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Sensitivity study of the reactive transport model for CO₂ injection into the Utsira saline formation using 3D fluid flow model history matched with 4D seismic

Audrey Estublier^{a,*}, Alexandre Fornel^a, Teddy Parra^a, Jean-Pierre Deflandre^a

^a IFP Energies nouvelles, 1-4 avenue de Bois-Préau, 92852 Rueil-Malmaison, France

Abstract

This article reports on the reactive transport simulation work dealing with CO₂ storage at Sleipner. The study has been initiated in the CO₂ReMoVe European project with one preliminary scenario and is carrying on in the CO₂CARE one with a sensibility study. Its main purpose was to simulate on the long term the CO₂ fate and to estimate the proportion of trapped CO₂ per trapping mechanisms over time. All the simulations were performed using a 2D reactive transport model based on a 3D fluid flow model history matched with 4D seismic.

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Keywords: CO₂ storage; reactive transport simulation; Sleipner

1. Introduction

An important aspect of establishing the practicability of CO₂ geological storage is the assessment of the likely performance of the storage system over relevant time-scales. There is much uncertainty associated with the evolution of CO₂ and subsurface parts of the CCS system, particularly in the long-term. This article presents the results of two complementary studies relative to long term CO₂ fate in the Utsira Sand formation of the Sleipner site. Since 1996, 1 Mton of CO₂ is injected per year through one well at a depth of about 1000 m into this formation, a major regional saline aquifer ([1],[2]). At this depth, the salinity is about 35 g/l, the in-situ temperature is closed to 37 °C and the pressure is about 10 MPa. At such reservoir conditions, CO₂ is supercritical[†]. The saline aquifer is a 200m-thick sandstone layer with

* Corresponding author.

E-mail address: audrey.estublier@ifpen.fr

[†] In the following article, the term “gas” will be used to denote carbon dioxide under supercritical conditions, which forms a phase separate from the aqueous or liquid phase (brine)

thin intra-reservoir shaly layers. Sandstone porosity is between 24% and 40% and the permeability between 1 and 3 Darcy. At the beginning of 2011 almost 12 million tons of CO₂ were stored.

The works presented here have been performed in two European projects. The CO₂ReMoVe project [3] was interested in the uncertainty of the CO₂ fate while addressing the long-term performance assessment. The present CO₂CARE project [4] issues concern site closure, transfer of responsibility (liability) to the competent authority and post-closure obligations. In these two studies, we are interested in site stabilization evidence over time. That requires to assess CO₂ proportion per trapping mechanism and to study the long-term evolution of potential risky zones. As part of the CO₂ReMoVe project objective, the approach has consisted in analyzing on the long term the reactive transport resulting from CO₂ injection. This work was based on a preliminary geochemical analysis and a 2D reactive transport simulation. It aimed at studying the fluid rock interactions resulting from pH decrease due to the presence of CO₂. It has been continued in the CO₂CARE project through a sensitivity study to analyze the impact of several uncertainties associated to both the geochemical model and the fluid flow one.

2. The initial 2D reactive transport simulation carried out in the CO₂ReMoVe project

2.1. The 0D geochemical simulations

We have considered two rock types to model the Utsira formation: the sand facies, a poorly cemented, fine-to-medium grained, moderately sorted sand and the shale facies (Table 1). The mineralogical composition of sand assessed by Pearce et al. [5] from core measurements is used as the reference one. Regarding the shaly layers, mineralogy is assumed to be equivalent to the cap rock one derived from Boe and Zweigel [6] because of a lack of data.

Table 1. Facies data and geochemical models: volume proportion (%)

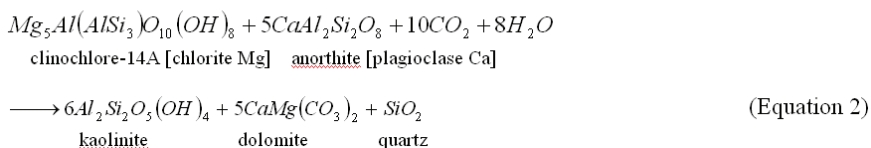
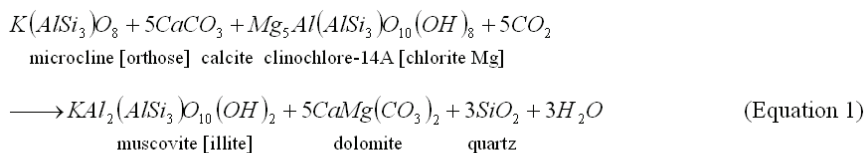
% volume	SAND volume proportion			SHALE volume proportion		
	Data	Fe chlorite model	Mg chlorite model	Data	Fe chlorite model	Mg chlorite model
quartz/chalcedony	76.33	76.50	76.50	22.8	26.03	26.03
K-feldspar/microcline/orthose plagiocase	6.93	6.95	6.95	2.3		
Na plagiocase/albite	3.01	2.96	2.96	13.2	7.53	7.53
Ca plagioclase/anorthite		0.06	0.06		7.53	7.53
mica/muscovite/illite	5.22	5.23	5.23	25.1	28.65	28.65
chlorite	1.33			4.4		
Mg-chlorite/clinochlore-14A			1.33			5.02
Fe (iron)-chlorite/daphnite-14A		1.33			5.02	
calcite	6.74	6.75	6.75	1	1.14	1.14
kaolinite		0.17	0.17	19.5	22.26	22.26
siderite				1.1		
pyrite	0.05	0.05	0.05	1.6	1.83	1.83
ilmenite	0.12					
zeolite	0.22					
ti oxides	0.03					
smectite				9		
apatite	0.02					

Regarding the geochemical model (Table 1), the mineralogical assemblage is as complete as possible considering the available mineralogy information. Twelve minerals are considered for characterizing the facies making two assumptions to mitigate with the lack of information. The first assumption concerns

the plagioclase mineral. An arbitrary composition is proposed to take into account both calcium (Ca) and sodium (Na) plagioclases in the sand and shale systems. For the second assumption, we decided to build two geochemical models for both sand and shale facies and to compare their impacts. One model considers the chlorite mineral only as iron chlorite (named Fe chlorite) and the other one, only as magnesium chlorite (named Mg chlorite). These two models differ from one to the other by the reactive pathway pattern. The Fe chlorite geochemical model is based on reactive pathways controlled by calcium and iron with a possible CO₂ mineralization through the calcite and siderite minerals. In the Mg chlorite model, they are controlled by calcium and magnesium with a possible CO₂ mineralization through the calcite, dolomite and magnesite minerals. Dawsonite was not chosen as a possible secondary mineral because of Utsira reservoir conditions. According to H. Hellevang [7], dawsonite is observed to form in natural reservoirs at high CO₂ pressures typically replacing Na-feldspars as pore-filling cement. He also adds that observation of dawsonite is few in quartz-rich shallow "cold" reservoirs such as Utsira.

The geochemical simulations, i.e. 0D simulations, were carried out with ARXIM[‡] software. It is a 0D fluid-rock interaction numerical simulator for estimating how mineral proportions and water compositions evolve over time. For these simulations only, the temperature is fixed at 37°C, the pressure at 10 MPa and the salinity at 35 g/l. ARXIM[‡] is based on the Arrhenius's law to model reaction rate. This equation depends on the reactive surface area and on the kinetics parameters. Due to the lack of data, the precipitation kinetics parameters are assumed to be the same to dissolution ones although one expects that precipitation is 10-100 times slower than dissolution. Mineral is modelled by a sphere and the evolution of the reactive surface is based on the radius evolution of the sphere. For the minerals, which are initially missing, the initial radius is fixed at 10⁻⁶ m. The reactive surface area is assumed to be the total (100%) surface area of minerals for the first simulated case carried out in CO₂ReMoVe because of the lack of information on it. However, interactions between water and minerals are generally expected to occur only at selective sites of the surface and the difference between total surface area and reactive surface area can be between 1 and 3 orders of magnitude [8]. This large uncertainty has been considered in the CO₂CARE project through the sensitivity analysis presented in the third part of this article.

With the Mg chlorite simulation, CO₂ is mineralized mainly as dolomite in both sand (Equation 1) and shale (Equation 2) facies. The necessary magnesium element is provided by the chlorite dissolution and the calcium element is from calcite and calcium plagioclase.



For both geochemical simulations, i.e. Fe and Mg models, we notice that sand is less reactive than shale in the Utsira formation. Thus, the sand porosity increases a little from 37% (the initial value) to maximum 37.21% whereas the shale one decreases from 10.25% (the initial value) to 6.75%. In the same

[‡] Trademark of School of Mines in Saint Etienne and IFPEN

way, the injected CO₂ mass, which has been minerally trapped over the simulated period of 10,000 years is only of 9 kg per 1 m³ of formation in the sand rock whereas it is seven to eight times bigger in shale. The comparison between the results of the two geochemical simulations shows there is a small difference. The minerally trapped CO₂ mass simulated by the Fe chlorite model is 0.8% bigger in sand and 5% smaller in shale than the one obtained with the Mg chlorite model. Regarding the porosity simulated by the Fe chlorite model, its increase in sand is 9% smaller and its decrease in shale is 0.5% bigger than the one obtained with the Mg chlorite model. The geochemical simulations show that the CO₂ mineralization occur in the first ten's of years i.e. between 10 and 100 years, which is quite fast. Such a high reactivity rate can be explained by the assumption of having 100% of the mineral surface to react. One can consider this primary result as an upper bound of mineral trapping by comparison with other simulation results presented here and using the same fluid flow model.

Table 2. Geochemical models: kinetics parameters and reactive area

	Kinetics parameters		Reactive surface area (cm ² /g)	
	pk=-log(k)	Ea(kJ/mol)	IFPEN (CO ₂ ReMoVe) model ^a	Audigane 2007, model ^b
quartz/chalcedony	13.4	90.9	1.13E+04	9.8
orthose (K-Felspar parameters)	12.41	38	1.18E+04	9.8
Na plagioclase (albite parameters)	12.56	69.8	12.0	9.8
Ca plagioclase (anorthite parameters)	9.12	17.8	10.9	not in the model
illite (muscovite parameters)	13.55	22	1.08E+07	151.6
Mg-chlorite/clinochlore-14A (chlorite parameters)	12.52	88	1.15E+04	9.8
Fe (iron)-chlorite/daphnite-14A (chlorite parameters)	12.52	88	1.15E+04	
calcite	5.81	23.5	11.1	equilibrium
kaolinite	13.18	22.2	1.14E+07	151.6
siderite (dolomite-dis parameters)	7.53	52.2	1.04E+04	9.8
pyrite	4.55	56.9	5.99E+03	not in the model
magnesite	9.34	23.5	1.02E+04	not in the model
dolomite	7.53	52.2	1.04E+04	9.8

^a Reactive surface area is 100% of the mineral surface

^b Reactive surface area is mainly 0.1% of the mineral surface excepted for Na plagioclase (100%) and for illite/kaolinite (0.001%)

2.2. The reactive transport simulation results

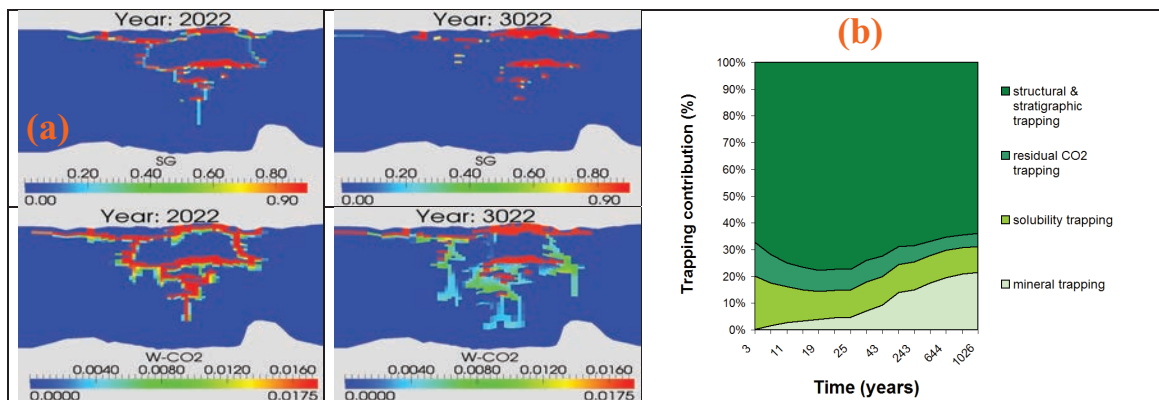


Fig. 1. 2D vertical reactive transport simulation: (a) CO₂ gas saturation (SG) and molar fraction of CO₂ dissolved in water phase (W-CO₂) for years 2022 and 3022 (b) CO₂ mass per trapping type in reactive transport simulation on a 1000-year period assuming 100% of the mineral surface to be reactive (upper bound for CO₂ mineralization with the geochemical model used)

The reactive transport simulations were performed with the coupling of the geochemical model and the fluid flow one based on a 2D vertical grid. In this study, we used a more complex fluid flow model compared to the current literature ([9] and [10]) since the heterogeneous Utsira formation is considered with a more realistic vertical alternation of sand and shale lens. In practice, the 2D fluid flow model was extracted from the 3D fluid flow one, which has been history matched by applying an innovative approach using 4D seismic inversion results [11]. To mitigate with very important computing time, a 2D vertical section extraction coupled to a vertical upscaling has been designed to get a smaller grid of 3239 cells ($dy=50$ m, $dz=5$ m). Boundary conditions were moved away at the basin scale to reduce their effect. Sand and shale facies have constant petrophysical properties [11]. Sand is characterized by a 37% porosity and a 3 darcys permeability with a threshold capillary pressure of 0.02 bar. Shale layers are assumed as semi-permeable with a porosity of 10.25% and a permeability of 10 mD with a threshold capillary pressure of 1.36 bar. Thus, water can migrate through shale layers but not the CO₂ gas due to the capillary barrier. The irreducible water saturation is arbitrary set to 20% because of the lack of information. With this model, the CPU time reaches 2 days for one thousand years of simulation.

In general, we found the same trends as for the 0D geochemical simulations in term of dissolution and precipitation for all minerals and regarding the excessive reaction rate. With the 2D model (Fig. 1a), we observe that at short term shales represent fluid flow barriers for CO₂ gas flow. As a consequence, they constitute the top of local intra-reservoir traps. On the long term, the main observation deals with the gravity effect of brine containing dissolved CO₂, this effect being important in Utsira due to its large thickness. While sinking, the CO₂-charged brine also exchanges with shale layers. Thus, this phenomenon makes at long term shale layers to become mineral trapping zones in this model. We observe (Fig. 1b) after 1,000 years that the reactive transport simulation using the Mg geochemical model stores up to 21.5% of the injected CO₂ mass through mineral trapping -mainly in sand rock. Regarding the three other CO₂ trapping mechanisms, 4.8% is capillary trapped (with a residual gas saturation of 5%), 9.7% is dissolved in water and 64% remains free. This high and unexpected proportion of minerally trapped CO₂ mainly results from the assumption on the reactive surface area (i.e. 100% here). But it is emphasized by the fluid flow effects since intra-shale barriers significantly contribute to intensify fluid flow exchanges.

The analysis of these preliminary results shows that, although the general trend is well simulated by the 0D model, the 2D model allows in addition to consider the CO₂ plume migration and its impact on CO₂ trapping. Thus, to better characterize storage performance and also possible risks of leakage, sufficiently accurate modelling of fluid flows is essential as it monitors the geochemical reactivity within the reservoir. However, due to modelling choices and parameter uncertainty, these results show a faster mineralization than suspected in the scientific literature at the origin of a high proportion of mineral trapping after 1,000 years. This preliminary case simulated in CO₂ReMoVe is one possible scenario (an upper bound one when considering mineralization). To complete and improve this result, we work in CO₂CARE at studying other possible scenarii while considering uncertainties linked to the models. Among them we look at the effect of the barrier capacity of shale layers, the heat model, the grid, the relative permeability curves, the permeability-porosity law, and the geochemical model.

Some other uncertainty sources have not been yet considered in the sensitivity analysis because of modelling challenges. They deal with model dimension, salinity transport model, shale layer distribution and capillary pressure.

3. Sensitivity analysis

The sensitivity analysis deals with six major issues. The characteristics of the models, which are built to simulate the impact of the selected uncertainties are detailed in Table 3. Fig. 2 compares the mineral trapping proportion over time for the different reactive transport models of Table 3.

Table 3. Characteristics of the models to simulate the impact of some uncertainties (in bold type, the characteristic names of the simulations)

Uncertainty sources	Characteristics of the simulations
Mg chlorite model	The initial reactive transport model simulated in CO ₂ ReMoVe (Mg chlorite). It uses the Mg chlorite geochemical model (Section 2). No heat model, no dynamic permeability, 100% reactive surface
(1) the barrier capacity of the shale layers	Two simulations: one case with impermeable shale layers ($K=10^{-4}$ mD and $P_c=27$ bar) and one with permeable shale layers ($K=10$ mD and $P_c=0.2$ bar)
(2) the heat model	Two simulations with heat transfer modelling: geological gradient of 0.033 °C/m and injected CO ₂ temperature (T=28°C , computed with a thermal fluid flow model for wells and T=58°C , computed with the assumption of adiabatic conditions)
(3) the grid	A Local Grid Refinement (LGR) of 9 cells ($dy=17$ m, $dz=1.7$ m) has been added to all the grid cells, which contained CO ₂ in the initial simulation based on the coarse grid at 1,000 years (Fig. 3(a))
(4) the relative permeability curves	Two uncertainties are taken into account: the irreducible water saturation Swi ([11],[12]) with the range [5%;10%] for sand and [5%;20%] for shale; the hysteresis phenomenon for relative permeability with a residual gas saturation of range [5%;25%]
(5) the permeability-porosity law	The permeability evolution law KPhi used is the Koseny-Carman equation
(6) the geochemical model	Three simulations: one with the Fe chlorite geochemical model (see section 2.1), one with the reactive surface area data from the Audigane's article [9] (area data 1,000 times smaller than the initial case - called Mg chlorite model), and one, which is the same as the previous one, with no Ca Plagioclase (plagioclase is only represented by Na plagioclase)

Fig. 2a shows the huge uncertainty on mineral trapping proportion due to the mineral assemblage uncertainty and to the reactive surface one. Regarding the chlorite type (magnesium or iron), its uncertainty has a very small impact on the CO₂ fate, with an absolute difference lower than 1%. On the contrary, it is huge regarding the reactive surface area when one compares the "Mg model" curve (i.e. 100% reactive surface) with the "reactive surface area, Audigane 2007" curve (i.e. 0.1% reactive surface). When reducing the reactive surface, we observe that about 5% of injected CO₂ is mineralized at 1,000 years -to be compared to 22% with 100% of reactive surface (Mg chlorite model). The third curve represents the impact of a geochemical model combining two assumptions (see the "reactive surface area Audigane 2007, no Ca plagioclase" curve): the reactive surface area is small (i.e. 0.1% reactive surface) and plagioclase is only represented by Na plagioclase. In that case, we observe that there is no CO₂ mineralization before 1,000 years (it only appears after 2,000 years in a proportion of 0.4% of injected CO₂). Indeed, Ca plagioclase is the main provider of the calcium element in shale and this element is necessary to calcite precipitation. Without this mineral, the CO₂ mineralization starts later with the dolomite precipitation thanks to the in-situ calcite dissolution.

In Fig. 2a, one can also notice a huge uncertainty on the mineral trapping proportion regarding the shale layer permeability. This is explained by the higher mineralization capacity of the shale compared to

the sand as explained in Section 2.1: 9 kg per 1 m³ of formation in the sand rock whereas it is seven to eight times bigger in shale. So, the impact of the shale layer permeability is linked to the shale mineralization capacity. Fig. 2b shows that by comparison with the geochemical model, the hysteresis phenomenon and the heat transfer model have a smaller impact on the mineralized CO₂ proportion (maximum absolute difference of 3% compared to the "Mg model"). As regards the irreducible water saturation and the dynamic permeability, their impact is very small, almost negligible.

The case with the Local Grid Refinement (Table 3 —line 3) is interesting because it allows to:

- decrease the numerical diffusion and so locally the dissolved CO₂ mass. The consequence is an increase of the free CO₂ gas mass available for vertical migration. That is why the CO₂ (supercritical, dissolved and mineralized) plume (Fig. 3, circle (b)) is more extended at the Utsira top. Unfortunately this simulation does not allow to totally conclude regarding the grid refinement impact. Indeed the plume reaches the lateral parts of the grid, which are very coarse and that could have contributed of the mineralized CO₂ mass increase (Fig. 2b).
- simulate gravity instabilities (Fig. 3, rectangle (a)), which are from local intra-reservoir traps. This phenomenon increases the CO₂ dissolution because it allows the mixing of two different brines: the sinking one because of dissolved CO₂ and the one going upward due to fluid circulation.

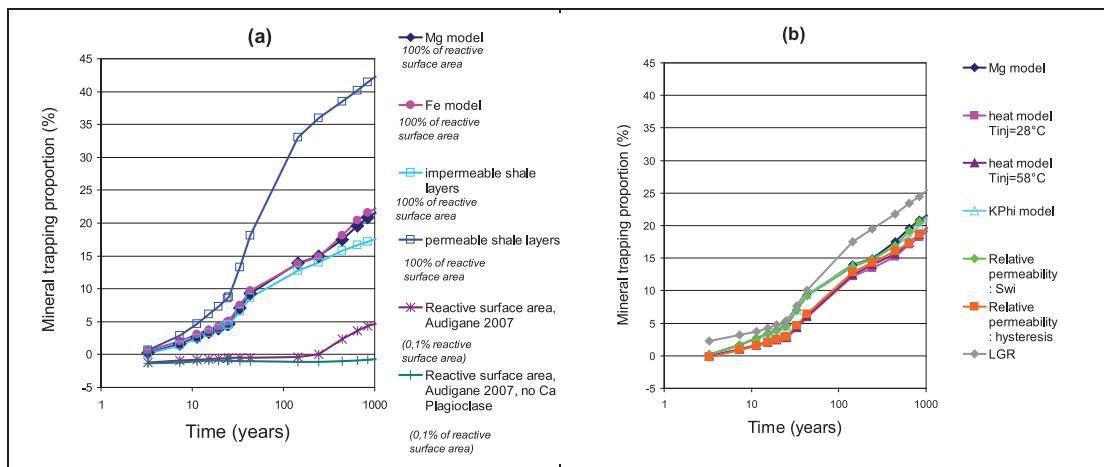


Fig. 2. Injected CO₂ proportion (%), which is minerally trapped (negative proportion means release of CO₂ through carbonate dissolution) over time - comparison of reactive transport models: (a) mineral assemblage (Fe and Mg with or without Ca Plagioclase) and reactive surface area permeability of shale layers (b) heat models, K-Phi law model, model with hysteresis, model with irreducible water saturation

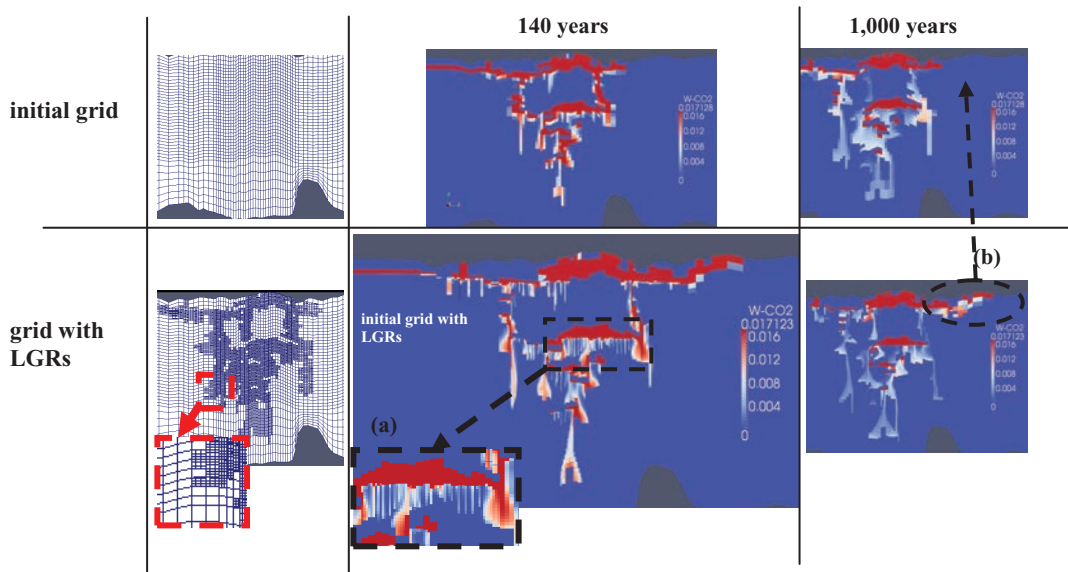


Fig. 3. 2D reactive transport simulations – grid and dissolved CO₂ molar fraction W-CO₂ at 140 and 1,000 years: with or without Local Grid Refinements (LGRs). (a) and (b) details explained in the text.

4. Conclusion

This article reports on reactive transport simulations performed for the Sleipner CO₂ storage. Reactive transport simulations were achieved by coupling the geochemical and fluid flow models in a 2D vertical grid. In this study, we considered different scenarios and paid a lot of attention to better consider the thin intra-reservoir shale layers that play an important role in fluid flows. In practice, the 2D fluid flow model was extracted from a 3D fluid flow model on which we applied an innovative approach based on the use of 4D seismic inversion results to history match the observed CO₂ plume. The first simulation shows a high and unexpected proportion of mineralized CO₂ (22% of injected CO₂ after 1,000 years), this partly results from the assumption made on the reactive surface area (i.e. 100% in that case) but it is also emphasized by the fluid flow effects since the thin intra-reservoir shale layers contribute to intensify fluid flow exchanges. We also looked to other scenarios to characterize the likely CO₂ fate, while considering the uncertainties linked to some aspects of the models and to data. The sensitivity analysis allowed to notice that the uncertainty on the geochemical model in combination with the mineralogical assemblage (chlorite and plagioclase) and to the reactive surface area [0.1%,100%] has a huge impact on the CO₂ mineralization that may range between 0% to 22% of the injected CO₂ mass. The uncertainty of the shale layer permeability also generates a huge uncertainty on the mineralized CO₂ proportion with a range of [17%,42%] in case of shale geochemical model with important mineralization capacity. Less impact is observed when looking to variations associated with uncertainties on: heat transfer model, permeability-porosity law, irreducible water saturation, hysteresis phenomenon and the grid. The resulting mineralized CO₂ mass proportion being between 19% and 25%.

The next step of this work is the 3D simulation of the reactive transport model to take into account formation heterogeneity created to better estimate the different trapping mechanism proportions over time. Following the sensitivity analysis, the key work would be an uncertainty analysis in order to

estimate the occurrence probability of a scenario. This requires to know the probability distribution (probabilistic law) of at least the most influential model parameters, which remains a challenge for numerous ones such as the reactive surface area of minerals. In conclusion, this work gives some general trends but highlights the importance of studying the impact of model uncertainties. It also shows it is necessary to reduce the model uncertainties with inputs from laboratory works, field tests and core measurements (prior and after CO₂ injection) in order to improve data base of geochemical codes and to build models with in-field data comparison.

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