

MODEL OF A THERMALLY INDUCED LIQUID INCLUSION MIGRATION IN A SINGLE CRYSTAL OF HALITES UNDER A HOMOGENEOUS TEMPERATURE GRADIENT FIELD

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Article received on December 23, 2021

The paper focuses on an evolution model developed for a liquid inclusion in halites under a temperature gradient field, taking into account surface phenomena. To demonstrate the safety of heat-generating radioactive waste disposal in salt rocks (halites), thermally induced migration of liquid inclusions should be studied. Based on the study, a software package was developed providing numerical implementation of this model. The diffusion equation with initial and interface conditions was approximated via the finite element method on triangular unstructured mobile grids. In case of weak formulation, the equations were discretized and solved in the FEniCS software. The study showed good agreement between the calculations and the experimental data.

Keywords: radioactive waste, temperature gradient, thermomigration, halites, liquid inclusions, chemical potential difference.

Introduction

Rock salt (halites) is a widespread rock that, despite its easy solubility in water, persists for hundreds of millions of years among geological formations of various continental structures. It is considered as the first and so far the only geological formation that has been recommended and actually used for underground RW disposal purposes. The IAEA papers present it as a low-permeability, plastic and almost unsaturated geological environment providing low-cost conditions for the construction of underground excavations [1]. Extensive raw material reserves of rock salt are available; therefore, salt formations can be used to site underground RW repositories.

Despite all the advantages of RW disposal in geological formations, long-term forecasts regarding the stability and the preservation of the main

barrier properties by the geological environment at RW disposal sites still remain an unresolved issue. To solve this complex scientific and engineering problem, data from various areas of the Earth sciences should be involved in the study, as well as the high-level information processing and interpretation methods that have recently evolved.

Problem definition

The study considers the evolution of a liquid inclusion in halite rocks given a field of a temperature gradient due to which the salt at the interface of the inclusion is unevenly dissolved and settled: at the “hot” interface of the inclusion, it dissolves, and at the “cold” interface, it settles. This effect makes the inclusion move along the temperature gradient

towards the heat source which is accompanied by a simultaneous change in its shape - stretching along the isothermal axis [2, 3].

Chemical potentials of salt in the solid phase and in the solution are set using the following expressions:

$$\mu_S = \mu_{0,S}(P_L, T) + \nu_S(P_S - P_L), \quad (1)$$

$$\mu_L = \mu_{0,L}(P_L, T) + RT \ln \alpha, \quad (2)$$

where P_L is the pressure in the liquid, P_S is the normal pressure in the solid, α is the activity of the salt in the solution, ν_S is the molar volume of the salt in the solid phase, $\mu_{0,S}$ is the chemical potential of the salt in the solid phase, $\mu_{0,L}$ is the chemical potential of the salt in the liquid phase.

Thermodynamic equilibrium corresponds to the condition $\mu_L = \mu_S$, but according to [2], dissolution and sedimentation of salt at the interface is a non-equilibrium process, therefore:

$$\mu_L = \mu_S + \Delta\mu, \quad (3)$$

where $\Delta\mu$ stands for some additional chemical potential being negative for the dissolving interface and positive for the interface on which the salt is precipitated from the solution.

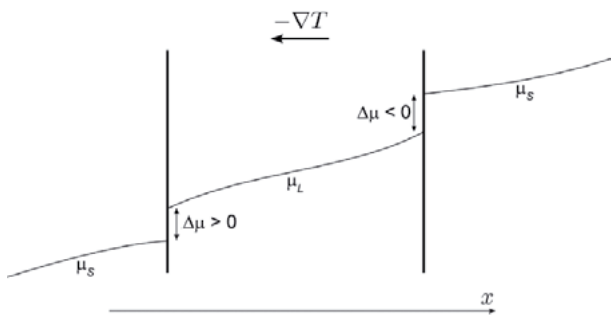


Figure 1. Chemical potential profile in an inclusion

Using (1)–(3) and assuming the activity equal to the salt concentration in the solution, the following expression can be obtained:

$$C = C_e(P_L, T) \exp\left(\frac{\Delta\mu}{RT}\right) \exp\left(-\frac{\nu_S \Delta p}{RT}\right). \quad (4)$$

Where $C_e(P_L, T)$ is the equilibrium concentration of a saturated salt solution.

In [2], $\Delta\mu$ is expressed as:

$$\Delta\mu = \begin{cases} -K_D & \text{for dissolving face,} \\ K_S & \text{for growing face} \end{cases} \quad (5)$$

In this case, $\Delta\mu$ on the end faces depends on the speed of their movement and in the steady period (when the speed of the end faces is equal to zero) it disappears.

For simplicity, we can assume that $\Delta\mu$ depends only on the projection of the face velocity onto its normal line and does not depend on its type (dissolving, growing, end face):

$$\Delta\mu(V) = \begin{cases} -K_D, & V \gg V_0 \\ K_S, & V \ll -V_0 \\ -V, & |V| \ll V_0 \end{cases} \quad (6)$$

Where V_0 is some characteristic velocity intermediate between the velocities of the growing and dissolving faces and the velocities of the end faces.

To provide a through (non-iterative) calculation, the piecewise constant function (6) was approximated by a continuous function:

$$\begin{aligned} \Delta\mu &= a + b \cdot \tanh(V / V_0 - x_0), \\ a &= \frac{1}{2}(K_S - K_D), \\ b &= -\frac{1}{2}(K_D + K_S), \\ x_0 &= \operatorname{arctanh}(a / b). \end{aligned} \quad (7)$$

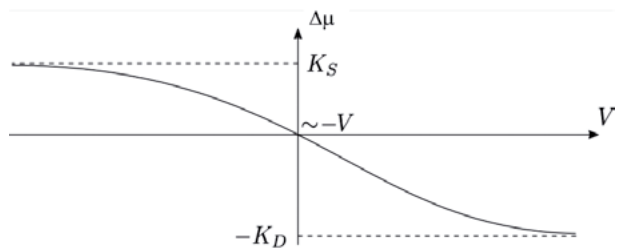


Figure 2. Function approximation for the kinetic potential difference

The computational domain can be represented as a rectangular inclusion with each of its faces moving at its own speed V_i (Figure 3).

The concentration C in the inclusion region Ω

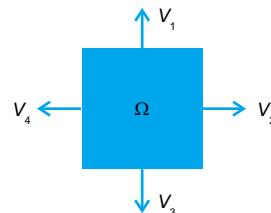


Figure 3. Computational domain and normal interface velocities

satisfies the diffusion equation:

$$\frac{\partial C}{\partial t} + \mathbf{V}\nabla C - D\Delta C = 0. \quad (8)$$

Boundary conditions:

$$C|_{\Gamma} = C_e(T) \cdot \exp\left(\frac{\Delta\mu}{RT}\right) \exp\left(-\frac{\nu_S \Delta p}{RT}\right). \quad (9)$$

The rate of changes in the interface area is governed by the following relation:

$$D \frac{\partial C}{\partial n} \Big|_{\Gamma} = -C_S V_n, \quad (10)$$

where C_S is the molar concentration of salt in the solid.

Initial conditions:

$$\begin{aligned} C(t=0) &= C_{e0} \\ T(t=0) &= T_0 \end{aligned}, \quad (11)$$

where C_{e0} is the equilibrium salt concentration in the brine at T_0 .

Numerical implementation

Expression (8) with boundary and initial conditions (9)–(11) is approximated by the finite element method on triangular unstructured moving grids, which are built using the AniAFT library from the Ani2D package [5].

Equation (8) is discretized by the Galerkin method:

$$\frac{\partial}{\partial t} \int_{\Omega} C \omega d\Omega + \int_{\Omega} V \nabla C \omega d\Omega - D \int_{\Omega} \Delta C \omega d\Omega = 0,$$

where ω is a test piecewise linear function.

From the Gauss-Ostrogradsky theorem:

$$D \int_{\Omega} \Delta C \omega d\Omega = D \int_{\partial\Omega} \frac{\partial C}{\partial n} \omega d\Gamma - D \int_{\Omega} \nabla C \nabla \omega d\Omega,$$

the boundary condition used to calculate the velocity (10) is substituted:

$$\frac{\partial}{\partial t} \int_{\Omega} C \omega d\Omega + \int_{\Omega} V \nabla C \omega d\Omega - C_S \int_{\partial\Omega} V_n \omega d\Gamma + D \int_{\Omega} \nabla C \nabla \omega d\Omega = 0.$$

The equation is approximated in time implicitly: C is the time-wise value at the n -layer (unknown), and \tilde{C} is the value of the previous step (known), then:

$$\frac{dC}{dt} = \frac{C - \tilde{C}}{\tau},$$

$$\int_{\Omega} \frac{C - \tilde{C}}{\tau} \omega d\Omega - C_S \int_{\partial\Omega} V_n \omega d\Gamma + \int_{\Omega} (D \nabla C) \nabla \omega d\Omega = 0.$$

Assuming that the computational domain retains a rectangle shape, the following method was used to calculate the velocities of the interfaces and rebuild the grid nodes: it was assumed that all nodes along the interfaces have the same corresponding normal velocity V_i :

$$\frac{1}{\tau} \int_{\Omega} C \omega d\Omega + \int_{\Omega} (D \nabla C) \nabla \omega d\Omega = \frac{1}{\tau} \int_{\Omega} \tilde{C} \omega d\Omega + C_S \sum_{i=1}^4 V_i \int_{\Gamma_i} \omega d\Gamma. \quad (12)$$

The linear concentration problem can be solved using the FEniCS software package.

To calculate C , a system of nonlinear equations obtained from the boundary condition (9) was solved:

$$\int_{\Gamma_i} C d\Gamma = \int_{\Gamma_i} C_e \exp\left(\frac{\Delta\mu}{RT}\right) \exp\left(\frac{v_S \Delta p}{RT}\right) d\Gamma. \quad (13)$$

The system of equations (12) and (13) is entirely responsible for the concentration and velocities of the interfaces. $\Delta\mu$ depends solely on the interface velocity, therefore, $\exp\left(\frac{\Delta\mu}{RT}\right)$ can be taken out from the integral sign. When calculating integrals of known quantities, a system of four nonlinear equations is obtained, which is solved by Newton's method.

To calculate the velocity in each node of the computational domain, the following assumptions were made: it was assumed that the fluid in the inclusion is incompressible $\text{div } \mathbf{V} = 0$, the flow is potential $\mathbf{V} = \text{grad } \varphi$. Therefore, the Poisson equation was used in this problem.

$$\Delta \varphi = 0 \text{ в } \Omega,$$

$$\frac{\partial \varphi}{\partial n} = V_n \text{ на } \partial\Omega. \quad (14)$$

The solution for the system of equations (13) was substituted as V_n .

Problem (14) was discretized based on the Galerkin method and solved using the FEniCS software package.

After finding the velocity of each node in the domain, the displacement vectors of the nodes were calculated.

In the calculations, the time step is adjusted so that the inclusion gets deformed by the order of the grid parameter h , i. e., the minimum grid size (to avoid some significant distortions). Its quality was also monitored: if it was found to be falling below a critical level, the grid was rebuilt using the AniAFT library.

Numerical methods were implemented using the open software environment FEniCS [4].

Results

Numerical solution of the problem has involved some constants and parameters that corresponded to the experimental conditions described in [2]. This article shows the evolution of four inclusions of different sizes. Figure 4 shows the transformation of one of them. Calculations were performed to compare them with the experimental data. Figure 5 presents the calculation results.

Comparison between Figures 4 and 5 shows that the numerical solution agrees qualitatively with the experimental results [2], but there is a significant quantitative discrepancy in the stabilization

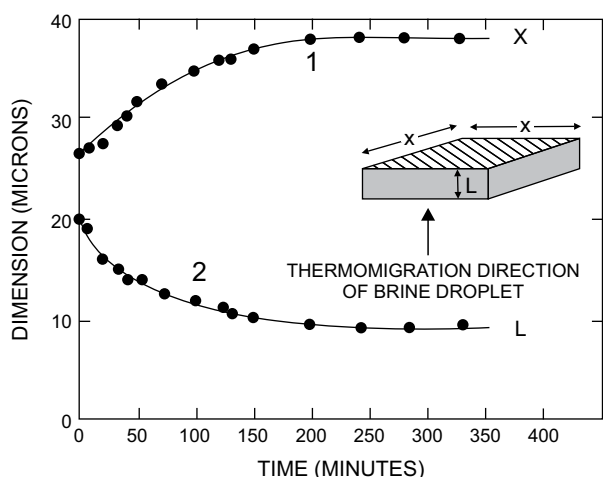


Figure 4. Evolution of vertical (1) and horizontal (2) dimensions of the inclusion [2]

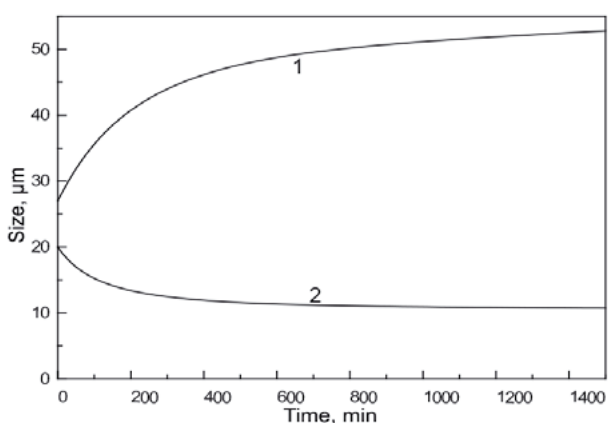


Figure 5. Evolution of vertical (1) and horizontal (2) dimensions of the inclusion

time, which can be explained by the following factors: firstly, the two-dimensional model problem definition, in contrast to the real-life three-dimensional experiment, and, secondly, $\Delta\mu$ approximation by function (7), which can be further replaced by a more appropriate one.

Conclusion

The paper considers a microscopic model of a liquid inclusion evolution in a temperature gradient field taking into account surface phenomena.

A software package has been developed for the numerical implementation of this model based on the finite element method and a moving adaptive grid.

Comparison of calculation results with experimental data [2] showed their good agreement.

The study was supported by the Russian Foundation for Basic Research (project 20-08-01192)

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Bibliographic description

Avetisyan A. R., Korchagina O. O., Matveev L. V. Model of a Thermally Induced Liquid Inclusion Migration in a Single Crystal of Halites under a Homogeneous Temperature Gradient Field. *Radioactive Waste*, 2022, no. 2 (19), pp. 100–104. DOI: 10.25283/2587-9707-2022-2-100-104. (In Russian).