup of the model enables one to investigate for example incomplete desorption phases. One can accept a trade-off between a preloading of the adsorber due to an incomplete desorption to decrease the desorption time. In addition, the model allows one to optimize the time and power of the resistive heating to minimize the total desorption time.

## Model validation

The model at hand is validated at two different sources [3, 4]. The first part of the validation focuses on the implementation of the adsorption as equilibrium reaction in the microscopic model and the implementation of the lookup tables for the multicomponent adsorption isotherms. Possible mass transfer limitations are neglected. Experimental so-called breakthrough curves (time dependent concentrations at an adsorber outlet) of a binary adsorption experiment [3] are compared with the simulation results, see figure 3.

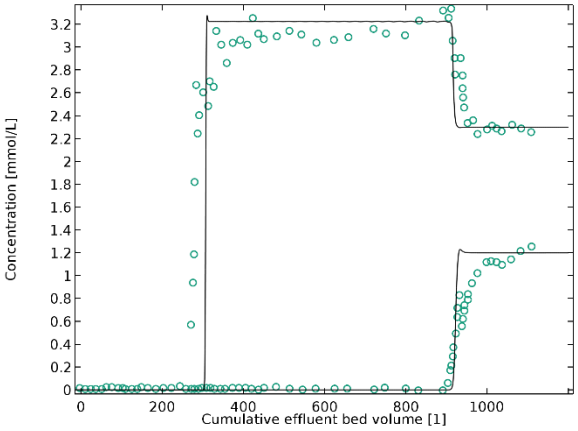


**Figure 3. Experimental (circles) [3] and simulated breakthrough curves of a binary mixture.**

Figure 3 shows that the simulated breakthrough curves exhibit the expected behavior. One can see the sharp concentration gradients due to the neglect of mass transfer limitations.

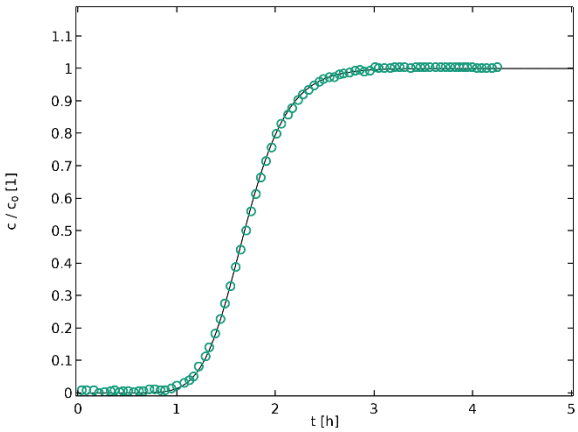


**Figure 4. Experimental (circles) [4] and simulated breakthrough curve of an adsorption experiment with mass transfer limitation.**

The second part of the validation focuses on mass transfer, too. Therefore, a Breakthrough curve of a single component experiment affected by mass transfer is used [4].

The experimental data points in figure 4 clearly show flat concentration gradients. A limiting pore diffusion coefficient and a limiting mass transfer coefficient are employed for the simulation. The agreement between the experiment and the simulation indicate a valid model.

## Conclusions

In summary, the outlined multiscale model uses the mass, energy and current conservation physic interfaces (Transport of Diluted Species in Porous Media, Chemistry, Electric Currents and Heat Transfer in Solids and Fluids) to rigorously model an electric swing adsorption. The implementation of lookup tables for pre-calculated multicomponent adsorption isotherms is based on the interpolation function of COMSOL.

The dynamic model of the adsorber is able to predict multicomponent breakthrough adsorption experiments and is able to consider mass transfer effects during the adsorption.

## References

1. M. Petkovska, D. Antov-Bozalo, A. Markovic, P. Sullivan, Multiphysics modeling of electric-swing adsorption system with in-vessel condensation, *Adsorption*, **13**, 357-372 (2007)
2. Austin Ladshaw, Sotira Yiacoumi,, Costas Tsouris, A Generalized Procedure for the Prediction of Multicomponent Adsorption Equilibria, *American Institute of Chemical Engineers*, **61**, 2600-2610 (2015)
3. Pei-Jen Lu, Chih-Sheng Chang, Jia-Ming Chern, Binary adsorption breakthrough curves in fixed bed: Experiment and prediction, *Journal of the Taiwan Institute of Chemical Engineers*, **45**, 1608-1617 (2014)
4. Hermann Dreher, Werner Kast, Analytische Näherungslösung für die Durchbruchskurve bei nichtlinearer Adsorptionsisotherme, *Chemie Ingenieur Technik*, **51**, 122-124 (1979)

## Acknowledgements

I wish to thank Stefan Schlüter and Torsten Hennig for their scientific support and their open door.