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Coupled Processes at Micro- and Macroscopic Levels for Long-Term Performance Assessment Studies of Nuclear Waste Repositories

Hua Shao [1,*], Eike Radeisen [1], Jürgen Hesser [1], Wenqing Wang [2] and Olaf Kolditz [2,3]

[1] Federal Institute for Geosciences and Natural Resources (BGR), 30655 Hannover, Germany
[2] Helmholtz Centre for Environmental Research (UFZ), 04318 Leipzig, Germany
[3] Faculty of Environmental Sciences, Technische Universität Dresden (TUD), 01069 Dresden, Germany
* Correspondence: hua.shao@bgr.de

Abstract: Performance assessment of nuclear waste repositories requires state-of-the-art knowledge of radionuclide transport properties. Additionally, the short-term development under thermal pulses and the long-term development of the near field—due to influences such as gas generation—must be evaluated. Key thermal-hydro-mechanical-chemical processes are strongly coupled on different spatial and temporal scales. To understand these coupling mechanisms, numerous material models and numerical codes have been developed. However, the existing constitutive approaches—which have been adapted to describe small-scale laboratory experiments and validated against real-scale field observations—are often unable to capture long-term material behavior with sufficient precision. To build the confidence, a more comprehensive understanding of the system at micro- and macroscopic scales is required. Most observed macroscopic processes result from microscopic changes in the crystal structure and/or crystalline aggregates, as well as changes in material properties under the influence of various factors. To characterize these physical fields in crystals, microscopic investigations, such as visualization, or geophysical methods are introduced to verify the understanding at the microscale. Two cases are demonstrated for the presented concept using microscale information: one deals with the mechanically and thermally driven migration of fluid inclusions in rock salt, the other with dilatancy-controlled gas transport in water-saturated clay material.

Keywords: understanding of coupled processes on the micro- and macroscopic levels; heterogeneity; advective gas flow in bentonite; fluid migration in rock salt; numerical code OGS (OpenGeoSys)

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1. Introduction

The assessment of long-term performance of nuclear waste repositories in geological environments is a crucial endeavor to understand the behavior of solute transport processes. Numerous investigations and studies have been carried out over recent decades to gain insight into the complex dynamics of solute transport over extended periods of time [1–3]. This multidimensional investigation involves various methods, including in situ experiments, laboratory experiments, microscale observations and numerical modeling. The fundamental processes of solute transport, e.g., diffusion, advection, sorption and retardation, are well understood on an experimental scale in the laboratory, and in the case of radioactive particles, even decay.

In nuclear waste repositories, the thermal, hydrogeological and mechanical conditions lead to complex interactions, which can significantly influence the transport mechanisms of solutes [4]. Mechanical stresses and strains within the repository can be influenced by thermal conditions. This thermo-mechanical coupling can alter the properties of geological materials, affecting porosity, permeability, and consequently, solute transport. Therefore, in recent years, demonstration experiments on coupled thermal-hydromechanical processes have been carried out in underground rock laboratories in all kinds of potential geological formations to investigate the changes in porosity and permeability due to coupled thermal and mechanical effects. Several large-scale tests, e.g., prototype repository in the hard rock laboratory Äspö (Sweden), FEBEX at the Grimsel test site (Switzerland), HE-E and FE in the underground rock laboratory Mont Terri, ACL in France's underground facility Bure (France), were successfully carried out in terms of implementation and long-term monitoring [5–8]. The temperature evolution can be reproduced and simulated very well in all experiments in the beginning. However, pore or fracture water pressure evolution, which is determined by porosity and permeability and represents an important variable for the transport mechanism, cannot be captured well. This mismatch can also impact long-term temperature performance.

Therefore, a comprehensive understanding of the coupled thermal, hydrogeological, mechanical and solute transport processes, supported by advanced modeling and validation through experiments at the different scales, is crucial for accurate long-term performance assessment. This type of validation of models at laboratory and field scales has been carried out extensively in different projects, e.g., the international collaborative project DECOVALEX (DEvelopment of COupled models and their VALidation against EXperiments in nuclear waste isolation) [4,9,10], European Commission (EC)-supported projects (EURAD) [11] and site-specific projects, such as the FEBEX experiment. Increasing attention is being paid to the question of how and to what extent the processes observed at laboratory and field scales are related to the microstructural changes under the coupled THM conditions. Hence, the results of the microscale investigations should be considered in the numerical modeling. Microscale observation techniques provide a finer resolution, allowing us to analyze the intricate details of the status or process taking place within the fractures and pores of rocks, as well as solids [12–14]. Such observations, which are currently limited to recording state images and may be extended in the future to recording dynamic processes, e.g., using acoustic emission (AE) techniques (R4), help bridge the gap between macroscopic studies at the field scale and microscopic processes at the pore scale, and they provide a broader perspective on long-term system performance.

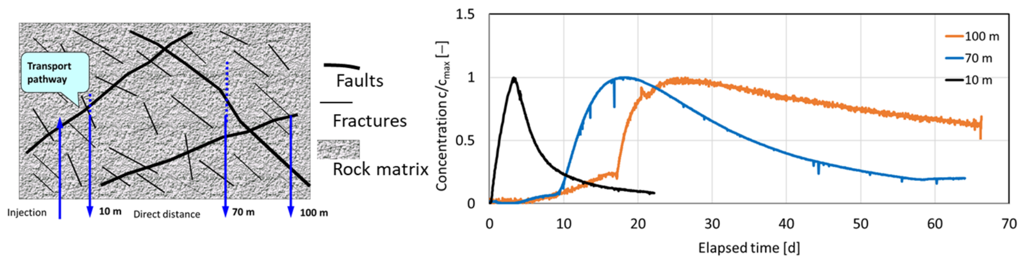
Based on the microscale study, pore-scale modeling proves to be a powerful tool in this field, allowing us to simulate and analyze processes at the microscopic level [15–17]. This method requires a detailed understanding of the physical processes occurring at the pore scale and a complete description of the morphology of the pore space. The pore bodies are interconnected by narrow spaces called pore throats, discretized by elements or cells depending on the numerical method used. By employing advanced mathematical, computational models and modern high-performance computer technologies, it is now possible to investigate the complex interactions between fluids and porous media and to facilitate the interpretation of experimental data. Nevertheless, there is still uncertainty and variability in the system. The inherent uncertainties in predicting long-term behavior under coupled conditions, such as variations (heterogeneity) in rock properties or future climate changes leading to changed boundary conditions, increase the complexity of solute transport assessments. The latter is not discussed in this article.

This article introduces some ideas for a comprehensive exploration of the multiple approaches to understanding coupled processes within fractured porous environments. By combining insights from in situ experiments, laboratory studies, microscale observations and pore-scale modeling, we can develop a better understanding of the intricate processes, which influence solute transport dynamics. We introduce two novel representations of microscale processes: (1) a stochastic parameter distribution for the microscale heterogeneity of gas migration in clayey materials, e.g., bentonite, and (2) a mixture-theory-based multiple modeling approach to understand fluid migration in rock salt. Both bentonite and rock salt are generally considered to be homogeneous materials. The migration processes observed in the experiments cannot be explained by a conventional continuum modeling concept [18].

2. Long-Term Solute Transport with Scale-Dependency in Space and Time

For a long-term safety analysis of a repository for radioactive waste in a deep geological formation, the modeling of groundwater flow and solute transport is important. Reactive transport is of particular importance in the safety analysis of final repositories. In granite, contaminant transport mainly takes place in the fractures or fracture networks, in other sedimentary rocks, such as claystone or rock salt, in the pore space, which can be observed as a microchannel network. The study of mass transport in fractured porous media is based on knowledge of the hydraulic properties of fractures at different scales and the transport mechanisms of the transported material, including advection, diffusion, retardation, sorption and decay.

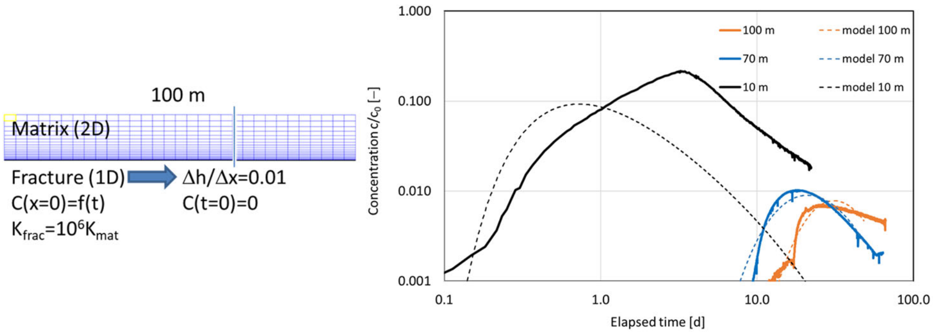
Scale-dependent tracer transport for a conservative tracer in fractured rock was extensively investigated at the Grimsel test site, Switzerland. The experiments were based on a dataset with more than 3800 fractures from 20 boreholes with a total drill core of 1183 m [3]. The entire dataset was evaluated by geo-statistical interpretation of fracture data, e.g., variogram analysis of fracture spacing and fracture density [19]. Three main fracture clusters were characterized and verified by hydraulic tests, including pumping and interference tests. On this basis, tracer experiments in a dipole configuration were performed. The direct transport distance varied from 10 m to 100 m between the injection borehole and observation boreholes. Three of them with different characteristic transport distances are plotted in Figure 1. Both the breakthrough time and the maximum concentration are strongly dependent on the transport distance, implying the properties of the influenced area, such as microcracks in the rock matrix.



(**a**) (**b**)

Figure 1. Scale-dependent solute transport in fractured rock: conceptual model including fracture network, fine fissures and matrix (not scaled) (**a**) and the normalized tracer breakthrough with different transport distances after a pulse tracer injection in all three cases (**b**).

A simpler model combining a 1D fracture and 2D matrix representing the fracture–matrix system was used to analyze the transport properties. A constant flow velocity was assigned, as defined in Figure 2, and in addition, a constant concentration for a certain time as a pulse injection was given. While the advective transport of this conservative tracer mainly occurs in the fracture zone, the diffusion process dominates due to the low permeability in the matrix, which typically plays only a secondary role. This model is not an interpretation model for the in situ tests performed and only presents systematic breakthrough curve styles. In the two large-scale tracer tests with 70 m and 100 m transport distances, both the arrival time and the longer tailing can be scaled well. However, the breakthrough curve in the 10 m tracer test does not agree well, indicating strong heterogeneity and multiple pathways of the small-scale system, since this test is performed under a lower pressure gradient, resulting in a transport distance of more than 10 m.



(**a**) (**b**)

Figure 2. Modeling of solute transport with a single fracture–matrix model (**a**) and characteristic curves for breakthrough (**b**).

In order to obtain satisfactory agreement, in most interpretation models, sensitivity analyses of the key parameters are often carried out. This exercise also helps to better understand the system's response to variations in these factors. A better fit to a certain tracer breakthrough can be obtained via the calibration of key parameters. The question is whether the overall scale effect of mass transport can be calibrated, i.e., can the result of small-scale tracer experiments be extrapolated to a large scale, even a million-year repository scale? A more comprehensive method is to look at the rock structure at the microscale.

3. Scale-Dependent Heterogeneity and Microscale Investigations

Heterogeneity, i.e., the variability or differences in the spatial properties of a material, exists at different scales of observation. Certain characteristics or patterns may be considered homogeneous at the macroscale, based on the concept of the representative elementary volume (REV) for porous media, but at the microscale, these systems are actually heterogeneous. Clayey materials (bentonite, Opalinus Clay) or rock salt, which are usually considered as homogeneous media in the interpretation of experimental data, are heterogeneous at the microscale. Bossart [20] showed that small-scale heterogeneities play a key role in the formation of the fracture network and that the anisotropy of the fabric may have a major influence on the final geometry of the excavation damaged zone. Thiemeyer et al. [13] addressed rock salt, which is characterized by heterogeneous grain shape structures with ellipsoidal grain shapes, and its impact on deformation and dynamics. Therefore, most behaviors observed at the macroscale cannot be fully understood if only one scale is studied.

Microscale investigation involves the study of phenomena or processes at a small scale, typically at the level of individual particles, crystals and minerals. This level of investigation allows us to understand the intricate details and interactions, which occur at the microscale, and provides valuable insights into the systems. To study the microscale, specialized analytical and imaging techniques, such as optical microscopy and electron backscatter diffraction (EBSD), have greatly enhanced our ability to explore and observe the microscale phenomena [13,17,21]. These investigations are currently limited to recording the steady state. Therefore, a dynamic picture of the mineralogical changes under macroscale test conditions is required, the information of which should be integrated into future numerical interpretation. However, when recording dynamic conditions under different thermo-hydromechanical conditions, additional challenges arise, such as the need for advanced technical instrumentation, the avoidance of artefacts in the measurements and the difficulty in transferring the results to larger systems.

To fully understand scale-dependent heterogeneity, researchers often need to integrate data from different scales. Computational models and simulations play a crucial role in bridging the gap between the microscale and larger scales (Figure 3). Due to limited computer capacity, microscale modeling with detailed information cannot be performed in a full-scale model. For this study, a multiple model concept was used, e.g., the boundary conditions applied in microscale modeling were determined by a macroscale simulation [22].

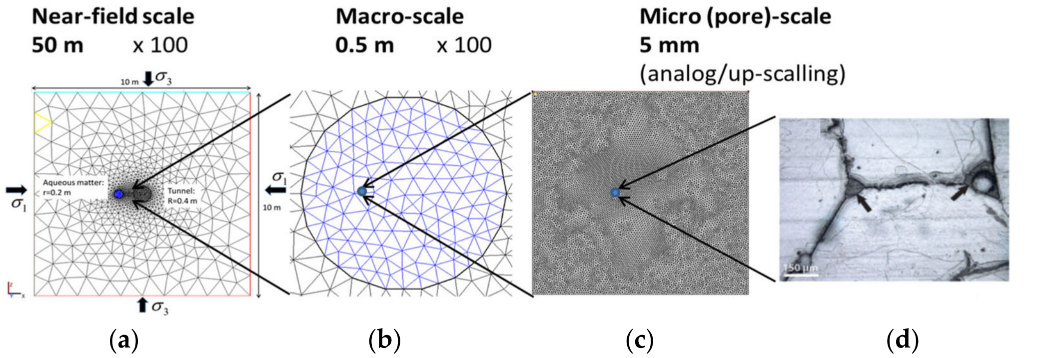


Figure 3. Scale-dependent simulation techniques for simulating fluid inclusion: Near-field scale model including a region with fluid (blue circle) under natural stress conditions (**a**); Enlargement of the mesh focusing on the fluid saturated zone (**b**); Enlargement of the mesh for a detailed consideration (**c**) and comparison with the information from microscale characterization with inclusions (arrows) [12] (**d**).

Numerical simulations, such as dynamic pore-scale simulations [15–17], allow us to manipulate the dynamic process, e.g., fluid–solid interaction or the percolation threshold at the (sub) pore size scale. At the microscopic (sub-pore) scale, the solid and fluid phases occupy different parts of the spatial domain and interact at their common interface [16]. Thus, the microscopic fields, describing the properties of the constituents, can be considered as homogeneous continua within a single (solid, liquid or gaseous) phase while exhibiting discontinuities at the interfaces between the phases. At such scale, the fluid flow with small Reynolds and Stokes numbers is governed by the Stokes equation rather than the Darcy equation used in porous media.

One way of addressing this type of heterogeneity is to evaluate the variability of the parameters. This variability can be represented by the element assignment of different material parameters in the different finite element meshes (Figure 4). Figure 4 shows an example of how a numerically coupled thermo-hydromechanical simulation is carried out on an Opalinus Clay sample from the Mont Terri research laboratory in Switzerland. The information on different minerals obtained from a scanning electron microscope (SEM) measurement is considered as different materials using color screening techniques. For a reliable analysis, the statistical distribution of the bandwidth should be taken into account.

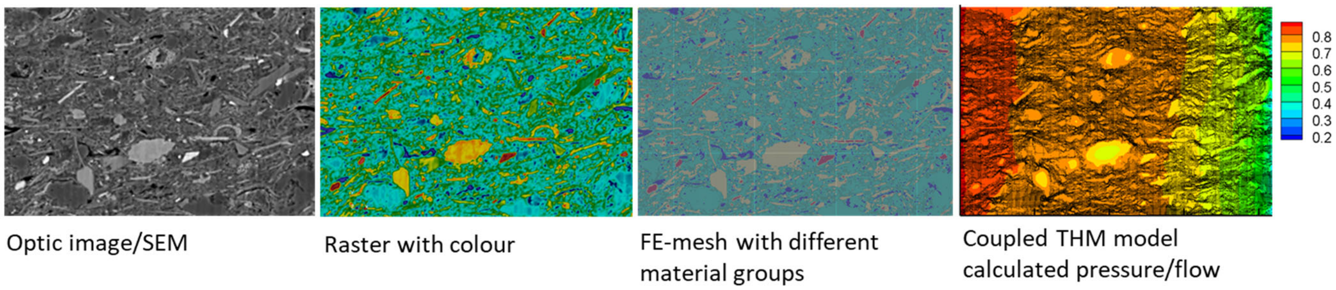


Figure 4. Microscale heterogeneity model (model size 0.09 × 0.07 mm).

4. Numerical Methods

In the field of coupled thermal-hydrological-mechanical-chemical (THMC) processes, the understanding of material behavior at both the microscopic and macroscopic level plays a crucial role in the accuracy of numerical simulations [23]. To bridge the gap between laboratory-scale experiments and real-field observations, it is essential to take into account the intrinsic heterogeneities of geological materials. In this context, stochastic parameter distributions prove to be a powerful tool for capturing microscale heterogeneity.

Microscopic variations in material properties, such as Young's modulus or permeability, can significantly influence the macroscopic behavior of the system. In stochastic approaches, randomness is added to these properties, reflecting the variability in natural formations. This randomness is often modeled by statistical distributions, which represent the range of possible material states. The objective is to develop numerical codes, which not only take into account deterministic properties but also account for the uncertainty of geological formations.

4.1. Stochastic Parameter Distribution for Microscale Heterogeneity

Understanding and implementing microscale heterogeneity are crucial for accurately representing the behavior of porous media. Stochastic parameter distribution methods offer a valuable approach for incorporating the variability observed at the microscale. In the context of fluid migration through dense, low-permeable porous media, heterogeneity can arise from variations in intrinsic microscale heterogeneity of the dry density, microfractures and other microscopic features.

Numerical treatments can be organized in three steps:

1. Deriving microscale parameter properties from either microscale or macroscale measurements or observations using inverse modeling techniques.
2. Spatial correlated heterogeneous distributions of material parameters using semi-variogram analysis.
3. Stochastic simulations to represent uncertainty, i.e., Monte Carlo simulations, Latin Hypercube Sampling or Markov Chain Monte Carlo.
4. Possible updating of any stochastic distribution parameters in an evolving system, such as developing fracture networks.

Various techniques can be used to derive microscale parameter properties from micro- or macroscopic properties. In the context of coupled TH2M models, microscopic properties such as permeability or gas entry pressure are often derived from variations in dry density or porosity [24–26]. These techniques represent the opposite of upscaling techniques, such as homogenization. The random spatial distributions generated can also be fitted to available data, such as fracture maps or anisotropic behavior, to account for observed patterns.

For example, the microscopically measured pore size distribution in bentonite can be used to derive microscopic material properties. Mercury intrusion porosimetry (MIP) is often used to measure pore size distribution, whereby pore sizes of 1 nm can be recorded. The measured pore size diameter d can then be used, for example, to calculate the gas entry pressure [27]:

$$p_b = \frac{4T_s\cos(\theta)}{d} + p_{l0} \tag{1}$$

where p_b is the so-called gas entry pressure; T_s is the surface tension of the wetting fluid; θ is the angle between wetting and non-wetting fluid; and p_{l0} is the initial liquid pressure. The saturation of the pore space can be used to estimate cos (θ), as shown in Figure 5. Under saturated conditions (case C in Figure 5), θ is 0° for the whole pore space, based on which it can be concluded that cos (θ) = 1 for all pores. Applying the pore size density measured by Seiphoori [28] on a MX-80 bentonite under saturated conditions to Equation (1), a stochastic distribution of the gas entry pressure can be derived, as shown in Figure 6. Other microscopic material properties related to the flow of fluids through dense and low-permeability materials include, e.g., permeability or stiffness. The derived stochastic distributions can be compared and validated with analytical solutions using simulation series in benchmark tests, as performed by Radeisen et al. [29].

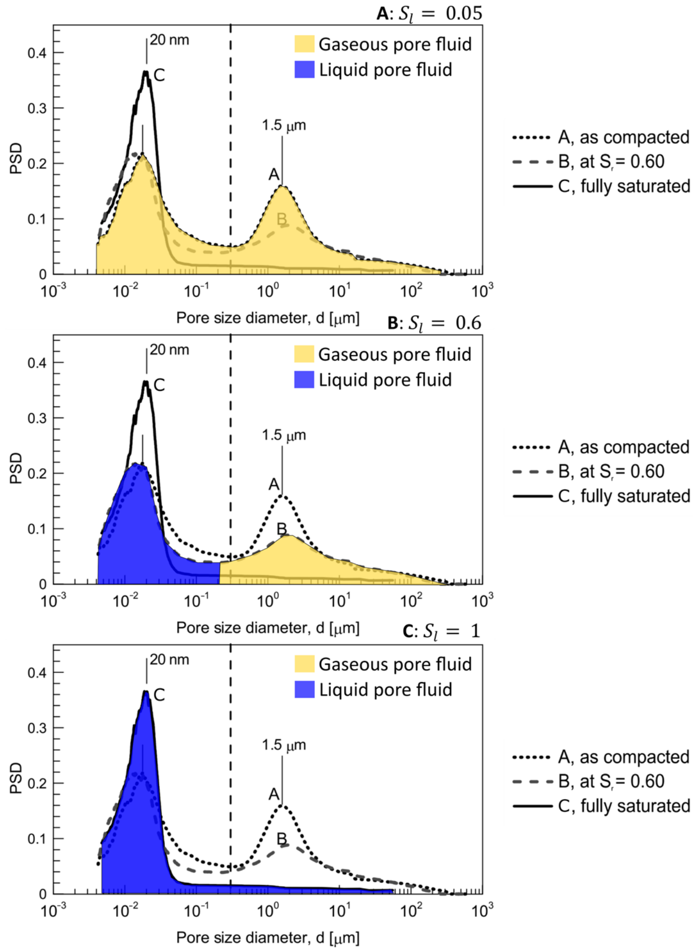


Figure 5. Pore size distributions (PSDs) of pore size diameter with corresponding pore fluid at three different conditions: (**A**) Compacted and dry with $S_l = 0.05$; (**B**) Partly saturated with $S_l = 0.6$; and (**C**) Fully saturated with $S_l = 1$. PSD measurements were performed by Seiphoori [28].

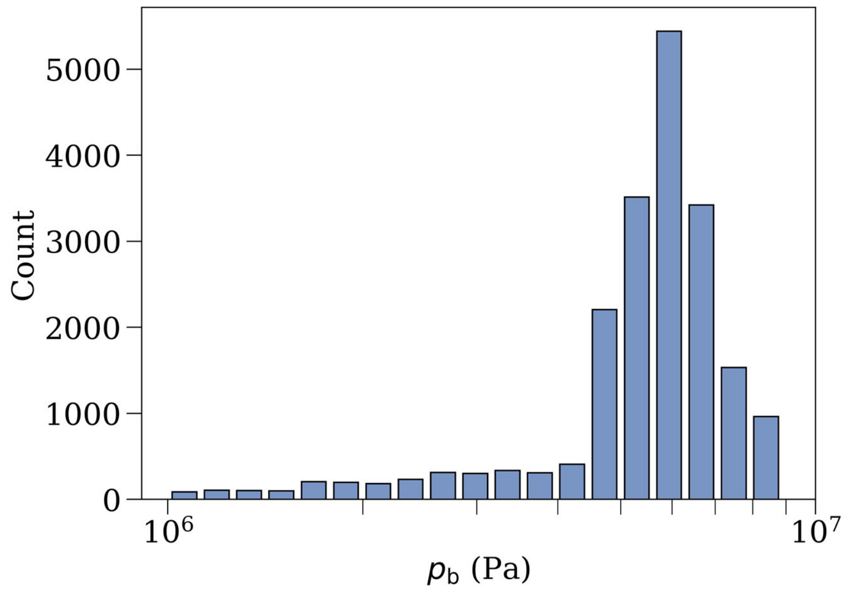


Figure 6. The derived distribution with the amount (count) of elements in a numerical model with the value of gas entry pressure based on microscale measurements of the pore diameter of an MX-80 bentonite applied in a gas test simulation [29].

4.2. Mixture Theory for Microscale Discontinuity

Generally, the mixture theory considers materials, which are not homogeneous but consist of different phases or constituents, such as a three-phase system with solid, fluid and gaseous components. These phases have distinct properties, and their interactions at the microscale can significantly influence the overall behavior of the total system. This approach is widely applied in fields such as material science, geophysics and biomechanics, where understanding the interactions and behavior of heterogeneous materials at the microscale is crucial.

The principle of the mixture theory is to determine effective properties of the material, such as effective stiffness, thermal conductivity or hydraulic permeability, which are key parameters in the coupled thermal-hydro-mechanical processes. This effective parameter can then be used to represent the overall response of a heterogeneous material to external forces or changes in conditions at the macroscale. The simplest way to determine this is to quantify the proportion of each phase or constituent within the material. These volumetric fractions are essential for calculating effective properties.

To demonstrate the mixture theory, a three-phase system in a porous medium with different thermal, hydraulic and mechanical properties is considered. Figure 7 shows the principle of the mixture theory, as an example, for hydraulic permeability in the x-direction. The permeability of the total system can be calculated based on the mass balance equation for flow (2).

$$Q_x = \sum_i Q_x^i = \sum_i v_x^i A_x^i = \sum_i K_{xi} \frac{\Delta P}{\Delta x} L_i B = K_x L \frac{\Delta P}{\Delta x} B,\ i = 1,2,3\ldots \tag{2}$$

where Q is the flow rate; v is the flow velocity; $A = L \times B$ is the area to be flowed through, indicating the volumetric fraction, with B representing the thickness of the area; K is the permeability; P is the pressure; and $\Delta P/\Delta x$ is the pressure gradient in the x-direction. The index i indicates the ith subsystem.

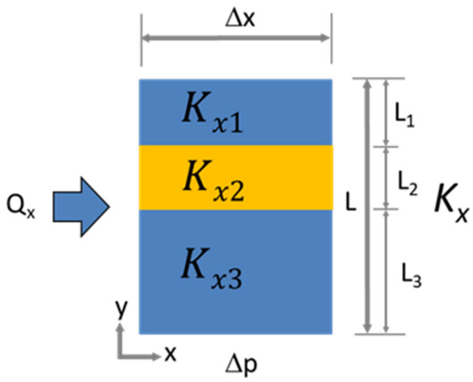


Figure 7. Mixture principle for flow parameter in the x-direction.

Simplifying Equation (2) results in

$$\sum_i K_{xi} L_i = K_x L \tag{3}$$

Equation (3) represents the rule of mixture for the permeability and the thermal conductivity in the x-direction, and the elastic modules in the y-direction. Similarly, the permeability and the thermal conductivity in the y-direction and the elastic modules in the x-direction can be calculated using the inverse rule of the mixture (4).

$$\sum_i \frac{L_i}{K_{yi}} = \frac{L}{K_y} \tag{4}$$

The mixture theory for microscale discontinuity provides a valuable framework for understanding and modeling the behavior of materials with heterogeneous microstructures. Using the effective properties calculated by Equations (3) and (4), thermal, hydraulic and mechanical processes in a microscale structure can be simulated using numerical methods, such as finite element analysis (Figure 3). The most important challenge may involve the understanding and modeling of interfaces between different phases. The behavior at these interfaces can have a significant impact on the overall properties of the material. Due to the complexity of interactions between phases at the microscopic scale and the lack of precise experimental data, these interactions are currently only speculative.

5. Applications

5.1. Gas Migration in Clayey Materials

Gas migration in clayey materials is a crucial aspect in assessing the long-term performance of nuclear waste repositories. The behavior of gases—especially under the influence of factors such as dilatancy—is a challenge, which requires a sophisticated understanding of both micro- and macroscopic processes. The use of numerical models to simulate gas migration in clayey materials is essential for clarifying the complex interrelationships in repository systems.

In this example, gas migration is governed by dilatancy-controlled processes in water-saturated clay materials. Dilatancy—the increase in volume of a material due to an increase in porosity—significantly influences the permeability and gas transport properties of the material. Understanding how these microscopic changes manifest themselves on a macroscopic scale is crucial for the development of accurate and reliable numerical simulations. In contrast to classical THM coupled simulations, the simulation presented here uses additional methods and concepts to simulate dilatancy-controlled fluid transport in clayey, expansive materials. The concepts are illustrated in Figure 8 and shortly explained in the following section.

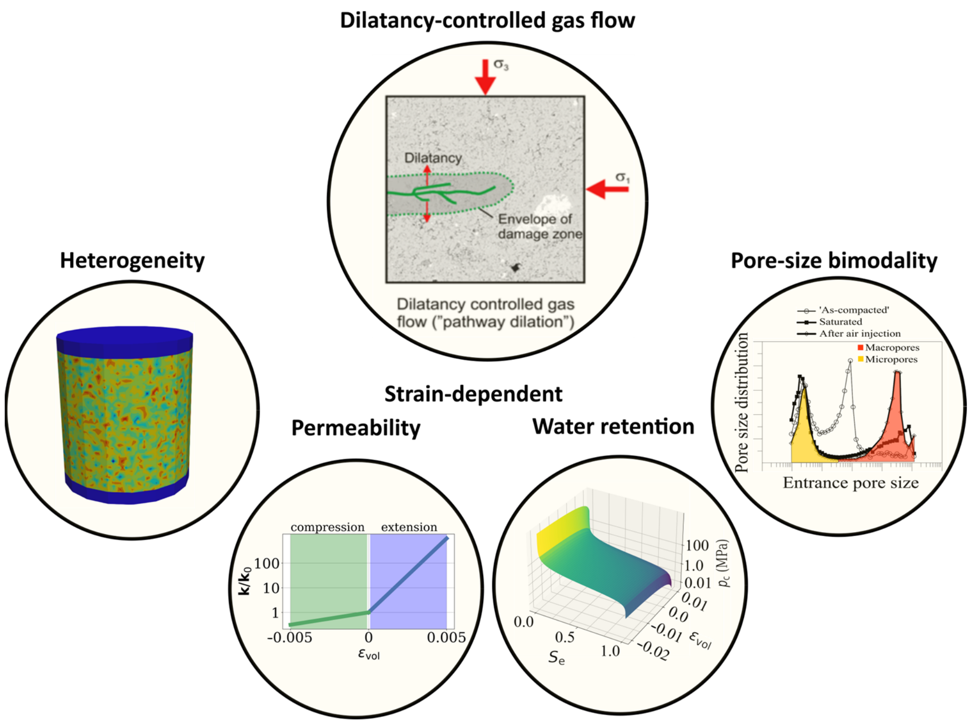


Figure 8. Illustration of the applied methods. Combined approaches for increasing the reliability of numerical models to represent dilatancy-controlled gas flow in clayey materials. The concept uses heterogeneity, strain-dependent permeability and water retention, in which permeability (**k**) and capillary pressure (p_c) are dependent on volumetric strain (ϵ_{vol}) and the pore size bimodality of clays.

The numerical treatment of gas migration in clayey materials goes beyond conventional approaches by incorporating stochastic parameter distributions to account for microscale heterogeneity. The microscale heterogeneity is then normalized to REV size to create a finite element mesh. The microscale heterogeneity is complemented by simple micromechanics, where strain-dependent permeability change and water retention are simulated based on a developed bimodal porosity approach typical for expansive bentonite [27]. A numerical model using these methods simulates a bentonite gas flow test. The conceptual model, based on a laboratory experiment in Ref. [30], consists of a bentonite sample enclosed on two sides by a filter material (Figure 9, left). A heterogeneous distribution of Young's modulus is derived from MIP data for the bentonite sample (Figure 9, right).

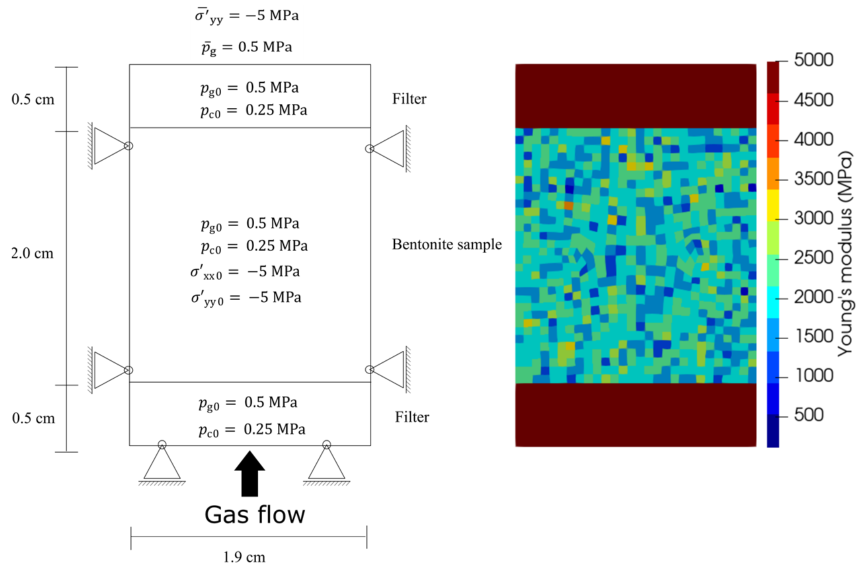


Figure 9. Conceptual model of a bentonite gas flow test (**left**), with initial and boundary conditions (with p_{g0} and p_{c0} representing initial gas and capillary pressure; σ'_{xx0} and σ'_{yy0} representing initial effective radial and axial stress; and $\overline{p}_g$ and $\overline{\sigma}'_{yy}$ representing boundary conditions for gas pressure and axial stress). A heterogeneous distribution of Young's modulus derived from MIP data for the bentonite sample was randomly distributed (**right**).

The case study on gas migration in clayey materials serves as a practical demonstration of the presented concept and shows the application of advanced numerical techniques to improve the understanding of coupled THMC processes. The knowledge gained from this application contributes to the broader objective of comprehensively assessing the long-term performance of nuclear waste repositories.

By applying stochastic measures combined with microstructural model approaches, preferential gas flow is simulated in a 2D axis-symmetric finite element model (Figure 10). The hydromechanical coupled model simulates an increase in macroporosity (Figure 11). The macroporosity has different material properties than the microporosity regarding permeability and water retention. An increase in macroporosity is simulated based on varying mechanical strain. After the gas breakthrough, the macroporosity reduces due to an elastic reduction in mechanical strain.

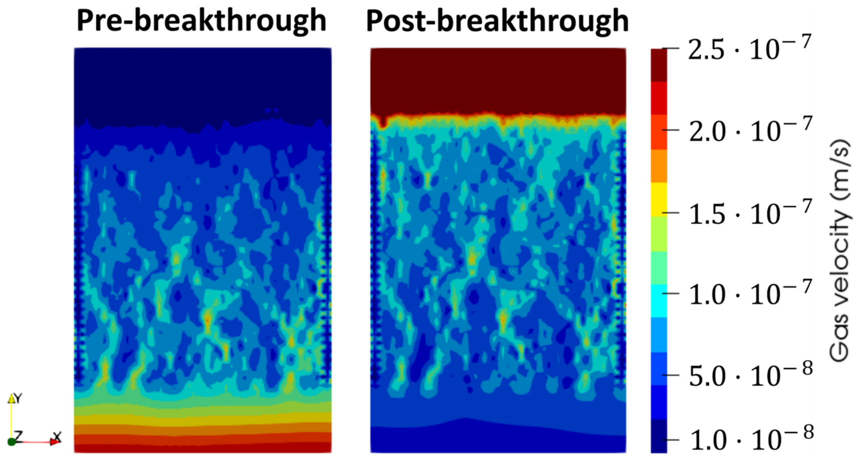


Figure 10. Contour plots of simulated gas flow through saturated bentonite before and after gas breakthrough.

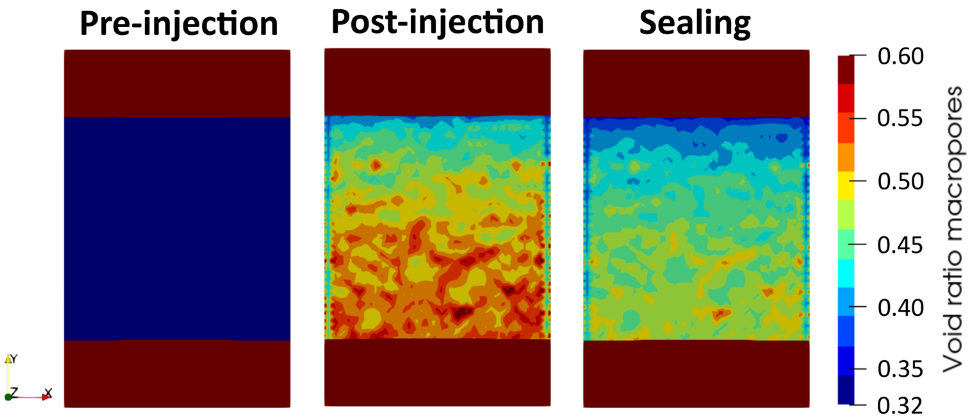


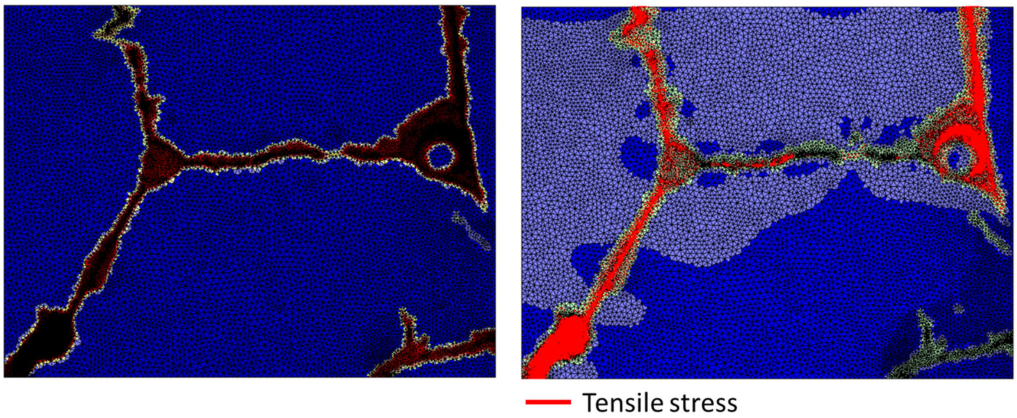
Figure 11. Calculated and simulated void ratio of macroporosity based on a micromechanical model.

5.2. Fluid Migration in Salt under Thermal, Hydraulic and Mechanical Gradients

Fluid can migrate if permeability exits. Rock salt is often considered impermeable or to have a permeability, which is unmeasurably low due to its creep deformation during formation. In order to construct an underground facility for a repository for high-level radioactive waste, shafts, tunnels and boreholes are excavated. Stress redistribution after excavation leads to the formation of an excavation damaged/disturbed zone (ED/dZ) in the near field of an opening, with significant changes in the hydraulic and mechanical properties. Further changes in properties can occur under thermal load during the heating period in the post-closure phase. Initially, more or less randomly distributed intragranular and intergranular fluid in low-permeability bedded or domed salt can then be mobilized and migrate toward the openings at a potentially significant rate under the altered hydro-mechanical and the coupled thermo-hydro-mechanical-chemical conditions.

In an underground facility in a salt dome in northern Germany, hydrocarbons are occasionally found on the tunnel surface and in the boreholes. To characterize the occurrence of hydrocarbons, twenty boreholes were drilled at locations where hydrocarbons were visible with either conventional artificial light or ultraviolet light [30]. All boreholes were equipped with a specially developed dual-line packer system in order to collect the gaseous and liquid hydrocarbons separately from the interval outside of the excavation damaged zone (EDZ) of the gallery [31]. Pressure build-up measurements were conducted for more than two years.

Microscopic investigations show that the void space with fluid inclusions along a grain boundary is not connected in a permeable condition [12,32]. However, the relatively low tensile strength of such zones compared to those of halite grains may lead to an opening of a pathway under the high fluid pressures associated with the imposed deviatoric stress state. Microscale modeling is based on the mixture theory, using two materials representing halite grain and the grain boundary to demonstrate how fluid inclusions can flow through the new pathway (Figure 12a) created by tensile stress (Figure 12b). In a study by Shao et al. [33], it was demonstrated that the rapid gas pressure build-up can be facilitated by the calculated high fluid velocity.



(**a**) (**b**)

Figure 12. Grain boundary simulation (model area 1 × 0.8 mm): Water with high pressure is located along the grain boundary (**a**) and distribution of σ1 and tensile stress vector (**b**).

Different positions in the near field of an opening (Figure 3) are subjected to different stress conditions. The farther away from the opening, the lower the deviatoric stress. Under the same initial hydraulic pressure conditions, different damaged or disturbed zones can be characterized. Since fluid inclusions do not occur continuously, the channel will close after the release of local inclusions, as long as no further fluid flows from inclusions under high pressure. In this case, the interval pressure remains constant and only increases again when other inclusions become active and are eventually released into the borehole. This analysis indicates that the fluid release in a borehole is inherently episodic and spatially localized, and it is accompanied by a dynamic microstructural adjustment of the salt [34].

Using a modified EDdZ model with different permeability values (permeability in EDZ up to 10^{-17} m^2, in EdZ 10^{-20} m^2, and in the large EDdZ model, a zone with a slightly increased permeability of 2×10^{-22} m^2), 430 pressure build-ups during a 938-day measurement period can be well predicted (Figure 13). In summary, the locally distributed high-pressure hydrocarbon with an increased permeability of four or five orders of magnitude at the grain boundary can flow as a microscale flow path. This study confirms the possible mechanism of pathway dilation due to the high fluid pressure of fluid inclusions under an excavation-induced deviatoric stress condition.

To investigate the fluid migration behavior under thermal load in salt, a test program called the brine availability test in salt (BATS) was carried out by Sandia National Laboratories (SNL) in the underground facility Waste Isolation Pilot Plant (WIPP), Albuquerque, USA [35]. In BATS, the thermal pressurization in the heated array is a significant process, which leads to an increase in the inflow rate compared to the unheated array. The inflow rate is up to four times higher under the thermal load than without temperature influence, which is due to the thermal expansion of solids and fluids [22], as well as an increase in the solubility of the salt in the liquid phase [36–39].

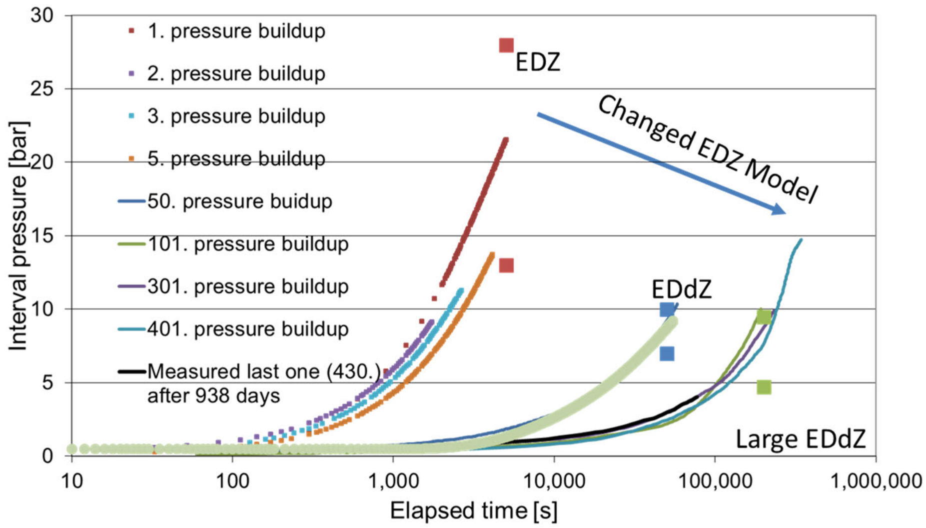
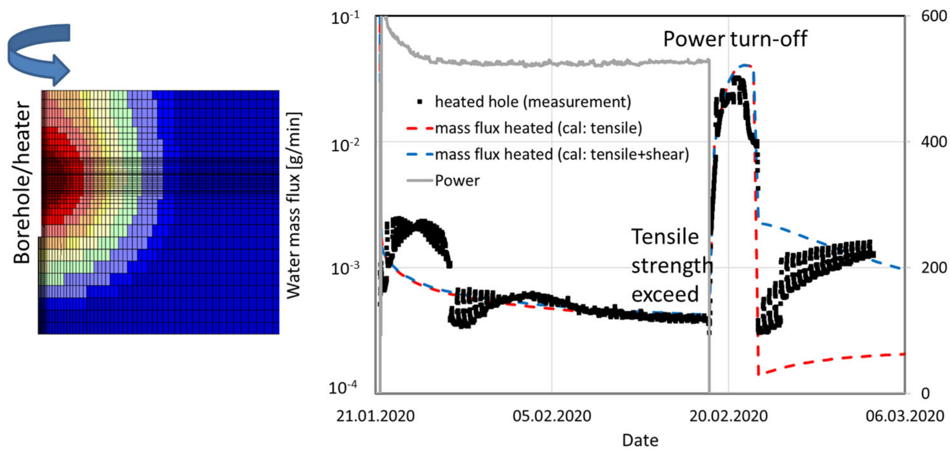


Figure 13. The changed EDZ model for the pressure build-ups.

As the power was turned off, a sudden increase in the inflow rate into the borehole was measured, which is referred to as a "spike" and is about two orders of magnitude. This "spike" is not relevant to the final disposal of radioactive waste, but it has particular significance for understanding the underlying processes. Similar behavior was also found in domed rock salt [40]. Conventional numerical models based on the continuum concept cannot predict this type of "spike", or only to a limited extent. The conceptual model based on the mixture theory, however, considers two continua at the micro scale: one represents the crystal matrix, and the other represents the brine-filled pore space, e.g., inter-/intragranular brine as total.

Due to the different thermal properties of these two materials (thermal conductivity for salt = 5.5 W/K/m and brine = 0.6 W/K/m), a sudden turning off of the heater leads to rapid temperature drop in the salt crystal, while the fluids continue to have a high temperature and therefore a high hydraulic pressure. During cooling, a contraction process occurs, leading to tensile stresses between particles or solid particles and fluid. This tensile stress can be higher than the tensile strength, resulting in the connectivity between microcracks with increased permeability. The permeability can be two orders of magnitude higher than the original one. This high permeability is responsible for the "spike" increase in the inflow (Figure 14).



(**a**) (**b**)

Figure 14. Radial model with temperature distribution for the heated borehole (**a**) and the comparison between calculated and measured water flux (**b**).

6. Conclusions

The predictive capabilities of current numerical models are limited in simulating the flow of fluids through geomaterials with low permeability (bentonite, claystone, salt) for long-term performance assessment studies for nuclear waste repositories. It is assumed that this is due to microscopic material heterogeneous properties and micromechanics, which may have strong effects on macroscopic fluid migration. The numerical simulation of solute transport in a fracture–matrix model clearly showed that measurements with a smaller range (10 m) are difficult to replicate due to a higher heterogeneity in the fracture network. With increasing distance (70 and 100 m), a better agreement between measurements and numerical results was achieved. This can be attributed to the reduced uncertainty of fracture networks in larger areas, as any outliers in smaller areas have less influence on the average permeability of the measured fracture network as the area increases.

It has to be taken into account that all materials are microscopically heterogeneous. In this study, some important characteristics of microscopic effects on macroscopic observations are exemplarily presented, e.g., dilatancy-controlled gas flow through clayey media and EDZ-related fluid transport in salt. These examples collectively demonstrate how microscopic solid variations influence macroscopic fluid flow. They emphasize the importance of considering scale-dependent heterogeneity and conducting microscale studies, highlighting the role of small-scale heterogeneities in the formation of fracture networks and their influence on system behavior. Microscale measurements using advanced analytical techniques, such as optical microscopy and electron backscattering, provide insights into the spatial variability, which is often overlooked in macroscopic interpretations.

The simulation of gas flow through an expansive bentonite sample uses different microscopically derived methods with strain-dependent water retention and permeability, heterogeneity and a bimodal pore size to simulate dilatancy-controlled gas flow. The numerical results show that microscopic hydro-mechanical behavior, such as preferential pathways and an increase in macroporosity, can be simulated. However, the determination of the values of the microscopically derived parameters is still subject to some uncertainty, also due to difficult measurement conditions.

Various numerical simulations of fluid flow through initially impermeable salt showed that microscopic strain may strongly influence the flow velocity. The constitutive models for EDZ and laboratory observations influenced the numerical models. As a result, a high-brine mass flow was predicted of about 100 times the initial flow rate, which had a good agreement with the in situ measurements.

The simulations show the relevance of research in practical scenarios, such as gas migration in clayey material and fluid migration in salt under different hydraulic, mechanical and thermal gradients. The numerical treatment of gas migration includes stochastic parameter distributions and micromechanics, which provide a nuanced understanding of dilatancy-controlled processes. The study of fluid migration in salt reveals the complexity of microstructural adjustments, episodic fluid releases and the potential impact on pressure build-up, and it demonstrates the practical implications of the research for real reservoirs. In summary, some important points can be formulated as follows:

- Microscale processes, such as fluid flow through materials with low permeability but low stiffness, can have a large impact on macroscale simulations.
- Microscale processes are usually smaller than the REV and need to be scaled up to the REV scale, taking into account stochastic features and governing principles, such as the mixture theory.
- Deterministic systems with heterogeneous distributions of material properties can be used to improve process understanding; simulation series with different stochastic distributions should be created for comprehensive safety calculations.

The comprehensive nature of this research, which integrates experimental, analytical and numerical approaches, taking into account microscale investigation of dynamic processes, contributes to improving our understanding of scale-dependent mass transport and heterogeneity in porous media. A promising example will be the planned project

"Hydro-micromechanics of clay rocks" within the framework of DECOVALEX-2027 [40,41]. The aim of the project is to understand the processes and mechanisms at the microscale governing the damage/deformation/dilatancy behavior of argillaceous rocks and the influence of this behavior on gas and water migration. These results will definitely have practical implications for the long-term safety assessment of radioactive waste repositories and provide a solid basis for further research and application in this field.

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References

1. Bear, J.; Tsang, C.F.; de Marsily, G. *Flow and Contaminant Transport in Fractured Rock*; Elsevier: Amsterdam, The Netherlands, 1993; ISBN 978-0-12-083980-3.
2. Kinzelbach, W. *Numerische Methoden zur Modellierung des Transports von Schadstoffen im Grundwasser*; Wasser·Abwasser 21 (RERO)1876869-41slsp 2. Aufl.; Oldenbourg: München, Germany, 1992.
3. Liedtke, L.; Shao, H.; Kull, H. Investigation of Two-Phase Flow in Fractured Rock. WM'98 Session: Status and Future Plans for Deep Underground Research and R&D Laboratories. 1999. Available online: https://archivedproceedings.econference.io/wmsym/1998/html/sess27/27-24/27-24.htm (accessed on 20 January 2024).
4. Birkholzer, J.T.; Tsang, C.F.; Bond, A.E.; Hudson, J.A.; Hudson, J.A.; Jing, L.; Stephansson, O. 25 years of DECOVALEX—Scientific advances and lessons learned from an international research collaboration in coupled subsurface processes. *Int. J. Rock Mech. Min. Sci.* **2019**, *122*. [CrossRef]
5. SKB The Äspö Hard Rock Laboratory. Available online: www.skb.com (accessed on 20 December 2023).
6. Nagra Felslabor Grimsel. Available online: www.nagra.ch (accessed on 20 December 2023).
7. Mont Terri Project. Available online: www.mont-terri.ch (accessed on 20 December 2023).
8. ANDRA. Available online: https://international.andra.fr (accessed on 20 December 2023).
9. Tsang, C.F.; Stephansson, O.; Jing, L.; Kautsky, F. DECOVALEX project: From 1992 to 2007. *Environ. Geol.* **2009**, *57*, 1221–1237. [CrossRef]
10. Birkholzer, J.T.; Bond, A.E. DECOVALEX-2019: An international collaboration for advancing the understanding and modeling of coupled thermo-hydro-mechanical-chemical (THMC) processes in geological systems. *Int. J. Rock Mech. Min. Sci.* **2022**, *154*, 105097. [CrossRef]
11. EURAD: Eurad Projects. Available online: https://www.ejp-eurad.eu/publications (accessed on 23 December 2023).
12. Thiemeyer, N.; Pusch, M.; Hammer, J.; Zulauf, G. Quantification and 3D visualisation of pore space in Gorleben rock salt: Constraints from CT imaging and microfabrics. *Z. Dt. Gers. Geowiss* **2013**, 15–25. [CrossRef]
13. Thiemeyer, N.; Zulauf, G.; Mertineit, M.; Linckens, J.; Pusch, M.; Hammer, J. Microfabrics and 3D grain shape of Gorleben rock salt: Constraints on deformation mechanisms and paleodifferential stress. *Tectonophysics* **2016**, *676*, 1–19. [CrossRef]
14. Akono, A.-T.; Werth, C.; Shi, Z.; Jessen, K.; Tsotsis, T.T. Advanced Geomechanical Model to Predict the Impact of CO_2-Induced Microstructural Alterations on the Cohesive-Frictional Behavior of Mt. Simon Sandstone. *Minerals* **2021**, *11*, 38. [CrossRef]
15. Al-Gharbi, M.S. Dynamic Pore-Scale Modelling of Two-Phase Flow. Ph.D. Thesis, Imperial College London, London, UK, 2004.

16. Catalano, E.; Chareyre, B.; Cortisy, A.; Barthelemy, E. A pore-scale hydro-mechanical coupled model for geomaterials. In Proceedings of the PARTICLES II International Conference on Particle-Based Methods—Fundamentals and Applications, Barcelona, Spain, 26–28 October 2011; Onate, E., Owen, D.R.J., Eds.; CIMNE: Barcelona, Spain, 2011.
17. Ferrari, A. Pore-Scale Modeling of Two-Phase Flow Instabilities in Porous Media. Ph.D. Thesis, University Lausanne, Lausanne, Switzerland, 2014.
18. Bear, J.; Cheng, A.H.-D. *Modelling Groundwater Flow and Contaminant Transport*; Textbook; Springer: Berlin/Heidelberg, Germany, 2010.
19. Himmelsbach, T.; Shao, H.; Wieczorek, K.; Flach, D.; Schuster, K.; Alheid, H.-J.; Liou, T.-S.; Bartlakowski, J.; Krekler, T. *Effective Field Parameter EFP*; NTB 03-13; Nagra: Wettingen, Switzerland, 2013.
20. Bossart, P. 10 Year MT Report. Swisstopo, Switzerland, 2006.
21. Thiemeyer, N.; Habersetzer, J.; Peinl, M.; Zulauf, G.; Hammer, J. The application of high resolution X-ray computed tomography on naturally deformed rock salt: Multi-scale investigations of the structural inventory. *J. Struct. Geol.* **2015**, *77*, 92–106. [CrossRef]
22. Shao, H.; Wang, Y.F.; Kolditz, O.; Nagel, T.; Brüning, T. Approaches to multi-scale analyses of mechanically and thermally-driven migration of fluid inclusions in salt rocks. *Phys. Chem. Earth* **2019**, *113*, 1–13. [CrossRef]
23. Chajed, S.; Singh, A. Acoustic Emission (AE) Based Damage Quantification and Its Relation with AE-Based Micromechanical Coupled Damage Plasticity Model for Intact Rocks. *Rock. Mech. Rock Eng.* **2024**, *57*, 2581–2604. [CrossRef]
24. Damians, I.P.; Olivella, S.; Gens, A. Modelling gas flow in clay materials incorporating material heterogeneity and embedded fractures. *Int. J. Rock Mech. Min. Sci.* **2020**, *136*, 104524. [CrossRef]
25. Rodriguez-Dono, A.; Zhou, Y.; Olivella, S. A new approach to model geomaterials with heterogeneous properties in thermo-hydro-mechanical coupled problems. *Comput. Geotech.* **2023**, *159*, 105400. [CrossRef]
26. Radeisen, E.; Shao, H.; Hesser, J.; Kolditz, O.; Xu, W.; Wang, W. Simulation of dilatancy-controlled gas migration processes in saturated bentonite using a coupled multiphase flow and elastoplastic H2M model. *J. Rock Mech. Geotech. Eng.* **2023**, *15*, 803–813. [CrossRef]
27. Radeisen, E.; Shao, H.; Hesser, J.; Naumov, D.; Wang, W.; Kolditz, O. Modelling of preferential gas flow in saturated bentonite using a bimodal, strain-dependent pore model. *Appl. Clay Sci.* **2024**, *249*, 107232. [CrossRef]
28. Seiphoori, A. *Thermo-Hydro-Mechanical Characterisation and Modelling of Wyoming Granular Bentonite*; Nagra Technical Report Issue; Nagra: Wettingen, Switzerland, 2015.
29. Radeisen, E.; Shao, H.; Pitz, M.; Hesser, J.; Wang, W. Derivation of heterogeneous material distributions and their sensitivity to HM-coupled two-phase flow models exemplified with the LASGIT experiment. *Environ. Earth Sci.* **2023**, *82*, 347. [CrossRef]
30. Gonzalez-Blanco, L.; Marschall, P.; Romero, E. Gas transport in granular compacted bentonite: Coupled hydro-mechanical interactions and microstructural features. *E3S Web Conf.* **2020**, *195*, 04008. [CrossRef]
31. Hammer, J.; Pusch, M.; Ostertag-Henning Ch Haeger, A.; Schlömer, S.; Scheeder, G.; Shao, H.; Paul, B.; Kus, J.; Hesser, J.; Blumenberg, M. Untersuchung von Kohlenwasserstoffen im Erkundungsbergwerk Gorleben; BGR-Bericht; 2018.
32. Paul, B.; Shao, H.; Hesser, J.; Ostertag-Henning, C.; Lege, C. *In-Situ Quantification of Hydrocarbon in an Underground Facility in Tight Salt Rock, Engineering Geology for Society and Territory*; Springer: Berlin/Heidelberg, Germany, 2015; Volume 6, pp. 893–896.
33. Shao, H.; Wang, Y.F.; Kolditz, O.; Nagel, T.; Yoshioka, K. Determination of permeability for hydrocarbon release due to excavation-induced stress redistribution in rock salt. *Int. J. Rock Mech. Min. Sci.* **2020**, *136*, 104525. [CrossRef]
34. Shao, H.; Hesser, J.; Wang, W.Q.; Kolditz, O. Modelling thermally-driven migration of fluid inclusions in rock salt. *Geomech. Energy Environ.* **2024**, *30*, 100542. [CrossRef]
35. Kuhlman, K.L.; Mills, M.; Jayne, R.; Matteo, E.N.; Herrick, C.; Nemer, M.; Heath, J.; Xiong, Y.; Choens, C.; Stauffer, P.; et al. *FY20 Update on Brine Availability Test in Salt*; SAND2020-9034R; Sandia National Laboratories: Albuquerque, NM, USA, 2020.
36. Anthony, T.R.; Cline, H.E. Thermal migration of liquid droplets through solids. *J. Appl. Phys.* **1971**, *42*, 3380–3387. [CrossRef]
37. Müller, E.; Zschätsch, B. Migration of brine inclusions through grain boundaries in rock salt. *Cryst. Res. Technol.* **1985**, *20*, K1–K3.
38. Roedder, E. The fluids in salt. *Am. Mineral.* **1984**, *69*, 413–439.
39. Rothfuchs, T.; Wieczorek, K.; Feddersen, H.K.; Staupendahl, G.; Coyle, A.J.; Kalia, H.; Eckert, J. Brine Migration Test; Final Report, GSF-Bericht 6/88; 1988.
40. DECOVALEX. Available online: https://decovalex.org/ (accessed on 1 March 2024).
41. Vo, U.; Fall, M.; Sedano, J.A.E.; Nguyen, T.S. A Multiphysics Model for the Near-Field Evolution of a Geological Repository of Radioactive Waste. *Minerals* **2023**, *13*, 1535. [CrossRef]