KINETIC MODEL SHOWING THE EVOLUTION OF LIQUID INCLUSIONS IN SALT ROCKS UNDER HIGH TEMPERATURE GRADIENTS

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The paper focuses on a kinetic evolution model developed for an ensemble of inclusions in halites in a temperature gradient field. To demonstrate the safety of heat-generating radioactive waste disposal in salt rocks (halites), thermally induced migration of liquid inclusions should be studied. A simplest approximation was assumed to describe the liquid inclusion evolution in halites: Boltzmann type equation was obtained for the distribution function depending on the isothermal size of the inclusion, taking into account coalescence and instantaneous equiprobable decay. The research allowed to develop a software module providing numerical implementation of the kinetic model.

Introduction

Radioactive waste (RW) disposal is seen as a most important task shaping the prospects for nuclear power development. Establishment of radioactive waste disposal facilities (DF) based on the principle of multi-barrier safety is considered a most preferable option for the final isolation of the entire RW inventory.

Currently, salt host rocks (halites) recommended by the IAEA are considered a potential medium for deep disposal facility siting. These host rocks are characterized with high thermal conductivity, which is seen as their advantage and is required to remove the heat released by RW with a high level of heat output due to their decay. Another advantage is their plasticity, which also reflects their ability to seal cracks and fractures. In addition, it is believed that these rocks have low water conductivity, which prevents the fluid flow (and, correspondingly, the contaminant flows) over long distances.

Even though groundwater is absent in the salt rocks, these rocks always contain small brine and/or gas inclusions. According to various literature sources, their relative volume fraction ranges from 0.5 % [1] to 2—3 % [2]. These inclusions are generated during the development of salt formations and remain in a stable (metastable) state provided that no external disturbances occur.

In case of RW disposal in halite, the residual heat release from radionuclide decay should be accounted for. Due to this factor, a non-uniform temperature distribution will be formed in sufficiently large areas around the repository, which may contribute to the migration of these inclusions [1—3]. In particular, if they contain only brine, then the resulting temperature gradient will cause their movement to hotter areas, i. e., to the disposal area. Thus, there's a risk that the brine available in the repository area may contribute to an aggressive environment

causing accelerated degradation of engineered barriers.

The paper considers a kinetic model representing intracrystalline migration of inclusions with brine in salt rocks [3—5] taking into account coalescence and instantaneous decomposition at high temperature gradients. The proposed model can be used as a component of a safety case for RW disposal in halite.

Physical model of liquid inclusion migration

In the temperature gradient field, all liquid inclusions in salt migrate along the gradient to the heat source, since the solubility of salt in inclusions increases along with temperature increase, which is higher on the surface of the inclusion being closer to the heat source than the one on the surface being far away from the source. Consequently, the salt dissolves along the hot surface of the inclusion and crystallizes along the cold one. Diffusion of salt from a hot to a cold surface forces the inclusion to move in the opposite direction, i.e., to a higher temperature (Figure 1).
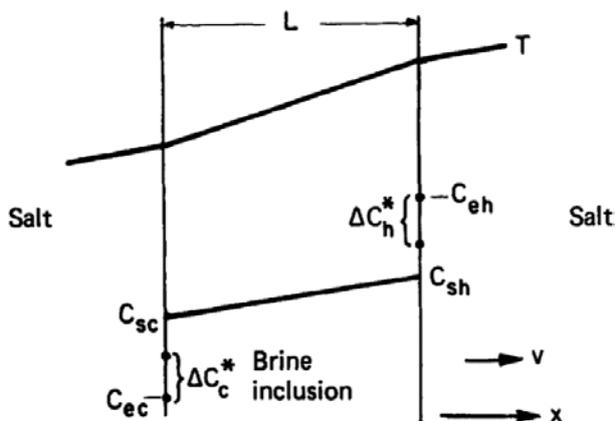

Figure 1. Mechanism of liquid inclusion migration in salts

Figure 1 schematically shows a flat-walled inclusion surrounded by solid salt. The salt concentration C_s in a liquid located close to a given surface is expected to be similar to the saturation concentration governed by the surface temperature. Since salt solubility increases along with temperature increase, the C_{sh} concentration near the hot surface is higher than C_{sc} concentration at the cold surface. This difference in concentration can cause the salt to diffuse through the inclusion. Such a simple mechanism can result in a linear change in the concentration of dissolved salt throughout the inclusion. The concentration profile may look like the one shown in Figure 1.

For a liquid inclusion (drop), the thermal migration process can be split into two periods: initial and steady [5]. At the initial stage, the shape of a drop with brine (it flattens along the isothermal dimension) and its velocity changes. In the steady period, these parameters come into equilibrium. For both periods, there is a minimum isothermal size of the inclusion, below which it stops migrating (R_{cr}^{min}). In the steady period, for any drop, regardless of its speed and shape, size L lined up along the temperature gradient gets stabilized. This size, as was experimentally demonstrated in [4], depends on the temperature gradient level. The time during which the inclusion comes to the equilibrium state (constant speed and shape) will be denoted as τ_1.

In [5], assuming that the velocity of any face of a drop can be calculated based on the flux of salt atoms emanating or falling on a particular face and taking into account the geometric identity of the cold and hot faces, an expression was obtained for the thermal migration rate of a liquid inclusion for the equilibrium period:

$$V = D\frac{C_l}{C_s}\left[\left(\frac{1}{C_E}\frac{\partial C_E}{\partial T}+\sigma\right)G_l - \frac{K_D+K_S}{(RT)L}\right], \quad (1)$$

where C_l is the salt concentration in brine, C_s is the molar concentration of salt in a solid, D is the diffusion coefficient of salt in brine, σ is the Soret coefficient, $G_l \sim \frac{R}{L}\nabla T$ is the thermal gradient in brine, C_E is the equilibrium solubility of salt in brine, K_D+K_S is the sum of kinetic potentials for the interface on the cold and hot walls; the values of all quantities were taken according to [5].

Thermal migration of an inclusion ensemble

The study was mainly aimed at calculating the amount of brine that may reach the RW disposal area, therefore, it was necessary to model the behavior of a liquid inclusion ensemble.

To describe it, $f(R, y, t)$ was used, which is a distribution function that depends on the isothermal size of the inclusion, its coordinates and time. In this study, it was assumed that $V(R)$ was the steady rate of inclusion migration depending on the isothermal size of the inclusion that could be calculated using the expression (1). The phenomena of coalescence and instantaneous disintegration of inclusions [4, 5] were taken into account.

A process of collision between two inclusions was considered. It resulted in a new large ($R_i > R_{cr}^{max}$), but nevertheless unstable inclusion, which immediately split into two. The distribution function would satisfy the following conditions:

$$\frac{df_i}{dt}+\frac{df_j}{dt} = \sum_{k+l=i+j} a_{kl}\left(\frac{df_k}{dt}+\frac{df_l}{dt}\right). \quad (2)$$

In such a process, the number of inclusions and their total volume should stay constant (in case of subcritical particles, only the volume remains constant). Assuming that all a_{kl} are equal, since the particles collide and are born in pairs, then $\dfrac{df_i}{dt} = \dfrac{df_j}{dt}$ and $\dfrac{df_k}{dt} = \dfrac{df_l}{dt}$. In addition, two new particles are considered to be obtained from two particles, therefore, $\dfrac{df_i}{dt} = N\dfrac{df_k}{dt} = v$, where N is the number of particle size options, v is the number of collisions per unit of time.

To describe the evolution of liquid inclusions in halite in the simplest approximation, an equation of the Boltzmann type was obtained for the distribution function depending on the isothermal size of the inclusion taking into account coalescence and instantaneous equiprobable decay [6]. In this case, its spatial distribution was not considered. Only the change in the size of inclusions due to collisions of small inclusions and the decay of large ones ($\geqslant 210$ μm) was taken into account. The migration rate was calculated through the expression (1). If we evenly divide the entire range of variation in the size of inclusions into N parts with a step Δr and consider the above assumptions, the kinetic equation may be represented as follows:

$$\frac{\partial f_i}{\partial t} = -f_i \sum_{j=0}^{I} a_{ij} f_j + \sum_{j,k=0}^{I} a_{jk} f_j f_k \begin{cases} \Delta r \delta\left(R_j + R_k - R_i\right), \ j+k < I+1-\dfrac{r}{\Delta r} \\[2mm] \begin{cases} \dfrac{1}{j+k+1}, \quad I+1-\dfrac{r}{\Delta r} \leq j+k \leq I, \ \ i = \overline{0, j+k}, \\[3mm] \dfrac{1}{2I}+1-(j+k), \quad I < j+k < 2I, \ \ i = \overline{j+k-I, I} \end{cases} \end{cases} \quad (3)$$

where f_i is the distribution function of liquid inclusions depending on the isothermal size R_i, V is the migration rate (1), $a_{i,j} = \Delta r S |V_i - V_j|$, $S = \pi(R_i + R_j)^2$ is the effective cross-sectional area for the inclusion interaction, $R_I = r + I\Delta r$, I is the index of the maximum stable inclusion.

The first item on the right side of the equation (3) is responsible for the decrease in the number of inclusions with isothermal size R_i, and the second one is responsible for its increase, which occurs due to coalescence and instantaneous binary equiprobable decay of large inclusions. It can

be seen from expression (3) that unstable inclusions are of two types: those obtained due to the merging of inclusions, the total index of which is less than or equal to the maximum index of stable inclusion I, and the total volume is greater than the maximum stable volume, and those whose total indices and volumes are greater than the maximum stable values.

Taking into account the spatial dependence of the distribution function, equations (3) may be expressed in the following way:

$$\frac{\partial f_i}{\partial t} - V_i \frac{\partial f_i}{\partial y} = -f_i \sum_{j=0}^{I} a_{ij} f_j + \sum_{j,k=0}^{I} a_{jk} f_j f_k \begin{cases} \Delta r \delta\left(R_j + R_k - R_i\right), \ j+k < I+1-\dfrac{r}{\Delta r} \\[2mm] \begin{cases} \dfrac{1}{j+k+1}, \quad I+1-\dfrac{r}{\Delta r} \leq j+k \leq I, \ \ i = \overline{0, j+k}. \\[3mm] \dfrac{1}{2I}+1-(j+k), \quad I < j+k < 2I, \ \ i = \overline{j+k-I, I} \end{cases} \end{cases} \quad (4)$$

To solve the system of equations (4) with given initial and periodic boundary conditions, the Euler method of the second order was used.

According to numerical calculations obtained from the kinetic equation with allowance for the spatial dependence of the distribution function (4), in the process of thermal migration, the inclusions tend to a certain isothermal size spectrum. Over time, the range narrows down and tends to reach the maximum stable size (Figure 2).

Considering the above model demonstrating liquid inclusion evolution, the coalescence of stable particles and the instantaneous equiprobable

binary decay of unstable inclusions, one may calculate their number available in the computational domain for a particular moment of time:

$$N = \int_{Y_0}^{Y_{max}} \int_{R_0}^{R_{max}} f(R,y,t)\,dR\,dy. \quad (5)$$

Conclusions

The paper analyzed the processes of liquid inclusion migration in single crystals of halite in the presence of a temperature gradient. A kinetic model has been developed to simulate the behavior
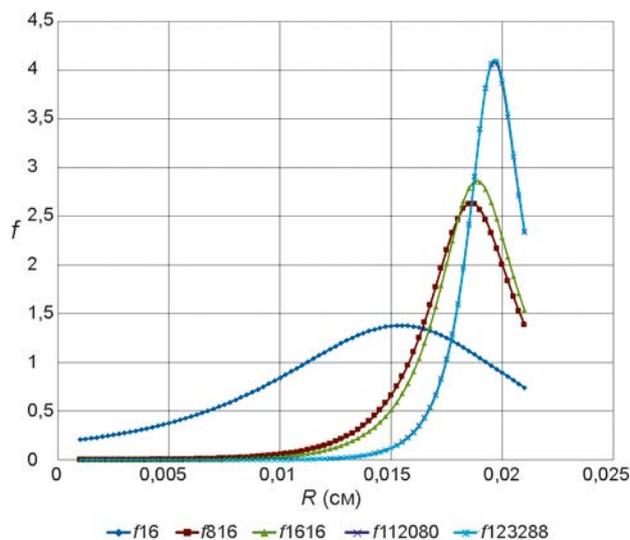
Figure 2. Dependences f(R) considering different times

of liquid inclusions in halite at high temperature gradients taking into account the coalescence and their instantaneous binary decomposition.

This model was used to perform calculations that showed that the thermal migration of liquid inclusions was ordered in its nature. With time, most of the inclusions, namely, their size tends to reach a narrow range around the maximum stable size.

The model can be also used to calculate the number of inclusions in the computational domain for a specific point in time, which is considered quite useful for further research.

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