Revisiting the salt dome problem: new insights with the salt dissolution and pollutant release–induced processes

Marwen Ben Refifa [1] · Lamia Guellouz [1] · Edouard Canot [2] · Ezzedine Laabidi [1] · Rachida Bouhlila [1]

Abstract

The salt dome problem is a well-known hydro geochemical challenge for density-dependent flow and transport modeling. The hydrogeologist community considers generally the salt rock as impervious. In reality, and even with its very low permeability, a water flow and salt transport inside the rock could be significant at large geological time scale. We use a coupled model of flow, transport, and geochemistry dealing with high concentrations, to simulate the coupled processes in the HYDROCOIN 4 classical problem to quantify these dissolution processes and their impact on the buried waste transport to the biosphere at large time scales. We quantify the time evolution of the rock porosity of the dome as well as the quantities of salt dissolved. We show that the porosity of the top band of the dome increases by a factor of 4.5 after 10,000 years. For the same duration, the top layer of the dome lost 50% of its salt mass. The amount of salt lost is not constant over time and the process of dissolution stabilizes after 8000 years keeping a constant rate of dissolution of 9.4 kg/year. A pollutant solute that migrates from the center of the dome, where waste burial facilities may have been stored, will reach the surface after 2200 years. We conclude that the salt dome is not impermeable and that the risk of migration of a pollutant through the dome is considerable.

Keywords Porous media · Geochemistry · Numerical simulation · Salt dome · Salt dissolution · Storage accident

Introduction

Salts domes are being considered by the scientific community for the burial of nuclear and other dangerous waste, regarding the very low porosity and permeability of the salt rock. The greatest risk is the escaping of the nuclear particles outside the dome which depends on several factors and parameters. In order to quantify this risk and to better understand the flow and transport processes (which are quite complicated, given the large non-linearity that characterizes their systems of equations), the problem of the salt dome has been developed in the last few decades by many researches. The most known studies are as follows:

– The study of the Mexico Gulf dome (Ranghathan and Jeffery 1988) dealing with the chemical and thermal characteristics of this problem and their influences on the configuration of the concentration and velocity fields.
– Shelkes et al. (1999) studied the risk of nuclear waste break out following fractures in rocks.
– Several studies and works took place under the HYDROCOIN project (Herbert et al. 1988) that began in 1984 and involved more than 20 organizations where 25 computer codes have been tested. These codes results were varied so as to be sometimes contradictory (Diersch and Kolditz 1998).

The salt domes are generally fully formed of the solid crystal form of the NaCl salt (Hart et al. 1981). In reality, the rock forming the salt dome is not totally impervious. Up to our knowledge, to this date, there are no exact measurements for

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✉ Marwen Ben Refifa
marwen_ben_refifa@yahoo.fr

Lamia Guellouz
lamiaguellouz@yahoo.fr

Edouard Canot
edouard.canot@univ-rennes1.fr

Ezzedine Laabidi
laabidimhe@yahoo.fr

Rachida Bouhlila
rjbouhlila@yahoo.fr

[1] LMHE, ENIT, Université Tunis EL Manar, Tunis, Tunisie

[2] IPR, UMR 6251, CNRS, Université de Rennes, Rennes, France

the evolution of the permeability within mining salt configurations. In situ measurements performed on salt drilling show that the permeability of the surface is 5 to 6 order of magnitude greater than that of the intact mass located away from the surface (Stormont et al. 1991). The permeability of this rock ranges from 10^{-15} and $10^{-21}m^2$ (Peach 1991; Stormont et al. 1991). Even if a permeability of $10^{-21}m^2$ is considered by hydrogeologists as impervious, this value over a period of time which can vary from few hundreds to thousands of years, depending on applied pressure, allows liquids to pass through the salt rock pores (Ghoryechi and Daupley 2003). On the other hand, rock salt is very soluble in water, up to 360 g/L (Haynes 2014), and on a very long time scale, the enhance of its permeability can lead to a significant leakage of the buried dangerous waste into the biosphere if the sarcophagus comes to give way. Therefore, and in view of the responsibilities we have towards future generations, for a better assessment of the security of the dangerous waste burial facilities, it is important to quantify, at rather geological time scales, the flow and the transport through these low permeability but highly soluble, media.

The problem test used by all these authors is that described by Herbert et al. (1988). It considers the salt rock as impermeable. In order to better understand the velocity field configurations, the salt concentration fields, and their possible evolution over time, a special attention should be paid to the top of the salt dome. Based on this domain configuration, several sensitivity studies are carried out such as that of (Younes et al. 1998; Bouhlila 1999) which analyzed the influence of dispersion and molecular diffusion on the occurrence of recirculation regions. Other studies were interested by the performance of the coupling methods of the flow and transport and by the methods used for spatial and time discretization (Freedman and Ibaraki 2002; Steefel and Yabusaki 1996). Among the numerical codes used in these studies, we can mention: SWIFT (Reeves et al. 1986) which is based on a finite difference method, NAMMU (Atkinston et al. 1985) and FEFLOW (Diersch and Kolditz 1998) which are based on finite element method and TVDV (Younes et al. 1998) which uses a mixed hybrid elements method.

With an objective of continuity and enrichment of the debate about the salt dome problem, a revisited problem dome is proposed by considering the long-term dissolution and its impact on the release of the buried substances in the biosphere. Comparing to the salt dome problem described by Herbert et al. (1988), the depth of the modeled domain is extended in order to include the upper part of the dome on the top of the waste burial site. Density-dependent flow and coupled reactive transport are simulated into this extended domain, with the known top and lateral boundary conditions but with salt dissolution in the bottom part of the domain representing the dome. The advantage of this configuration is that it will allow us to study the evolution of the porosity and the permeability

of the rock over several tens of thousands of years. The variation in porosity can not only affect the salt transport, but also modify the transfer of a contaminant, initially stored in the dome rock and can thus increase the risk that this pollutant reaches the surface of the soil.

Referring to our former study (Ben Refifa et al. 2016), simulating the extended domain allows the dissolution process on the top of the dome. This result showed that the dissolution of the dome cannot be neglected.

The novelty of this work is the development and the extension of the previous work by taking into consideration different laws of permeability, viscosity, and porosity as well as a deterministic model of the solution density. The new results are the estimation of the mass balance, the dissolution rate of salt, and the period of dissolution of the entire dome. In addition, we simulate a radioactive storage accident within the salt dome.

The majority of the works presented in the literature (Herbert et al. 1988; Younes et al. 1998) modeled the salt dome by using a traditional density–dependent model and does not consider the effect of the dissolution process. All the cited work shows that a stationary regime is reached after about 1000 years. In this work, we consider the effect of the dissolution reaction and the effect of this latter on the porosity variation we find out that the dissolution process is perpetual and no stationary regime is reached.

In this work, the simulations are performed using the GEODENS code (Bouhlila 1999). The assembling and resolution methods of this code have been recently improved (Laabidi and Bouhlila 2015; Ben Refifa et al. 2016). The first part of this work deals with the fact of considering the dissolution of the salt dome. In the second part, a description of the model and the computing strategy are presented. Finally, simulations results of a radioactive storage accident are presented.

The dissolution of the salt dome

The mineralogy of the salt dome is quite simple, except that its formation process as well as its geological age which may exceed 600 million years (Fries 1987; Evans 2003; Michelsen 2003) generated a quite complicated internal structure. In general, the dome rock is mostly composed of Halite (NaCl) (90%) in addition to some anhydrite (CaSO4), silica, and carbonates (Hart et al. 1981). The coexistence of rock salt (solid NaCl) and the hydrological circulation of groundwater call into question the phenomenon of dissolution. Evidence of the existence of the dissolution has been the subject of several investigations such as that of the Hainesville dome (USA) where Jackson and Seni (1983) proved the existence of salt loss of the order of 160 km^3. An amount of dissolved salt was detected at the Palestine dome site in Texas by the U.S. Nuclear regulatory Commission in 1981 (Hart et al. 1981).

Kloppmann et al. (2001) also proved that on the site of Permian salt dome in the North of Germany, the phenomenon of dissolution does exist.

The phenomenon of dome dissolution is therefore highlighted but it is very complex. Indeed, the domes that have been studied as well as the others are still existing and do not change in shape or size. The research carried out leads to two hypotheses in order to explain this contradiction: the first is that the phenomenon of dissolution took place in the past and then it stopped. The second is the existence of a barrier that protects the dome, which is its impermeability (Hart et al. 1981). The first hypothesis is immediately rejected since the quantity of dissolved salt continues to increase (Larsson 1992). The second hypothesis was adopted by the vast majority of the scientific community: the dome is considered impermeable and its porosity is zero. Therefore, there is no flow or transport in the salt rock, which has fixed properties over time. We carried out previous works within this hypothesis (Bouhlila 1999; Ben Refifa et al. 2016). However, in the present work, we are interested in exploring the flow and transport in the dome rock, and in emphasizing on their influences on the future of the dangerous waste storage.

In the ideal case, the dome is entirely formed by rock salt, and this rock is characterized by a permeability which varies from 10^{-15} to 10^{-21} m^2 (Peach 1991; Stormont et al. 1991). The values of permeability of domes measured in situ, range from $9.8 \ 10^{-20}$ to 1.10^{-17} m^2. Determined in laboratory, these values, for the same samples, can reach 10^{-12} m^2 (Kelsall and Nelson 1985). Porosity measurements were performed at four different sites in the USA and Canada, and the values vary between 0.62 and 7.17% (Aufricht 1961). These measurements show that the porosity of the dome is not zero and that the permeability, for the weakest case, is around 10^{-19} m^2. These values, although quite low, allow the circulation of water in the rocks (Ghoryechi and Daupley 2003) that could lead to a significant mass transfer at a geological time scale. Indeed, we can consider salt domes not impervious and admit small water circulation, which can change the physical characteristics, porosity, and permeability, as well as the general shape of the salt dome.

Physical and mathematical models

The problem is inspired from an actual geological configuration in the north of Germany (OECD, f 1988). The salt dome is located 300 m below the surface. A circulation of fresh water takes place above it. The water dissolves the salt and moves it downstream (Fig. 1). The physical phenomena occurring are essentially water flow coupled with solute transport in the porous medium. Since the salt dissolution is important, the solute concentration increases so that the density-driven flow cannot be ignored.

To sum up, the physical model deals with density-driven water flow coupled with solute transport in porous media, taking into account salt dissolution or precipitation. The flow equation is given in the (Eq. 1) (Bouhlila 1999):

$$\frac{\partial}{\partial x_i}(q_i) = \frac{\partial(\rho\Phi)}{\partial t} \qquad (1)$$

With

$$q_i = u_i\Phi = -\frac{k_{ij}}{\mu}\left(\frac{\partial p}{\partial x_j} - \rho g_i\right) \qquad (2)$$

with k_{ij} the intrinsic permeability [m^2] which depends on Φ, p [kg m^{-1} s^{-2}] the local fluid pressure, μ [kg m^{-1} s^{-1}] the viscosity, ρ [kg m^{-3}] the fluid density, g_i [m s^{-2}] the gravity in the i direction, Φ the porosity ($0 \leq \Phi \leq 1$), u_i [m s^{-1}] the effective velocity in the porous media, q [m s^{-1}] is Darcy's velocity, and i and j are space axes. The solute conservation equation determined by its mass fraction C_i is:

$$L(c_i) + Qc_i + \Phi f_{gi} = \frac{\partial(\rho\Phi c_i)}{\partial t} \qquad (3)$$

With L an operator that encompasses the processes of convection, dispersion, and diffusion (Eq. 3). Q [mt^{-1} l^{-3}] is the water mass flow per unit volume of porous media, which represents the external inputs or source. f_{gi} [mt^{-1} l^{-3}] is the flux of species i due to the different geochemical reactions that concern this species per unit volume of porous medium. It should be noted that this writing of the mass balance induces, in particular, that the possible geochemical reactions are uniformly distributed in the elementary volume representative of the porous medium.

These equations are closely coupled since the density driven term (calculation of ρ, the fluid density) depends on the solute concentration C, and on the other hand, the velocities (u_i) appearing in the solute conservation equation (Eq. 4) are those calculated in equation (Eq. 2).

$$L(C_i) = \frac{\partial}{\partial x_i}\left\{\rho\Phi D_{ij}\frac{\partial C_i}{\partial x_j} - \rho\Phi u_i C_i\right\} \qquad (4)$$
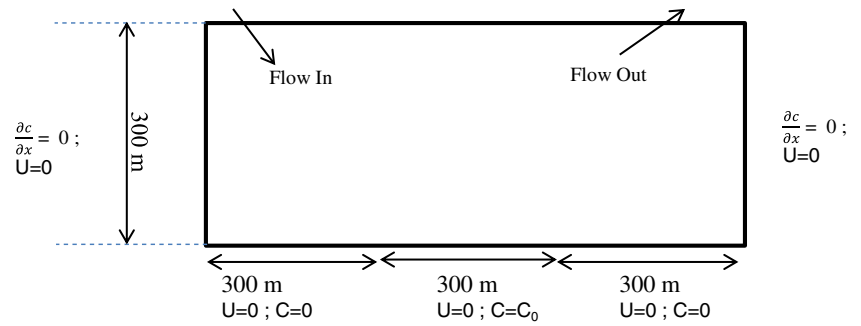
With D_{ij} the diffusion-dispersion term, it is expressed as follows:

$$D_{ij} = (d + \alpha_L u)\delta_{ij} + (\alpha_L - \alpha_T)\frac{u_i - u_j}{u} \qquad (5)$$

With d [m^2 s^{-1}] the molecular diffusion, α_L [m] and α_T [m] are the longitudinal and transverse dispersion, respectively, δ_{ij} is the Kronecker symbol, and C_i is the mass fraction of the chemical species i.

The calculation of the geochemical flow f_{gi} goes through the characterization of the solution in each point of the field of study and at every moment. It is the sum of the contributions

Fig. 1 Geometry and boundary conditions of salt dome problem

Flow In   Flow Out

300 m

$\frac{\partial c}{\partial x} = 0$;
U=0

$\frac{\partial c}{\partial x} = 0$;
U=0

300 m
U=0 ; C=0

300 m
U=0 ; C=C_0

300 m
U=0 ; C=0

of all the reactions that can take place locally (Bouhlila 1999). If Nr is the number of these reactions and if S_i is the concentration of the chemical species i it comes:

$$f_{gi} = \sum_{r=1}^{Nr} \rho \left(\frac{ds_i}{dt} \right)_r \tag{6}$$

With S_i the concentration of the chemical species i.

The calculation of this term is carried out with the values of the concentrations in the preceding time step, thus according to an explicit procedure. The geochemical flow of species i to node I is then:

$$\Phi F_{gi}^I = \rho \Phi \frac{c_i^I(t) - c_i^I(t-\Delta t)}{\Delta t} \tag{7}$$

With c_i^I the corresponding equilibrium concentration.

Computing strategy

The code GEODENS is a numerical model designed for the resolution of density coupled flow and transport in porous media. Built on a finite element method with Galerkin approximation, the reference elements are six-node-based triangles. This choice originates from the weighted residuals method developed by (Bouhlila 1999) for the calculation of the terms of gravity in Darcy's law. A Semi-implicit Euler scheme is used for the time discretization. Temporal first derivatives which appear in the system of equations are approximated at time $(t + b \Delta t)$ by finite difference expressions.

$$X_{t+\Delta t} = X_t + \Delta t X'_{t+b\Delta t} \tag{8}$$

With X_t and $X_{t+\Delta t}$ respectively the values of the unknown variable X (in our case pressure or concentration) at the instant t and $(t + \Delta t)$. b varies within 0 and 1. X' is the time derivative of X.

GEODENS simulates the evolution in time of salt-brine systems. It calculates, over time, the concentrations of ions in the solution, the density of the brine, and amounts of salts, precipitated or dissolved, in contact with the solution. These calculations are based on the determination of the water and aqueous species activities presents in solution. This code uses the Pitzer model of the ions interaction (Monnin 1989) that best fits with the brines problem. Six different aqueous species are currently considered: Na^+, K^+, Ca^{++}, Mg^{++}, Cl^-, and $SO4^=$. These elements represent the major species in the class of treated problems. In the conceptualization of the Pitzer model, these species are not associated in the solution. Therefore, there is no reaction in the aqueous phase to be taken into consideration: the only phenomenon to be considered will be the precipitation or dissolution of minerals. GEODENS coupled the flow and transport at each node using density.

It calculates the density by taking into account the existence of solute and/or solid salt. Once the geochemical process detects the existence of one or more precipitated and/or dissolved salts, it updates the values of the appropriate concentrations and returns the precipitated and/or dissolved amounts to the flow and transport processes to use it to update the values of the physical parameters (porosity, permeability...).The amount of solid salt in the pores varies continuously due to the dissolution and precipitation reactions. At each time step,

Table 1 Physical parameters of the salt dome problem (Herbert et al. 1988)

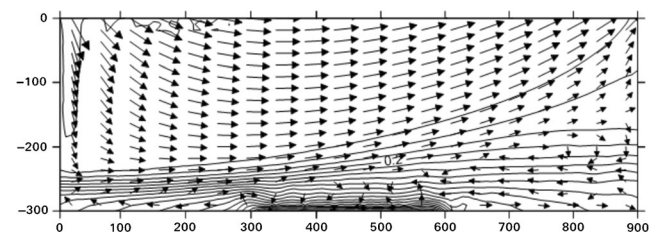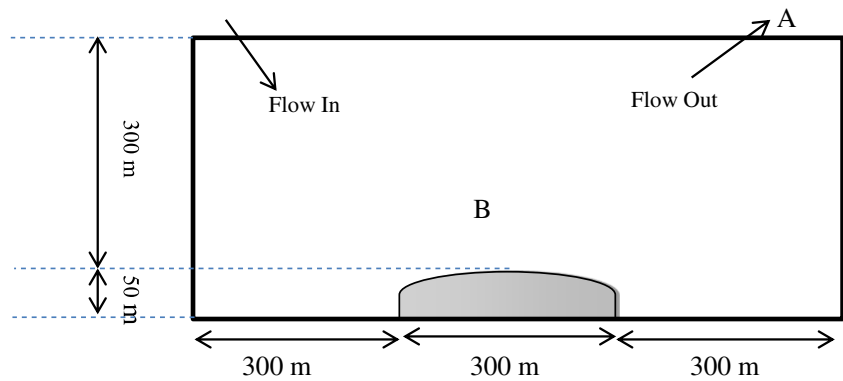|	
Permeability	$K = 1.10^{-12}$ m^2
Porosity	$\Phi = 0.2$
Longitudinal dispersion	$\alpha_L = 20$ m
Transversal dispersion	$\alpha_T = 2$ m
Diffusion	$D = 2.78\ 10^{-9}$ m^2/s
Brine density	$\rho_s = 1200$ kg/m^3
Fresh water density	$\rho_0 = 1000$ kg/m^3

Fig. 2 Concentration and velocity fields above the salt dome ($T = 1000$ year).

Fig. 3 Adapted geometry of the salt dome.

the amount of salt dissolved or precipitated is quantified, and porosity and permeability are calculated at each node of the mesh (Ben Refifa et al. 2017). This new approach has been tested and validated through simple configurations and analytical solutions (Ben Refifa et al. 2016).

Simulating the classic dome problem

In order to compare the results of the dissolving dome to those of the classic problem, we present in the current section the simulations of the dome problem without dissolution. The computing domain is a vertical section of soil, of 300 m high and 900 m wide (Fig. 1). The boundary conditions are as presented by (Herbert et al. 1988). On the upper face of the domain, a zero concentration ($C = 0$) on the section ($0 \leq x \leq 200$) is imposed for the transport, and the remainder of the surface is characterized by a Neumann condition (where $\partial c / \partial z = 0$). For the flow, a pressure condition which decreases linearly, from 10^5 in the left, to 0 Pa in the right, is imposed. On the bottom side, the concentration C_0 (saturation value) is imposed different from zero only for x ranging in [300,600] m. The physical parameters are given in Table 1.

The simulations are performed using a six nodes triangular mesh with a total number of 1681 nodes. The steady state is attained when convergence is reached, i.e., when the evolution of the relative norm of the vector of unknowns $\frac{\|\Delta X\|}{\|X\|}$ (with ΔX is the difference between X at time step "N" and X at time step $N-1$)

for t_{n+1} is less than 10^{-9}. In order to perform appropriate comparison with the previous work, we conduct a calculation over 1000 years. In our case, the steady state is reached at 600 years which agrees with a time found by Younes et al. (1998).

The results (Fig. 2) perfectly match the work of Herbert et al. (1988) presented in the final report (OECD, f 1988). These results prove the existence of a large recirculation in the bottom of the field; however, we notice the existence of a small difference in the final distribution of the concentration lines. This difference is explained by the behavior of the numerical methods used in the various codes which have addressed this problem (Konikow et al. 1997).

New approach: simulating the dome dissolution

The salt dome is a hydrogeological problem characterized by a duration that can exceed millions of years. Our previous work, which adopts a zero permeability of the dome, was limited to simulation times of the order of 1000 years. Indeed, the steady state is established after a similar duration (Younes et al. 1998). In order to study the phenomenon of dissolution of the salt dome, we are interested in much longer simulation times. We are interested in highlighting this problem by studying the variations of porosity and permeability within the dome. The salt dome rock is composed mainly by sodium chloride which is characterized by a molar mass of 58.44 g/mol
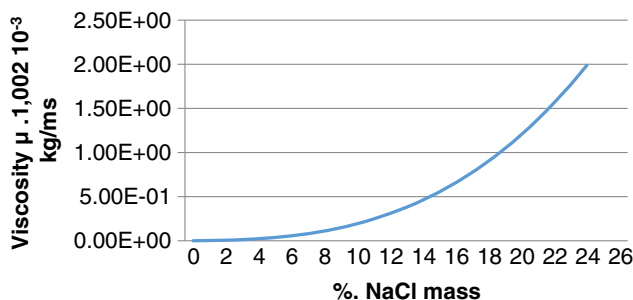

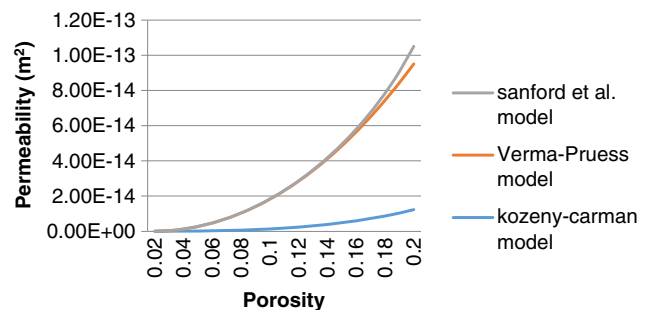**Fig. 4** Dynamic viscosity (μ) of NaCL solutions

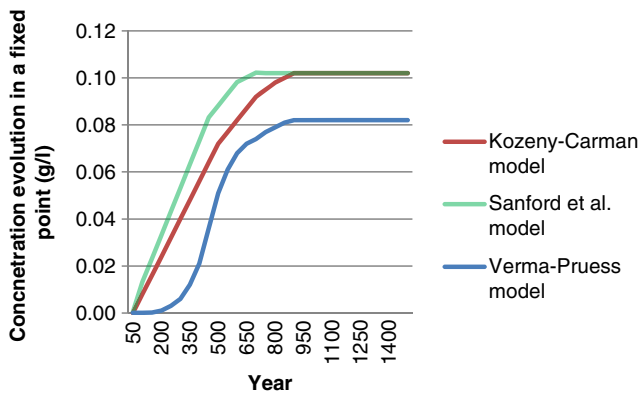**Fig. 5** Permeability variation with porosity

Fig. 6 Evolution of the concentration according to the law of permeability

and its solubility in water is about 359 g/L (Hart et al. 1981). The equilibrium reaction of the sodium chloride is:

$$Na^+ + Cl^- \rightarrow NaCl \qquad (9)$$

Salt dome permeability ranges from 10^{-15} and $10^{-21}[m^2]$. The idea is to expand the computational domain to include a portion of the dome (Fig. 3). We have expanded the computational domain high by 50 m. New problem dimensions are (350 m × 900 m). The physical parameters of the aquifer domain are the same as those used in the original problem (Table 1). We suppose that the salt dome domain is a porous medium characterized by porosity equals to 0.2 and that the dome is composed by 90% of solid salt (NaCl) and 10% of a solid matrix. The effective porosity of the rock is equal to 0.02. Its initial permeability is 10^{-17} [m²].

Parameters evolution laws

Viscosity

The adopted viscosity is that of water with different salt concentrations. The viscosity of the NaCl solutions at mass

fractions ranging from 0 to 26% is given in the Handbook of Chemistry and Physics (1972–1973) (see Fig. 4). Adjusting a law to these values yields a polynomial of degree 3, depending on the mass fraction normalized by the maximum mass fraction of 0.26 (C therefore varies from 0 to 1) (Herbert et al. 1988).

$$\mu(c) = \mu_0\left(1 + 0.4819C - 0.2774C^2 + 0.7814C^3\right) \qquad (10)$$

With μ_0 the initial viscosity and C is the mass fraction normalized by the maximum mass fraction.

Density

The density of the mixture ρ is the coupling element between the different code processes. Dome problems are generally characterized by high concentrations with the existence of salts precipitates. The calculation of the density is no longer a simple linear relationship, applicable only to low concentrations. For that purpose, this variable is calculated using a geochemical process as mentioned in the section computing strategy.

Porosity

The porosity is following a law of variation, depending on the existence of the precipitation/dissolution reactions, according to the mass conservation of different species. The variation law of porosity ϕ between the time (t) and ($t+\Delta t$) for each element "e" of the mesh is given by:

$$\Phi(t + \Delta t) = \Phi(t) + d\Phi \quad \text{with} \quad d\Phi = dN\frac{V_m}{V_e} \qquad (11)$$

With dN the difference in salt mole number precipitated ($-$) or dissolved ($+$), V_m is the molar volume of the salt, and V_e is the volume of the mesh element.
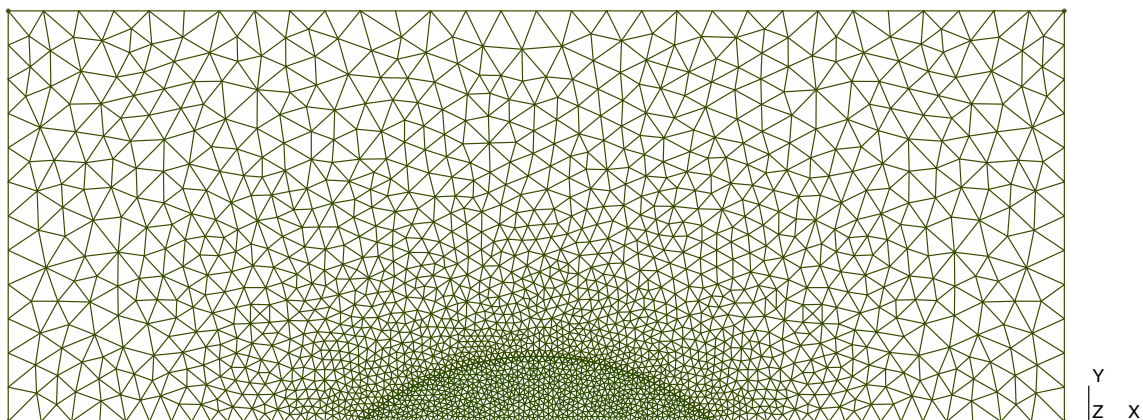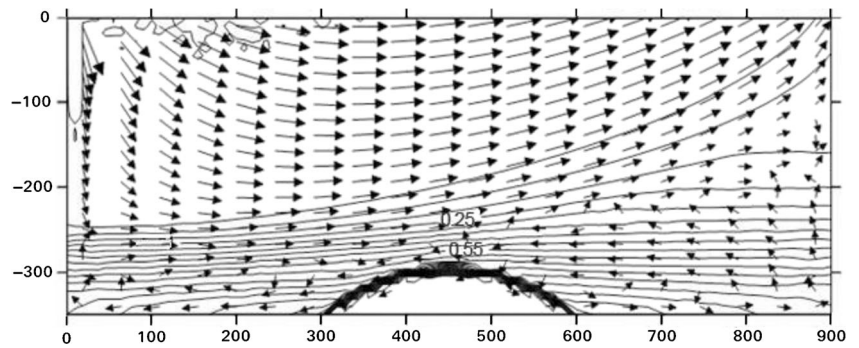

Fig. 7 Adopted mesh for the salt dome problem: 2136 elements

Fig. 8 Velocity and concentration fields for a simulation of 1000 years

Permeability

The relationship between porosity and permeability is complex in natural environments. The variation of the permeability is neither automatically in the same direction nor proportional to the volumes of precipitated or dissolved minerals. We are not able to study the distribution of porous spaces because it requires a laborious experimental work. On the other hand, we are interested in the variation of the law of permeability as a function of porosity. Capillary models or plane cracks models allow for unambiguous relationships between porosity and permeability. We particularly study the well-known capillary model of Koseny-Carman and compare results with those given by Verma-Pruess model and a logarithmic relationship between permeability and porosity.

– Koseny-Carman model expresses the intrinsic permeability k [L^2] depending on the porosity Φ:

$$\frac{k}{k_0} = \left(\frac{\Phi}{\Phi_0}\right)^3 \left(\frac{1-\Phi_0}{1-\Phi}\right)^2 \quad (12)$$

With Φ_0: initial porosity and k_0: initial permeability.

– Verma and Pruess (Verma and Pruess 1988) developed a permeability-porosity relationship from a porous body in which the permeability can be reduced to zero with a remaining critical porosity.

$$\frac{k}{k_0} = \left(\frac{\Phi-\Phi_c}{\Phi_0-\Phi_c}\right)^2 \quad (13)$$

With $\Phi_c = 0.9\ \Phi_0$ is a critical porosity under which the permeability reaches zero.

– Logarithmic relationship for k in [m^2] and Φ (Stanford and Konikow 1989), consisting of:

$$\log(k) = a\Phi - b \quad (14)$$

with a: constant of the order of 20. The constant b is determined from the initial values of k and Φ measured in the field. Either for rock salt:

$$\log(k) = 20\Phi - 19 \quad (15)$$

The evolutions of the permeability with respect to these three laws are given on Fig. 5. We notice that the permeability follows the same behavior for the Verma-Pruess and Stanford models, but its evolution is less important for Kozeny-Carman's law. Therefore, the permeability-porosity relationship can affect the concentration field of the studied problems.

In order to study the influence of the law of permeability on the concentration of the salt dome problem, we compared the concentration (in terms of percentage with respect to the initial concentration) at the outlet of the domain to the coordinates (00.00; 850.00), point "A" of Fig. 3 for the three laws (Fig. 6). The concentration reaches the same final value for the two
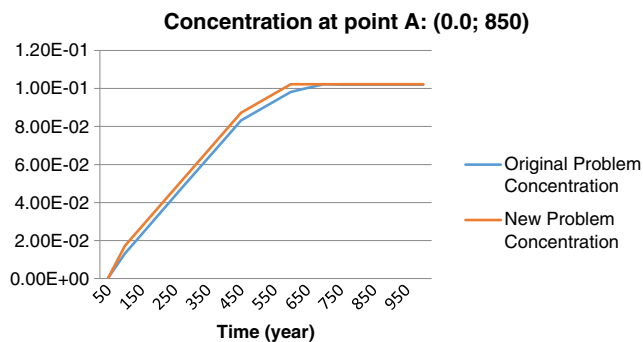

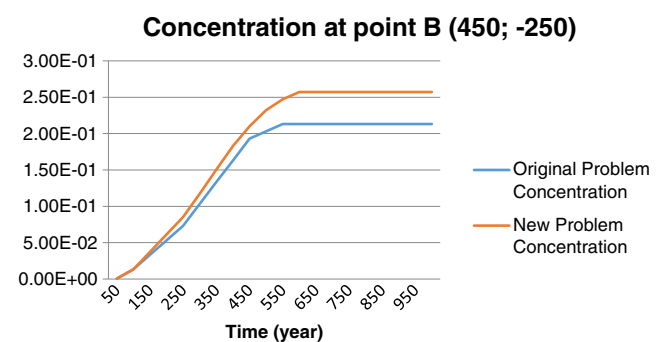**Fig. 9** Evolution of the concentration at point A: $x = 0$; $y = 850$

Fig. 10 Evolution of the concentration at point B: $x = 450$; $y = -250$
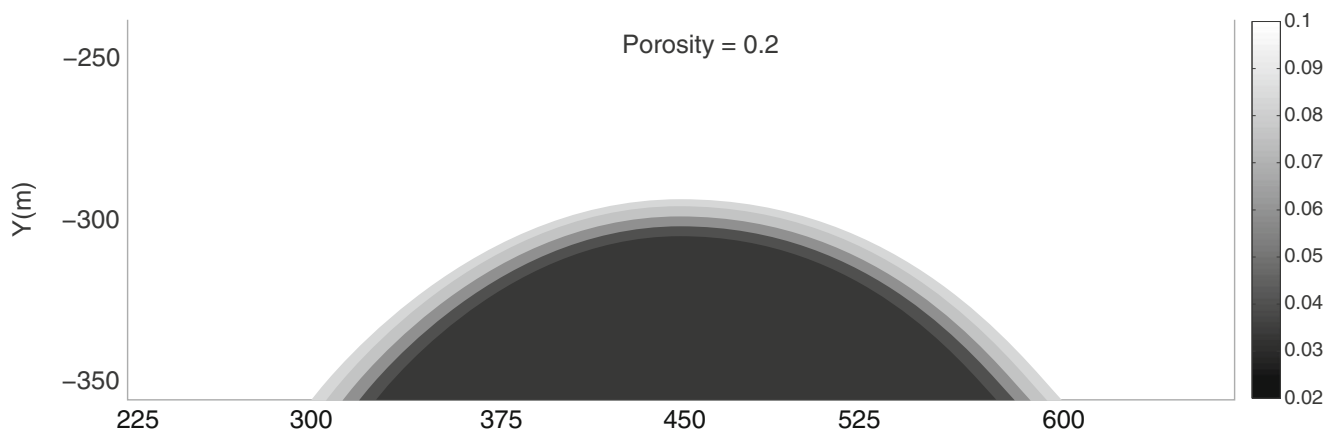
Fig. 11 Porosity fields for a simulation of 10,000 years

laws of Kozeny-Carman and Stanford (0.1 or 10%). The evolution of the concentration for the third law is less intense; indeed, it reaches a maximum value of 8% of the initial concentration. The steady state (in term of evolution of concentrations) reaches more rapidly with the law of Stanford et al. (650 years) than with those of Kozeny-Carman and Verma-Pruess (800 years).

To study the dissolution of the dome, we would better choose the law that represents the worst-case scenario in terms of natural risk. It is the law that favors the evolution of porosity and permeability in the dome. So we choose Stanford's law (Eq. 11).

Computing results

This section deals with the study of the salt dome threatened by dissolution. The computing domain is presented in Fig. 3. The adopted mesh consists of 2136 elements and 4391 nodes (Fig. 7). We adopt a refined mesh for the dome in order to minimize the numerical instabilities generated by the strong contrast of the physical parameters between the dome and the rest of the domain. The physical parameters are the same used for the original problem. To compare the results of this problem with those of the original one, we conducted a simulation over a period of 1000 years (Fig. 8).

We present first some previous results; the velocity field is identical to that of the original problem in terms of norm. We notice very low velocities at the top layer of the dome (1.10^{-10} m/s). The configuration of the concentration field is similar to that of the original problem. Recirculating cells exist on both sides of the salt dome. These cells are larger than those found in the original problem. Indeed, previous results of salt dome are seen as a representation of a horizontal sectional view along the dimension $y = -300$ m (Ben Refifa et al. 2016, 2017).

In a second place, we compare the concentration fields of the adapted problem with those of the original problem. To do

this, we compare the evolution of the concentrations during the simulations for the following points:

PointA : $x = 0; y = 850$ PointB : $x = 450; y = -250$

The concentration at the outlet of the domain (point A) follows the same behavior for both configurations and reaches the same maximum value which is equal to 0.1 (10%) of the maximum concentration (Fig. 9). This result is in perfect agreement with the results mentioned in the final report of the HYDROCOIN project, where the scientific community has stated that the final concentration, at steady state at the outlet, is equal to 10% of the initial concentration.

The concentration at the second point (point B: 50 m above the top of the dome) is higher for the new problem (Fig. 10). Indeed, it rose to 0.25 after a period of 600 years against 0.21 for that of the original problem. This increase is explained by the influence of the existence of the dome, a large amount of salt, especially that we use a chemical code for the calculation of the concentration and the density, unlike the previous works which use a linear law for the density calculation.

Porosity and mass balance

Because of the low permeability of the dome rock, fresh water dissolves its surface. The porosity of the upper layer elements of the dome has increased from 0.02 to 0.04 for a simulation of 1000 years. By considering a domain thickness of about 1 m, the amount of dissolved salt is of the order of $1.2\ 10^5$ kg including 400 kg which circulates in the computational domain.

The objective is to quantify the evolution of the salt dome porosity. For this, we have extended our simulations over a time of 10,000 years. The porosity of the upper layer of the rock has reached 0.09 (Fig. 11). It has increased by a factor of 4.5. The dissolved salt (lost) amount is of the order of $3\ 10^5$ kg which represents 50% of the amount of initially existing salt

Fig. 12 Evolution of the amount of dissolved salt in the upper band of the rock dome: **a** quantity of global dissolved salt; **b** quantity of dissolved salt per year
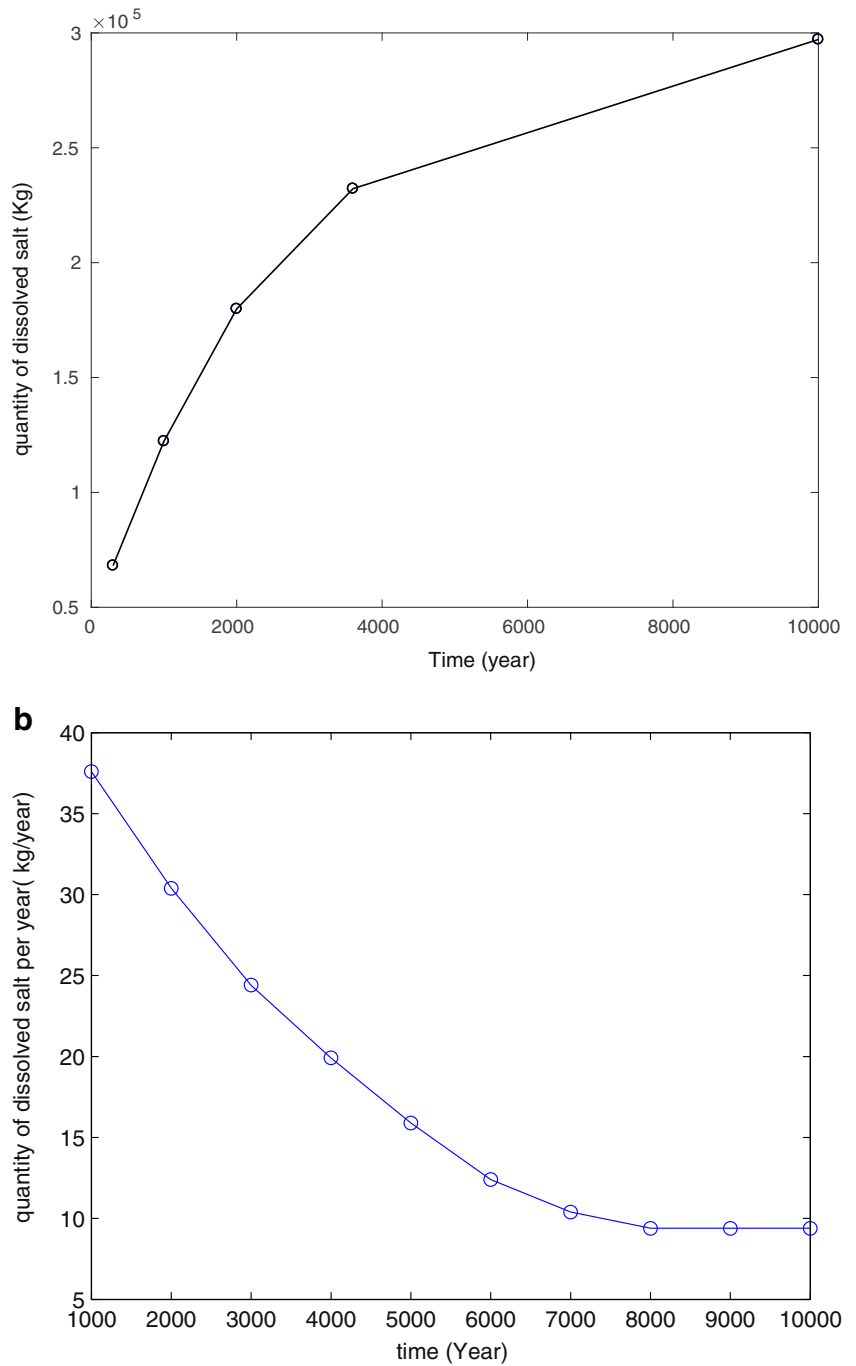
on this upper layer of dome. The porosity of the elements in the second range, increased slowly until reaching a factor of 2.

Fresh or unsaturated water (in salt) dissolves the upper layer of rock salt. The pores of this first layer are gradually released from existing salt and the permeability, therefore,

Table 2 Physical parameters of the proposed problem

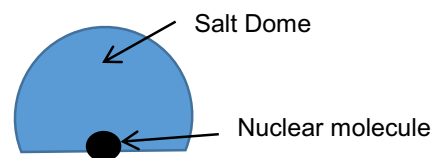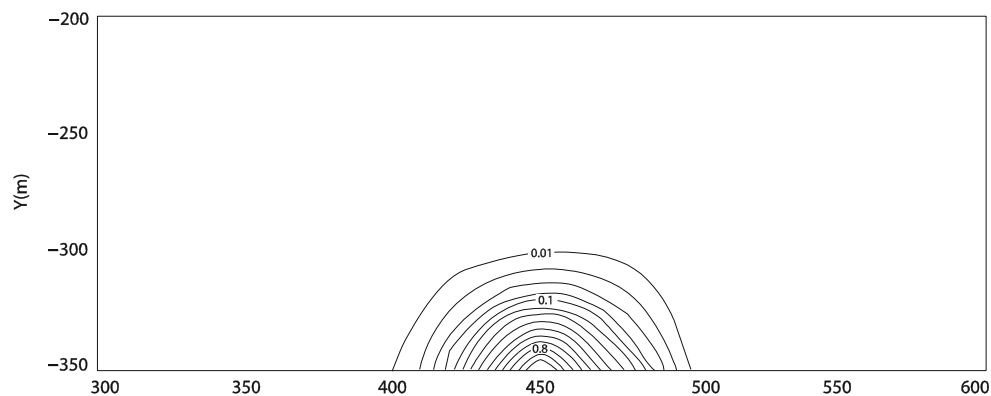|	
Permeability	$K = 1.10^{-17}$ m^2
Porosity	$\Phi = 0.02$
Longitudinal dispersivity	$\alpha_L = 0$ m
Transversal dispersivity	$\alpha_T = 0$ m
Diffusion	$D = 2.78 \ 10^{-9}$ m^2/s

Fig. 13 Proposed scenario

Fig. 14 Diffusion of the tracer in the dome at 2000 years

increases. These two factors promote circulation within the dome and the water tends to penetrate more and more into the pores of the lower layers. The amount of salt lost is not constant over time (Fig. 12). The dissolution process is much faster for the first 1000 years. After the establishment of the steady state, the water circulating in the area is more concentrated and the dissolution process becomes slower. This process of dissolution stabilizes after 8000 years keeping a constant rate of dissolution of 9.4 kg/year.

The amount of the dissolved salt percentage, in the top surface of the dome (8 m thick), is about 50% of the initial amount. By comparing the total stock of dissolved salt, the dissolution process has the same behavior and the same speed for different grid meshes (Ben Refifa et al. 2016). The quantity of salt dissolved remains the same per meter of depth irrespective of the adopted mesh. We estimate a complete disappearance of the dome top layer after 42,000 years. This layer can be seen as a band of 8 m in depth.

Simulating a case of radioactive storage accident

Deep geological disposal is assumed to be the reference solution for the definitive management of medium and high-level long-lived waste (IRSN Institute of Radiation Protection and Nuclear Safety). The Waste Insulation Pilot Plant (WIPP-USA) has been receiving, since 1999, military waste containing long-lived radioactive elements (transuranic) in facilities dug in a layer of salt. Morsleben (Germany), a former salt mine, was used as a radioactive waste storage facility until 1998.

The salt dome is considered to be one of the most suitable formations that are retained for radioactive storage, thanks to the unique properties of this material (Djahanguiri and Matthews 1985), which contribute to its containment capacity. Rock salt has several advantages that favor the storage of radioactive waste such as its low permeability, low porosity, and relatively simple geological structure (Massal 1988).

Despite the advantages of salt rock for this type of storage, the chance of escape of radioactive molecules is not zero. That is why this subject has marked the center of the various research projects over the last two decades (Bouc and Auclair 2007). We are interested in this section in an escape accident scenario of a high radioactivity molecule initially placed at the bottom of the dome (Fig. 13).

The problem to be studied, therefore, consists in following the evolution of the radioactive molecules within the dome. These molecules have very high radioactivity, therefore life of the order of thousands to millions of years. In order to compute the worst case, we do not take into account the degradation and we consider the nuclear molecules as a tracer moving under the effect of molecular diffusion. The physical parameters are presented in the Table 2.

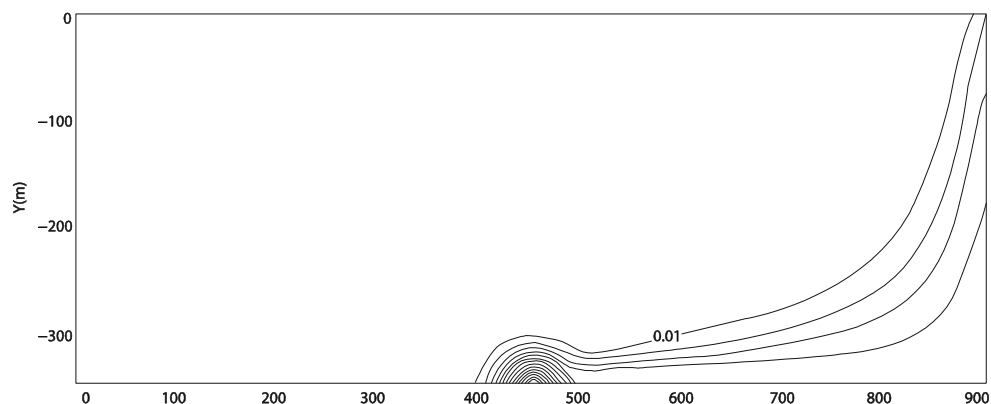**Fig. 15** Diffusion of the tracer in the dome at 2112 years

Table 3 Tracer migration time in the dome

Diffusion D (m^2/s)	$2.78 \ 10^{-9}$	$1.39 \ 10^{-9}$	$1 \ 10^{-10}$	$1 \ 10^{-11}$
Numerically calculated time (year)	2106	4212	58,110	581,100
Analytically calculated time (year)	2200	4400	61,168	611,686

The tracer diffuses in a regular way within the dome. Under the effect of pure diffusion, the molecule will reach the surface of the dome and leave the domain (Figs. 14 and 15). For a diffusion of 2.78 10^{-9} m^2/s, the calculated time, needed for the tracer to reach the surface of the dome, is about 2100 years. This value has been confirmed by an analytical calculation by solving the pure diffusion equation.

The low permeability and porosity of the salt dome on one hand, the difference in size between the diameter of the dome and the molecule on the other hand are arguments that can justify the choice of the analytical solution of pure diffusion with constant diffusion coefficient, in a semi-infinite domain. The solution for a semi-infinite domain, under the initial condition $C = C_i$ in the whole domain and the boundary condition $C = C_0$ at the end of the domain is (Philibert 1985):

$$\frac{C_0 - C}{C_0 - C_i} = \text{erf}\left(\frac{|x|}{2\sqrt{Dt}}\right) \tag{14}$$

With

x The length of the domain (Dome radius)
D the molecular diffusion coefficient
t the time

The analytically calculated time is equal to 2200 years, which is in perfect match with the numerical result. This time depends essentially on the diffusion coefficient.

In Table 3, we compare the computed and calculated times for different values of diffusion. The migration time can reach 600,000 year for a molecular diffusion of 10^{-11} m^2/s.

Conclusion

The salt dome problem is one of the most interesting hydrogeological applications regarding its use for the burial of nuclear or dangerous waste. According to our simulations results, the salt rock that forms the dome cannot be considered as totally safe for long time scale. The porosity of the top band of the dome has increased by a factor of 4.5 after a simulation of 10,000 years. For the same duration, the top layer of the dome lost 50% of its salt mass. The dissolution process is quite slow but we proved that it cannot be neglected. The diffusive movement, inside the dome, will increase with the increase of the permeability of the upper layer. This process will speed up the salt dissolving process in the higher layers of the dome and enhance the transport processes of the buried dangerous products in case of leak. The simulated scenario of the storage accident estimates that the nuclear molecule can escape the dome within 2200 years. This estimation is based only on the diffusion process and does not take into account water circulation in the dissolving dome due to increase in permeability.

References

Atkinston K, Herbert AW, Jackson CP & Robinson PC (1985) NAMMU user guide,. United Kingdom: AERE-R–11364; DOE-RW–85.065; ISBN 0-7058-1421-1; Worldcat; CONTRACT PECD-7/9/124-35/83; H.M. Stationery Office, London

Aufricht WR (1961) Salt characteristics as they affect storage of hydro-carbons. Soc Petrol Eng 13(08):733–738. https://doi.org/10.2118/23-PA

Ben Refifa M, Canot E, Guellouz L, Bouhlila R (2016) Modeling of coupled processes: flow, transport and geochemistry, application to the dissolution of salt dome. J Hydrogeol Hydrol Eng 5(4). https://doi.org/10.4172/2325-9647.1000143

Ben Refifa M, Canot E, Guellouz L & Bouhlila R (2017) Salt dome dissolution: Coupling flow, transport and geochemistry. Tendances dans les ApplicationsMathématiques en Tunisie, Algérie et Maroc, (pp 475–480). Hammamet

Bouc O & Auclair S (2007) Critéres de securité pour les analogues industriel au stockage géologique du CO2. 3 ill,2 ann: BRGM/RP-55840 –FR

Bouhlila R (1999) Ecoulements, Transports et Réactions Géochimiques Couplés Dans le milieu Poreux. Ecole National d'Ingenieur de Tuins, Tunisie

Diersch HJ, Kolditz O (1998) Coupled groundwater flow and transport: 2.Thermoline and 3D convection systems. Adv Water Resour 21: 401–425

Djahanguiri F & Matthews S (1985) Geotechnical considerations for design of a nuclear repository in bedded salt in the U.S. In Sixth International Symposium on Salt. Alexandria Va: the Salt Institute, pp 561–584

Evans D (2003) The millennium atlas: petroleum geology of the central and northern North Sea. Geological Society of London, London

Freedman V, Ibaraki M (2002) Effects of chemical reactions onon density-dependent fluid flow: on the numerical formulation and the development of instabilities. Adv Water Resour 25(4):439–453. https://doi.org/10.1016/S0309-1708(01)00056-2

Fries G (1987) Étude de l'éventualité de création d'un dôme. Direction générale Science, recherche et développement. LUXEMBOURG: COMMISSION DES COMMUNAUTÉS EUROPÉENNES. ISBN 92-825-7443-1

Ghoryechi M & Daupley X (2003) Devenir à long terme d'exploitation abandonnées de sel en France. Colloque International Après-mine 2003. Nancy, France

Hart P, Jhonson RL, Nicholson TJ, Quinn E & Wrigth JR (1981) Survey of salt dome investigations. U.S Nuclear Regulatory Comission

Haynes W (2014) CRC handbook of chemistry and physics, vol 94. CRC Press LLC, Boca Raton

Herbert AW, Jackson CP, Lever DA (1988) Coupled groundwater flow and solute transport with fluid density strongly dependent upon concentration. Water Resour Res 34(0):1781–1795

Jackson M, Seni SJ (1983) Geometry and evolution of salt structures in a marginal rift basin of the Gulf of Mexico, east Texas. Geology 11(3): 131–135. https://doi.org/10.1130/0091-7613

Kelsall P, Nelson JW (1985) Geologic and ingeneering caracterizations of gulf region salt domes applied to underground storage and mining (Vol. 1). Sixth international symposium on salt

Kloppmann W, Negrel P, Casanova J, Klinge H, Schelkes K, Guerrot C (2001) Halite dissolution derived brines in the vicinity of a Permian salt dome (N German Basin). Evidence from boron, strontium, oxygen, and hydrogen isotopes. Geochim Cosmochim Acta 65(22): 4087–4101. https://doi.org/10.1016/S0016-7037(01)00640-8

Konikow LF, Sanford WE, Campbell PJ (1997) Canstant concentration boundary conditions:lessons from the hydrocoin variable density groundwater benchmark problem. Water Resour Res 33(10):2253–2261

Laabidi E, Bouhlila R (2015) Nonstationary porosity evolution in mixing zone in coastal carbonate aquifer using an alternative modeling approach. Environ Sci Pol 22(13):10070–10082. https://doi.org/10.1007/s11356-015-4207-2

Larsson A (1992) The international projects INTRACOIN, HYDROCOIN and INTRAVAL. Adv Water Resour 15(01):85–87. https://doi.org/10.1016/0309-1708(92)90034-Y

Massal P (1988) Rheologie,proprieté physiques de sel gemme et phenoménes de migration dans le sel. Bureau de Recherche geologiques et minière, BRGM, Orléans

Michelsen ON (2003) Jurassic lithostratigraphy and stratigraphic development onshore and offshore Denmark. Geological Survey of Denmark and Greenland. The Jurassic of Denmark and Greenland, Denmark

Monnin C (1989) An ion interaction model for the volumetric properties of natural waters: density of the solution and partial molal volumes of electrolytes to high concentrations at 25°C. Geochim Cosmochim Acta 53(6):1177–1188

OECD, f (1988) The international Hydrocoin Project,Level 1,code verification. Paris: Rep.71617

Peach CJ (1991) Influence of deformation on the fluid transport properties of salt rocks. In Geologica Ultraiectina. Faculteit Aardwetenschappen der Rijksuniversiteit te Utrecht, pp 1–238

Philibert J (1985) Diffusion et transport de matière dans les solides (Vol. 01). (É. d. Physique, Ed.) Monographies de physique. 2868830048, 9782868830043

Ranghathan v, Jeffery HS (1988) Density-driven groundwater flow near salt domes. Chem Geol 74:173–188

Reeves M, Ward DS, Johns ND & Cranwell RM (1986) Theory and implementation of SWIFT II,the sandia waste isolation flow and transport model for fractured media, release 4. 84. United States: Sandia National Labs., Albuquerque, NM (USA)

Shelkes K, Klinge H, SchildKnecht F & Weber JR (1999) Water-conducting features in a geologic system consisting of a salt dome and overlying unconsolidated sediments. Water-conducting features in radionuclide Migration, Proc. GEOTRAP Workshop. Paris: OECD/NEA, pp 145–156

Stanford W, Konikow LF (1989) Simulation of calcite dissolution and porosity changes in saltwater mixing zones in coastal aquifers. Water Resour 25:655–667

Steefel CI & Yabusaki S (1996) os3D/GIMRT Software for Modeling Multicomponent-Multidimensional Reactive Transport USER MANUAL & PROGRAMMER' S GUIDE. Department of Geology, University of South Florida Tampa, Florida 33620 U.S.A. Richland, Washington: Pacific Northwest Nati. PNNL-11166

Stormont JC, Howard CL & Daemen JK (1991) Changes in rock salt permeability due to nearby excavation, Rock Mechanics as a Multidisciplinary Science. In Proceedings of the 32nd US Symposium. Balkema, Brookfield: The University of Oklahoma, Norman, Oklahoma, pp 899–907

Verma A, Pruess K (1988) Thermohydrological conditions and silica redistribution near high-level nuclear wastes emplaced in saturated geological formations. J Geophys Res 93(B2):1159–1173. https://doi.org/10.1029/JB093iB02p01159

Younes A, Ackerer P, Mose R (1998) Modeling variable density flow and solute transport in poros medium:2 Re evaluation of the salt dome flow problem. Transport in poros media 35:375–394