A NOVEL CONCEPT FOR LONG-TERM CO₂ SEALING BY INTENTIONAL SALT CLOGGING

Laura J. Wasch, Jens Wollenweber, Tim J. Tambach TNO Princetonlaan 6 3508 TA, Utrecht, The Netherlands e-mail: laura.wasch@tno.nl

ABSTRACT

Well abandonment after CO₂ storage demands a mitigation strategy for CO₂ leakage along the wellbore. We propose forming a salt seal around the wellbore after CO₂ has been injected into a depleted gas field, preventing possible CO2 transport to the surface. The placed salt plug will protect the wellbore material from contact with supercritical CO₂ and acid brine. We propose the concept of brine-alternating CO₂ injection for intentional salt clogging of the reservoir. Injection of CO₂ will evaporate water from previously injected brine, causing salt precipitation. The formation of a dryout zone is a known process occurring during CO₂ injection in saline aquifers or depleted gas fields that leads to unintentional clogging and possibly injectivity issues.

Modeling is performed with TOUGH2, simulating injection of brine and subsequently CO₂. Our model is based on the K12-B depleted gas field, using the elevated pressure conditions of a CO₂-filled storage reservoir. The model results indicate that injecting multiple cycles of brine and CO₂ could be used for controlled precipitation of salt in the reservoir. The injection procedure results in a 40 cm thick salt bank around the well, with the precipitated salt reducing the porosity from 10 to 8% at the location of maximum salt precipitation. This porosity decrease causes complete permeability impairment. We conclude that brine-alternating CO₂ injection could pose an effective method for intentional salt-clogging of the near-wellbore area.

INTRODUCTION

Carbon capture and storage (CCS) has great potential for reducing CO_2 emissions into the atmosphere and mitigating climate change (IPCC, 2005). Geological storage reservoirs are commonly pierced by multiple wells, especially reservoirs previously used for oil and gas production. Wells are potential CO_2 leakage pathways to the surface. Hence, reservoirs for CO_2 storage require a long-term wellbore seal to prevent CO_2 leakage to the surface (Bachu, 2003).

A major concern is leakage along the wellbore after extended time intervals caused by mechanical loads and/or chemical degradation of well-cement. CO₂ partially dissolves in the formation water, resulting in acid conditions in the storage reservoir. Wellbore materials react under acid conditions (e.g., Kutchko et al., 2007), possibly affecting its containment properties. Techniques previously suggested to ensure well integrity in CO₂ storage environment include, among others: improvement of cement design (Benge, 2009) and clogging of the reservoir by polymers (Hou et al., 2011) or by biomineralization (Cunningham et al., 2011). We propose salt for sealing the reservoir section of the well, since salt has proven sealing capacities with respect to gas and may "naturally" precipitate during gas storage or production.

Salt clogging of the reservoir or wellbore is generally regarded as a possible problem in gas and oil production (e.g., Kleinitz et al., 2003), CO₂ storage in saline aquifers (e.g., André et al., 2007), and depleted gas fields (e.g., Giorgis et al., 2007, Tambach et al., 2011 and Tambach et al., 2012). Gas production and injection may lead to salt precipitation by the evaporation of water from the brine into the gas phase. Salt precipitation in the pore space reduces flow, affecting negatively gas injectivity or productivity. We propose to reverse this problem using this phenomenon for intentional salt clogging (Figure 1). Simulations are performed to investigate controlled salt precipitation by brine-alternating CO_2 injection. A practical injection strategy must be designed to facilitate the formation of an effective wellbore salt-seal while keeping operational time as short as possible.



Figure 1. Illustration of the brine alternating CO_2 injection concept forming a salt seal around the wellbore.

SIMULATION DETAILS

Simulations are performed with the flow simulator TOUGH2, using its ECO2N module for CO_2 and brine mass transfer (Pruess and Spycher, 2007). Permeability changes are modeled with a tubes-in-series porosity-permeability model, assuming 20% porosity clogging to result in complete permeability impairment (Verma and Pruess, 1988). Precipitation is studied by the pore-space fraction occupied by salt, the "salt pore volume fraction." Chemical rockfluid, capillary pressure, permeability hysteresis and nonisothermal effects are neglected.

A simplified one layer model is built after the K12-B depleted gas field (Audigane et al., 2007), a Dutch CO₂ storage demonstration site. The model assumes a homogeneous sandstone reservoir of fluvial facies. The sandstone is characterized by a porosity of 0.11 and a permeability of 22.4 mD. A 1D radial reservoir model is used, described by 59 grid cells with grid refinement towards the well interface. The 20 m near-well model extends from 3640 to 3780 m depth. Ambient conditions applied are a 90°C reservoir temperature and 300 bar pressure after CO₂ storage. The reservoir is assumed dried out after CO₂ injection (no brine) and CO₂ filled (no methane). No initial salt due to previous gas

production or storage (e.g. Kleinitz et al., 2003) is assumed for the base-case model.

Injection details are shown in Table 1. The injection strategy is designed with a brine/CO₂ ratio allowing complete evaporation of injected water into CO₂ at reservoir conditions. Note that over 200 times more CO₂ than brine is injected in order to evaporate the injected brine. The injection rates versus times are chosen arbitrarily while respecting the brine/CO₂ ratio. A 0.1 NaCl mass fraction brine (XNaCl) is used.

Table 1. One cycle of brine-alternating CO_2 injection.

Phase	Time (hr.)	Injection rate (kg/s)	Injected mass (kg)
Brine	0.2	1.4	945
CO_2	82.5	0.7	207347

RESULTS

Brine alternating CO₂ injection

Two cycles of brine-CO₂ injection are modeled to assess the effect of repeated injection cycles on salt precipitation. Simulations results are shown in Figure 2 and 3. The injection cycle starts with brine injection. At the end of the cycle, brine has moved 0.23 m into the reservoir. Subsequent CO₂ injection displaces the brine ~ 0.1 m further into the reservoir. While pushing the brine forward, CO₂ evaporates water from the brine at the CO₂-brine contact. Precipitation occurs in one cell at each time step when dissolved salt reaches the solubility limit.. This precipitation front moves the reservoir inward with time as more CO_2 is injected, leaving a salt bank behind. Salt precipitation progresses from the well, forming a 0.37 m thick salt bank at the end of the cycle (Figure 2, 82.7 hr.).

The second injection cycle starts again with brine injection. Since the brine is NaCl undersaturated, the salt bank formed in the first cycle is redissolved (Figure 2, 82.9 hr.). Brine salinity increases with flow through the reservoir as salt is progressively dissolved. This results in a higher salt mass fraction at the brine injection front (Figure 3, 82.9 hr.). Upon CO_2 injection, the brine is pushed forwards and dissolves the final part of the salt bank (~0.1 m). In addition, CO_2 evaporates brine at the CO_2 -brine contact. This results in a high salinity peak at both the CO_2 and brine injection front (Figure 3, 120 hr.). CO_2 injection evaporates the brine starting from the well reservoir inwards, again forming a continuous salt bank. Furthermore, since the brine at the brine injection front has a higher salt content, a salt peak is formed (Figure 2, 165.4 hr.). This salt peak is located at 0.3 m distance to the injection well, has a thickness of 0.2 m, and clogs up to 3.0% of the original pore volume.



Figure 2. The fraction of pore space occupied by salt with distance from the well. The salt content is illustrated for different times, showing the built up of a salt bank (cycle 1) and dissolution followed by re-precipitation of the salt bank, now forming a salt peak (cycle 2).



Figure 3. The NaCl mass fraction of brine during cycle 1 and 2. The NaCl mass fraction is close to the initial value of 0.1 in cycle 1. Brine injection in cycle 2 shows an increase of salinity at the front. During CO₂ injection the NaCl mass fraction shows the movement of brine and evaporation from the back increasing the value.

Intentional clogging of the near-well area

Modeling of two cycles of brine alternating CO_2 injection indicated increased pore clogging within the second cycle. To increase the effectiveness of clogging multiple injection cycles are modeled and more salt is added to the system with every brine- CO_2 injection cycle. The modeling will help assess the amount of cycles required for effective clogging.

The model results for the salt content at the end of a cycle are shown in Figure 4. They indicate that the salt peak grows with every cycle while salt clogging near well remains constant. Furthermore, the width of the peak and extent of the salt bank into the reservoir are equal after each subsequent cycle. The salt content nearwell remains constant since this salt precipitated out of newly injected brine with constant salinity every cycle. Only the first part of the injected brine dissolves salt, yielding more salt precipitation with each cycle. Because the same amount of brine is injected each cycle, the salt bank reaches an equal distance from the well following drying out at the end of every cycle.



Figure 4. The fraction of pore space occupied by salt with distance in the reservoir. The salt content for eight subsequent Brine-CO₂ injection cycles is illustrated. With every subsequent cycle, salt is added to the salt peak while the near-well part of the salt bank remains constant.

Following the porosity-permeability relationship of Verma & Pruess (1988), the permeability is effectively impaired after a 20% reduction in pore space. Modeling predicts that eight cycles of brine-CO₂ injection are required to reduce the porosity from 10% to 8% and effectively clog the reservoir (Figure 5). The porosity is roughly gradually decreased with subsequent cycles (as also shown in Figure 4). In contrast, permeability slightly decreases at first but reduces significantly when approaching 20% clogging. Note that the TOUGH2 simulation stops before reaching zero permeability, since the pressure increases to the limit upon complete clogging.



Figure 5. Porosity and permeability development with subsequent cycles of brine and CO₂ injection. Note that the permeability scale is logarithmic. Permeability calculated after the implemented porosity-permeability relationship (Verma and Pruess, 1988).

Sensitivity of the number of cycles

Brine-alternating CO_2 injection is used to increase salt precipitation around the wellbore with every cycle. The number of cycles required for clogging depends on the amount of salt added per cycle (brine salinity and injection rate/time), the amount of salt initially in the reservoir (solid salt or brine), and the spatial distribution of the injected salt. To assess the sensitivity of the predicted clogging towards these factors, two scenarios are run with a higher NaCl mass fraction of 0.2 and with initial salt present (salt mass fraction 0.0125).

The results are compared with the base-case model results. The first cycle already shows clear differences (Figure 6). A higher NaCl mass-fraction brine yields a salt bank with a higher percentage pore clogging (2.5% vs. 1.7%). Initial salt gives a similar salt bank near well, since the injected brine salinity and hence resulting salt is equal. However, this model already forms a salt peak in the first cycle because salt is initially present that is dissolved and re-precipitated further in the reservoir. For initial salt in the reservoir, the shape of the salt

peak remains equal to the base case for subsequent stages (Figure 6, cycle 4). Conversely, a higher salinity brine increases the width of peak and enhances clogging in the near-well area. Both scenarios yield faster clogging than the base-case model and require seven instead of eight cycles for effective clogging (Figure 7).



Figure 6. The fraction of pore space occupied by salt with distance from the well, presented for cycle 1 and 4. The salt content is shown for the base case and two scenarios with higher salinity and initial salt in the reservoir.



Figure 7. The fraction of pore space occupied by salt with distance from the well, presented for the final cycles yielding 20% salt clogging. The salt content is shown for the base case and two scenarios with higher salinity and initial salt in the reservoir.

DISCUSSION

The model results indicate that controlled CO_2 injection into a brine-containing reservoir (after brine injection) can result in intentional salt clogging. Furthermore, the salt precipitation induces significant permeability impairment. This is in agreement with studies on CO_2 induced salt precipitation. Muller et al. (2009) report permeability reductions of up to 60% for 16% salt clogging. Experimental work by Bacci et al. (2011) showed an 86% permeability decrease for a ~30% porosity reduction. Although significant permeability reduction is observed, complete clogging could require more (or less) precipitation than the 20% salt saturation assumed. Knowing the amount of precipitation required for effective clogging is crucial for modeling and designing a clogging strategy. Consequently, accurate porosity-permeability relations are required (Bacci et al., 2011).

Modeling predicts that the salt bank has a lateral extent of 0.4 m into the reservoir at the end of a brine-CO₂ injection cycle. A broader salt bank could be formed for longer intervals of brine and subsequent CO₂ injection or higher injection rates. Special care must be taken to design a practical and economic strategy of injection rates and times. Further research is required to study the desired width of the salt bank. This width will depend on the mechanical and chemical stability of the salt bank.

The model predicts the presence of a lower-saltcontent zone around the well, while complete clogging is modeled further in the reservoir at the salt peak. Clogging the reservoir up to the well-reservoir interface provides higher safety in terms of leakage prevention along the wellbore and well-material protection. Salt precipitation can be enhanced near the well by increasing the salinity of the injected brine. For example, injecting saturated brine prevents redissolution of the salt bank with a subsequent injection cycle. This allows the buildup of a salt bank from the wellbore interface. Although increased salinity is favorable for clogging up to the wellbore-reservoir interface, high salinity brines can also lead to unintended salt precipitation, which can clog the well before a proper salt plug has been placed. Alternatively, the remaining open pore space around the well could be sealed by injection of a synthetic polymer (Hou et al., 2011).

The current model consists of a homogenous reservoir. However, heterogeneities do exist in the reservoir (vertical and lateral) such as changes in lithology and fractures. Heterogeneities influence the flow of CO_2 and the porosity-permeability relationship and consequently the

location of salt precipitation (Pruess and Müller, 2009). Hence, heterogeneities should be considered to insure effective clogging around the entire wellbore in future work.

Currently, capillary-pressure effects are ignored. However, capillary pressure can affect salt precipitation and already precipitated salt by inducing backflow of brine towards the evaporation front (Pruess and Müller, 2009, Alkan et al., 2010, Ott et al., 2011, Zeidouni et al., 2009). Brine backflow enhances precipitation by adding salt to the system. On the other hand, backflow could lead to dissolution of the salt bank after its emplacement. Intentional salt clogging appears therefore particularly suitable for application to depleted gas fields that have low water content and accordingly little chance of salt seal redissolution by the backflowing water. Future modeling work is required regarding the longterm stability of the salt bank and its resistivity to redissolution. This process will eventually define the size of the salt bank required for effective long-term sealing of CO₂ injection wells.

CONCLUSIONS

Intentional salt clogging is proposed to protect wellbore cement against corrosive CO_2 and to mitigate leakage of stored CO_2 to the surface. Brine-alternating CO_2 injection has been found to be effective for intentional clogging of the near-well area. TOUGH2 modeling predicts that multiple cycles of brine- CO_2 injection are required for effective clogging and permeability impairment of the reservoir. For the presented injection strategy and model characteristics, the formed salt bank extends 0.4 m into the reservoir. The salt bank clogs 3% of the original reservoir porosity near the well, whereas the salt peak is responsible for 20% porosity clogging further in the reservoir.

The current injection strategy requires eight cycles of brine-alternating CO_2 injection for complete permeability impairment. However, the number of cycles required for effective clogging depends on the brine salinity and amount of injected brine. Additionally, initial salt or brine present in the reservoir reduces the amount of salt that has to be introduced to the reservoir for clogging. Moreover, when changing the porosity-permeability relationship, more or fewer stages are required as more or less salt clogging is required.

The presented injection strategy predicts effective clogging in relatively short time periods. Nevertheless, experimental or field case tests are required to assess if intentional salt clogging can be practically applied without, for example, clogging the wellbore. In conclusion, intentional salt clogging potentially provides a practical solution for protecting the wellbore against corrosive CO_2 attack, as well as blocking the flow of CO_2 along the wellbore and mitigating leakage to the surface.

ACKNOWLEDGMENT

We thank Pascal Audigane for providing the K12-B TOUGH2 model. The work was performed within the EU FP7 project "CO2CARE–CO₂ Site Closure Assessment Research" (Grant Agreement no. 256625).

REFERENCES

- Alkan, H., Cinar, Y., Ülker, E.B., Impact of Capillary Pressure, Salinity and In situ Conditions on CO₂ Injection into Saline Aquifers, *Transp.Porous Media*, 84(3), 799-819, 2010.
- André, L., Audigane, P., Azaroual, M., Menjoz, A., Numerical modeling of fluid-rock chemical interactions at the supercritical CO₂-liquid interface during CO₂ injection into a carbonate reservoir, the Dogger aquifer (Paris Basin, France), *Energy Convers.Manage.*, 48(6), 1782-1797, 2007.
- Audigane, P., J. Lions, I. Gaus, C. Robelin, P. Durst, B. Van der Meer, K. Geel, C. Oldenburg, and T.Xu, 2009, *Geochemical modeling of CO₂ injection into a methane gas reservoir at the K12-B field, North Sea*, in M. Grobe, J. C. Pashin, and R. L. Dodge, eds., Carbon dioxide sequestration in geological media—State of the science: AAPG Studies in Geology 59, p. 499–519.
- Bacci, G., Korre, A., Durucan, S., Experimental investigation into salt precipitation during CO₂ injection in saline aquifers. *Energy Procedia.* 4, 4450-4456, 2011.
- Bachu, S., Screening and ranking of sedimentary basins for sequestration of CO₂ in geological media in response to climate change, *Environ.Geol.*, 44(3), 277-289, 2003.

- Benge, G., Improving Wellbore Seal Integrity in CO₂ Injection Wells. 2009.
- Cunningham, A.B., Gerlach, R., Spangler, L., Mitchell, A.C., Parks, S., Phillips, A., Reducing the risk of well bore leakage of CO₂ using engineered biomineralization barriers. *Energy Procedia.* 4, 5178-5185, 2011.
- Giorgis, T., Carpita, M., Battistelli, A., 2D modeling of salt precipitation during the injection of dry CO₂ in a depleted gas reservoir, *Energy Convers.Manage.*, 48(6), 1816-1826, 2007.
- Hou, M.Z., Wundram, L., Wendel, H., Meyer, R., Schmidt, M., Kretzschmar, H.J., Schmitz, S., Franz, O., Developing, modeling and in-situ-testing a long-term wellbore seal in the framework of the CO₂. *Energy Procedia.* 4, 5291-5298, 2011.
- IPCC, 2005. IPCC Special Report on carbon dioxide capture and storage. Prepared by Working Group III of the Intergovernmental Panel on Climate Change [Metz, B., Davidson, O., de Coninck, H.C., Loos, M., Meyer, L.A. (Eds.)]. Cambridge University Press, Cambridge, United Kingdom/New York, NY, USA, 442 pp.
- Kleinitz, W., Dietzsch, G., Köhler, M., Halite scale formation in gas-producing wells, *Chem.Eng.Res.Des.*, *81*(3), 352-358, 2003.
- Kutchko, B.G., Strazisar, B.R., Dzombak, D.A., Lowry, G.V., Thauiow, N., Degradation of well cement by CO₂ under geologic sequestration conditions, *Environ.Sci.Technol.*, *41*(13), 4787-4792, 2007.
- Ott, H., De Kloe, K.D., Marcelis, F., Makurat, A., Injection of supercritical CO₂ in brine saturated sandstone: Pattern formation during salt precipitation. *Energy Procedia*. 4, 4425-4432, 2011.
- Pruess, K. and Müller , N., Formation dry-out from CO₂ injection into saline aquifers: 1. effects of solids precipitation and their mitigation, *Water Resour.Res.*, 45(3), 2009.
- Pruess, K. and Spycher, N., ECO2N A fluid property module for the TOUGH2 code for studies of CO₂ storage in saline aquifers, *Energy Convers.Manage.*, 48(6), 1761-1767, 2007.

- Tambach, T., Koenen, M., Van Bergen, F., Geochemical evaluation of CO₂ injection into storage reservoirs based on case-studies in The Netherlands. *Energy Procedia*. 4, 4747-4753, 2011.
- Tambach, T.J., D. Loeve, C. Hofstee, W.-. Plug, and J.G. Maas, Effect of gas field production and CO₂ injection on brine flow and salt precipitation, Proceedings TOUGH Symposium 2012, Lawrence Berkeley National Laboratory, Berkeley, California, September 17-19, 2012.
- Verma, A. and Pruess, K., Thermohydrological conditions and silica redistribution near high-level nuclear wastes emplaced in saturated geological formations, *J.GEOPHYS.RES.*, 93(B2), 1159-1173, 1988.

Zeidouni, M., Pooladi-Darvish, M., Keith, D., Analytical solution to evaluate salt precipitation during CO2 injection in saline aquifers, *Int.J.Greenh.Gas Control*, 3(5), 600-611, 2009.