MODELLING OF HALITE FORMATION IN NATURAL GAS STORAGE AQUIFERS

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ABSTRACT

The applicability of TOUGH2 using the EWASG module for the modelling of halite formation in natural gas storage aquifers is investigated.

First TOUGH2 and the EWASG module are properly modified. An additional NCG is implemented into EWASG which allows input of user defined gas properties. Additional curves for relative permeability and capillary pressure are implemented, in which user defined input values are linearly interpolated. To improve the capability of EWASG a new correlation is established calculating the enthalpy of aqueous NaCl-solutions for temperatures below 100 °C. In the 2nd step classical sample problems from the ECLIPSE-code are calculated with TOUGH2 to demonstrate that the requirements of the oil and gas industry are met. In the 3rd step existing laboratory experiments on the dissolution of salt by injection of dry gas into a sandstone sample are simulated. The results agree well with the experiments within the expected range of accuracy. In the last step salting out for a generic aquifer gas storage model is simulated for the case of injection, production and a cycle of injection followed by production. The aim is to check if the observed behaviour of salting out in the field can be simulated qualitatively and if the conceptual model implemented in EWASG is suitable for this problem.

Results indicate a better understanding of the processes and conditions for halite formation. From the technical point of view TOUGH2 is suitable for handling the problem of halite formation in the gas industry. In the next step further qualification of the code by application on a real gas aquifer will be performed.

INTRODUCTION

During operation of natural gas storage aquifers and reservoirs precipitation of dissolved salts can occur near as well as in the wellbore, if saline groundwater is present. This may reduce the wellbore performance drastically and causes considerable problems. Halite formation occurs during injection of dry gas as well as during production from the well. The physical processes for the case of injection are well understood, whereas the driving forces for production of halite formation are still not completely known. Up to now the relevant physical processes can be described only qualitatively. An overview about the problem of halite formation is given by Kleinitz and Tölke (1982). Kleinitz et al. (2001) provide a literature review.

In practice halite formation is resolved by fresh water injection. Although this method is successfully used, further understanding of the mechanism and prognosis of formation behaviour is needed for optimising the fresh water injection and for minimising the potential of long term damage. The need for prognosis of halite formation is rising because this phenomenon increases with operational age of the storage aquifer.

Commercial codes used in oil and gas industry like e.g. ECLIPSE or CMG consider non-isothermal conditions not in their standard versions and only for special applications as Enhanced Oil Recovery. The phase changes of water are taken into account, but it is not possible to consider phase changes of NaCl or salt as an additional component and their effect of thermodynamic properties. Therefore up to now the problem of halite formation in gas aquifers can not properly be simulated numerically. TOUGH2, Version 2 (Pruess et al., 1999) with the EWASG module (Batistelli et al., 1997) seems to be suitable to fill this gap.

The requirements of the oil and gas industry on computer codes concerning validation and user defined properties are very high. One of the main differences between TOUGH2 and codes used in the oil and gas industry is the handling of thermodynamic data. In TOUGH2, thermodynamic properties are directly implemented into the code by the different EOS modules. On the other hand the oil and gas industry demands the possibility of introducing user defined properties. Therefore some modifications of TOUGH2, Version 2 and of the EWASG module are needed before an application on halite formation in aquifer gas storage.

This work is focused on the modifications performed on TOUGH2, Version 2 to model the halite formation

in aquifer gas storage and on the qualification of the modified code rather than on the results.

TOUGH-MODIFICATIONS

Enthalpy of Aqueous NaCl-Solutions

Operation temperatures of gas storages are usually between 40 and 100 °C. For this temperature range the existing commercial version of EWASG is not applicable because of the limitations of the equation for the determination of the enthalpy of aqueous NaCl-solutions.

Palliser and McKibbin (1998) give a literature review concerning the enthalpy of aqueous NaCl-solutions. The enthalpy equation for the NaCl-solution implemented in the EWASG module is developed by Michaelides (1981) on the basis of the data of Haas (1977). These data are given for the vapor pressure of the NaCl-solution, for a temperature range between 80 and 300 °C and up to 30 wt-% NaCl. Haas (1977) also gives extrapolated data for up to 325 °C and up to halite saturation. Pitzer et al. (1979) provide data from 0 to 300 °C and NaCl-concentrations up to 25 wt-% for water vapor pressure. These data are an extract of a comprehensive literature survey and evaluation of the thermodynamic properties of aqueous NaCl-solutions presented by Pitzer et al. (1984). Tanger and Pitzer (1989) give data for temperatures above 300 °C for different pressure values.

Figure 1 illustrates the temperature and concentration range of the available enthalpy data together with the temperature dependent solubility of NaCl according to Chou (1987), which is used in the EWASG module.
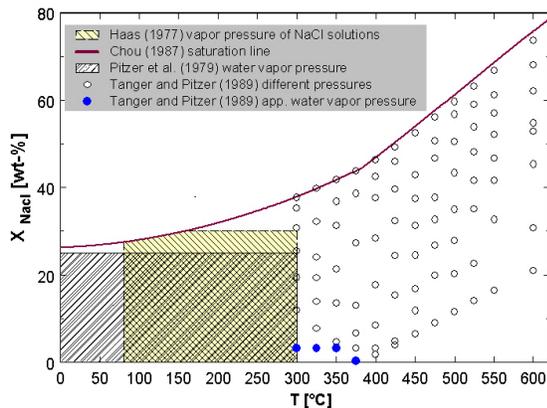
Figure 1. Illustration of available data for the enthalpy of aqueous NaCl-solutions

One of the main problems to fit data dependent on more than one variable is to determine the adequate type of equation. Based on the basic laws of thermodynamics the following equation of the type

Taylor-series is developed for the residual enthalpy Δh between the enthalpy at the specific conditions and a reference value:

$$\Delta h = R \cdot T \cdot \left(\frac{B - B'}{v} + \frac{C - C'}{2v^2} + ... \right) \quad (1)$$

Therefore the enthalpy depends on the temperature T, the gas constant R, the specific volume v, the virial coefficients B, C, etc. and the derivatives of the virial coefficients B', C', etc..

$$B' = T \left(\frac{\partial B}{\partial T} \right)_v \quad (2)$$

Substituting the reciprocal of the specific volume v by the mass concentration of NaCl, collecting the constant values and rearrangement yields the following type of equation to fit enthalpy data dependent on temperature T and NaCl-concentration x:

$$\Delta h(x,T) = \sum_{i=0}^{n} a_i(T) \cdot x^i \quad (3)$$

The coefficients a_i can be physically interpreted as a combination of constant values and the temperature dependent virial coefficients.

The temperature dependent enthalpy of water $h_{water}(T)$ according to IFC (1967) as used in TOUGH2 is set as reference enthalpy. In the limiting case of zero NaCl-concentration eq.(3) fits the enthalpy of water when the constant term is omitted. A 3rd degree polynom is identified as sufficient for the number of terms n. The equation for the enthalpy of aqueous NaCl-solutions therefore becomes

$$h_{Solution}(x,T) = h_{Water}(T) + \sum_{i=1}^{3} a_i(T) \cdot x^i \quad (4)$$

The equation type for the coefficients a_i are fitted without any physical meaning. For these coefficients also a 3rd degree polynom was evaluated for each coefficient a_i to

$$a_i(T) = \sum_{j=0}^{3} b_{i,j} \cdot T^j \quad (5)$$

The values for the coefficients $b_{i,j}$ are listed in Table 1. The temperatures T are in °C and the NaCl-concentration x in wt-%.

The maximum deviation from the data from Pitzer et al. (1979) are 6 J/g for a temperature of 300 °C and a NaCl-concentration of 20 wt-%. The maximum relative deviation is 2 % for temperatures between 25 and 300 °C. At 0 °C the relative deviations are up to 600 % for low salinity and up to 10 % for high

salinity. This high deviation is caused by the enthalpy values very close to zero at 0 °C.

Table 1. Coefficients $b_{i,j}$ in eq.(5) to fit enthalpy data given by Pitzer et al. (1979)

a_i	$b_{i,0}$	$b_{i,1}$	$b_{i,2}$	$b_{i,3}$
a_1	$2.985 \cdot 10^{-1}$	$-7.819 \cdot 10^{-2}$	$3.479 \cdot 10^{-4}$	$-1.203 \cdot 10^{-6}$
a_2	$-7.257 \cdot 10^{-2}$	$2.169 \cdot 10^{-3}$	$-1.809 \cdot 10^{-5}$	$5.910 \cdot 10^{-8}$
a_3	$1.071 \cdot 10^{-3}$	$-3.343 \cdot 10^{-5}$	$3.450 \cdot 10^{-7}$	$-1.131 \cdot 10^{-9}$

Extrapolation for higher salinities and temperatures

Pitzer et al. (1984) stated that it would serve little purpose to make detailed comparisons with earlier publications. Nevertheless application of eq.(4) and (5) to the data presented by Haas (1977) offers the possibility to check extrapolation above 25 wt-%. The application for temperatures between 80 and 300 °C and up to 25 wt-% shows differences between 2 and 9 %. Note that no pressure correction has been made. Application of the equation on the data of Haas (1977) up to 30 wt-% yields a maximum deviation of 8.6 % at 125 °C and 29 wt-%. Therefore it can be concluded that extrapolation of the equation for enthalpy for higher salinities is appropriate. Note for that purpose that the parameters $a_{i,j}$ does not depend on salinity.

Application of eq.(4) and (5) on the data presented by Tanger and Pitzer (1989) yields very good agreement for the values which are approximately at water vapor pressure with low salinity, where the enthalpy of the solution is close to the enthalpy of water. For the other data from Tanger and Pitzer (1989) differences between the measured pressure and the water vapor pressure are too high for a useful comparison without a pressure correction.

User Defined NCG in EWASG

An additional NCG is implemented in the EWASG module, in which user defined properties can be read from an additional file. In the current status discrete data provided by the file are linearly interpolated. For thermodynamic properties without a data base it can be chosen, from which of the implemented NCG's the relevant property shall be used.

Linear Interpolation of Discrete Capillary Pressure and Relative Permeability Data

Another modification on TOUGH2, Version 2 is the implementation of additional functions to determine the relative permeability and the capillary pressure.

In both cases discrete data points for given saturation values are provided from an additional file. The data are linearly interpolated to determine the capillary pressure and the relative permeability, respectively.

The capillary pressure values are set to zero for liquid saturation above the highest value and to the maximum pressure for liquid saturation below the lowest value. For the relative permeability, values are set to zero and the maximum value, respectively, outside of the tabulated range.

The implementation includes a check for data consistency, sorting of the discrete data for monotonic ascending values of liquid saturation, transformation of different units of capillary pressure and transformation into negative capillary pressure data, as they are used in TOUGH2.

NUMERICAL MODELLING

Code Comparison

A sample problem from the ECLIPSE code, GASWATER, is used for a direct code comparison. It is a simplified 3-D water saturated aquifer model with one injection and one production well. First dry gas is injected with a constant rate for 3000 days, then water is injected with constant rate over 3000 days. Production is modelled with constant bottom hole pressure option.

For a convenient comparison the thermodynamic properties of water in TOUGH2 are appropriately modified to be equal to the properties used in the sample problem. Furthermore, solubility of the gas is omitted. Because TOUGH2 does not include the option of hysteresis for relative permeability, calculations in TOUGH2 are performed using the curves for imbibition (the first 3000 days).

Figure 2 shows the results of the comparison. During the injection of gas both codes show identical results. The difference at the bottom hole pressure during the injection of water is most probably due to the modelling of the relative permeability without hysteresis in TOUGH2.
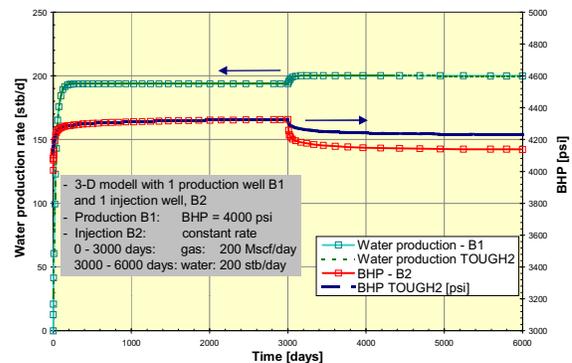
Figure 2. Comparison of TOUGH2, Version 2 with ECLIPSE. Water production rate and bottom hole pressure (BHP) for the sample problem GASWATER

Simulation of Laboratory Experiments

The numerical simulation of existing experimental data (Meyn and Pusch, 1997) are performed to qualify the conceptual model underlying EWASG.

The experimental setup used to investigate the halite formation during injection shows Figure 3. A dry high permeable Bentheim Sandstone ($k = 2 \cdot 10^{-12}$ m²) in the inner core is used for the gas flow. This core is surrounded by a brine saturated low permeable Obernkirchner Sandstone ($k = 7 \cdot 10^{-15}$ m²) which serves as brine reservoir. Nitrogen is injected to the inner core from the top and a mixture of gas and vapor is released from the bottom. The experimental setup consists of 4 measurement segments along the height.
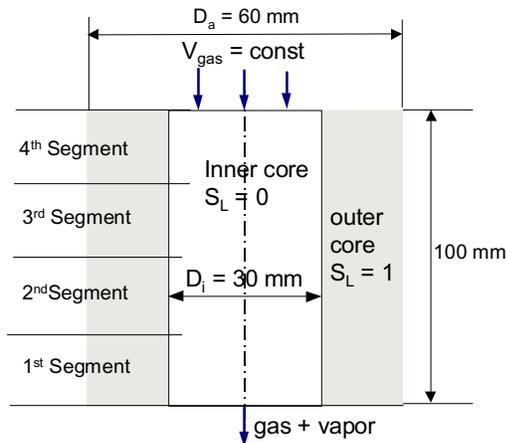
Figure 3. Experimental setup for investigation of halite formation during injection

Experiments were performed for 5 various conditions with different gas injection rates, duration and temperature. The experimental results relevant for a comparison with the numerical simulation are the liquid and the solid saturation in each segment of the inner and of the outer core at the end of duration. Furthermore, the measured permeability reduction caused by halite precipitation was given. These data are used to determine the parameters of the function implemented in EWASG to model permeability reduction.

Because of the relatively small dimensions of the experimental setup one main question for the numerical investigation is the discretisation of the model or in other words the adequate size of the representative elementary volume (REV). Therefore simulations with three different grid resolutions are performed. For coarse resolution each segment of each core is one element, for the fine resolution each segment of each core is divided into 16 elements. Figure 4 shows exemplary the results of the solid saturation in the inner core for the different grid resolutions compared to the experimental results.
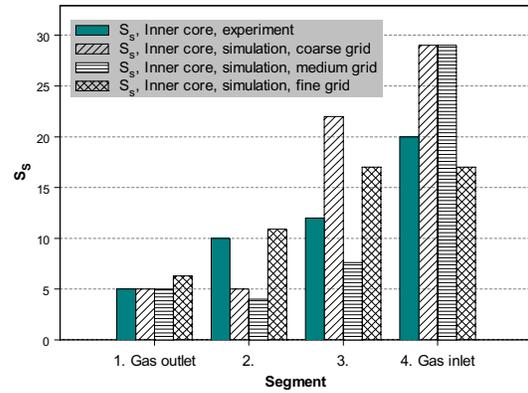
Figure 4. Halite precipitation in the inner core, experiment No. 3, comparison of experimental and numerical results

The results of the numerical simulations agree qualitatively very good with the experiments. The followed basic transport processes and mechanism from the experimental results can be deduced. First brine is transported from the outer to the inner core, then water evaporates until halite saturation is reached and precipitation occurs. Halite formation occurs mainly in the inner core. These processes could be visualized from the numerical simulations in detail. The simulations show also the experimentally observed gradients of liquid and solid saturation.

The quantitative comparison with the experiments is in the expected order of magnitude. The absolute values show partially higher deviations. The reasons for the deviations can be explained by

- the band width of input parameters for which no experimental data are documented (e.g. heat capacity of rock),
- differences in the boundary conditions (e.g. constant volume flow in experiments, constant mass flow in simulations, possible boundary effects),
- the numerical discretisation, especially because of the relatively small dimensions of the experimental setup,
- measurement accuracy.

In summary, the comparison of the measured and simulated results demonstrated that the conceptual model underlying EWASG is appropriate to model halite formation during gas injection.

Modelling of a Generic Natural Gas Storage Aquifer

The halite formation in a generic natural gas storage aquifer is investigated. The simulations serve for qualification of the conceptual model under realistic reservoir conditions for injection as well as for production and for the code modifications performed.

Model description

Two 3-D models of the generic storage aquifer are used, one with a vertical and the other with a horizontal well in the middle of the grid network. The dimensions, two-phase flow parameters and the initial conditions for pressure, temperature and liquid saturation are set as close as possible to realistic conditions. The main parameters of the grid and the initial conditions are listed in Table 2.

Table 2. Generic storage aquifer grid parameters and initial conditions

elements and dimensions	x-y-z direction:
- horizontal well	15x17x3 (565x520x15 m)
- vertical well	15x15x3 (565x565x15 m)
grid blocks for well	
- horizontal well	9 elements
- vertical well	2 elements
initial temperature	80 °C
initial reservoir pressure	
- injection	20 MPa
- production	30 MPa
initial gas saturation	80 %

The thermodynamic properties of the NCG are as follows. The density and viscosity are user defined. Their values are close to data for a realistic storage aquifer. For the enthalpy of the gaseous component, the enthalpy of the dissolved gas, and the Henry constant, the properties of CH_4 as implemented in EWASG are used.
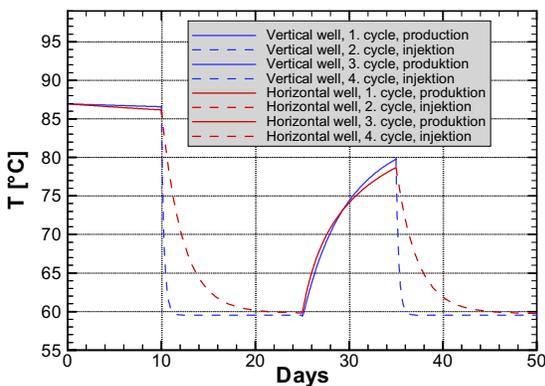
Figure 5. Temperature history of a selected element in the center of the model for the generic storage aquifer
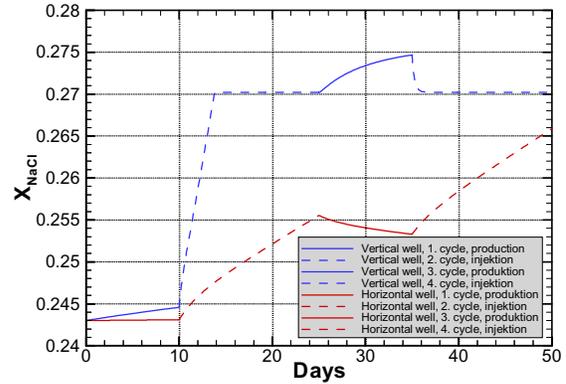
Figure 6. History of NaCl-concentration in the liquid phase of a selected element in the center of the model for the generic storage aquifer
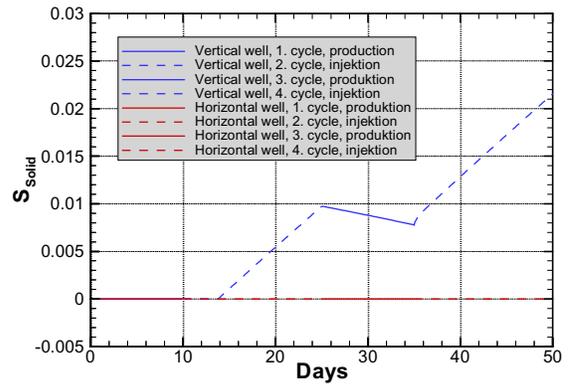
Figure 7. History of solid saturation of a selected element in the center of the model for the generic storage aquifer

The simulated scenarios are pure injection of dry gas, pure production and a cycle of production, followed by injection. The initial salt content of the brine is varied between $x_{NaCl} = 0.243$ and halite saturation (0.27). Constant mass flow rate is assumed for injection and production. Injection gas temperature is 60 °C.

At the boundaries no flow conditions are set which inhibit the inflow of brine as well as heat conduction from outside the aquifer.

Figure 5, 6 and 7 show exemplary the results of simulations for the cycle, both, for the vertical and the horizontal well.

In summary the results of the simulation of the generic aquifer meet the field experiences quite good considering the simplified configuration and data basis.

The simulations of the pure injection into the generic aquifer storage comply with the experiences on real aquifer storages. In that case precipitation occurs locally close to the well, which is also the experience in real systems.

The simulations for pure production show also precipitation, when the initial salt saturation is high enough. In that case halite formation occurs along the whole aquifer due to the pressure drop in the aquifer. Therefore, local solid saturation is rather low, but the total amount of precipitation is much higher than in the case of pure injection. This is not in contradiction to field experiences.

The simulations for the cycles show a higher tendency of precipitation with time. This is plausible, because the results indicates that the history of the aquifer may play an important role in the adequate simulation of halite formation.

CONCLUSIONS

TOUGH2 Modifications

The performed modifications enable the application of TOUGH2, Version 2 to the problem of halite formation in the gas industry which can not be handled by commercial codes yet. Additional work is still open to meet the requirements completely.

The new equation for the enthalpy of aqueous NaCl-solutions enhances the temperature range of EWASG down to 25 °C. The estimation quality decreases considerable for 0 °C. Since the absolute values at 0 °C are very close to zero, this does not allow an exact evaluation of the quality of the fit. Here additional measured values of the enthalpy between 0 and 25 °C would be helpful. The reference enthalpy of water in the equation limits the application of the equation to the critical temperature of water (350 °C). This choice is only arbitrary and probably does not make much sense if enthalpy values above 350 °C have to be fitted. Nevertheless the type of equation still can be used, only the parameters and the degree of the polynoms may change.

Numerical Modelling of Halite Formation

The laboratory experiments could be simulated appropriately and the simulations of the pure injection of the generic aquifer storage also comply with the experiences on real aquifer storages. Therefore it can be concluded that the conceptual model underlying EWASG is appropriate to model the halite formation in the case of pure injection.

The simulations for pure production indicate the presence of precipitation if the initial salt saturation is high enough or duration is long enough. However, in this case the level and the speed of precipitation in the performed simulations is too low to extract certain conclusion about the underlying processes. It can not be excluded that the no flow boundary conditions may play an important role on the simulated temperature and pressure field.

The results of the cycle-simulations indicate that the aquifer history may also play an important role for an adequate simulation of halite precipitation.

In real aquifers differences between horizontal and vertical wells are more pronounced, especially in the temperature history. It is assumed that the reason is the enthalpy used for CH_4 for the gaseous phase as implemented in EWASG. Here CH_4 is treated as an ideal gas, in which the Joule-Thomson effect is neglected. This causes that the temperature gradient behaves independent from the pressure drop. In fact pressure drop in horizontal and vertical wells can differ considerably which may yield to greater temperature differences.

Outlook

It has been demonstrated that the modified version of TOUGH2, version 2 is a promising tool to model the halite formation in natural gas storage aquifers.

Further code qualification is still required. This will be performed by a stepwise approach to a real natural gas storage aquifer. The first steps are the implementation of complete thermodynamic properties of the gaseous component and the evaluation of the effect of the boundary conditions. The latter means the investigation of the effect of lateral heat conduction and brine inflow. Final conclusions can be drawn if real field cases are simulated in future.

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