State Of the Art Report in the fields of numerical analysis and scientific computing. Final version as of 16/02/2020 deliverable D4.1 of the HORIZON 2020 project EURAD.

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Work Package DONUT

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Executive Summary

This document provides an assessment of the current state-of-the-art for the DONUT work package. It is intended as a reference for the involved actors in EURAD and will be updated at the end of the project as new information becomes available. The prupose of this work package is to improve/develop methods or numerical tools in order to go a step further in development of (i) relevant, performant and cutting-edge numerical methods that can easily be implemented in existing or new tools, in order to carry out high-performance computing to facilitate the study of highly coupled processes in large systems. These methods and their implementation in tools will be mainly applied to reactive transport, 2-phase flow, and THM modelling in porous and fractured media; (ii) numerical scale transition schemes for coupled processes (meso1 to macro scale), supporting the study of specific multi-scale couplings such as chemo-mechanics; (iii) innovative numerical methods to carry out uncertainty and sensitivity analyses coupled processes.

The readers have to keep in mind that this report is not reviewing all the existing codes, methods or tools that are available in the literature. It is rather written and oriented in the perspective of the research program that will be conducted by the different partners within DONUT. While some general statement are given, the research direction that will be followed by partners are outlined.

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1. Introduction

Demonstrating safety of a nuclear waste disposal over time comparable to geological timescales relies on a rigorous, complex and iterative scientific approach referred to as “long-term behavior science” that is based upon three pillars: experiments, modeling, and natural/archaeological analogs (see Bildstein et al (2019) for a review). In that context, improvement of numerical methods and tools for modelling multi-physical Thermo-Hydro-Mechanical-Chemical coupled processes (THMC) occurring in radioactive waste disposal is a major and permanent issue to support its optimization of design. Indeed, numerical simulations are necessary either to make predictive multi-physical assessments, at time frames and space scales larger than experiments can cover,or to model the experiment itself to verify, calibrate and improve numerical models. These numerical simulations require integrating, in a consistent framework, an increasing scientific knowledge acquired for each of the individual components of a system for radioactive waste disposal. This implies to consider couplings of different and non-linear processes, applied to a wide range of materials with contrasting properties as a function of time and space in ever-larger systems.

In this context, the development of cutting-edge and efficient numerical methods is thus necessary, in the scope of development and use of powerful and relevant numerical tools for assessments. In parallel, it is also necessary to manage the uncertainties associated to the input data that feed the models, and to the representation of the processes, in order to assess the range of variability of the results and to identify the main parameters and processes that drive the behaviour of the systems of interest. Managing uncertainties in these complex systems requires the improvement and the development of innovative, appropriate and efficient methods.

Within the EURAD joint programming initiative, the work package **DONUT** (**D**evelopment and Improvement **O**f **NU**merical methods and **T**ools for modelling coupled processes) ambitions to go a step further in the development:

- of relevant, performant and cutting-edge numerical methods that can easily be implemented in existing or new tools, in order to carry out high-performance computing to facilitate the study of highly coupled processes in large systems. These methods and their implementation in tools will be mainly applied to reactive transport, 2-phase flow, and THM modelling in porous and fractured media;
- of numerical scale transition schemes for coupled processes (meso[1] to macro scale), supporting the study of specific multi-scale couplings such as chemo-mechanics;
- of innovative numerical methods to carry out uncertainty and sensitivity analyses;

This report describes briefly the starting point of the concept that will be developed within DONUT. Within DONUT a task is also devoted to the set-up and the achievement of benchmark exercises, on representative test cases, to test the efficiency of developed methods (robustness, accuracy, time computational) on relevant tools. This part is not developed in the present report as the benchmark are not yet existing and will be develop in 2020. However we would like to recall here that we are aware (and even taking part) of existing benchmark initiative like DECOVALEX (Birkholzer, 2019) or reactive transport benchmarks for subsurface environmental simulation (Lagneau and Van Der Lee, 2010; Steefel, Yabusaki, et al., 2015). Of course the knowledge gained within such exercise will be used to set up efficient and relevant benchmarks.

2. *Numerical methods for high performance computing of coupled processes.*

Task 2 of the DONUT work-package is devoted to the development of new methods and new tools to efficiently solve numerically highly coupled problems arising in the context of waste repository management. In Section 2.1, we briefly comment on the coupled processes to be considered in this task. Then in Section 2.2, we present the foundations of the numerical methods to be developed within this task. Finally, Section 2.3 is devoted to the presentation of the existing tools (or codes) that will serve as a basis for further developments.

2.1 Coupled processes

This section is devoted to the description of the physical models to be studied

2.1.1 Spatial coupling of gas free flows and two-phase porous media flows

Flow and transport processes in domains composed of a porous medium and an adjacent free-flow region appear in a wide range of industrial and environmental applications. This is in particular the case for radioactive waste deep geological repositories where such models must be used to predict the mass and energy exchanges occurring at the interface between the repository and the ventilation excavated tunnels. Typically, in this example, the porous medium initially saturated with the liquid phase is dried by suction in the neighborhood of the interface. To model such physical processes, one needs to account in the porous medium for the flow of the liquid and gas phases including the vaporization of the water component in the gas phase and the dissolution of the gaseous components in the liquid phase. In the tunnel, a single phase gas free flow can be considered assuming that the liquid phase is instantaneously vaporized at the interface. This single phase gas free flow has to be compositional and non-isothermal to account for the change of the relative humidity and temperature in the tunnel which has a strong feedback on the liquid flow rate at the interface.

If many works have been performed to model and discretize the coupling of single phase Darcy and free flows, there is very little work on the coupling of a two phase gas liquid compositional Darcy flow with a single phase compositional free flow. Such a coupled model has been recently proposed in Mosthaf et al. (2011) and Vanderborght et al. (2017) using proper matching conditions at the interface between the porous medium and the free flow regions. This model will be the starting point of our work and will be simplified taking into account the physical characteristics of our problem focusing on the drying processes at the interface between the nuclear waste repository and the ventilation excavated tunnels.

To obtain our simplified model, we will assume that the longitudinal dimensions of the tunnels are large compared with their diameters allowing to reduce the model in the tunnel to a 1D free flow. In the spirit of Birgle et al. (2018) and Beaude et al. (2019), we will extend to the non-isothermal case the approach proposed in Brenner et al. (2016).

2.1.2 Non-zero gas entry pressure and hysteresis

The study the performance of the host rock and engineering barriers materials (seals like bentonite, backfill…) is crucial for radioactive waste confinement in a deep geological disposal (e.g., safety case of CIGEO project developed by ANDRA, France). Reliable models are needed for water and gas transport in a clay host rock formation (COx: Callovo-Oxfordian), and in any clay-based engineered materials used as barriers in the disposal, as well as their impact on radionuclide transport within the disposal. There is a substantial experimental evidence showing the existence of non-zero gas entry pressure and hysteresis in hydraulic properties of these materials (Amri 2018), but their modelling by exact mathematical formulations, and their impact on two-phase (water-gas) flow have never been studied. The impact of non-zero gas entry pressure and hysteresis on maximal gas pressure and on desaturation range around the repository during the gas transitory period also has to be assessed. IRSN started to study the effect of air-entry pressure first at smaller scale (dozen of meters and several days). Therefore, the work focused on ANDRA PGZ-experiment where data of both measured COx-hydraulic properties, and measured gas (N_2) pressure inside a borehole are available. Data of the COx-hydraulic properties measured have been collected from literature and were analyzed in an innovative way, by introducing an adequate van Genuchten-Mualem (VGM) model accounting for air-entry pressure (Vogel et al., 2001; Amri et al., 2020).

2.1.3 Thermo-hydro-mechanics for porous media flows

We shall start with the analysis of hydro-mechanical (HM) processes, which occurs in rocks as well as in buffer and sealing materials in potential deep underground nuclear waste repository. The basic model is the Biot's type poroelasticity, which couples linear elasticity and Darcy's flow in saturated media. Even this simplest HM model shows that there are important questions about the robustness of the discretization (in space and time) and efficient solution of the arising linear systems. Those points will be discussed in further details in Section 2.2. Many applications require to solve HM problems in porous media with fractures. The flow can be modelled by a system which combines porous media flow in the porous matrix with the flow in fractures, which are modelled as lower-dimensional objects (Martin et al., 2005; B rezina and Stebel, 2015; Berre et al., 2019). This approach was consistently applied in the software Flow123d developed by TUL (B rezina et al., n.d.) for the solute and heat transport and (un)saturated Darcy flow. The mechanical part is frequently solved by penalty (Garipov et al., 2016), used for the ease of implementation, but the need for accuracy can make the resulting stiffness matrix severely ill-conditioned, making difficulty for solvers. It motivates the use of Lagrange multipliers for avoiding penetration, see e.g. Franceschini et al. (2016). Even in the simplest setting, the HM problem in continuum with fractures involves two types of nonlinearity due to dependence of fracture conductivity on aperture, non-penetration condition and proper techniques have to be adopted for the solution. Further extension was done in Ganis et al. (2014) where the poroelasticity model is considered on the continuum accompanied by the flow model on the fractures. The similar model but including the contacts on fractures, a fracture growth model and using the extended finite elements is considered in Khoei (2014). Recently the same model but using a conforming FEM – FV discretization and discussing parallelization was described in Settgast et al. (2017). To the best of our knowledge, there is no work dealing with the full continuum-fracture poroelastic model with the consistent elasticity model on fractures. This case is particularly important the context of waste repository where fracture domain is used to describe larger fracture zones, which are nonempty and can be considered as a continuum with diminished strength.

Thermal effects also induce important effect than can often not be neglected, requiring the use of thermo-hydro-mechanical (THM) coupled models. The research teams at CU and CTU have developed and numerically implemented a coupled multiscale THM model suitable for describing the behaviour of expansive soils under non-isothermal conditions (Mašın, 2017). In particular, the model is intended to be applied to bentonite buffers in deep geological nuclear waste repositories. The model uses the theory of soil hypoplasticity and a double-structure approach (Alonso et al., 1999) – backed by various micro-mechanical studies – in which soil macrostructure (silt-size aggregates of clay particles) and microstructure (internal structure of the aggregates) are treated separately. Four coupled components are thus described: a mechanical model for the microstructure, a hydraulic model for the microstructure, a mechanical model for the macrostructure, and a hydraulic model for the macrostructure. These models are coupled at each structural level, and the behavior of the two structural levels is linked through the double-structure coupling function (Mašın, 2013). In particular, the mechanical behaviour of the macrostructure accounts for partial saturation, and its hydraulic response is based on a void ratio-dependent water retention model (improvements to this model will be proposed during the project). The microstructure is considered as fully saturated, with its mechanical behavior being reversible volumetric, governed by the Terzaghi effective stress principle. This simplification has proven to limit the model performance in some cases, such as during oedometric swelling; therefore, an improved description of the microstructural behaviour will be sought during the project.

Each of the structural levels responds differently to temperature change, suction change, and mechanical action. The developed model thus considers a temperature dependence for all four model components. As for the mechanical behavior of the macrostructure, temperature influences the position of the normal compression lines of a soil. The normal compression line represents the maximum void ratio of the soil at the given mean stress and temperature; therefore, a heating-induced shift in the position of the normal compression line implies heating-induced soil compaction of loosely compacted soils (Mašın, 2017). This effect is considered in the model by assuming that the asymptotic state boundary surface depends on temperature. Temperature also affects the mechanical response of the microstructure. It influences both the

basal spacing and thus the amount of crystalline water and also the diffuse double-layer thickness. Depending on conditions and clay type, the combinations of these effects can result in either swelling or contraction. This is accounted for in the model by a single parameter, calibrated through experiments.

In the water retention behavior, it is difficult to distinguish experimentally the response of the macrostructure and the microstructure. Because the microstructure may be considered saturated up to large values of suction. temperature-induced microstructural swelling also implies an increase in water retention capacity, and microstructural shrinkage implies a decrease in water retention capacity. Therefore, a simplified approach is chosen, in which the effect of temperature on water retention capacity of the macrostructure is considered to depend solely on the change in surface tension of water with temperature, without considering the effect of variable grain-water interface contact angle (Mašın, 2017). Global water retention capacity is thus implied both by the retention of the macrostructure and by the specific volume of the (saturated) microstructure, which are linked through the double-structure coupling framework.

The model is included into the balance equations, where variable number of components can be taken into account, i.e. grains, liquid water, vapour, dry air or their combinations. The grains and liquid water can be assumed compressible or incompressible.

Since their first introduction by about 20 years ago, the model of thermo-hydro-mechanical coupling undergo a constant improvement with the version evolution of finite elements software Code_Aster developed by EDF. Today's open source version of the code provides users with a family of laws of THM behavior for the saturated and unsaturated mediums. Chosen modeling approach is based on the presentation of the porous environments elaborated in particular by Coussy et al. (2004). Constitutive equations of the behavior law are obtained starting from thermodynamic considerations where each component of the porous environment such as a mechanical, hydraulic and thermal ones are supposed to be a state variables. Then the relations describing the hydraulic and thermal quantities are introduced by taking into account strong couplings between these two phenomena together with the mechanical deformations through the hypothesis of Bishop effective stress (Bishop 1959). The main advantage of adopted approach ([R7.01.10], n.d.; [R7.01.11], n.d.; [R7.01.16], n.d.; [R7.01.18], n.d.; [R7.01.24], n.d.) is that the obtained relations could be coupled with any mechanical behavior law paving the way to the description of highly nonlinear phenomena. We mention here two most relevant mechanical behavior laws available in Code_Aster that are designed to represent the rock formation and soils. Geomaterials show asymmetric traction-compression curve and deform plastically under lower tension, the hydrostatic compression making them more "elastic" under shear stress. Therefore, the yield function is usually written with help of hydrostatic stress invariant, which is antisymmetric under stress reversal. Drucker-Prager type behavior laws ([R7.01.16], n.d.) have the most simple linear yield function, they are commonly used to represent rock/soil behavior under significant compression. These laws find their limitations for the situations where some stress component's values approach the traction loading domain as it is the case for tunnel excavation. The LKR behavior law ([R7.01.16], n.d.; Kleine, 2007; Raude, 2015) is a viscoplastic law developed by EDF to model the behavior of rocks, and, in particular those of the nuclear waste storage project Cigéo in the East of France. The plasticity criterion in this model is based on quadratic Hoek_Brown yield function ([R7.01.16], n.d.; Hoek and Brown, 1980) which allows to model more accurately traction loading domain.

The principal industrial application within the framework of nuclear waste disposal management is related to the simulations of tunnel excavations. The particularity of this kind of simulation is laid in the complexity of loading pass of vault top of the tunnel which undergo the evolution from high compression to bi-traction during the excavation. As the principle physical phenomena which should be taken into account are nuclides diffusion in cracked or highly plastified environment, it seems that currently available in Code_Aster THM models/laws are perfectly up to date to capture these main phenomena. At the same time they suffer from poor robustness and numerical efficiency which would be our main focus inside the current work.

2.1.4 Thermodynamically consistent modeling of corrosion

A mechanistic model of electrochemical corrosion in deep geological storage of nuclear waste has been proposed in Bataillon et al. (2010). This model named Diffusion Poisson Coupled Model (DPCM) assumes

that the carbon steel canister is covered by a dense spinel iron oxyde layer. The oxyde layer is in direct contact with the oxydizing medium or with a layer of corrosion products obtained by a precipitation mechanism, which can be modelled as a saturated porous medium, and with the undamaged steel. A fine description of the anaerobic corrosion process arising at the surface of the steel canister is necessary to predict both the evolution of the canister along time, as well as to predict the evolution of the chemical composition of the fluid in the porous medium (e.g., dihydrogen concentration) to be coupled with reactive transport models. While the corrosion process impacts the chemical composition of the fluid, the evolution of the fluid via reactive transport models impacts the boundary conditions of the corrosion models which depend on the fluid composition, for instance through its pH.

From a mathematical point of view, it consists in a drift-diffusion-Poisson system of equations on a moving domain with strongly coupled boundary conditions. The main difficulty for the mathematical analysis comes from the moving boundaries whose velocity depends on the concentrations of the chemical species and on the electrical potential. Only results on simplified models have been obtained so far. Existence of solutions for some simplified models which do not take into account the moving boundary interfaces have been obtained in Chainais-Hillairet and Lacroix-Violet (2012, 2015). The existence of pseudo-stationary states is established in Chainais-Hillairet and Gallouët (2016) under some simplifying assumptions on the model.

In opposition to the simpler case of concrete carbonation (Aiki and Muntean, 2011; Zurek, 2019), no Lyapunov functional has been found so far for the DPCM model despite it is expected that the model fulfils Onsager's reciprocal relations which ensures that the physical energy decays along time (and is thus a Lyapunov functional). Besides, there has been recent advances in the mathematical description of models with generalized gradient flow structure (roughly speaking, model satisfying Onsager's reciprocal relation). Otto's seminal works (Jordan et al., 1998; Otto, 2001) has been a key step to understand the mathematical structure of generalized gradient flows where the evolution of the physical configuration relies on transport of chemical species, as it is the case for corrosion models. The Poisson-Nernst-Planck system has been shown in Kinderlehrer et al. (2017) to have the generalized gradient flow structure highlighted in Jordan et al. (1998). A first extension of this approach on moving domains was performed in Portegies and Peletier (2010) and extended to a toy model for corrosion in Zurek (2019). Finally, a framework was recently proposed in Mielke (2011) and Mielke et al. (2016) allowing to go beyond Onsager's framework, i.e. when the relation between the forces and the evolution are no longer linear.

2.1.5 Non-Darcian, non-Fickian properties of nanoporous media

Nanoporous media consist of homogeneous or heterogeneous porous material in which a significant part of the pore size distribution lies in the nanometer range. Clayey rocks, sediments or soils are natural nanoporous media, and cementitious materials, the most widely used industrial materials in the world, are also nanoporous materials. The bulk fluid transport properties and the in situ chemical reactivity properties of nanoporous media are notoriously difficult to characterize. Because of their large specific surface area, most of the fluid volume in nanoporous media is influenced by the close proximity of mineral surfaces, which explains the very low transmissivity of these materials. As a consequence, the experimental characterization of their permeability requires special techniques (Neuzil and Person, 2017). Also, the large specific surface area of nanoporous material provides them with very high adsorption capacity. The strong adsorption and resulting retardation of many contaminants by nanoporous material make them ideal for use in natural or engineered barrier systems or in filtration technologies. A good understanding of their chemical reactivity coupled to their transport properties is necessary to predict the long-term evolution of these properties of interest as a function of a range of physical and chemical conditions and processes. In this regard, reactive transport modeling can help bridging the gap between current process knowledge and predictions of the long term evolution of natural and engineered nanoporous materials in geological and industrial settings. However, nanoporous media exhibit a remarkable array of macro-scale properties with marked departures from those observed in "conventional" porous media such as permeable aquifers, for the study of which reactive transport models and codes have been historically developed. These properties arise from the interactions of charged mineral surfaces with water and solutes present in the nanopores, which leads to coupling between flux terms in the presence of a so-called diffuse layer at clay mineral surfaces. These

couplings manifest themselves in macroscopic observations that have intrigued geologists for more than one century, such as geologic ultrafiltration, i.e. the accumulation of solutes on the inflow side of clay-rich lithologies (Lynde, 1912; Neuzil and Person, 2017).

The vast majority of published reactive transport studies dealing with clay and cement materials are related to the evaluation of the long-term stability of surface and underground radioactive waste storage systems (see Claret et al. (2018) and references therein). In these types of simulation, the modeling effort has been focused primarily on the reactivity of the nanoporous materials rather than on their transport properties. Traditionally, Fickian diffusion has been typically considered, i.e. without taking into account advection, and without taking into account the anomalous transport properties of nanoporous media. In the last decade, special capabilities have been developed in a limited number of reactive transport codes that make them able to model part of these unconventional properties, with consideration of coupled processes that go beyond the traditional coupling between advective flow, dispersion, diffusion and reactions.

A recent review showed that the current implementations in reactive transport codes of coupled transport processes in the presence of a diffuse layer are in agreement with the theory of irreversible thermodynamics processes (Tournassat and Steefel, 2019). These implementations available in only two codes – CrunchClay and PHREEQC – are currently limited to the modeling of purely diffusive systems, i.e. without advective flow where there is a need to consider flow with that portion of the pore structure affected by the diffuse layer. It is possible to model electro-migration in PHREEQC. The multi-component capability of these codes makes it possible to overcome many limitations encountered with simplified approaches that are limited to single salt transport. In particular, the transport of electrolyte background ions and tracer ions can be simulated simultaneously, demonstrating the existence of highly coupled processes during their transport through nanoporous media. In addition, reactive transport codes are not limited to the simulation of systems with fixed surface charge and fixed diffuse layer dimensions, and can incorporate microstructural information in dual- to multi-continuum descriptions of the porosity. Applications of these capabilities to the simulation of clayey and cementitious material properties highlighted the prominent role of the diffuse layer in the transport properties of ions. The above mentioned review paper provided some insights to advance reactive transport modeling in the direction of a full implementation of multi-component advective flux in the presence of a diffuse layer. In particular, the osmotic efficiency parameter must be revisited so that ion and neutral molecule specific properties can be simulated simultaneously.

2.2 Numerical methods

This section briefly described the state of the art on the numerical methods for the suitable approximation of the physical processes mentioned above.

2.2.1 Numerical coupling strategies for multi-physics processes

When one aims at simulating coupled Multiphysics processes, different strategies can be relevant. One can either solve the full system all at once in what one would call a fully-coupled or fully implicit strategy, or one can solve each physical process apart and sequentially. This second approach should provide similar results as the fully coupled one if one iterates the sequential procedure until convergence, while allowing for a modular approach where each module is devoted to one physics.

Reactive transport modeling (RTM) plays a crucial role in the understanding of coupled thermal, hydraulic and chemical (THM) processes for the performance assessment of geological disposal of radioactive waste (Claret et al., 2018; Bildstein et al., 2019). In some situations of interest this context, such processes are governed by non-isothermal multiphase multicomponent flows with reactive transport in porous media. At the continuum scale, reactive transport models for porous media rely on constitutive equations that make it possible to describe the porous medium with respect to its macroscopically measurable properties such as permeability, dispersivity, diffusibility, and so on (Steefel et al., 2014). The significance of reactive transport constitutive equations and their treatment in saturated or non-saturated conditions have been recently reviewed by Claret et al. 2018. From the numerical point of view, modeling THC phenomena leads to a

highly nonlinear coupled system of degenerate partial differential equations (PDEs) governing the compositional multiphase flow to algebraic or ordinary differential equations (ODEs) modeling respectively equilibrium and kinetic reactions. Several approaches can be found in the literature to deal with coupling between flow, transport, chemistry. The first approach is a fully coupled procedure and consists in solving a nonlinear system gathering all equations at each time step (Brunner and Knabner, 2019). The second approach is sequential and more widespread since it is used in most of the numerical codes cited in (Claret et al., 2018; Bildstein et al., 2019). We can mention non-exhaustively several sequential codes HYTEC (Lee et al., 2003), TOUGHREACT (Xu et al., 2012), CORE^{2D} V4 (Samper et al., 2012), Crunchflow (Steefel, Appelo, et al., 2015) or OpenGeoSys (Kolditz et al., 2012) that have been intensively used for reactive transport modeling applied to the management of radioactive waste. In this case, flow, reactive transport (or possibly, flow, transport and chemistry) are solved sequentially at each time step, possibly within an iterative loop. No comparative study exists yet to quantify the accuracy loss for this approach in comparison with fully implicit approach, but its gain in implementation and saving in computing time are fairly obvious. However, sequential approaches introduce operator splitting errors and restrictions on the time step are mandatory to ensure mass conservation for instance. In the context of the simulator DuMu^{X} (Flemisch et al., 2011), a sequential approach was developed and implemented for coupling two-phase flows and reactive transport, in a high performance computing context. More precisely, in Ahusborde et al. (2017), a sequential approach was developed to study the migration of hydrogen produced by the corrosion reaction in deep geological radioactive waste repository. This approach splits the global problem into two sub-problems. The first sub-problem computes a two-phase compositional flow where only species present in both phases are treated implicitly. The second sub-problem calculates a reactive transport problem where flow properties are given by the first step. Both subproblems were solved using a fully implicit manner (Ahusborde et al., 2018) but in a sequential procedure. However, some questions about the coupling between all the physical phenomena described above have also to be studied to assess if sequential approaches are suited to model very coupled phenomena or if fully implicit strategies would be more relevant. For example, when simulating fast kinetics or strong chemical interaction, higher levels of coupling, implicitness may be useful as this can eliminate operator-splitting error and expedite nonlinear convergence. Obtaining solutions to these large systems of equations calls for a massively parallel implementation to reduce both computation time and memory requirements. In this context, in (Ahusborde, El Ossmani, et al., 2019), a fully implicit approach for a single phase multicomponent flow with reactive transport has been developed and validated via numerous 2D and 3D test cases including high performance computing. In Ahusborde et al. (2019), the global implicit approach developed in Ahusborde, El Ossmani et al (2019) was extended to deal with reactive two-phase flow for a one-dimensional test case proposed in Seigneur et al. (2018). This test case deals with porosity and permeability changes due to dissolution/precipitation dissolution/precipitation of minerals that can be linked to a process such as the atmospheric carbonation of partially saturated concrete.

Besides, THM problems are solved in SIFEL (Koudelka et al., 2017, 2018) by two approaches: (a) a staggered approach, in which transport and mechanical parts are solved independently and in sequence (an iterative procedure is used to eliminate unbalanced forces); (b) a fully coupled approach, in which a complete thermo-hydro-mechanical stiffness matrix is assembled and solved using a Newton-Raphson iterative scheme in a single step. For both approaches, research is currently devoted to eliminating unwanted pore water pressure oscillations that arise in the model when full saturation is approaching.

Concerning the spatial coupling between free gas flows in the galleries and coupled liquid gas Darcy flows in the surrounding matrix, domain decomposition appears to be a natural approach compatible with a modular implementation. A nonlinear domain decomposition algorithm based on the previous works (Masson et al., 2016; Birgle et al., 2018) will be designed.

2.2.2 Linear and nonlinear algebraic solvers

Space and time discretization of HM and THM problems provides time-stepping numerical scheme with linear or nonlinear systems to be solved in each time step. These systems have a block structure corresponding to the selection of basis state variables. In the case of large problems, it can be more efficient to solve such systems iteratively by some Krylov space method with suitable preconditioner (Saad, 2003). A general way for construction of preconditioners begins with exploiting the block structure, keeping some diagonal blocks but replace others by the corresponding Schur complements (Ferronato et al., 2010; Haga et al., 2010; Axelsson et al., 2012; White et al., 2016). Such preconditioners have to be subsequently modified by simplification of the Schur complements. For implementation, subsystems arising from the diagonal blocks are usually solved by suitable inner iterations. Specific problems are the robustness of such techniques with respect to oscillations and contrast in problem coefficients (Hong and Kraus, 2018) when materials are heterogeneous. Another issue is parallel computation which can be applied for solving several block subsystems in parallel and, especially, it should be used at the level of the inner iterations (Blaheta, Luber, et al., 2018). Similar preconditioners can be used for solving nonlinear system arising e.g. for flow in partly saturated media, when the problems are linearized by various Newton-type techniques (Bergamaschi and Putti, 1999). The stochastic character of the problems arising in many geotechnical applications brings another challenge to be solved (see Task 4).

In the numerical approximation of the THM problem provided by SIFEL (Koudelka et al., 2017, 2018), the construction of the stiffness matrix – to be consistent with the global Newton-Raphson iterative scheme for the adopted Runge-Kutta adaptive model integration – is carried out by two alternative approaches: (a) approximation using the linear part of the thermo-hydro-mechanical hypoplastic model; (b) numerical estimation of the stiffness matrix using perturbation. As neither of the approaches offers quadratic convergence because of various numerical problems, the convergence can be rather slow. Hence efforts are being made to find a reliable way to construct the stiffness matrix to obtain robust and fast converging simulations.

The Newton-Raphson method is used in the staggered as well as in fully coupled formulation. There are two implementations of the method. The classical method, where the system matrix is updated at every iteration, is usually more time consuming because of permanent matrix factorisation. On the other hand, the modified method uses the same system matrix for several steps, and the matrix factorisation is needed only when the matrix is updated. The modified Newton-Raphson method usually requires a shorter computing time.

Reactive transport modeling leads to a highly nonlinear coupled system of degenerate partial differential equations to algebraic or ordinary differential equations. Due to the strong coupling of the flow and reactive transport equations, a specific attention was paid by many author to improve the convergence of the nonlinear and linear solvers, even more when considering fully implicit approaches. For instance, in Fan et al. (2012), after the Jacobian matrix assembly, a series of algebraic reductions (Schur complements), are performed to reduce the number of discrete equations to be solved. This strategy is commonly used in compositional reservoir simulations. In Brunner and Knabner (2019), the reduction technique described in Hoffmann et al. (2012) for reactive single phase flow was extended to the case of two-phase reactive flow. Unlike the classical strategy, their general transformation method does not only eliminate unknown equilibrium reaction rates. It also potentially reduces the nonlinear coupled part of the problem, allowing the use of large time steps and avoiding the potential drawbacks of sequential approaches.

2.2.3 Discretization methods

Finite Volume methods are natural to discretize physical balance laws in a locally conservative way. Mathematical analysis tools have been developed in the last decades to assess their robustness and accuracy (see for instance Eymard et al. (2000). It is now well understood that the Scharfetter-Gummel numerical fluxes (Scharfetter and Gummel, 1969), dedicated to the numerical approximation of convection-diffusion fluxes, allow to capture in an accurate way the long-time behaviour of drift-diffusion systems (Chatard, 2011; Bessemoulin-Chatard and Chainais-Hillairet, 2017). This motivated the choice of

Scharfetter-Gummel based fluxes in (Bataillon et al., 2012) for the simulation of the DPCM corrosion model (Bataillon et al., 2010). These particular fluxes were also used in the numerical investigations on reduced models (Chainais-Hillairet and Lacroix-Violet, 2015; Chainais-Hillairet and Gallouët, 2016; Zurek, 2019).

When a system of partial differential equations is cast under a gradient-flow structure, this gradient-flow structure suggests canonical time-discrete approximations and provides a context for the design of numerical methods which preserve structural properties Minimizing movement schemes (Jordan et al., 1998) provide naturally stable time-discretizations of generalized gradient flows. They can be seen as generalizations of the Backward Euler scheme and are now common for studying generalized gradient flows (Ambrosio, Gigli et Savaré 2008). The exploitation of such fully discrete minimizing movement schemes for numerical purpose is quite recent and goes back to Benamou et al. (2016). Then several improvements have been proposed to improve the efficiency of such approaches (Cancès et al., 2019; Carrillo et al., 2019; Li et al., 2019).

Most of the methods that have been designed in order to maintain their thermodynamical consistency require some strong regularity assumptions on the mesh that are not suitable in the context of porous media flows in complex geometries. Such frameworks motivated the development of advanced finite volume methods, among which the nodal Vertex Approximate Gradient (VAG) scheme introduced by Eymard et al. (2012). The discretization of the 3D non-isothermal compositional liquid gas Darcy flow is based on VAG scheme (Xing et al., 2017). The coupling with the 1D free gas flow will be based on non-matching meshes between the 3D Darcy flow domain and the 1D domain in order to account for complex geometries of the tunnels.

Another type of discretization method is the lattice Boltzmann discretization method. (Higuera et al., 1989; Qian et al., 1992). It is a special discretization of the kinetic Boltzmann equation with applications of LBM range from diffusion dominated reactive transport simulations in porous media to multiphase fluid dynamics of turbulent flow (Succi, 2001; Kang et al., 2002; Chen et al., 2003). Recently, it has been implemented to study the flow properties of a compacted illite digital rock (Pazdniakou et al., 2018)u , as well as for the cross scale modelling of dissolution and precipitation . Implementation of the classical nucleation theory allows to trace the formation of first critical nuclei up to the micrometer scale as was shown for the case of barite and Celestine precipitation (Prasianakis et al., 2017). Models that account for nucleation and growth of particles are very sensitive to microscopic parameters like the surface tension and the size of first critical nuclei (Noguera et al., 2010). An application to radioactive waste disposal and more specific the Radium-bearing barite precipitation has been recently presented (Curti et al., 2019). Upon upscaling the pore-level algorithm can be used to investigate the coupled advection-diffusion-reaction-precipitation processes (Churakov and Prasianakis, 2018). The precipitation and the dissolution of minerals has a direct effect to the mass transport and vice versa (Prasianakis et al., 2018; Poonoosamy, Klinkenberg, et al., 2019). A decrease in the porosity will result to increased resistance to flow, which under a hydraulic gradient could lead to increase in stresses. A direct coupling of lattice Boltzmann simulators for fluid flow with mechanics simulators still remains a challenging task. .

As concerns the discretization of HM, it was even very early observed that two field (displacement-pressure) P1-P1 finite element (FE) discretization (i.e., both the displacement and the pressure are approximated by piecewise linear fields on simplex meshes) is not robust and can produce spurious oscillations due to a locking (e.g. (Lewis et al., 1998)). P2-P1 elements (i.e, the approximate displacement field is now piecewise quadratic) help but they are more expensive in computations. The reason for these effects is nowadays explained by inheriting saddle point structure. Later, three field (displacement-velocity-pressure) formulation was used with discretization e.g. by P1-RT0-P0 elements (RT0 denotes the lowest order Raviart-Thomas element, and P0 denotes piecewise constant fields). This discretization provides conservativeness for flow, but it was recently shown that a fully stable discretization with lowest order FE for elasticity requires abandoning conformity and using stabilized Crouzeix-Raviart elements or discontinuous Galerkin formulations, see Yi (2013). It is also possible to discretize elasticity by mixed FE and use four field formulation, see Bean et al (2017). The IGN team is testing robust discretization with the consecutive aim to develop robust and efficient solvers for these variants. Some preliminary results will be reported soon. For the time discretization, we tested the higher-order stable discretization of the Radau type, see Axelsson, Blaheta and Kohut (2015) and Axelsson, Blaheta and Luber (2015).

Capillary effects play a crucial role for the migration of multiphase fluids in heterogeneous media (Duijn et al., 1995; Cancès, 2010; Bourgeat et al., 2013). Discontinuities of the conservative variables caused by spatial heterogeneities make it necessary to design appropriate strategies concerning the discretization (Hoteit and Firoozabadi, 2008; Cancès, 2009; Bastian, 2014; Eymard et al., 2014) and the nonlinear solvers (Wang and Tchelepi, 2013; Brenner et al., 2017). As proposed in Hoteil and Firoozabadi (2008) and Bastian (2014), a natural choice for the primary variable is the capillary pressure rather than the saturation, requiring adaptation of the existing tools like TOUGH2-MP (Pruess et al., 1999). These adaptation are already at the basis of the recent contribution (Amri et al., 2020). Future work will be dedicated to the validation of this new version of TOUGH2-MP on PGZ-experiment by adding an adequate hysteresis model in the compositional two-phase flow and transport equations.

2.2.4 Error control

Ensuring convergence of a nonlinear numerical simulations on large scale structures are usually rather complex task. In practice in the case of difficulties, one could opt for relaxing convergence criterion together with refinement of the loading steps. This basic engineering craftiness (or hints) become trickier once it comes to HM (or THM) simulations. The point is that the latter case corresponds to the multi-physical coupling phenomenon and the relaxation in convergence criterion necessarily leads to physical results alternation. But if in pure mechanical (or pure hydraulic) simulation one could easily have a reasonable guess on the results validity, in the case of coupling it becomes almost impossible mission as all the phenomena intercalate. In particular, one could obtain physically relevant results from mechanical point of view (as deformations of the structure) but at the same time getting wrong outputs for pressure or hydraulic fluxes distribution. The main reason for that is related to the fact that in the standard finite elements code the convergence errors sources, i.e. hydraulic, mechanic or temporal refinement one, are all put together to produce a single scalar stopping criterion. The idea is then to separate all these errors in order to be able to perform a more subtle analysis. More than that we could provide the finite elements code user with a heuristic algorithm, which stop automatically the code execution once the best possible convergence for the given mesh is attained.

There exists two main approach to tackle an error estimation problem. A classical *a priori* one has a disadvantage of relying on the problem regularity, which is not known a priori for most of the industrial applications. On a contrary *a posteriori* error analysis (see for example Ainsworth and Oden (2000)) seems to be more relevant as the solution regularity is “reconstructed” based on the current finite elements solution. In most recent developments, one reconstructs the associated fluxes (stress for displacement, hydraulic flux for pressure) which dispose some important mathematical properties (smooth elements transition, symmetry, Riedlbeck et al. (2017)). The principle advantage of the following technique is the easy comparisons of all errors sources. The last property leading to possible implementation of different stopping algorithms.

A posteriori error estimation based of flux reconstruction on patches have been tested in development version of finite elements Code_Aster software (not available to end users up to now) in order to evaluate efficiency of current technic for industrial cases. The result obtained for the tunnel excavation-like test-case are encouraging as close to perfect efficiency is attained for error estimation compared to reference study on highly refined mesh (Riedlbeck et al., 2017; Botti and Riedlbeck, 2018). Nevertheless, the currently high computational cost of error estimation is blocking extension of tested error estimation strategy to the large scale industrial calculations. We have identified two main reasons of this “poor” numerical performance. First one is related to data-processing architecture incompatibility with sub-optimal memory usage, the second is due to pure sequential programming.

Finally the convergence of the nonlinear and linear solvers can be improved using *a posteriori* estimators to achieve computational savings by stopping timely the linear and nonlinear solvers (Vohralík and Wheeler, 2013; Riedlbeck et al., 2017; Ben Gharbia et al., 2019), but all these procedures need to be parallelised in order to meet industrial needs in numerical simulations.

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2.2.5 Going parallel

The development of more powerful and energy-efficient computer architectures is widening the use of large scale high performance computing techniques. Notable increases in computational power are being achieved by many core systems with powerful accelerators such as general-purpose graphics processing units (GP-GPUs). These systems also include more complex hierarchical cache/memory designs which permit to provide the needed data flow required by computing units. These new hardware designs pose a challenge for the implementation and scalability of current simulation algorithms and, at the same time, offer an opportunity for the development of new ones.

In the near future, supercomputers are expected to reach a peak performance of one exaflop/s, which implies a 100 times improvement with respect to current supercomputers. This improvement will not be based on faster processors, but on a much larger number of processors (in a broad sense). This situation will certainly have an impact in large scale computational science and engineering. Parallel algorithms will be required to exhibit much higher levels of concurrency, keeping good scalability properties. When dealing with transient problems, since information always moves forward in time, one can exploit sequentially. However, the tremendous amounts of parallelism to be exploited in the near future certainly motivates to change this paradigm. One of the motivations to exploit higher levels of parallelism will be to reduce the time-to-solution.

Different iterative methods have been considered so far. Lions et al. (2001) used a time discretization of a partial differential equation (PDE) that allows for parallel implementations. The method, based on an Euler scheme, combines coarse resolutions and independent fine resolutions in time in the same spirit as standard spatial approximations. Emmett and Minion (2012) also applied a parallelization of numerical methods for PDEs in the temporal direction. The method was iterative with each iteration consisting of deferred correction sweeps performed alternately on fine and coarse space-time discretizations. With regard to direct methods, time-parallel methods can be found in Gander et al. (2015). In general these methods can exploit low levels of concurrency. Moreover, Badia and Olm (2018) proposed a parallel-in-time solver for ordinary differential equations that relies on the well-known Schur complement method in linear algebra. Chen et al. (2019) used multipoint flux approximation (MPFA) method to discretize the spatial variables on staggered grid in order to accurately describe the flow behaviour in the porous media whose permeability is anisotropic, in which case the permeability tensor is full. For staggered grid in 2-D space, pressure values were defined at the centre of the cells and velocity components were located on edges. They were considered the permeability fields which contain different anisotropy angles in the simulation and fractures are represented explicitly by volumetric grid cells and the numerical algorithm is parallelized in order to reduce the simulation time. The algorithm is implemented in parallel using FORTRAN and the linear system arising from the pressure equation is solved by the aggregation-based algebraic multigrid solver (Notay, 2010; Napov and Notay, 2012).

As was already mentioned in 2.1.3, the computation of HM or THM model is usually performed by a time-stepping scheme with the solution of linear or nonlinear systems in each time step. The time-stepping itself is sequential so that the space for parallelization arises in the solution of the arising systems. Iterative solution of these systems provides natural space for parallelisation. The crucial point is then a sparse approximation of the Schur complement systems and parallelization of the preconditioners. The preconditioners can involve inner iterative solvers for which efficient parallel solution methods are available. In this respect, some experience of the IGN team was gained with in house FE software GEM using Schwarz domain decomposition method and FORTRAN/MPI parallel programming (Blaheta et al., 2010) and PERMON as a tool for solving quadratic problems with or without constraints, which support the FETI domain decomposition and use of PETSc library (Hapla et al., 2016).

Application of the finite element method in solution of coupled thermo-hydro-mechanical problems leads to a relatively large number of unknowns because displacements, pore pressures, and temperature are defined in each node. Therefore, the total number of unknowns in systems of algebraic equations is higher than in the system corresponding to a pure mechanical problems where the sole displacement would be discretized. Moreover, the high number of unknowns in each node is connected with a very broad bandwidth of the system matrix. These two reasons lead to the necessity of parallel processing in the case of large and complex engineering structures. Parallelisation of a finite element code can be done efficiently with the help of either domain decomposition methods (DDM) or alternatively splitting method. In both cases the domain solved is split into smaller subdomains which are treated independently on particular processors. In DDM method each subproblem is solved independently and the continuity on the borders of subdomains is then enforced. In splitting method the decomposition is used mainly in order to reduce memory consumption due to parallelisation of elementary calculation (element by element in finite elements method calculation), the total stiffness matrix is then assembled as a whole and finally the global algebraic equation is then solved using second level parallelisation relied on other parallelisation scheme. With respect to highly nonlinear and nonsymmetric systems of algebraic equations, the most suitable DDM is the Schur complement method, which is a variant of the Gaussian elimination. Splitting method may be less efficient then the DDM scheme but it allows more versatile usage while the second level parallelisation of linear algebraic system is completely independent. For the latter task one could rely on external expertise such as know-how of worldwide community of software developers proposing a highly stable and scalable open source solvers as MUMPS (MUltifrontal Massively Parallel Sparse direct Solver, (Amestoy et al., 2001, 2006)) or PETSc (Portable, Extensible Toolkit for Scientific Computation, (Balay et al., 2019)).

Since 2019 all new versions of Code_Aster (called Aster_xx) are distributed as fully parallelizable. The adopted parallelization is the splitting method described above. While all elementary (in the sense of finite elements method) calculations are naturally parallelizable, the parallelization of a posteriori errors that we will address in the current work necessitate in deep comprehension of Code_Aster architecture. Error estimator that seems to be suitable for hydro-mechanical coupling problems described in the recent works (Riedlbeck et al., 2017; Botti and Riedlbeck, 2018) is based on patch reconstruction scheme. It means that each elementary calculation is not a simple mesh element integral as in the standard finite element procedure, but a patch calculation, patch being an agglomeration of mesh elements connected to the given node. The parallelization scheme adopted in Code_Aster must then be modified in order to allow new kind of elementary calculation be more efficient. Given the usually rigid architecture of industrial finite elements code the full parallelization of a posteriori error estimation is a rather challenging task. Nevertheless we are confident that successful development would be made in order to benefit all the end users of our open source finite elements code in its future stable release expected in 2021.

Because the NRG/ORCHESTRA contribution focuses on the chemical solver, which operates on single cells, there are no spatial / discretization topics in this contribution. The ORCHESTRA chemical solver is very well suited for parallelization, as it has a very small memory footprint, and is prepared for the existence of multiple instances in a single memory space.

The coupled liquid gas Darcy and free gas flows will be implemented in the open source parallel code ComPASS (Xing et al., 2017; Beaude et al., 2018; Lopez et al., 2018). The COMPASS parallelization is based on mesh partitioning and MPI communications. It uses the Petsc and Hypre libraries for the solution of the linear systems. Since the size of the 1D free gas flow model is negligible compared with the size of the 3D Darcy flow model, the domain decomposition approach will allow a simple parallel implementation of the coupled model.

2.3 Available Tools

Let us describe the available tools that will be at the basis of the developments carried out in the context of Task 2.

CALIPSO is a Fortran90 code for the simulation of corrosion of iron based alloys relying on the discretization presented in (Bataillon et al., 2012) of the DPCM model (Bataillon et al., 2010). Its development was initiated

by ANDRA in 2008. A stabilized version has been registered in 2014 at the French Agency for Program Protection (https://www.app.asso.fr/) under reference IDDN.FR.OO 1. I 60009.000.s.P.20 I 5.000.30000. It is currently maintained by Christian Bataillon (CEA) and Laurent Trenty (ANDRA).

COMPASS http://www.anr-charms.org/page/compass-code is an open source parallel code co-developped by the partners of the ANR CHARMS project BGRM, UCA-CNRS-Inria-LJAD, MdS, Storengy and LJLL (Xing et al., 2017; Beaude et al., 2018; Lopez et al., 2018). It has the following features:

- **Physics**
  - Multiphase compositional thermal Darcy flow model
  - 2D discrete fracture or fault network coupled with the surrounding 3D matrix
  - Multi-branch well model
- **Numerical methods**
  - Parallel Vertex Approximate Gradient (VAG) scheme on polyhedral meshes accounting for the flow in the 3D matrix and the 2D fault network.
  - Fully implicit Coat's type formulation of multiphase compositional thermal models.
  - GMRES or BiCGStab linear solver with CPR-AMG preconditioner
- **Computer science**
  - SPMD Paradigm
  - Fortran 2003 + C/C++ + MPI
  - Libraries Metis, Petsc + Hypre, vtk, HDF5
  - High level python API

ORCHESTRA is a (currently) Java code that was developed by Johannes Meeussen to develop new chemical adsorption models and to combine these with reactive transport processes. It is available from orchestra.meeussen.nl. Its unique property is that all chemical and physical model code is defined in object oriented text files outside the executable code. These text files are read as input during runtime and can be extended or adapted by end users. As a results the calculation kernel is very small and relatively fast. This approach / structure has been described in (Meeussen, 2003). It contains a large set of chemical models, including a large number of state of the art adsorption models, and has proven to be very calculation efficient in reactive transport benchmarks (Steefel, Appelo, et al., 2015). This framework is written in Java which allows ORCHESTRA to run on different operating systems, but makes it more difficult to combine modules with C++/FORTRAN code. It can be used in a so-called split operator approach in which chemical equilibrium is solved separately from the transport equations.

DuMu^x: For THM numerical modeling, several sequential (Ahusborde et al., 2017, 2018) and fully implicit (Ahusborde, Amaziane, et al., 2019; Ahusborde, El Ossmani, et al., 2019) modules have been developed and integrated in DuMu^x framework. DuMu^x (DUNE for Multi-{Phase, Component, Scale, Physics, ...} Flow and Transport in Porous Media) is a free and open-source object-oriented software written in C++ \cite{DUMUX}. The environment is equipped with efficient solvers and massively parallel computation capability provided by DUNE (Bastian et al., 2008).

TRIAX – The coupled thermo-hydro-mechanical model for expansive clays is implemented in an inhouse element test driver developed since 2001 at Charles University and elsewhere by David Mašín and co-workers. The driver covers a wide range of constitutive models and testing conditions. Virtually, any element test can be simulated using the procedure generated by (Janda and Mašın, 2016); the majority of standard and less-standard testing conditions are pre-defined (including complex thermo-hydro-mechanical tests in unsaturated conditions, rate-dependent loading, plotting response envelopes, hypoplastic asymptotic state boundary surfaces, and elasto-plastic yield surfaces). Loading stages can be combined to form complex loading paths. The driver can also run any constitutive model implemented in *umat* format. The driver can be freely downloaded at https://soilmodels.com/triax/. The download package includes the source code, pre-compiled Windows and Linux executables, and a wide range of input files associated with various publications.

ExCalibre is s an online tool (freely accessible at https://soilmodels.com/excalibre/) which enables users to automatically calibrate advanced soil constitutive models (Kadlıcek et al., 2018). It adopts a unique model-specific calibration algorithm which remains consistent with the physical meaning of material parameters

rather than using a blind parameter optimisation. It thus reproduces the thinking of an experienced engineer calibrating the model, leading to a reliable parameter set to be used in geotechnical simulations. After calibration, the user can manually change the parameters to fine-tune the calibration or to investigate the effect of individual parameters on model predictions. A simple Excel spreadsheet based on the provided template is used as an input. In addition to automatic calibration, ExCalibre also acts as an element test driver simulating standard laboratory tests.

SIFEL (**SI**mple **F**inite **EL**ements) is an open source finite element code which has been developed since 2001 at the Department of Mechanics of the Faculty of Civil Engineering of the Czech Technical University. SIFEL comprises several components which can be used as independent computer codes, or can be linked to obtain additional functionalities. All parts are released under a GNU General Public Licence (GPL) and can be downloaded at http://mech.fsv.cvut.cz/~sifel/. Within the SIFEL package, in particular, MEFEL is the component which deals with mechanical problems, such as linear and non-linear statics, eigen-vibration analysis, forced dynamics, time dependent problems of rheology. The coupled transport of heat, moisture and other species can be modelled in the part TRFEL. Namely, it contains heat transport, moisture transport, coupled heat and moisture transfer, moisture and salt transfer. Coupled thermo-hydro-mechanical models, including models for expansive clays, are implemented in the part METR which interacts with the MEFEL and TRFEL. All parts of the code are equipped with one, two and three-dimensional finite elements and material models. The material models can be isotropic, orthotropic or generally anisotropic.

Code_Aster : Today, all the poromechanical models used in EDF are capitalized in Code_Aster (EDF code in Open-Source: www.code_aster.org). Code_Aster is integrated in multifunctional finite elements platform Salome_Meca. Code_Aster offers a full range of multiphysical analysis and modelling methods that go well beyond the standard functions of a thermomechanical calculation code: from seismic analysis to porous media via acoustics, fatigue, stochastic dynamics, etc. Its modelling, algorithms and solvers are constantly under construction to improve and complete them (1.200.000 lines of code, 200 operators). Resolutely open, it is linked, coupled and encapsulated innumerous ways. Code_Aster provides full thermo-hydro-mechanical coupling framework for stationary and transient calculations. It relies on highly efficient external linear algebra solvers. It worth to mention that the software is under quality assurance QA (independent validations, reference of 2.000 test cases, 13.000 pages of documentation, source management, qualification of version, etc.).

GEM-Selektor: (http://gems.web.psi.ch/GEMS3/) is a graphical user interface of GEMS3K solver of chemical equilibria (Wagner et al., 2012; Kulik et al., 2013). It greatly facilitates configuring the chemical system, the input of composition recipes, the computation of single equilibria or process simulations, viewing/exporting the results as tabulated data or plots, and managing the thermodynamic data. Chemical system definitions can be exported per mouse click into GEMS3K i/o files for use in coupled codes such as GEMSFITS and reactive-transport simulators Comsol-GEM, OpenGeoSys-GEM and CSMP++GEM. Simplified 1-D reactive transport models can be performed directly in GEMS-Selektor using its GEM2MT module. GEMS is open-source software, available at https://bitbucket.org, with binaries distributed from http://gems.web.psi.ch as freemium together with the built-in PSI-Nagra and SUPCRT98 chemical thermodynamic databases. In addition, third-party thermodynamic databases (e.g. Cemdata'18, HERACLES, Mines'19), maintained by their owners (https://www.empa.ch/cemdata; http://tdb.mines.edu), are available as plugins for GEM-Selektor. The GEMS Development Team currently involves 11 members from 6 institutions, and strives to implement innovative concepts, modern algorithmic frameworks, and tools to improve thermodynamic data, all in order to ensure the state-of-the-art functionality of GEMS for the next decade.

PERMON is a set of solvers combining quadratic programming and domain decomposition methods. It makes use of and extends the PETSc framework for numerical computations. The core of PERMON is formed by the PermonQP package which is able to solve large scale quadratic programming (QP) problems. Among the main applications are contact problems of mechanics and Support Vector Machines (SVM) machine learning. Contact problems can be decomposed by means of TFETI (Total Finite Element Tearing and Interconnecting) non-overlapping domain decomposition method implemented in the PermonFLLOP package. SVM can be solved by PermonSVM. Both PermonFLLOP and PermonSVM make use of

PermonQP to solve the resulting QP problems. For more information see https://www.mcs.anl.gov/petsc/ with reference to http://permon.vsb.cz/index.htm.

Flow123d (https://flow123d.github.io/) is a simulator of underground water flow, solute and heat transport in fractured porous media. Novelty of this software is support of computations on the mixed meshes consisting of simplicial elements of different dimensions. Therefore, we can combine continuum models and discrete fracture network models. Current development version includes: mixed-hybrid solver for (un)saturated Darcy flow, finite volume model and discontinuous Galerkin method for solute transport of several substances, DG method applied to the heat transfer and poroelasticity coupling. Several kind of reaction processes are supported including dual porosity, sorption, decays and simple reactions. Computations can be run in parallel using MPI and PETSc with scalability up to hundreds of processors. The input interface based on YAML file format allows specification of general space-time dependent data for any physical parameter that does not compromise performance. Program supports output into GMSH and VTK formats. Implementation use C++ and Python, software is published under use GNU public licence v3.

OpenGeoSys (OGS) (www.opengeosys.org) is a scientific open-source initiative for the numerical simulation of thermo-hydro-mechanical/chemical (THMC) processes in porous and fractured media. The basic concept of OGS consist on providing a flexible numerical framework, using primarily the Finite Element Method (FEM) for solving multi-field coupled processes with application in different scientific and technical disciplines. For example, OGS has been successfully applied in the fields of regional, contaminant and coastal hydrology, technical and geothermal energy systems, geotechnical engineering, energy storage, CO2 sequestration/storage and nuclear waste management and disposal. The actual version OGS-6 is providing complete workflows starting from data integration, HPC for coupled process simulation and using virtual reality (VR) concepts for data analytics. OGS-6 is developed and maintained platform-independently using professional software engineering tools such as version management (GitHub) and containerization (e.g. Docker, Singularity). A strict code review is conducted for quality assurance completed by unit testing and comprehensive benchmarking. OGS provides open interfaces for combining with other simulators (e.g. GEMS, iPhreeqc for geochemical processes) including Python bindings. A recent overview of OGS software engineering can be found in Bilke et al. (2019).

CORE^{2D} V5 (Fernández, 2017) is a code for transient saturated and unsaturated water flow, heat transport and multicomponent reactive solute transport under both local chemical equilibrium and kinetic conditions in heterogeneous and anisotropic media. The flow and transport equations are solved with Galerkin finite elements and an Euler scheme for time discretization. The solute transport equation accounts for advection, molecular diffusion and mechanical dispersion. The chemical formulation is based on the ion association theory and uses an extended version of the Debye-Hückel equation (B-dot) for the activity coefficients of aqueous species. The following chemical reactions are considered: aqueous complexation, acid-base, redox, mineral dissolution/precipitation, cation exchange, surface complexation and gas dissolution/exsolution. CORE^{2D} V5 relies on the "*com*" thermodynamic database of EQ3/6 (Wolery, 1992). The code also allows the use of other thermodynamic databases. CORE^{2D} V5 is based on the sequential iteration approach to solve for chemical reactive solute transport. Iterations are repeated until some prescribed convergence criteria are attained (Javier Samper and Yang, 2009). The Gaines-Thomas convention is used for cation exchange. Surface complexation is modelled by using three types of protonation/deprotonation sites, S^{S}-OH, S^{W1}-OH and S^{W2}-OH, as proposed by Bradbury and Baeyens (1997). CORE^{2D} V5 takes into account the changes in porosity due to mineral dissolution/precipitation reactions and their feedback effect on the flow and transport parameters under isothermal and nonisothermal conditions (Fernández, 2017). This version also was updated to use available public-domain post-processing tools.

INVERSE-FADES-CORE V2 (Mon, 2017) is a finite element code for modelling non-isothermal multiphase flow, heat transport and multicomponent reactive solute transport under both chemical equilibrium and kinetic conditions in deformable media. The code takes into account the mass balance of water, air, solid and enthalpy; the transport of solids and the mechanical equilibrium. The solute transport equation accounts for advection, molecular diffusion and mechanical dispersion. INVERSE-FADES-CORE V2 solves both forward and inverse multiphase flow and multicomponent reactive transport problems in 1-, 2- and 3-D axisymmetric

porous and fractured media. The state variables of the forward model include the liquid and the gas pressures and temperature, which are solved with a Newton-Raphson method. The inverse problem is solved by minimizing a generalized least-squares criterion with a Gauss-Newton-Levenberg-Marquardt method (Dai and Samper, 2004). The forward routines of INVERSE-FADES-CORE have been widely verified with analytical solutions and other reactive transport codes. Mon (2017) implemented reactive gas transport in INVERSE-FADES-CORE V2 by including additional mass balance equations for the reactive gaseous species in the gaseous phase. INVERSE-FADES-CORE V2 has been benchmarked with other codes for benchmarking problems such as: 1) The chemical interactions of the concrete liner with the compacted bentonite of the engineered barrier and the clay host rock; and 2) The carbonation of concrete in unsaturated conditions during the operational period of a repository.

3. Scale transition schemes for coupled processes

Scale transitions with related, subsequent models are the only way to convey multi-physics, chemical as well as microbiological process information between different scales of interest (Bilke et al., 2019). Due to coupling effects and non-linear phenomena, upscaling methods are a particular challenge for coupled THM/C processes. In general we can distinguish between different directions in multi-scale modeling, e.g. (1) development of inherent upscaling methods (e.g. coarse graining), (2) building connected model chains (see Figure 1), (3) using computational power (i.e. High-Performance-Computing) to increase model domains (Link to Task 2). As there is no perfect way for scale transition, we have to follow several directions in Task 3.

3.1 Multiscale Modelling

Figure 1 serves as an explanation for (2) building connected model chains. It starts from modeling at the process scale using methods down to the atomistic, molecular scales such as Lattice-Boltzmann, Lattice-Element approaches, Molecular Dynamics or Particle Methods. These modelling techniques are able to represent fundamental processes at very small (atomistic) scales, but computationally very expensive and difficult to upscale to measurement scales. The opposite end of the scales is represented by so-called complexity-reduced models. At this scale, questions of long-term environmental impacts of radioactive waste repositories have to be addressed. To this purpose data from various sources have to be combined including geological and geographical information as well as socio-economic aspects. For large scale analysis, stochastic approaches and network simulation are being used. Only a small portion from small scale simulations can be used at this scale. However, fundamental process understanding will be a key for more reliable long-term predictions.

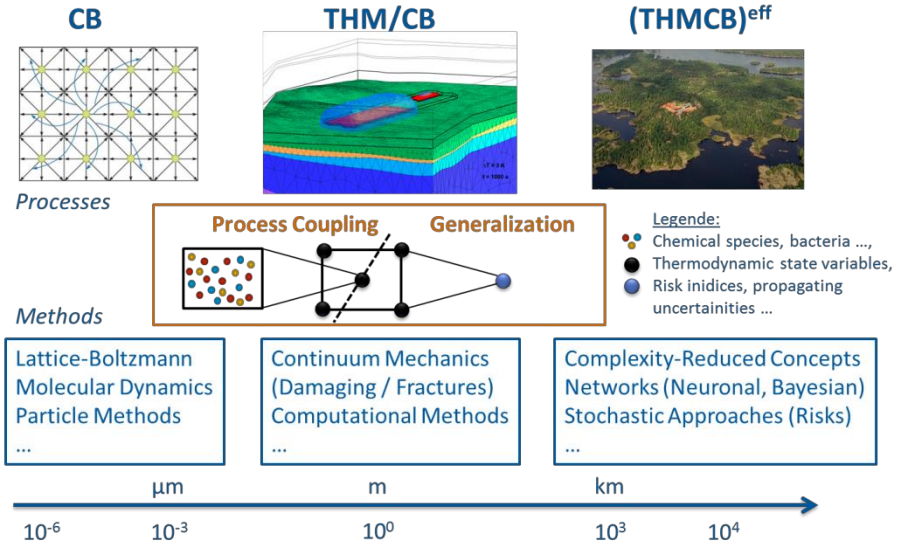

Figure 1: Vision for scale transition schemes: from process to management scales (figure sources: https://developer.nvidia.com, BGR (OGS simulation), Google Earth).

A possible link between the small and large scales, in particular for multi-physics problems such as THM/C, is continuum mechanics as it is based on first-order principles (conservation laws of mass, momentum and energy). On the other side continuum mechanics invoke thermodynamic consistent, constitutive laws, which may describe complex material behavior at multiple scales (e.g. micro-mechanics). In addition to fundamental continuum mechanical approaches, numerical methods using significant computational power (HPC) are available for multi-scale purposes. However, it has to be clearly stated that computational mechanics for sure can handle a certain range of process scales but not the whole required scale chain as shown in Figure 1. Therefore, the development of reliable and computational efficient model chains is important to further improve performance and safety assessment tools of radioactive waste management.

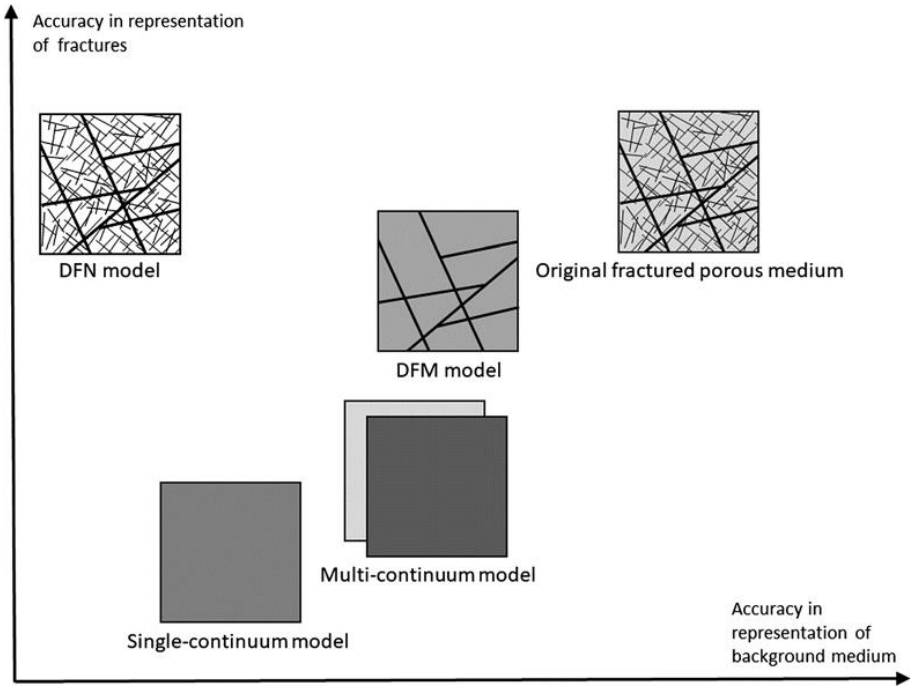

Figure 2: Conceptual approach for fractured porous media (Berre et al., 2019).

In addition to building model chains over scales, as mentioned above, the second challenge for THM/C modeling is the structural complexity of potential host rocks for nuclear waste repositories. A key structural element for geological formations is the presence of discontinuities (e.g. faults, fractures, cracks). Characterizing the state of the stress (i.e. spatial stress directions and magnitudes) in a field is, if not impossible, extremely difficult. Even if the general stress field of a potential site is reasonably estimated, it is nearly impossible to map a precise description of the existing fractures and faults and to predict their potential evolution when temperatures, pore pressures, mechanical stresses are changing. For these reasons, characterization of the earth model is typically combined with stochastic approaches involving multiple realizations in order to bracket the reality. For upscaling, so-called fractured-porous media, different methods have been developed in past (Berre et al., 2019), Figure 2): (1) Discrete-Fracture-Network (DFN) models explicitly take into account the presence of discontinuities, (2) Single and Multi-Continua approaches use effective, statistically representative parameters for describing fracture behavior (REV concept). The main idea of Multi-Continua models is to couple multiple continua via exchange functions, where every continuum is representing a typical part, i.e. fracture systems, rock matrix. (3) Discrete-Fracture-Matrix (DFM) models are combining both discrete and continual behaviors. Discrete approaches (1&3) require very precise numerical methods for accurate representation of exchange processes between fracture and matrix compartments.

3.2 Multi-scale experiments

Dissolution and precipitation of minerals and its impact on the transport of fluids and solutes in porous media is a key factor for the long-term behaviour of natural and engineered systems in the subsurface. The implementation of such coupled processes into numerical reactive transport codes requires a mechanistic process understanding and model validation with quantitative experiments (Tartakovsky et al., 2008; Katz et al., 2011; Poonoosamy et al., 2015; Poonoosamy, Klinkenberg, et al., 2019). These experiments are usually based on a simple chemical setup with well-defined kinetic and initial transport parameters. The discrepancies between experimental observations highlighted the limitations of continuum scale models when it comes to predicting changes in the transport properties in porous media due to mineral precipitation and consequent porosity changes (Chagneau et al., 2015; Poonoosamy et al., 2015; Poonoosamy, Klinkenberg, et al., 2019). The primary cause for these discrepancies is that reactive transport operates at the continuum scale and considers upscaled parameters. Consequently, the interdependencies between

porosity and transport parameters for instance, are accounted for in numerical models by using empirical relationships developed on the continuum scale (Hommel et al., 2018). These relationships were either fitted to experimental data or based on theoretical considerations (Poonoosamy, Klinkenberg, et al., 2019; Seigneur et al., 2019). However, solute transport depends on more complicated pore space changes (pore geometry, pore connectivity) than the upscaled parameter "permeability" as accounted for in continuum-scale reactive transport approaches. Moreover, the averaging process also includes geochemical parameters such as reactive surface area and reaction rates. The heterogeneity in porous media likely controls the mass fluxes and residence time in the reactive media and therefore are known to control the effective rates of mineral dissolution by reducing effective mineral surface areas physically (e.g. in fractures, or at partial saturation) and chemically (Berkowitz et al., 2016; Deng, Molins, et al., 2018; Deng, Steefel, et al., 2018; Jung and Navarre-Sitchler, 2018a, 2018b). Moreover, the reactive surface area of minerals constituting the porous media is not directly experimentally accessible and is often used as a fitting parameter in reactive transport simulations (Noiriel et al., 2009).

Because of the heterogeneity of porous media, defining a representative evolution of effective properties of the porous medium is a challenge. In-situ imaging of the evolution of the porous media and visualization of transport process using advanced experimental methods. Such a method is the X-ray computed tomography (Spanne et al., 1994; Coles et al., 1996, 1998; Lindquist et al., 2000; Noiriel et al., 2004; Blunt et al., 2013). The neutron radiography (Shafizadeh et al., 2015), the magnetic resonance imagery (Fridjonsson et al., 2011), and the positron emission tomography (PET) (Kulenkampff et al., 2018) provide details at different scales. Finally, the microfluidic experiments (Harrison et al., 2017; Soulaine et al., 2017, 2018; Poonoosamy, Westerwalbesloh, et al., 2019) in combination with pore scale modelling can be versatile approach to gain mechanistic process understanding and provide new methodologies for upscaling. Pore scale modelling can be used to investigate physicochemical processes (nucleation mechanism, passivation of surfaces, creation of unconnected porosities etc.) that are not resolved in continuum scale models (Prasianakis et al., 2017, 2018).

The combination of experiments and simulations can provide a fundamental understanding on the sub-continuum scale processes in the pore space, which is needed for the derivation of permeability-porosity and diffusivity-porosity or other porosity dependent empirical relationships. However, studies using macroscopic experiments to provide a microscopic understanding of mineral precipitation and feedback to transport properties of porous media have only been started recently and are scarce so far (Poonoosamy, Klinkenberg, et al., 2019).

3.3 Scale transitions schemes for hydro-mechanical processes

In the following section we briefly describe numerical methods which are actively developed by the project partners in order to tackle scale transition problems

3.3.1 Two common approaches for numerical simulation of fractures Generalized/Extended Finite-Element (G/XFEM) and Variational Phase Field (VPF) methods

3.3.1.1 Generalized/Extended Finite-Element-Method

In recent years, generalized (Strouboulis et al., 2000) or extended (Belytschko and Black, 1999) finite element methods (G/XFEM) and phase-field models (Bourdin et al., 2000) for the description of existing and developing discontinuities and singularities within continuum mechanical approaches have established (Belytschko et al., 2009) themselves ahead of all others in recent years. Both methods differ fundamentally and have their own strengths and weaknesses. G/XFEM locally extends the approach and test function space by formulations that can map the discontinuous course of the solution and introduces corresponding additional local degrees of freedom. Usually, this approach is combined with so-called level set methods, which help to localize the discontinuity and thus ultimately determine in which elements the solution space has to be extended. This approach allows the approximation of discontinuous solutions on comparatively coarse grids, but requires extensive programming infrastructures for the treatment of flexible additional

degrees of freedom, level sets and other aspects, which require a not inconsiderable implementation effort, especially in branched crack systems. In contrast, the phase field method represents a sharp interface such as discrete crack with a smoothly varying scalar field which is regularized by the internal length. Thus the discontinuity is not described as a low-dimensional unit, but by the means of an additional variable defined on the entire solution area, which originates in certain characteristics of an energetically motivated variational formulation in (Francfort and Marigo, 1998). This approach can be implemented into existing continuum codes without additional algorithmic effort where it is immediately available for solving arbitrary-dimensional problems. Crack formation, propagation, branching and unification also occur without additional effort.

G/XFEM (Belytschko and Black, 1999) was originally developed for crack propagation problems and was also applied in the geotechnical context, e.g. for multiphase flows (Chessa and Belytschko, 2003; Mohammadnejad and Khoei, 2013) and heat transport (Khoei et al., 2012; Shao et al., 2019). Current developments of generalized and extended finite element methods in the context of hydraulic stimulation are mainly concerned with the efficient coupling of solid-state and flow-mechanical problems (Yazid et al., 2009; Watanabe et al., 2012; Meschke and Leonhart, 2015).

3.3.1.2 Variational Phase-Field models for fractures

The theoretical basis of the so-called phase-field model for fracture was incepted in the work by Francfort and Marigo (1998) that revisted Griffith's fracture criteria as an energy minimization problem (variational approach), and later on its numerical solution was proposed by regularizing the energy functional with a smoothly varying scalar (phase-field variable) and a regularization length parameter (internal length) by Bourdin et al. (2000). Although their work had gone relatively unnoticed in the mechanics community for a decade or so, its popularity has been surging since a ceremonial publication by (Miehe et al., 2010) who coined the method "phase-field model". Its brief narrative history can be found in a summary article in (Bourdin and Francfort, 2019). In recent years, the phase-field model enjoys its wide applicability ranging from brittle and cohesive fracture (Bourdin et al., 2008; Amor et al., 2009; Hakim and Karma, 2009; Kuhn and Müller, 2010; Pham et al., 2011; Verhoosel and Borst, 2013; Vignollet et al., 2014; Ambati, Gerasimov, and Lorenzis, 2015; Marigo et al., 2016; Wu, 2017; Sargado et al., 2018; Tanné et al., 2018), to ductile fracturing (Ambati, Gerasimov, and De Lorenzis, 2015; Miehe, Hofacker, et al., 2015; Kuhn et al., 2016; Alessi et al., 2017), fatigue (Alessi et al., 2018), desiccation fracture (Maurini et al., 2013; Cajuhi et al., 2018), dynamic fracturing (Bourdin et al., 2008; Borden et al., 2012; Schlüter et al., 2014; Li et al., 2016), and hydraulic fracturing (Bourdin et al., 2012; Miehe, Hofacker, et al., 2015; Yoshioka and Bourdin, 2016; Ehlers and Luo, 2017; Lee et al., 2017; Santillán et al., 2017; Chukwudozie et al., 2019).

A set of crack (Γ) in a domain (Ω) are discontinuities where a jump in quantities such as displacement is observed (Figure 3). Therefore, when represented in a discretized form (e.g. finite element), the discretization must honor the discontinous geometry of Γ, and it can get easily untractable when the crack set evolves with time (Figure 3, left). In phase-field models for fracture, the discontinuty (Γ) is represented by a smoothly varying scalar (v as in Figure 3, right) so that the discretization does not need to track the geometry change but the change in the scalar field instead. Naturally a special care must be taken in setting up such a scalar field in order to preserve the energy equivalance in the two systems and the convergence of the phase-field representation to the discrete fracture as the regularization approaches zero (Chambolle, 2004; Bourdin et al., 2008).

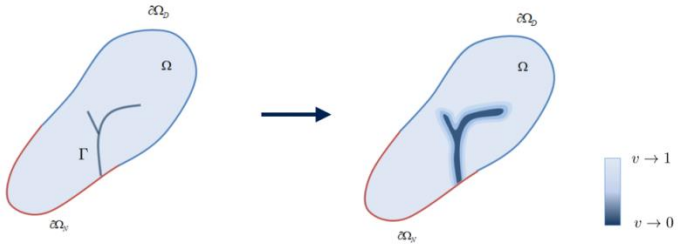

Figure 3: Concept of the Variational Phase Field (VPF method) for handling discontinuities in the context of coupled processes.

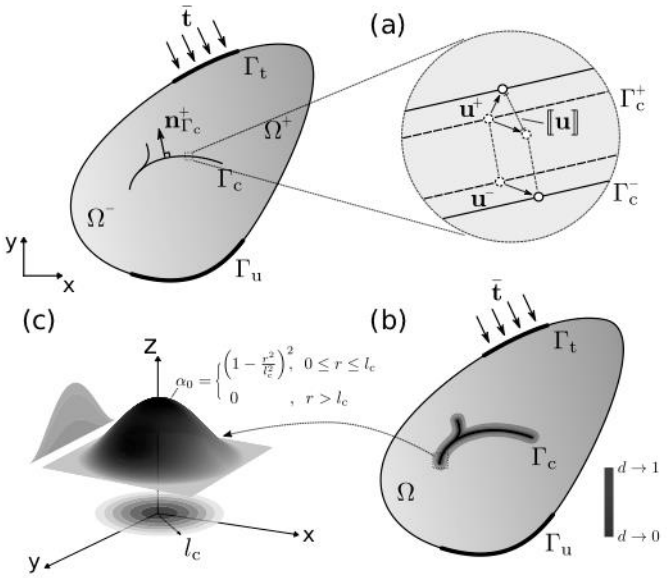

Figure 4: Graphical comparison of numerical methods for description of discontinuities (Yoshioka et al., 2019)

First extension of the phase-field model to hydraulic fracturing was proposed in (Bourdin et al., 2012) by adding the fluid driven pressure work on the crack lips and Wheeler et al. (2014) further adapted it by adding the poro-elastic effects. In these models, while the pressures in the non-fractured medium ($\Omega\backslash\Gamma$) and the fracture (Γ) are distinguished, their profiles are considered spatially uniform within the domains. Phase-field fracture models coupled with Darcy-Reynolds flow system were developed in Mikelic et al. (2015a), Miehe et al. (2015), Miehe and Mauthe (2016), Santillan et al. (2017), Xia et al. (2017) and Lee et al. (2017) and Darcy-Stokes type in Wilson and Landis (2016), Ehlers and Luo (2017) and Heider and Market (2017). Other "homogenization" approaches include a model that uses the phase-field variable as an indicator in Mikelic et al. (2015b) or as a weight function for permeability averaging in Miehe et (2016). Heider and Market (2017) and Ehlers and Luo (2017) developed the phase-field fracture model within the framework of the Theory of Porous Media (TPM) and considered the phase-field variable as a weight between the Darcy flow in porous media and the Stokes flow in fracture. Alternatively, coupling with fluid flow has been simplify accomplished by linking to an external standalone fluid flow simulator in Wick et al. (2016) and Yoshioka and Bourdin (2016) where a phase-field variable was used to differentiate the fracture from the non-fracture domain. Instead of using the phase-field variable as an indicator or a weight, Chukwudozie et al. (2019) applied the phase-field calculus to homogenize the system while conserving the mass.

Another mass conserving approach was proposed by Santillan et al. (2017) where an independent 1D domain is generated dynamically out of the main computational domain based on certain the phase-field threshold and the fluid flows are solved separately and iteratively

Figure 4 depicts a graphical representation of various numerical methods for the description of evolution of discontinuities, i.e. (a) lower-interface-method (LIE), (b) variational-phase field method (VPF), and non-local-deformation (NLD) methods, which will be also used in the context of various EURAD work packages (e.g. WP-GAS).

3.3.2 Multi-continua methods for hydro-mechanical processes

3.3.2.1 Modelling approach

So-called multi-continuum models are often used to better determine the heterogeneity of fractured or karstied aquifers. For example, double-continuum models take advantage of the different hydraulic properties of the rock matrix and the fissure system. While fissure systems are characterized by high permeabilities and low storage capacities, the rock matrix has low permeabilities and high storage capacities. Thus, the fissure system and the rock matrix are attributed to separate "overlapping" continua, which are coupled with each other via source-sink terms in the entire model area (see Figure 2).

Similar to the multiphase concept of a porous medium, several state variables are defined at each point in the multi-continuum model. In addition, the individual continua also have different material parameters. If one wanted to use the category of phase for a double-continuum model, it would be an allocation from a macroscopic point of view: thus not in the microscopic sense fluid and solid, but fissure system and matrix. The key point in multi-continuum modelling is the description of the interaction between the individual continua. In a sense, the exchange relationships represent non-equilibrium conditions. The recording of the local dynamics of exchange processes (gradient approaches) is problematic. Compared to the classical single-continuum approaches, multi-continuum models are better able to map heterogeneities. The differentiated continua have to process smaller structural variabilities, which has an advantageous effect on the averaging technique (e.g. size of the REV). Average state variables can be determined separately for the fissure and matrix continuum rather than an average state variable for the entire fissure-matrix system.

The numerical implementation of multi-continuum models can be done on the one hand by coupling separate equations for fissure and matrix media via exchange terms (Bibby, 1981; Huyakorn et al., 1983; Birkhölzer et al., 1993a, 1993b) or on the other hand by simultaneously solving the equation systems using iterative techniques (Pruess and Narasimhan, 1982; Neretnieks and Rasmuson, 1984).

Multi-continuum models are widely used, for example, in the interpretation of pump tests in solid rock or the delayed hydraulic response to precipitation events in karst aquifers (Sauter et al., 2006), the hazard assessment of groundwater deposits in near-surface solid rock areas and the exploration of oil and gas deposits (Gilman et al., 1983). An advantage for the model calibration is the justifiable exploration effort to adapt the limited number of model parameters. Typical geological formations for which classical single-continuum models may fail and require multi-continuum approaches include porous sandstones, karst aquifers, loose rocks with clay intercalations or silty portions, and fissured bedrock. The application potential of the multi-continuum concept to process simulation in hierarchical fissure or pore systems (e.g. intergranular pores of the grain skeleton and micropores in the rock grains) seems to be interesting. In the process, fracture systems of different scale sizes are to be assigned individual continua (Clemens et al., 1996). More recent efforts in model development also concern the coupling of discrete fissure models with multi-continuum models.

3.3.2.2 Hydromechanical Problems

Dual and multiple continua for hydromechanical problems follows from original models for this kind of conceptual representation of individual processes of solute transport with dead-end pores or mechanics of composites. More details of development of the porous/fracture flow and solute transport models is given in the section (3.3.2.). Interaction of stress and deformation with fluid flow, the general used theory is by Biot (1941) and the generalisation into the dual porosity concept of poroelasticity is by Aifantis (1977).

The recent work aims to various applications, with shares of reservoir engineering, hydraulic fracturing, and deep geological repository. More frequent are hierarchical porous media than upscaled fractured rock. Ashworth and Doster (2019) compares various formulas of previous authors, especially 3 works in details,

including a table form of coefficient formulas list, and evaluates the importance of coupling. Example calculation is with Mandel problem (Nguyen and Abousleiman, 2010) – Figure 5.

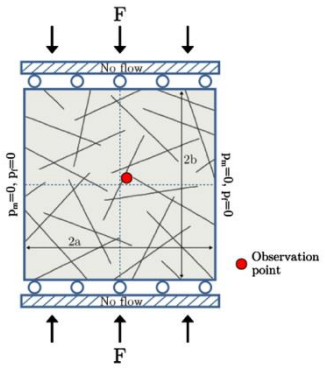

Figure 5: Problem used for benchmarking dual-continuum poroelastic models (Nguyen and Abousleiman, 2010).

Liu et al. (2018) derive theoretical formulation and analytical solutions to the inclined wellbore problem and axisymmetric Mandel-Type problem of dual-porosity, dual-permeability poro-chemo-electro-elasticity. The model is able to explain phenomena which could seem anomalous without consideration of the coupling effects. Kim et al. (2018) simulates hydraulic fracturing process by means of 3D thermo-poro-mechanical model including the dual porosity approach. The movement of water is retarded with respect to the fracture propagation. It also theoretically demonstrates which of the effects can be detected by electromagnetic geophysical methods. Vasilyeva et al. (2019) fits the method to perforated domains, which establish quite active sub-topic within the dual-continuum upscaling. Non-local multi-continuum method (Chung et al., 2018) is used which is specific in construction of local basis functions capturing also the non-local effects. Special attention is to generalisation of the boundary conditions on perforations. Choo et al. (2016) and Sanchez et al. (2016) deal with unsaturated conditions of the dual-porosity media and model the hydro-mechanical and thermos-hydro-mechanical processes, respectively. Kim et al. (2015) suggest an algorithm for coupling of chemo-thermo-poro-mechanics, for fractured rock. Wang et al. (2014) simulates the fractured rock, with effect of stress on flow and transport, as an issue of the deep geological repository safety. The transport is represented by multiple continua (MINC) and the hydro-mechanics by Oda (1986) crack tensor theory, denoting the method as Ex-MINC (extended) - Figure 6. The benchmark problem was also used to successfully compare this upscaled homogenization concept with several solution on full fracture network (small-scale view).

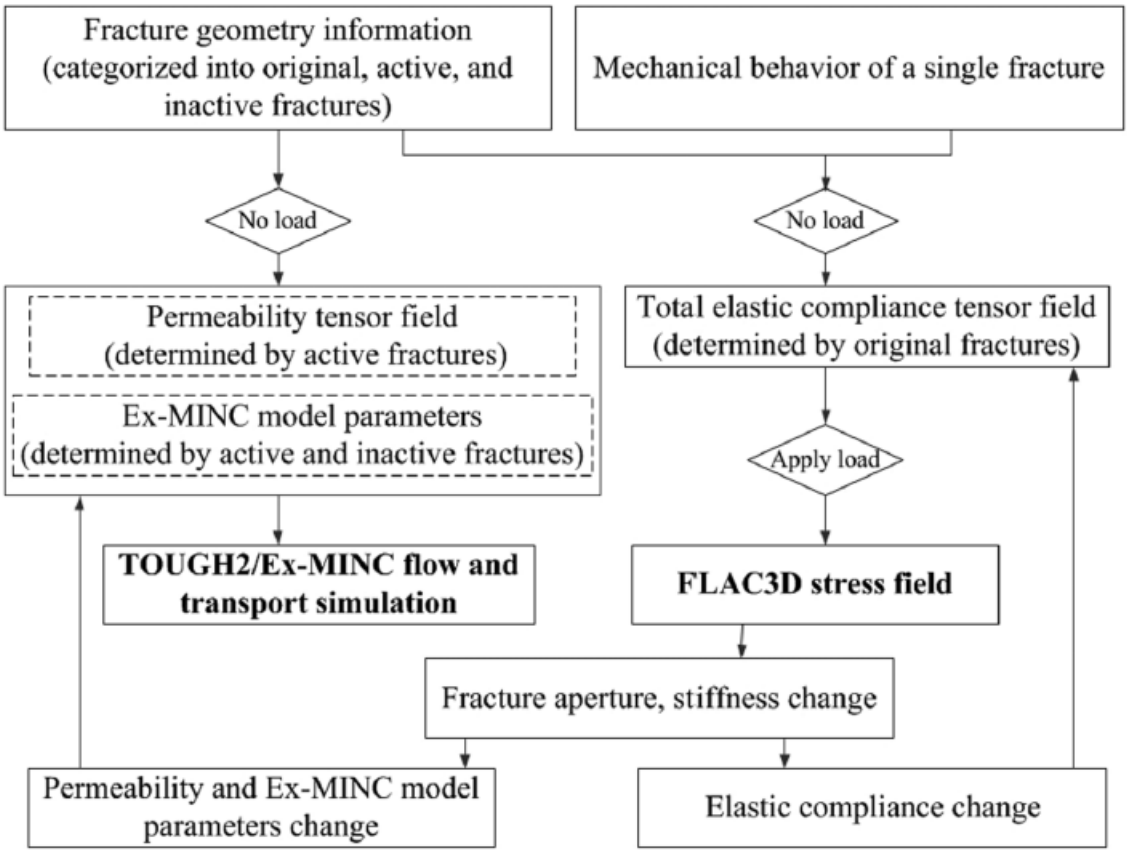

Figure 6: Scheme of the extended multiple interacting continua (Ex-MINC) method (Wang et al., 2014).

3.3.3 Lumped Parameter Modeling

The term Lumped Parameter Modeling (LPM) is not yet used in context of repository models, but many of them are built on these principles. The meaning is that a system is represented by a single value of certain quantity (either a parameter or a state variable) for whole or a component, in an average or integral meaning, contrary to point values representing infinitesimally small volume. The classical use of LPMs is to create the model based on system observation, capturing its behavior as a link between “input” and “output”, or control and response quantities – understanding the system as “black-box”. Alternatively, LPMs can be seen as an upscaling method, using simplified and numerically efficient model with less degrees of freedom whereas the lumped behavior is based on solution of detailed equations controlling the system (physically-based model). The principal ingredient of such approach is then a procedure of integral postprocessing of quantity fields resulting from the controlling equations.

Besides LPM, also a term “compartment model” is used in similar meaning, but including flexibility such that the meaning of a compartment can be either a lumped subsystem, or a discretization element (Figure 7). Such case is the concept in Goldsim software (Golder Associates, 2014), which is also considered as one of the options in Czech performance assessment of spent nuclear fuel repository (Vopálka et al., 2006).

There are not many recent studies using lumped models for flow and solute transport, which are meant also as basis for the performance assessment models. Therefore the references cover a longer period and contain work which was not further developed in the recent 5-10 years. The state of the art report is limited to the partial topics of how the continuous field quantities of 2D/3D models are integrated into the lumped parameter models and how the models with various features related to LPMs are used in performance assessment.

Figure 7: Scheme of a compartment capturing a solute transport process in the lumped-parameter model sense (Golder Associates, 2014).

The principle of "integrating" the physics into the compartment block in/out behavior in Goldsim software (Golder Associates, 2014) is the use of analytical solution, which calculates a "breakthrough" of radionuclide controlled by homogeneous fracture and rock parameters for the respective compartment. Similar concept of analytical solution is introduced by (Chopra et al., 2015), which improves the referred existing (mostly widely used) solutions with adding the proper hydrodynamic dispersion solution.

A lumped model denoted as Tank model for regional groundwater flows is proposed for a case study of the groundwater flow system (Kazumba et al., 2008). The aquifer is divided into Tanks within which the average values of the groundwater levels are assumed to be representative in the Tanks. For each Tank the mass-balance equations are written. The Quasi–Newton optimization technique together with Akaike's Information Criterion, AIC, are employed in order to balance the model/observation fit and reliability of the parameter estimation. The results suggest disconnection of some regional aquifers as well as inflow of seawater to the deeper layers.

The case study (Hagedorn et al., 2018) examines the nitrate pollution vulnerability of groundwater in sedimentary aquifers of California's South Coast Range using stepwise logistic regression modelling. The study highlights the utility of two parameters, Dissolved Oxygen and Modern Water Proportion, for predicting nitrate contamination vulnerability. Authors conclude a possibility to use the results to guide field investigations in areas with little data but similar hydrogeology and land use.

Overview paper (Rechard et al., 2014) summarizes modelling of radionuclide transport in the unsaturated and saturated zone conducted between 1984 and 2008 to evaluate the performance of Yucca Mountain disposal system. The listed models start at 1-D transport for a single porosity media without lateral dispersion in both the saturated and unsaturated zone for the first assessment in 1984. Gradually the models progressed until 2008 to the 3-D transport model the saturated zone, combined with 1-D transport results, which evaluated decay of radionuclides, in order to evaluate compliance with groundwater protection requirements. Uncertainty in flow within the unsaturated and saturated zone was generally important to explaining the spread in the individual dose performance measure.

For Nuclear fuel cycle and nuclear waste disposal decisions, (Huff, 2017) developed the Cyder disposal environment and repository module, which implements models of medium likelihood of hydrological radionuclide to support assessment suitable for fuel cycle simulation simulator "Cyclus". By means of four modules with close-form solution of physics it is possible to reduce the computational need.

The lumped Rapid Saline Groundwater Exfiltration Model (RSGEM) was developed to support operational water management of freshwater resources in coastal lowlands, a need exists for a rapid, well-identifiable model to simulate salinity dynamics of exfiltrating groundwater (Delsman et al., 2017). The RSGEM was applied to a field site in the coastal region of the Netherlands, parameter estimation and uncertainty analysis were performed using generalized likelihood uncertainty estimation. The model showed good correspondence to measured groundwater levels, exfiltration rates and salinity response. Moreover, RSGEM results were very similar to a detailed, complex groundwater flow and transport model previously applied to this field site.

The model representing the groundwater pathway in risk assessment or performance assessment developed by (Robinson and Chu, 2013) is called the Residence Time Distribution (RTD) Mixing Model (RTDMM), allows for an arbitrary distribution of fluid travel times to be represented, to capture the effects on the breakthrough curve of flow processes such as channelized flow and fast pathways and complex three-dimensional dispersion. Mathematical methods are derived directly from the theory of residence time

distributions. A simple mixing model is presented, along with the basic equations required to enable an arbitrary RTD to be reproduced using the model. The advantages include easy incorporation into a multi-realization probabilistic simulation; computational burden similar to 1D model with the same number of grid cells; and straightforward implementation into existing flow and transport modelling codes. Examples include cases of a radioactive decay chain, dual porosity transport and sorption.

Kawasaki and Ahn (2008) also employs the residence time distribution and includes stochastic methods like random walk. The evaluation can be modularised and so it convenient e.g. for efficient representation of multiple waste packages. It also avoids artificial numerical dispersion for coarse discretisation.

Murakami and Ahn (2011) develops a model with array of compartments, among which a transition probability matrix describes radionuclide transport (Markov-chain process). Transition probabilities are upscaled from finite-element flow and transport solutions. Authors suggest efficiency of the method for modelling effects of parameter changes. Paper of Cadini et al. (2010) brings another example of compartment model with stochastic features, where detailed local-scale modelling results “feed” a global-scale analysis of the repository, at reasonable computational expenses.

The older work of Gylling et al. (1997) represents a reference case of performance assessment model which has features of both distributed and lumped model. The channel network model (CHAN3D) for solute transport in fractured granite uses direct input of data (not as upscaling), but the channels are defined with strong abstraction of physical meaning (regularized geometry with transforming of parameters to the same effective properties).

3.4 Scale transitions schemes for hydro-chemical processes

3.4.1 Lattice Boltzmann Method (LBM)

The lattice Boltzmann method (LBM) is a special discretization of the Boltzmann equation operating at the level of velocity probability distribution functions f_i, with focus on fluid dynamics and mass and heat transport phenomena. At every distribution function corresponds a discrete velocity vector (Frisch et al., 1986; Succi et al., 1989). Different discretization schemes lead to different number of discrete velocities, there exist different lattice models (Higuera et al., 1989; Qian et al., 1992). For the recovery of Navier-Stokes fluid motion equations, for two-dimensional simulations the standard D2Q9 square lattice with 9 discrete velocities is usually implemented, and for three dimensional simulations the D3Q27 cubic lattice with 27 discrete velocities, due to their simplicity and robustness in complex geometry domains (see Figure 8). The diffusion equation can also be recovered on the same or smaller velocity sets.

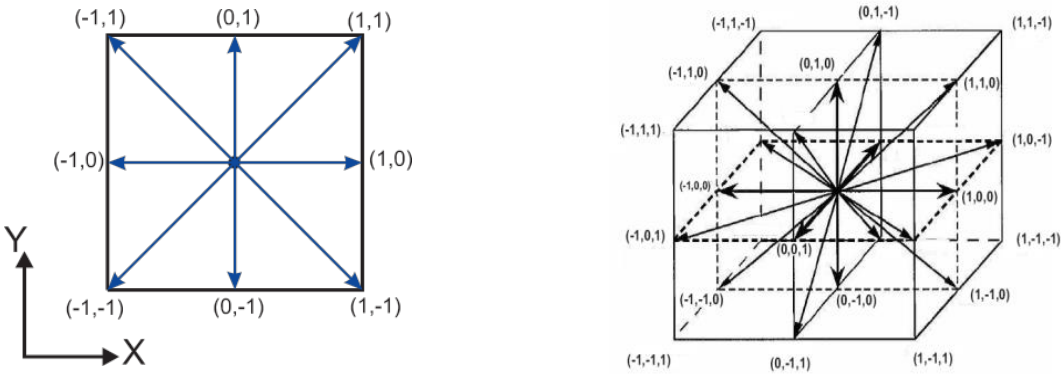

Figure 8: Standard lattices (left) two dimensional 9-velocity lattice (D2Q9), (right) the three dimensional 27-velocity lattice (D3Q27)

For reactive transport simulations in geological relevant flows, the mass is transported via diffusion and advection. For the modelling of the advection-diffusion equation the passive scalar approach is usually implemented. Such a model is composed of a basis fluid medium (a population set which has the dynamics as described by the Navier-Stokes equations , plus a passive scalar coupled population set that simulates

the diffusion of ions. The basic lattice Boltzmann discretization approximates the Navier-Stokes equations to a certain accuracy, which for very high flow velocities can become significant and can manifest itself as the loss of Galilean invariance. There are several ways to overcome this issue. For example, the isothermal guided equilibrium nine-velocity model (D2Q9 lattice), can be selected as the basis model for the advective flow field as well as for the field that advect and diffuse (Prasianakis et al., 2009, 2017). This model provides enhanced Galilean invariance and rotational isotropy on the standard lattices.

The discrete velocities of populations f_i for i=0-8, are c_i = (0, 0) for i = 0, c_i = (±1, 0) and (0, ±1) for i = 1–4, and c_i = (±1, ±1) for i = 5–8 (Qian et al., 1992). The following population-moments correspond to the density of the solution ρ and the momentum j_a in the direction a = x, y:

$$\sum_{i=0}^{8} f_i = \rho \ , \ \sum_{i=0}^{8} c_{ia} f_i = j_a \tag{1}$$

The guided equilibrium populations f_i^{eq} are given in a closed form, where T_0=1/3:

$$f_i^{eq} = \rho \prod_{a=x,y} \frac{\left(2c_{ia}^2 - 1\right)}{2^{c_{ia}^2}} \left(c_{ia}^2 - 1 + c_{ia}u_a + u_a^2 + T_0\right) \tag{2}$$

The Boltzmann BGK equation is solved:

$$\partial_t f_i + c_{ia}\partial_a f_i = -\frac{1}{\tau}\left(f_i - f_i^{eq}\right) \ , \tag{3}$$

where τ is the relaxation parameter and $\mu = \tau\rho T_0$ is the resulting macroscopic dynamic viscosity. BGK stands for the Bhatnagar-Gross-Krook collision model as depicted in the right hand side of the aforementioned equation and describes the relaxation of populations f_i to their equilibrium state f_i^{eq} with relaxation time τ. For the advection-diffusion reaction equations of the reactive species a D2Q9 model can also be implemented. The solutes (primary and secondary species) will advect and diffuse along the streamlines of the flow field. The equilibrium populations ξ_i^{eq} for the reactive species ξ_i are given:

$$\xi_i^{eq} = C_i \prod_{\alpha=x,y} \frac{\left(1 - 2c_{i\alpha}^2\right)}{2^{c_{i\alpha}^2}} \left(c_{i\alpha}^2 - 1 + c_{i\alpha}u_\alpha + T_0\right), \tag{4}$$

where C_i is the concentration of the considered ions and u_α is obtained from the basis model. The relevant population moment that corresponds to the concentration is:

$$\sum_{i=0}^{8} \xi_i = C_i \tag{1) (5}$$

Advantages of the LBM for reactive transport geochemical applications are the ability to simulate evolving geometries of high complexity (due to dissolution or precipitation), the local mass and momentum conservation, as well as the parallelization efficiency. The high scalability of LBM codes is related to the locality of the mesh based particle-particle interactions, which requires only next neighbor communication between the grid points. To that, it should be added that LB codes are usually simple in terms of coding and they do not depend on external libraries. This allows the incorporation of additional physics (electrochemical, multiphase etc.) in a relatively fast pace, as soon as the theoretical model is expressed in the lattice Boltzmann framework.

3.4.1.1 Classical nucleation theory embedded in a cross scale LB model for precipitation

Mineral dissolution and precipitation is a multiscale process that covers several length and time scales. According to the classical nucleation theory, at the molecular level, when a supersaturation level is exceeded the ions start forming critical nuclei which serve as the bases of crystal formation. The energetic barrier that has to be overcome is different depending on the mechanism of precipitation. Heterogeneous precipitation (HET) proceeds on an existing template, and the formation of critical occurs at lower supersaturations, compared to the homogeneous nucleation (HON). Homogenous nucleation takes place in the bulk solution and requires no substrate, but higher level of supersaturation. In

Figure 9 (left) a schematic representation of this mechanism is plotted. The blue curve shows the HET nucleation path and the red curve the HON. In Prasianakis et al. (2017), the classical nucleation theory was implemented within the LBM framework. Once the pore structure is known (e.g. x-ray tomogram), the reactive surface areas for heterogeneous nucleation are resolved at the micrometer level. For the homogeneous nucleation, a sub-lattice model has been implemented. The formation of critical nuclei depends on saturation index. The number of molecules that form the critical nuclei and induction time are modelled by CNT, and the precipitation proceeds on spherical particles which grow until they fill the space. The surface area for precipitation is the sum of the surface of the spheres. A schematic of this process is shown in

Figure 9 (right). At the moment only 2D simulations have been conducted, and only for barite and celestite minerals whose precipitation follows the classical nucleation paths. While challenging, this model can be potentially applied for cases where minerals follow non-classical nucleation paths. The parametrization of HET and HON nucleation mechanisms takes into account also the contact angles of different phases and the surface tensions.

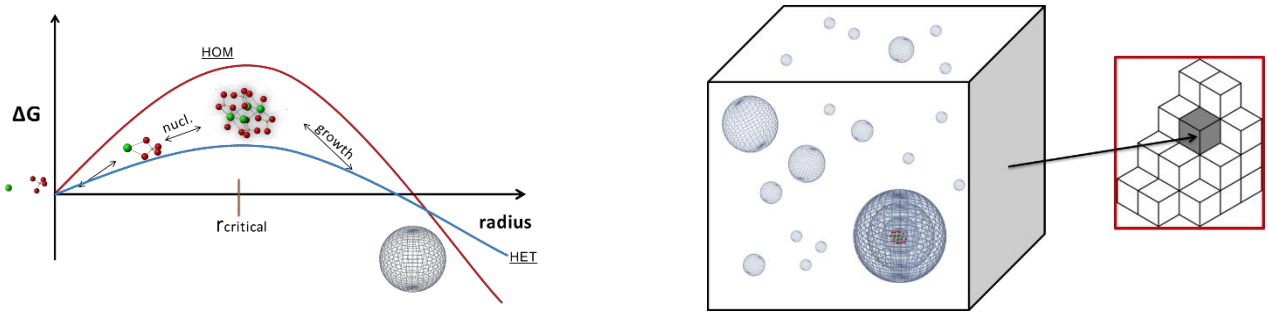

Figure 9: Cross scale precipitation modelling. (left) Classical nucleation Theory and energetic barriers to produce the critical nuclei for homogeneous (HON) and heterogeneous (HET) nucleation mechanisms. The two dimensional 9-velocity lattice (D2Q9), (right) Sub-lattice model for homogeneous nucleation which allows to model the evolution of reactive surface areas.

3.4.1.2 Electrochemical couplings at Lattice Boltzmann framework

The dominating pores for ion transport in tight porous media (such as high-performance concrete and compacted clays) are normally at several nanometers size. At this scale, the interface effect, especially the electrical double layer effect, has an important role on ion transport. For example, the relative effective diffusivities of different species in cement paste follow the order $Cl^- \approx HTO/O_2 > Na^+ > Cs^+$ (Yang et al., 2019). However, this is completely different from the order: $Cl^- < HTO/O_2 < Na^+$ obtained by the classical Nernst-Planck equation:

$$\frac{\partial C_i}{\partial t} + \nabla \cdot \boldsymbol{J}_i = 0\,, \tag{2}$$

$$\boldsymbol{J}_i = -D_{i,0}\nabla C_i - D_{i,0}\frac{z_i e C_i}{kT}\nabla \psi\,, \tag{3}$$

where C_i, $D_{i,0}$, $\boldsymbol{J}_i$, ψ and z_i denote the aqueous concentration (mol/m^3), the diffusion coefficient in free water (m^2/s), flux (mol/m^2/s), local electrical potential (V) and valence of the i^{th} species, respectively; t is time (s), e the absolute charge of the electron (C), *k* the Boltzmann constant (J/K) and *T* the absolute temperature (K). This classical model cannot accurately capture the ionic steric effect and the ion-ion correlation effect (Gillespie, 2014). Therefore in order to consider the steric effect and the ion-ion correlation effect, an excess chemical potential μ_i^{ex} term is usually added to the classical chemical potential as:

$$\boldsymbol{J}_i = -D_{i,0}\nabla C_i - D_{i,0}\frac{z_i e C_i}{kT}\nabla\psi - D_{i,0}\frac{C_i}{kT}\nabla\mu_i^{ex}. \tag{4}$$

This equation is the modified Nernst-Planck equation. However, the estimation of excess chemical potential in the Stern layer is still developing. The model was set up by Bikerman in 1942 to correct the ion concentrations in the Stern layer. After Bikerman's model, numerous continuous-scale theoretical models were developed to calculate the excess chemical potential in the Stern layer, but still each theory has its limitations (Lu and Zhou, 2011; Gillespie, 2014; Qiao et al., 2016). The exact ion distribution in the EDL at an arbitrarily charged surface or ionic strength of the electrolyte solution can be obtained by molecular-scale simulation. Therefore, the statistic information obtained by the molecular-scale simulation can be used to calculate the dimensionless excess chemical potential μ_i^{ex}/kT:

$$\frac{1}{kT}\mu_i^{ex}(x) = -\frac{ez_i}{kT}\psi^{GCMC}(x) - \ln\left(\frac{C_i^{GCMC}(x)}{C_{i,\infty}}\right), \tag{5}$$

where the equilibrium ion concentration distribution $C_i^{GCMC}(x)$ and the electrical potential distribution $\psi^{GCMC}(x)$ away from the solid surface are obtained by the grand canonical Monte Carlo scheme at atomic scale. This dimensionless excess chemical potential μ_i^{ex}/kT from the grand canonical Monte Carlo simulation is then applied to fitting with the nonlinear functions $F(x, C_{i,\infty})$ to get the relationship of μ_i^{ex}/kT with respect to the distance *x* and bulk concentration $C_{i,\infty}$. This upscaling strategy combines pore-scale modeling with atomic-scale modeling by adding the correction term for the excess chemical potential into the classical Nernst Planck equation to account for steric and ion-ion correlation effects. After this, the modified Nernst Planck equation is solved by the Lattice-Boltzmann method to simulate ion transport in tight porous media at pore scale and to calculate the effective diffusion coefficient of each species.

3.4.2 Multi-continua methods for hydro-chemical processes

Similar conceptual models of solute transport are covered by slightly variable terminology – double/dual porosity, double/dual continuum, two-region model. Contrary, the terms describing the configuration can be the same used as related to various physical processes. An exception is often such that dual-permeability and dual-porosity distinguish a relation to either the fluid flow or the solute transport.

Examples of early work are Coats and Smith (1964) with explanation of stagnant water in dead-end pores and, from another application field, Bedford and Stern (1972) developing theory for mechanics of composite material. The common idea for conceptual models is that a heterogeneous material is represented macroscopically by two or more interacting continua, each with separate degrees of freedom for a state physical quantity in a geometric point of space, understood in sense of representative elementary volume. Various terminologies are for denoting the meaning of continua, for solute transport it is typically the mobile and immobile zone.

In the context of upscaling, the multi-continuum models are used as homogenized counterpart of fractured rock, capturing the different behaviour of fracture volume and matrix pores, as introduced e.g. by Barenblatt and Zheltov (1960) and Warren and Root (1963).

One of the further direction of concept development was the "multiple interacting continua" (MINC) by Pruess and Narasimhan (1982). It assumes a nested sequence of continua which represent a virtual distance from the fracture into the matrix, i.e. one degree of freedom for the immobile zone is multiplied to represent the micro-scale heterogeneity more accurately. The models are in quite continuous interest in time and it is a live topic both for application of methods for specific case studies and for further developments and extensions of theory or calculation efficiency. The number of relevant publications in the last five year creates a significant share of the total number over the history (up to 30%).

The recent studies are oriented to various types of improvements. A lot of studies is related to the reservoir engineering especially gas/oil in shales, CO2 enhancing or CO2 storage, with effort to suit the methods and data for specific type of rocks.

Examples of generic topics (Figure 10):

- Procedures to obtain dual- or multi-continuum parameters from local
- Relevant pore-scale properties which are (controlling) for determination of macro-scale parameters
- Selection of number of continuum levels and their association to the material
- Extension of methods from standard flow and transport equations to mode complex processes (non-linear, coupled problems)
- Numerical efficiency improvement

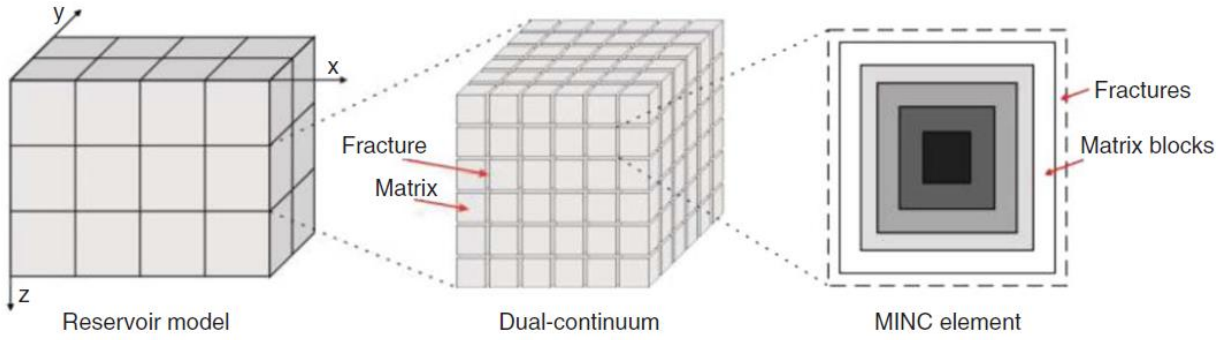

Figure 10: Concept of the multiple interacting continua (MINC) method ((Jiang et al., 2016) redesigned form of original (Pruess and Narasimhan, 1982)).

3.4.2.1 Phenomena capturing, conceptual improvement

Vogler et al. (2018) compares a hierarchy of upscaled dual porosity models and by comparison with tracer experiment they show that the dominant error arises from neglecting of local convection and that the properly upscaled model can explain the experiment without need to consider non-Fickian diffusion (see section 3.3.6). Wan et al. (2016) introduce a transient transfer function which is based on discrete fractures. Wang et al. (2020) derives a new scheme based on the Generalized Multiscale Finite Element Method (Efendiev et al., 2013) and establish a triple-continuum model for fractured-vuggy carbonate reservoirs. The method improves efficiency. This is accomplished by systematically generating an approximation space through solving a series of local snapshot and spectral problems. The resulting eigenfunctions can pass the local features to the global level when acting as basis functions in the global coarse problems.

3.4.2.2 Small scale models / upscaling parameters

Chen and Liu (2019) develop a method to measure the dual porosity parameters, in relation to hydrocarbon sources in shales. It is improved with respect to the previous work through different upstream and downstream. Steiner and Mittermeir (2017) evaluate the matrix-fracture transfer rate, from a numerical calculation on a fine scale single matrix block model. They conclude that a general formula it not yet established but the suggested type is perspective for further generalisation. Ceriotti et al. (2019) introduce a

technique to obtain two effective parameters of the upscaled dual porosity model through numerical solution of a simplified unit cell (

Figure 11). Porta et al. (2015) evaluate experiments in “synthetic” porous media (glass beads) and uses a pore-scale 3D image. They resolves the former limitation that relying solely on information about the geometrical attributes of the pore space obtained through NMR and electric resistivity measurements does not lead to a reliable characterization of the full set of model parameters.

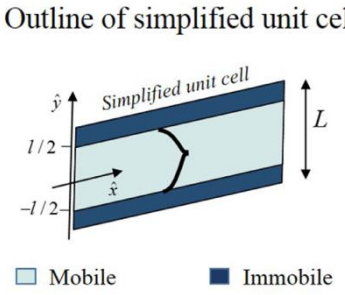

Figure 11: Example of small-scale cell used for calculation of macro-scale dual porosity parameters (Ceriotti et al., 2019).

3.4.2.3 Numerical methods

Ghahfarokhi (2017) shows that a structured mesh orientation has effect on the Oda’s method accuracy even when the known correction is used. Wang et al. (2017) applied the “proper orthogonal decomposition”, which is useful for model reduction and introduced a new version with mass conservation. Next, Wang et al. (2018) improves the solution algorithm to ensure the global mass conservation. Karimi-Fard and Durlofsky (2016) introduced a special technique of fine-grid discretization, upscaling, and coarse-grid discretization, which can be used with “arbitrary” simulator for accurate solution with little computing requirement. Chung et al. (2018) provide rigorous and accurate non-local solution (in the oversampled region).

3.4.3 Non Darcian and non-Fickian transport processes at the continuum scale

This subject has been introduced in section 2.1.4 already, here we explain some aspects related to sale transitions such as effective parameterization by averaging procedures and flux coupling schemes, respectively. Nanoporous materials such as clays and cement materials show a remarkable array of macro-scale properties such as very low permeability, partial or complete ion exclusion, and a strong coupling between geochemical, mechanical, and osmotic properties. These properties arise from the interactions of charged clay nano-pore surfaces with water and solutes, which lead to highly nonlinear coupling between flux terms. Consequently, predictive models of flow and ion transport in nano-porous charged media face severe scientific and computational challenges. Various Darcy scale models describing water and ion transport in clay media have been proposed in the literature that rely on the description of a representative elementary volume by volume averaging the local Nernst-Planck and Stokes equations. These types of models are necessary to conduct predictive large scale calculations aiming at coupling THMC processes. However, until now, Darcy scale approaches were mostly restricted to cases where (i) the solution composition was described by a symmetric electrolyte, and where (ii) the porosity could be assimilated to a homogeneous domain with average properties. In addition, these models are often restricted to the study of stationary states. These restrictions have many limitations. For example, the first restriction prevents the use of these models to predict the coupling of the transport properties of the dominant ions in the electrolyte and those of other ions present at trace concentrations, a case that is typically encountered in the simulation of radionuclides diffusion. This modeling shortcoming can be avoided by using state-of-the-art reactive transport codes (PHREEQC and CrunchClay) that account for the presence of a charged diffuse layer whose volume can be dominant over the bulk water volume in the porosity. Reactive transport code developments have been limited however to the case of purely diffusive systems. The modeling of transient states involving the advective movement of water and gas in clay media necessitates to extend these code capabilities to include

advective processes and related off-diagonal coupling terms. The current state of the art of this development is available in a recent review (Tournassat and Steefel, 2019).

3.4.4 Coarse graining

Coarse-grained models, aim at simulating the behavior of complex systems using a simplified (coarse-grained) representation. Coarse-grained approaches are widely used in atomistic / molecular modeling. Simplification is essential, as the large numbers of atoms / molecules cannot be represented. Coarse-grained models are also often used for multiscale modeling. Coarse-grained approaches are attempting to bridge neighbor scales in a consistent continuum-mechanical way.

We provide a few recent examples to elucidate the status-quo and potential of the multiscale methodology (Tahmasebi and Kamrava, 2018). Recently, Churakov and Prasianakis (2018) presented a review on a holistic process-based approach for reactive mass transport with an emphasis on mineral reactions in porous media. The paper reports the current status quo of mineral-fluid interface chemistry at molecular level and develops a road map for the general challenge: How to translate atomistic phenomena translate into macroscopic levels for practical model applications?

Gurikov et al. (2016)(Gurikov et al., 2016) present a cellular automaton model for large-scale simulations at molecular level. They attempt to describe mass transport processes in porous media, i.e. diffusion, adsorption and directed flow in porous media, with large representative elementary volumes (REV). The framework could be successfully used to model diffusion processes in disordered porous structures with phase separation phenomena. Essential features of the molecular processes could be represented for physically meaningful time scales.

Recently, Schmidt and Steeb (2019) introduced a multiscale approach for hydro-mechanical coupling of fluid-filled fractures using a hybrid-dimensional element formulations with non-conformal meshes. In their hybrid approach they use parabolic velocity profile with the fracture to capture the fluid mechanics behavior at small scale and avoiding an explicit discretization. The fracture dimension is reduced by one order and implemented as interface elements to the modeling domain. The mass and momentum balances are described by modified balance equations, respectively. Different coupling schemes (staggered vs. monolithic) have been implemented and tested for testing the accuracy of the hybrid scheme. A number of test cases have been used for validation of the newly developed approach. Alyaev et al. (2019) using a similar approach for simulating single-phase flows in porous media with inertial effects based on a control volume heterogeneous multiscale method.

Coarse-grained methods are also used in related areas such as sediment transport, when modeling flow over mobile porous media. Biegert et al. (2018) couple particle and fluid phase flow with a coarse-grained approach. They conducted a grain-resolved direct numerical simulation of multiphase flow for the coupled particle-fluid systems. The particle phase is introduced by a boundary method where particle interactions be can taken into account via a discrete element method. The governing equations for fluid flow and stress budgets are derived by double averaging procedures.

3.4.5 Porosity permeability alterations in reacting environments and upscaling approaches

Precipitation and dissolution reactions in porous media dominate and control a large number of geochemical processes and industrial applications. The precipitation and dissolution of minerals from aqueous solutions alters the pore space and its connectivity. This has a strong effect on the mass convection and diffusion through the porous medium. For example, when a mineral precipitates/dissolves on the reactive porous surface, the overall porosity decreases/increases leading to a subsequent decrease/increase in permeability and effective diffusivity. At the same time the connectivity of the pores can also change in a way to block or to facilitate the mass diffusion processes.

It is common that the field scale simulations (continuum scale) use a simplified description of the processes that actually take place at the pore-level scale (microscale). For example, the change in permeability and diffusivity of a porous medium due to reactions is correlated to a global change of the bulk porosity in the

medium, using empirical laws. It has been demonstrated, that smooth simplified relations are frequently inadequate to predict accurately the temporal evolution of the system of interest (Steefel et al., 2015).

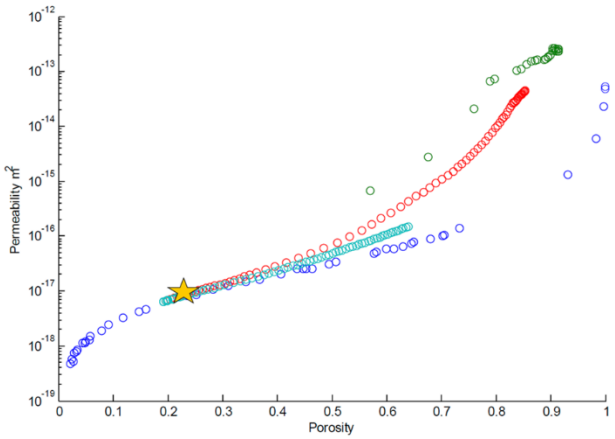

Figure 12: Evolution of permeability for different reactive transport dissolution scenario in a porous medium. Porosity permeability correlations can be extracted via fitting for use with macroscopic codes. Blue points represent the random set of geometries permeabilities that can serve as starting points (star). A geometry with 20% porosity is selected. Temporal evolution of the structure for three different cases were Peclet and Damkohler number vary.

The predictability of the models can be greatly improved after resolving experimentally and numerically the physical phenomena and process mechanisms occurring at the microscale, followed by an upscaling procedure to the continuum scale (Noiriel and Daval, 2017; Deng, Molins, et al., 2018; Prasianakis et al., 2018; Molins and Knabner, 2019). For example, depending on the conditions, a calcite rock can be dissolved following a face dissolution mechanism or via wormhole mechanism (Hoefner and Fogler, 1988; Portier et al., 2009; Menke et al., 2016). The resulting porosity-permeability correlation for the same rock can be greatly different (Prasianakis et al., 2018). The flow conditions can be described by the Peclet and Damkohler numbers. In Figure 12 such an example is shown, where the LBM framework has been used. Different flow conditions on the same rock have been implemented, which resulted in distinct permeability evolution paths.

At the moment, it is computationally very challenging to extract such correlations, since three dimensional reactive transport calculations are needed.

Upscaling of mineral precipitation and dissolution processes in porous media affecting the pore structure and subsequently the flow properties in the continuum scale is a challenging task to be considered in mathematical models. To model such processes, one needs conservation laws for mass, momentum and solute in time-dependent domains where the evolution of the sharp interface separating two domains (mobile and immobile) is not known a priori. Therefore, a scheme accounting for moving/free boundary conditions, where the boundaries and hence the domains evolve, needs to be considered.

In modelling such coupled processes, two spatial flow scales are normally considered: pore (Stokes/Reynolds) and macro (Darcy) scale (Noorden, 2009) Precipitation/dissolution reactions are considered using reactions rates where multiple chemical components could play a role. One of the most promising numerical schemes of recent is to use the phase-field model to represent moving sharp interfaces (Xu and Meakin, 2011) and it has been extended to porous media (Bringedal and Pop, 2019).

4. Tools and methods to quantify/derive uncertainties induced by coupled processes

4.1 Understanding of uncertainties in disposal systems and quantification of uncertainties

4.1.1 Introduction

The subject of couplings between the thermal, hydrological and mechanical processes in rocks has become an important subject mainly due to the modeling requirements for the design and performance assessment of radioactive waste repositories and other engineering fields such as gas/oil projects, geothermal energy exploitation, and environment impact evaluation in general. Coupled THM (thermo-hydro-mechanical) and coupled THMC (thermo-hydro-mechanical and (geo)chemical) processes are the ones most often required in the numerical models (Hudson JA., 2005; Hudson, 2017).

The requirement of coupling the processes in a geological system can be derived from processes that affect each other, which occurs mainly in two ways. The first is direct coupling in which one process induces directly the development of another process, represented by a cross term involving both processes in the governing equations. The other way is indirect coupling in which one process changes the parameters controlling another process.

A key process in the early stage of repository construction is the creation of an excavation damaged zone (EDZ) resulting in increased permeability around the tunnels. Performance assessment requires the knowledge of spatial-temporal evolution of EDZ properties. During the Exploitation Stage, decay heat from radioactive waste may induce large temperature gradients in the buffer zone and in the near-field rock. At the same time, resaturation in the near-field occurs which is a slow process controlled by low rock permeability and buffer characteristics. Heat generation may also lead to water vapor convection near the heat sources. This process induces transient pore pressure build-up resulting from differential thermal expansion. The pressure build-up affects effective stresses in the engineered and natural barrier system. Heating may impact rock strength and may induce chemical reactions with impact on rock properties as well. The Long-Term Post-Closure Stage is the subject of long-term performance and safety assessment. The key process is the likely failure of the engineered barriers that may lead to the release and subsequent transport of radionuclides and any nonradioactive contaminants. The transport characteristics in the tunnel backfill and near-field rock during this period depend on the coupled processes and effects that occurred during the previous stages.

Main sources of uncertainties related to radwaste disposal and their assessment that numerical modelers must consider:

- The extremely long (geological) timescale over which system evolution must be evaluated and predicted. Thus, the long-term evolution of safety functions of the repository cannot be well supported by comparably short-term laboratory, in-situ and/or URL investigations.
- The inherent variability of geological features makes the characterization of host rock formations challenging at the required level of detail based on small-scale experiments and laboratory tests.
- One must consider the coupled behavior of thermo-hydraulic-mechanical as well as (geo)chemical processes to be investigated with respect to spatial scale and timeframe of investigation.
- The perturbation effect of relatively rapid (geological) changes, e.g. climate change, earthquake as well as heat generation on the long-term evolution of the repository system. From a numerical modelling point of view, this requires either hybrid modelling approaches or the availability to dynamically change the time-stepping algorithms of the models.
- The applicability of the models used for assessing the system evolution. Thus, the risk related to uncertainty regarding the conceptual model used to describe interconnected physical processes must be evaluated.

In this state-of-the-art report, we provide a brief overview of coupled processes and the associated uncertainties with respect to underground nuclear waste repositories. First, we summarize the relevant THM

and THMC processes and their general method of resolution. This is followed by characterizing the uncertainties of these phenomena. The report ends with conclusions regarding the status of handling these uncertainties.

4.1.2 Description of the coupled processes in the radioactive disposal system

4.1.2.1 Thermal-hydro-mechanically and chemically coupled processes and their resolution

One of the main difficulties in the prediction of the spatial-temporal evolution of a radioactive waste disposal is to consider all relevant processes and their couplings (Millard, 2017). These include mechanical, hydraulic, thermal as well as geochemical processes which may be extended to gaseous and biological phenomena. The THMC-processes and their connections are illustrated in Figure 13.

The basis of mechanical processes is that rock mechanical properties are changed due to repository excavation; and the rock mass may contain water. Thus, under loading, the stresses developing in the rock medium are linked to both solid and fluid components, leading to the concept of effective stresses.

The hydraulic part depends on rock saturation. In the simplified case of a saturated rock, water flow is not only controlled by pore pressure gradient (Darcy flow), but also by pore volume change induced by rock deformation. Water expansion due to temperature change may be considered as well.

The evolution of temperature is governed by heat release from the waste. This is described by heat diffusion due to developing temperature gradients and advection associated with fluid flow. Chemical processes are dominated by dissolution and precipitation enhanced by thermal gradients.

Changes in water pressure and chemical reactions affect water and gas flow paths and contaminant transport characteristics.

Gaseous processes include gas generation, pressure build-up as well as gas lift. Furthermore, extensive pressure build-up may lead to the failure of some parts of the Engineered Barrier System (EBS) further enhancing the release of radionuclides from the repository. Gas (mainly methane) could be generated as a result of biodegradation of organic material, hydrogen gas generation is foreseeable from the corrosion of metallic components and some gas could be generated from hydrolysis.

Consequently, the assessment of radioactive waste disposal evolution requires the knowledge of physical variables controlling thermo-hydro-mechanical-chemical processes at any time. Thus, the resolution of a coupled system of partial differential equations is required. There are two types of approaches to resolve such problems (Ismail-Zadeh A, 2010).

In the first type, all variables are solved simultaneously. In simultaneous solution methods, all the equations are considered as part of a single system. However, direct solution of these equations is time consuming, especially when these equations are non-linear, and the problem is three-dimensional, which is the case generally for radioactive waste disposal systems. When the equations are linear and fully coupled, the simultaneous approach is the best.

However, the second method, the iterative approach may be computationally much more efficient. The equations can be so complex and non-linear that coupled methods are too challenging to apply. It may then be preferable to treat each equation as if it has only a single unknown explicitly, temporarily treating the other variables with the currently available values for them. The equations are then solved in turns, repeating the cycle until all equations are satisfied. Since some terms, e.g. the coefficients and source terms that depend on other variables change as the simulation proceeds, iterative solvers are much more efficient and so these are preferred. Iterations performed on each equation are called inner iterations (e.g. explicit integration scheme for solving the equation of motion). In order to obtain a solution that satisfies all the equations, the coefficient matrices and source vector must be updated after each cycle and this process is then repeated. These cycles are called outer iterations.

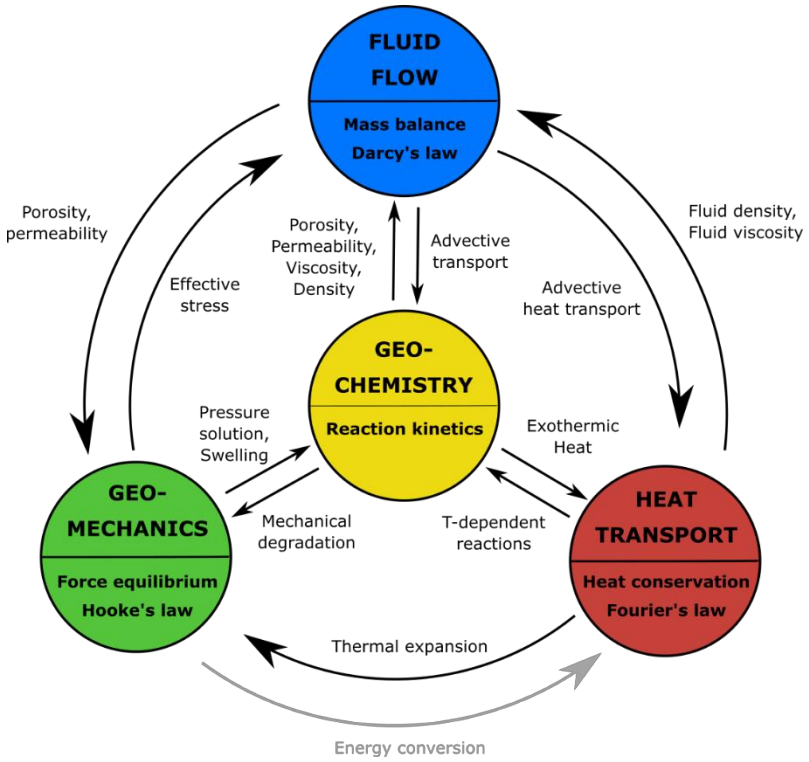

Figure 13 – Schematic illustration of the coupled thermal-hydro-mechanical-chemical processes relevant to safety assessment of nuclear waste repositories (modified from Gaucher et al. (2015))

4.1.2.2 Sources of uncertainty in coupled processes associated with radioactive waste repositories

For the performance assessment of a radioactive waste repository, analyses of the coupled THM(C) phenomena must be considered. However, many of these processes are non-linear and the constitutive relationships between them are not well known. Thus, various constitutive relationships are used that are applicable for different geological settings. What coupled THM or THMC processes should be included in a simulation model depend on site-specific safety criteria and scenarios. Therefore, one should characterize uncertainty arising from conceptual model approaches.

Birkholzer (2019) point out that the various sources of uncertainty associated with numerical modelling of evolution of radioactive waste repositories lead to the realization of validating numerical modelling results against laboratory and field data. For this purpose, several laboratory experiments have been carried out using core samples of different sizes ranging at cm and m scale. Field measurements are also conducted at experimental sites, either at locations where repositories are proposed to be situated or at areas with representative geologic conditions. Several subsurface facilities, referred to as underground research laboratories (URL) are designed to conduct such tests. The goal of these tests is to study coupled processes and check modeling results often at the spatial scale envisioned for the waste emplacement. Numerical simulations with different modeling approaches are then performed to test the capability of the model against measured data. The goal of these simulations is to improve modelling approaches, to increase model confidence as well as to estimate uncertainty of various parameters governing coupled processes. However, it must be pointed out that geological settings and site characteristics cannot be known completely due to limitations associated with site observations and field tests.

Another source of uncertainty lies in different key processes governing the evolution of the disposal system at different stages of the repository and spatial scales (**Erreur ! Source du renvoi introuvable.**). Birkholzer (2019) and Tsang (2012) summarize key processes and issues for different phases of repository lifetime, such as early repository implementation phases (Construction and Open Drift Stage), the period of thermal disturbance after waste emplacement (Exploitation Stage) and a long-term period where much of the early

perturbations are no longer valid and the system is approaching a late time equilibrium (Long-Term Post-Closure Stage).

4.1.3 Uncertainties of coupled THCM models for the EBS & needs for further research

THCM models of the EBS have conceptual model uncertainties, regarding the identification of the relevant processes, parameter uncertainties and uncertainties in the initial and boundary conditions. Usually, these uncertainties are analysed by means of sensitivity analyses. The relevance of physical and chemical processes are analysed by performing simulation runs with and without the consideration of a given physico-chemical process. Similarly, the uncertainty in THMC parameters is commonly ascertained by performing systematic sets of sensitivity runs in which the values of the parameters are increased and decreased one-at-a-time.

Amongst others example of the remaining uncertainties of the coupled THMC models of the EBS that that need further research include:

1. Multiphase flow:
    a. The complex porosity structure of the bentonite which requires the use of dual and triple continuum models (Marcelo Sanchez and Olivella, 2005, 2012, 2016)
    b. The retention curve of the bentonite: hysteresis and temperature dependence
2. Reactive solute transport
    a. Relevance of reactive gases such as CO2(g) and O2(g)
    b. Solute diffusion from the EBS into the host rock
    c. Alternative conceptual models of solute diffusion in the EBS:
        i. The single interlayer-porosity Donnan equilibrium model (Birgersson, 2017)
        ii. The electrochemical model based on the Nernst-Planck equation (Idiart and Pkekala, 2016)
        iii. The multiple porosity (interlayer, double layer and free) model (Samper et al., 2008; Zheng and Samper, 2015).
        iv. The use of different effective diffusion coefficients for each chemical species while preserving charge balance
3. Geochemistry
    a. Precipitation of the secondary mineral phases at the canister-bentonite interphase such as Fe-phyllosilicates (chlorite, cronstedtite and berthierine); Fe-rich smectites and zeolites such as phillipsite, chabazite and merlionite;
    b. Precipitation of secondary mineral phases at the concrete-bentonite interphase such as CSH, CASH, zeolites such as phillipsite, chabazite and merlionite, and iron silicates
    c. Fe reduction in the bentonite
    d. The uncertainties in the kinetic rate laws, rate constants, reactive surfaces and catalytic effects of kinetic mineral dissolution/precipitation. The dynamic update of the reactive surface areas of the minerals;
4. Coupled effects
    a. Changes in porosity and other transport and chemical parameters caused by mineral precipitation/dissolution reactions
    b. Simulating canister corrosion in a more realistic manner by adopting a dynamic corrosion front;
    c. The coupling of the evolution of the bentonite microstructure with the changes in the composition of the exchange complex

To date Global sensitivity and uncertainty quantification methods have been widely used in performance assessment studies to quantify the prediction uncertainties caused by uncertainties in flow and transport parameters (Bonano and Cranwell, 1988; Helton et al., 2012). A few studies have analyzed the uncertainties in using reactive transport models. Matsumoto and Neyama (2008) developed a preliminary uncertainty analysis tool for performance assessment calculations considering uncertainties in the groundwater concentrations of Na, Ca, HCO3 and Cl. Criscenti et al. (1996) and Denison and Garnier-Laplace (2005) applied Monte Carlo methods for geochemical calculations. Their focus were: (a) the significance of uncertainties in predicted pH values for a simple buffer solution and for calcite saturation indices, and (b) the

effects of database parameter uncertainty on uranium(IV) equilibrium calculations (i.e. uranium(IV) speciation), respectively. Cerotti et al. (2017) presented a model to estimate the uncertainty associated with the amounts of CO2 generated by carbonate-clays reactions in subsurface systems. Recently, Ayoub et al. (2020) presented a comprehensive uncertainty and sensitivity analysis of Cs sorption reactive transport modeling with 3 cation exchange sites in the near field of a deep geological repository for nuclear waste based on the Morris and Sobol method. Ayoub et al. (2020) analyzed the uncertainties in: 1) Cation exchange selectivities and 2) Clay pore water cation concentrations. They were able to identify the most important uncertain parameters affecting the transport of Cesium and the combination of parameters values leading to the maximum Cesium concentrations at a specified location using classification trees. Finally, they proposed a Cesium isotherm and a Kd uncertainty range based on a large number of numerical simulations.

4.1.4 Approximation of the probability density function

The classical Monte Carlo is an attractive method for uncertainty propagation due to its noninvasive nature. However this flexibility is paid by the slow convergency and resulting computational costs.

The Multilevel Monte Carlo (MLMC) method introduced by Giles (2008) provides an efficient way to estimate the expectation of the variable that is result of an approximation. Basic idea is to do high volume of samples using a less accurate but cheap approximation and much fewer samples of the difference between the observable and its approximation. If the difference have much smaller variance we obtain the estimate of the same accuracy but at the fraction of costs. The MLMC method has been already studied and analyzed in connection with variety of PDE problems: elliptic equations (Barth et al., 2011; Cliffe et al., 2011; Abdulle et al., 2013), parabolic equations (Barth et al., 2013), multiphase flow (Muller et al., 2013; Lu et al., 2016).

However, application of MLMC to the continuum-fracture models, e.g. Flow123d (see paragraph 2.3), has not yet been explored.

While MLMC can be used to efficiently estimate generalized moments of a variable X, the maximum entropy method provides reconstruction of PDF. The theoretical basis of the method was founded in Barron and Sheu (1991). Recently these results were applied in the context of MLMC, see Bierig and Chernov (2016). Yet few problems prevents practical application of this method. First, it leads to an ill conditioned nonlinear system that is difficult to solve. Second, for moment estimates of a lower precision the resulting PDF approximation is highly oscillating.

A methodology for addressing uncertainties in a broader context as required in PA will be confronted with (as outlined above) fragmentary information, with heterogeneous spatial (and sometimes also temporal) geochemical composition (minerals, solution, hydrodynamic characteristics), and even with occasional scientific dissent about governing processes and other boundary conditions. This applies (with increasing magnitude) for the parameterization of thermodynamics of solids (and solid solutions), ion-ion interactions, and properties of accessible / reactive surfaces. Strategies to overcome such problems are very diverse and depend to a high degree on the efforts necessary to perform additional lab experiments or field sampling / characterization. Ideally it builds on previous knowledge of phenomenology and processes in the respective geochemical systems. The range of strategies given below is sorted (approximately) from very “cheap” to “expensive” - in the sense of fiscal, temporal and personal resources requested. Starting at the 2nd alternative, a variety of pdfs is usually considered. For (namely aqueous) systems with sufficient independent parameter determinations Gaussian (or log Gaussian) distributions or as .more robust alternatives trigonal or uniform pdfs are used, the latter often being the only sensible approach when dealing with bimodal parameter distributions. Moreover, in the not so exotic case of numerical data themselves only being estimations (or even expert guesses), only the first three tiers are applicable at all.

1. “Do nothing” option: ignore the respective uncertainty as the underlying process may hopefully not be essential (should of course be verified) and the harm of ignoring may be smaller than using some estimates not really defensible.
2. “Expert guess” which can be anything between pure assumptions and the application of well-established informal rules based on general geochemical principles.
3. “Estimates” stemming from pure correlations in the sense of empirical fits, over chemical analogs (an often exploited analogy covers Ln(III) vs. An(III)), the usage of structure-based incremental

systems, linear free energy relationships, or quantum chemical approximations (to name just the most prominent examples).

4. "Re-computation" of parameters from other (hopefully similar) models (e.g. going from the Pitzer-approach to SIT parameters and vice versa – this direction being the more challenging one).
5. "Re-fitting" of a pool of already existing raw data with uncertainties as by-product.
6. Setting up an experimental program to derive the missing parameters in the lab.

4.1.5 Examination of uncertainties in the geochemistry

In the following the international status with respect to the assessment of uncertainties within the field of geochemistry is highlighted, sketching also some possible development lines. The first three sub-chapters describe the types of uncertainty usually encountered in conventional geochemistry. Thereafter, consequences with respect to PA applications are outlined. Here it must also be mentioned that within EURAD, the strategic study "Uncertainty Management multi-Actor Network (UMAN)" is addressing these issues from a more general point, too.

4.1.5.1 Aqueous speciation

Uncertainties for thermodynamic data are available in general, but often only describing the reproducibility of experimental data, ignoring systematic errors that are of course more difficult to quantify. Moreover, the formulations of chemical reactions, i.e. which educts react to form which products, are sometime mere postulations. It may even turn out, that a chemical system can be described by two mutually incompatible sets of species that represent available experimental findings equally well. Thus we have an overlap of numerical and conceptual uncertainty. Another problem arises from the fact that in natural systems some reactions are strongly kinetically hindered (especially if reactions involve re-structuring of molecules / ions). This is regularly observed in redox systems, e.g. involving Pu, Se, U or Tc species. Finally, the uncertainty related to thermodynamic data at temperatures higher than 25 °C is also of major concern. Here, very few data (derived from calorimetric studies or speciation experiments as f(T)) are available at all, and the temperature dependence itself is often described by purely empirical T-functions.

Concerning the parameters required in activity models to describe ion-ion-interactions in highly saline solutions, the situation differs for the two major approaches. Whereas in the SIT (Specific Ion Interaction Theory – Ciavatta (1980)) the most comprehensive compilation(s) contain also uncertainties, the situation in the Pitzer model (Pitzer, 1991) is more complicated. Due to the high connectivity / interdependence of the various types of binary and ternary Pitzer coefficients usually no uncertainties are given at all, i.e. so far no commonly agreed-upon procedure is established to yield sensible uncertainties for them. This may be of concern in highly saline environments such as salt rock or certain clay formations, whereas in all other host rock scenarios the SIT approach is the best choice.

4.1.5.2 Solids (minerals)

The situation here is rather similar to the one described for aqueous species. A further source of uncertainty is the exact stoichiometry of the solid and their physical properties such as specific surface area (SSA). This is connected to the problem that dissolution experiments to yield solubility constants usually start from well-crystalline, aged phases. In nature, however, many of these phases do either not form at all under hydrothermal conditions or undergo a transition from amorphous intermediates (with much higher solubilities) to eventually well-crystalline ones. Thus it is helpful to distinguish between those processes that involve the dissolution of "matured" well-crystalline phases and the precipitation (and dissolution) of "fresh" amorphous phases. As for a variety of phases solubilities for both variants have been determined, these values scan be used for correlations to close respective data gaps.

With respect to T-functions the picture is better than for the aqueous solutions (and much better than for surface species) as many enthalpic and entropic data are available for solid phases.

When it comes to minerals also spatial heterogeneities (including accessible surfaces) must be discussed. Whereas nearly all uncertainties relevant for geochemical processes are epistemic ones, a few – and namely those associated to mineralogical heterogeneity – are aleatoric. Other uncertainties where additional experiments may not reduce uncertainties are pore size distributions, accessibility of pore volumes or reactive surface areas.

4.1.5.3 Surface speciation

Here, the publishing of uncertainties is not so well established. If so, the same restrictions apply as already outlined above for aqueous and solid species. Surface reaction suffer from a further limitation: there is a variety of mutual competing models available describing sorption via surface complexation models and ion exchange, such as Non-Electrostatic, Diffuse Double Layer, Constant Capacitance, Triple Layer or Basic Stern models (Stumm, 1992). The respective parameters are usually not directly convertible, thus the parameter pool to derive probability density function (pdfs) is diminished. Another drawback is the relation to mineral characteristics (such as SSA or surface binding site density) being themselves not well established, e.g. surface protonation or again specific surface area. An overview about the data situation can be found in Ochs (2012) as well as in the mineral-specific sorption database RES³T (V. Brendler, 2003), also accessible under www.hzdr.de/res3t. Data scatter evidenced as multi-modal distribution patterns is often related to alternative experimental methods. A classical case is the determination of SSA of clay minerals by either BET measurements or via the EGME method.

4.1.5.4 Established uncertainty assessment methodologies

Despite the above listed difficulties encountered in geochemistry there is at least a well-established procedure available how to treat parameter uncertainties. Annex C “Assigned uncertainties” being part of all volumes issued so far of the Thermochemical Database of the Nuclear Energy Agency (NEA TDB), with the most recent one treating iron (Lemire et al., 2013, 2020) contains very comprehensive procedures how to compute uncertainties when combining / averaging several independent sources of parameter values. Last but not least the derivation of uncertainties when re-formulating chemical reaction equations (to accommodate e.g. switches in the master species set required in different speciation codes) has to follow the common procedures of error propagation. The uncertainties given for the thermodynamic values recommended by the NEA TDB are provided in a consistent and traceable way thus setting international standards. The same holds for the values provided by the most recent PSI/Nagra TDB (Thoenen, 2014) which to a large degree actually relies on the NEA TDB but follows a different approach with respect to SIT coefficients. Other TDBs regularly utilized in the context of nuclear waste disposal such as ThermoChimie (Giffaut, 2014), the Yucca Mountain Project TDB (G.E. Gdowski and Rosenberg, 2002) or the HKL TDB (Helgeson, 1981) do either follow the general lines set by the NEA TDB, or do not report any uncertainties at all.

4.1.5.5 Predictive modelling

A further drawback is to mention here: for the time being no geochemical speciation or reactive transport codeis exploiting available information about uncertainties. To do so, separate routines must be written (or re-used from codes such as UCODE (Poeter, 2014) or PEST (Doherty, 2010), “enclosing” the proper code and adding such functionalities (Stockmann, 2017). The transfer of geochemically induced uncertainties to PA is usually done by lookup tables (e.g. for solubilities or distribution coefficients). More recent developments try to tackle the problem of multidimensionality also by exploiting uncertainty and sensitivity analysis, the introduction of surrogate functions, neural networks or other machine learning approaches, see chapters 4.2 and 4.3 for more details.

A special problem is treatment of strongly dependent parameters, i.e. the correlation matrices obtained from multiple parameter fits are usually not considered in geochemistry. This may even lead to the situation that uncertainties are agglomerated (Meinrath, 1997) eventually yielding significantly exaggerated combined uncertainties.

4.1.6 Derivation of procedures for pdf of corresponding parameters for chemical models

Wide set of data for radionuclide transport processes is available for central European crystalline massive conditions. Specific conditions of transport processes specific for Bohemia crystalline rock massive conditions were described in Havlová (2013), defining that Central European crystalline rock massifs are specific by groundwaters of lower salinity and rock structures and textures less distorted by orogenic long-term processes in comparison with Swedish and Finish crystalline rock or with Alpine granites as in Grimsel Test Site. Studies of radionuclide transport properties on crystalline rock has been performed in Czech Republic since late 90's (Passas and Bunatova, 1996; Buňatová, 1998; Havlová, 1999) being focused namely on radionuclide sorption (Cs^{I}, Sr^{II}, Np^{III}, I^{-I}) on generic granites under synthetic groundwater conditions. Those studies, mostly using batch sorption experiments on crushed materials, were complemented by studies focused on aerobic/anaerobic behaviour of redox sensitive species (Se, Tc), using more comprehensive methods, like surface scanning and spectroscopic studies for element (Se) specific sorption at the rock surface (Videnská, 2012, 2014a, 2014b).

Migration of radionuclides in dynamic system is described with the transport parameters such as the retardation and distribution coefficients (R and K_d), Peclet number (Pe) or hydrodynamic dispersion coefficient (D). 2D- or 3D-models deal with longitudinal hydrodyna-mic dispersion coefficient. These parameters can be determined using dynamic column technique, which has many advantages in comparison with static batch experiments. Namely, it enables: (i) direct determination of contaminant retardation coefficients dissolved in aqueous phase, (ii) closer approximation of the real conditions in the natural system, (iii) study of less disturbed samples, (iv) study of desorption process following immediately after sorption, and so on. Retardation of both non-sorbing species (HTO, Cl^{-I}, $Se^{IV/VI}$) and sorbing species (Sr^{II}) in dynamic granitic column system has been presented (Palágyi and Vodičková, 2013; Videnská, 2013, 2015; Štamberg and Havlová, 2014).

Systematic approach toward potential site selection brought into the focus some of the potential DGR sites since 2015. The most comprehensive approach to the rock condition characterisation under conditions close to the DGR emplacement in the depth of 500m brought Bukov URL survey, including a wide set of sorption and diffusion experiments for several radionuclides (Cs^{I}, Sr^{II}, Se^{VI}, U^{VI}, HTO, Cl^{-I}, I^{-I}) (Bukovská, 2017). The above mentioned study has been complemented with more generic diffusion studies, being performed either in generic Czech massive granite samples (Havlová V., 2012) or on URL samples from Grimsel Test Site (Soler J.M., 2015).

Direct tool for potential assessment of fracture flow quantification and sensitivity analyse is available in ÚJV laboratory. The physical model of granite block (dimensions: 80×50×40cm) was assembled and the generated artificial fracture was characterised by means of 3D laser scanner. The goal of the tracer tests, using different types of tracers (Cl^{-I}, I^{-I}, HTO, Pb^{II}) has been to develop tools for evaluation of migration and retention of potential contaminants in the rock environment as the key input values for the safety assessment of anthropogenic activities and also to enable test mathematical codes and assess the uncertainty of reaction transport processes determination. Subsequently the block was instrumented with on-line measuring devices (conductivity, pressure), allowing performance of migration experiments. Detailed description of the fracture parameters (*Figure 14*) serves as a key input for modelling purposes (Jankovec J., 2019; Jankovský F., 2019a) using both conventional software (MODFLOW, FEFLOW) and in-house code (Flow123d). However, natural fracture flow represents more complex environment, including surface coatings and miscellaneous topographic domains within the surface (Jankovský F., 2019b). Here WP FUTURE will play a bridging role with WP DONUT, producing data from flow-through experiments and batch sorption experiments with fracture infill.

However, laboratory experiments are not able to reflect the real conditions in the rock massive all the time. Therefore, the tracer tests both under static or dynamic conditions in-situ are to look into upscaling the transport processes. Such an experiment can be focused both on studying radionuclide diffusion into the rock matrix (Soler J.M., 2015) or on advective flow experiments in fracture, including modelling in MODFLOW/MT3D code (Zuna and Souček, 2017).

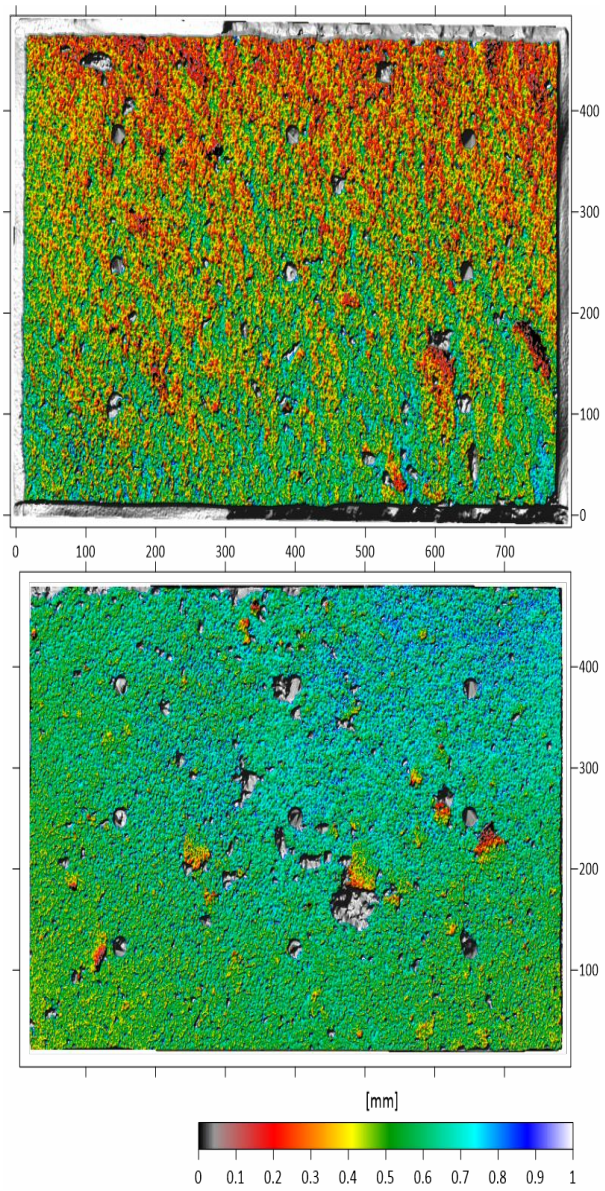

Figure 14 – Colored fracture aperture distribution at artificial granite block fracture (Jankovec J., 2019)

4.2 Methods to evaluate uncertainties

4.2.1 Adjoint state methods

Adjoint state (AS) methods have been widely used in optimization and inverse problems in: 1) Groundwater flow models (Townley and Wilson, 1985; Sun, 2013) and conservative solute transport models (Samper, 1986; Ackerer et al., 2014). The adjoint state variables play the role of Lagrange multipliers. The advantages of the AS method include: 1) The calculation of the adjoint state requires a single calculation of the forward problem, regardless of the number or parameters; 2) The method is well suited for computing derivatives or local sensitivities for a large number of parameters; and 3) The AS equations can be derived for the continuous formulation or for the discretized numerical equations. Delay et al. (2017) presented a comparison of discrete and continuous adjoint states for groundwater flow in heterogeneous dual porosity systems. They concluded that both discrete and continuous adjoint states provide similar solutions. They claimed that the continuous adjoint state is the most versatile form as its non-intrusive character allows for plugging an inversion toolbox quasi-independent from the code employed for solving the forward problem.

There are no reported applications of the AS method for reactive transport modelling and its application within the framework of geological HLW disposal. There is the need to derive both the continuous and the discrete AS equations for multicomponent reactive transport, study the properties of the AS equations and test the method with several test cases.

4.2.2 Statistical metamodeling

4.2.2.1 State of the art compilation of efforts regarding emulation with machine learning techniques for reactive transport modelling.

Although the resulting benefits are potentially huge in terms of computational savings, emulation of reactive transport models has been scarcely addressed so far. Jatnieks (2016) presented a short comparison of a range of nonlinear regression methods for the emulation of the geochemical component of a relatively simple RTM. Leal (2017) demonstrated for a simple reactive transport problem how the geochemical calculations can be efficiently replaced by an online first-order Taylor approximation around the closest previously simulated point by the original geochemical solver. Huang (2018) proposed to use a lookup table to also replace the geochemical solver of an RTM for a 1D reactive transport problem with two parameters. The studies by Huang (2018), Jatnieks (2016) and Leal (2017) are thus concerned with replacing the geochemical component of an RTM by a computationally cheap approximation while assuming the geochemical model parameters to be fixed. Since geochemical calculations commonly take most of the running time of an RTM, this can be very useful when the goal is to perform one or more otherwise CPU-intensive simulation(s) with fixed model parameters. Nevertheless, this strategy is not suited to tasks involving varying the model parameter values such as sensitivity analysis (SA), uncertainty quantification (UQ), and model calibration (inversion). To address these situations, Laloy (2019)(Laloy, 2019) compared different machine learning methods, namely polynomial chaos expansion (PCE), Gaussian processes (GP) and deep neural networks (DNN), for the emulation of the full RTM response to (some) model parameter and boundary condition changes. Here the considered problem was the 1D reactive transport of Uranium VI through a soil column with 8 to 13 uncertain model parameters. The strategy of emulating the full RTM response permits emulation-based SA, UQ, and inversion (model calibration), and has the advantage of being non-intrusive: no modifications to the RTM need to be made. Note that in principle, one could plug an emulator of the geochemical solver within an RTM and try to build a new emulator on the fly for each evaluated model parameter set within a SA, UQ, or inversion procedure. The technical feasibility and tractability of such approach remain to be investigated though. The work by Sun (2012) reports on the calibration of 4 first-order reaction rates using a rather simple RTM that does not account for thermodynamic equilibrium, inter-species interactions, and sorption processes. In addition, Sun and coworkers do not mention which emulation method they use exactly. Keating (2016)(Keating, 2016) resort to multivariate adaptive regression splines to emulate a model of the multi-phase transport of CO_2 with non-reactive transport of associated solutes. Keating (2016) use their emulator for an UP application that consists of predicting the ensemble behavior of some given model outputs across the considered parameter space.

4.2.3 Experiences on metamodelling based on deep neural networks.

Uncertainty is a common denominator in modelling real-world phenomena. Uncertainty arises from the variability of the input and internal parameters of the model. Also, there may be uncertainties related to the structure of the model. Various frameworks exist to quantify these uncertainties (see overview by Borgonovo (2016), for example). In the following, some of the most common sensitivity analysis frameworks are briefly presented.

FAST and EFAST

Fourier Amplitude Sensitivity Test (FAST) is a variance based sensitivity analysis method by Cukier RI (1973) and KE. (1973). Outputs of the model are converted into coefficients using multiple Fourier Transforms that represent conditional variance. After that, the multidimensional Fourier coefficient integral is converted into one-dimensional integral using the theory developed by Weyl (1938). The frequencies are usually irregular

non-integer frequencies and to reduce computation, a set of integer frequencies (multiples of base frequency) are selected. Integer frequencies are selected in a way that enables the error to be controlled. By using integers, the resulting function of one-dimensional integral is periodic and only a single period is needed for evaluation. The continuous integral function can be sampled using Nyquist-Shannon sampling theorem and the resulting sampled function is evaluated from the summation of function values. FAST method is so-called "first-order sensitivity index" method.

Extended Fourier Amplitude Sensitivity Test (FAST) is a modification of FAST proposed by Saltelli A (1999). The EFAST method estimates total effect indices by computing the variance of a complementary set to that introduced in FAST. Also, more flexible sampling scheme is introduced when compared to the FAST algorithm. EFAST algorithm can produce both "first-order" and "total-order" sensitivity indices.

RBD

Random Balance Design (RBD) was first proposed for regression problems by Satterthwaite (1959). Tarantola (2006) modified the RBD method by combining it with the FAST method. While FAST algorithm uses integer multiples of frequencies, RBD uses random permutations of the selected frequencies to generate scrambled set that covers the input space. RBD is a "first-order" sensitivity method.

EASI

Effective Algorithm for Sensitivity Indices (EASI) was developed by Plischke (2010). EASI algorithm is basically similar to FAST and RBD but the frequencies are obtained by sorting and shuffling the input samples and not the frequencies of the Fourier transform. Input samples are sorted and output samples are re-arranged based on sorted input samples. Sorted data is then analyzed using power spectrum. EASI is a "first-order" sensitivity method, but it can be extended to higher order method by implementing multidimensional search curve to the search algorithm.

SOBOL

In Sobol method, the output variance of the model is decomposed into summands of input parameter variances for increasing dimensionality (Sobol, 1993; Zhang X-Y, 2015). Sobol sensitivity analysis determines the contribution of each input parameter and their interactions related to the output parameter variance. While the FAST method uses sinusoidal components of the Fourier method for pattern search, Sobol uses Monte Carlo simulation for that purpose. Sobol algorithm can produce both "first-order" and "total-order" sensitivity indices.

We have recently compared the described sensitivity analysis methods for evaluating a radionuclide transport compartment-type model. The model considers a lake-farm scenario at a site of nuclear waste repository. There were altogether 34 probabilistic variables describing distribution coefficients (K_d) of the model compartments, features of the modelled lake (area, volume), nutrient root uptake of plants etc. We found that for ^{36}Cl , for example, when considering the first-order sensitivity, all the methods ranked the same parameter – K_d of the soil – as the feature contributing most to the output variability (i.e., the variability of the dose to humans living in the modelled site). Also, the three next contributing parameters were the same for all methods. However, the contribution of the respective input variables varied significantly for different methods (from 54.8 % to 83.8 % for K_d of soil). When applying total-order sensitivity analysis (EFAST and SOBOL methods), the difference was even higher (the most contributing variable, K_d of soil, contributed 28.3 % in EFAST and 71.1 % in SOBOL analysis).

Sensitivity analysis using the frameworks described above, requires the model to be executed repeatedly with parameter valued drawn from the corresponding distributions (i.e., Monte Carlo simulation). This is often not feasible as the implementation of the physical models may require excessive computational power. The physical model can, however, be replaced by a metamodel, emulating the input-output relationship of the true model. In case the metamodel is capable of approximating the model function sufficiently well, it can be used to further explore the behaviour of the modelled phenomenon. Also, the metamodel can be used for sensitivity analysis through Monte Carlo simulation (Figure 1). Neural Network based metamodel has been used in Shing Chan (2018), for example, to emulate a geological Multiscale Finite Volume model.

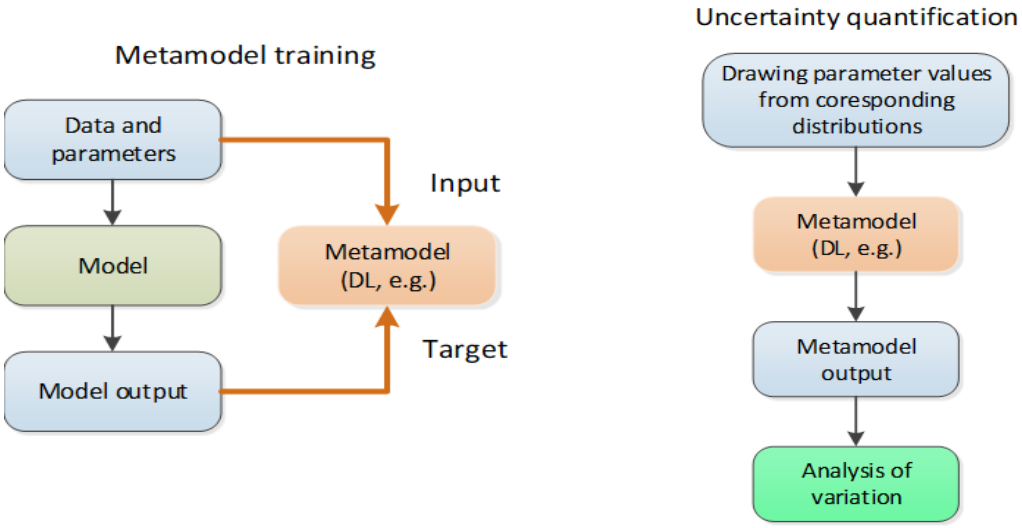

Figure 15 – Using metamodeling in sensitivity analysis

Recently, a methodology has been developed to trace back the input variables contributing most to the output of a trained deep neural network (Montavon and Müller, 2018). For example, in the case of applying a Convolutional Neural Network to a 2-D input data matrix, the method enables to determine the significance of each input pixel to the network output. The methodology is generally applicable to all neural network type structures.

A step forward is:

- to compare different sensitivity analysis frameworks in the evaluation of THM(C) models used for modelling the phenomena related to geological repositories of nuclear waste
- to develop and evaluate Deep Neural Network based metamodels in emulating physical THM(C) models for sensitivity analysis trough Monte Carlo simulation
- to apply the Relevance Propagation technique proposed by Montavon and Müller (2018) to the sensitivity analysis of THM(C) models using metamodeling.

4.2.3.1 Surrogate model construction

Uncertainty quantification (UQ) techniques have been extensively developed in the last decade in order to estimate the uncertainties in a Quantity of interest (QoI) caused by uncertainties in model inputs. A cornerstone in UQ is an accurate estimation of the changes in the uncertain QoI caused by changes in the uncertain inputs. The goal is generally to estimate the statistics of the QoI, such as its mean and its variance, and this can be achieved using Monte Carlo technique (and its variants) whereby an ensemble of simulations is performed with randomly selected input data. The convergence rate of the Monte Carlo method is independent of the dimension of the uncertain space but is rather slow (the error in estimating the mean, for example, decreases as the square root of the number of realizations). As a consequence, the number of ensemble members needed to estimate the statistical moments with high accuracy increases dramatically with the number of uncertain parameters and becomes nearly impractical when the forward model is computationally expensive and can only be run hundreds of times (as opposed to millions of times).

So, for a computationally expensive model, the use of a surrogate can be the only way to do an uncertainty propagation problem. The principle of a surrogate (a.k.a. metamodel, emulator, proxy or response surface) is to build, from a finite (relatively slow) number of samples, an approximation of the original forward model able to estimate the response of the QoI to changes in the input. The main advantages of a surrogate are (i) to efficiently propagate the uncertainties in the model inputs in order to compute the uncertainties in the model output, (ii) to identify the dominant contributors to the output uncertainties through the sensitivity analysis, and (iii) to perform parameter calibrations efficiently (the so-called backward propagation) if observational data is available. From a practical point of view, the surrogate is built unintrusively (i.e., without changes to the forward model that is treated as a “black box”) at the cost of performing ensemble calculations. The input-output relationships of the ensemble members are in fact exploited to obtain a simplified

representation of the model output whose computational cost is negligible in comparison with forward model evaluation.

Two of the most popular and commonly used surrogate construction techniques are **polynomial chaos** (PC) expansions and **Gaussian process** (GP) regressions. On the one hand, PC expansion (Ghanem and S.D., 1991; Le Maître and Knio, 2010) represents the QoI by a finite sum of orthogonal polynomials. Many works have been devoted to improve the computation of the PC expansions including for instance (Blatman and Sudret, 2011; Doostan and Owhadi, 2011; Conrad and Marzouk, 2013; Alemazkoor and Meidani, 2017). On the other hand, in GP regression (Rasmussen and Williams, 2005), a.k.a. kriging (Matheron, 1973) in geostatistics, the QoI is modeled as the realization of a stochastic process. As well, an abundant literature exists for the GP (Williams, 2001; Lawrence, 2003; Conti and O'Hagan, 2010; Hensman, 2013; Gramacy, 2015). Some recent studies (Le Gratiet L, 2016; Owen NE, 2017; Roy, 2017; Sochala and Iskandarani, 2019), have showed that the two approaches lead to a similar level of accuracy, the differences being on the computational complexity and flexibility.

Suitable methods for surrogate model construction include the polynomial approximation via the stochastic collocation method (Babuška, 2007; Nobile, 2008) and the radial basis functions interpolation (Powell, 2001; Chen, 2005). Both of these types of surrogate models have been successfully utilized for the acceleration of Markov chain Monte Carlo methods, see e.g. Bliznioukˇ (2008). John and Martins (2018) discusses the construction of surrogate models based on random data and points out problematic situations caused by high number of random parameters and high number of available snapshots.

Motivation for the development of surrogate models comes from the Bayesian solution of inverse problems. Numerical realization of the Bayesian inversion typically involves the use of Markov chain Monte Carlo (MCMC) methods which provide random snapshots of an underlying numerical model. For the acceleration of the Bayesian inversion, it is beneficial to construct a surrogate model from these snapshots (as explained within the next topic „Uncertainty treatment and sensitivity analysis"). So far IGN constructed surrogate models based on the approximation using complete polynomials and using radial basis functions, see Domesová (2018). IGN also have experiences with the use of intrusive surrogate models within the Bayesian framework, see the stochastic Galerkin aproach in Blaheta (2018).

4.3 Uncertainty treatment and sensitivity analysis

4.3.1 Compilation of the recent efforts taken for developing sensitivity analysis methods

Sensitivity analysis is a means of treating uncertainties that is generally considered essential in repository performance assessment. By analysing the effects of input variation to the output of performance assessment (PA) models, it allows the identification of the most relevant uncertainties or, from the reverse point of view, the exclusion of practically irrelevant uncertainties from further investigation. It has to be kept in mind, however, that sensitivity analysis is performed on the model, not on the reality. Therefore, its results describe the sensitivities of the model and must not be misinterpreted as statements about the actual sensitivities of the repository system.

Sensitivity analysis can be performed in different ways. While local sensitivity analysis investigates the behaviour of the model system under variation of one specific model condition (typically a parameter value) keeping all others constant, in a global sensitivity analysis one varies all conditions or model parameters that are considered uncertain simultaneously, executing a higher number of model runs, and analyses the outcome using appropriate methods.

Local sensitivity analysis is normally performed by executing a few model runs with deterministically selected conditions. This is often done if these conditions are not describable by a simple parameter but represent more complex situations, such as scenario variants, but also for dedicated investigation of the influence of a specific parameter. Global sensitivity analysis, however, is technically limited to parameter uncertainties. The usual approach to global sensitivity analysis is the probabilistic approach. It is assumed that the uncertainties of the model input parameter values can be represented by statistical distributions. According to these

distributions, sample values are drawn and combined in such a way that the total parameter space is covered as well as possible. The model has then to be calculated with each of the sampled parameter combinations. The collected outcome of these calculations can be mathematically analysed in view of model sensitivity to individual parameters.

Coming along with the increase of computer capabilities, probabilistic sensitivity analysis has been a subject of specific interest in mathematical research during the recent two or three decades. A variety of more or less sophisticated mathematical methods have been developed, which produce numerical sensitivity measures or graphically coded information (Saltelli, 2008; Borgonovo, 2016). Different types of methods act differently to models with different properties. Repository PA models often exhibit a distinctly nonlinear behaviour or even discontinuities and the model output is typically distributed over several orders of magnitude. As a consequence, such models should not just be analysed using some standard method as the results might be misleading or do not reveal the actually interesting details of sensitivity.

The methods of sensitivity analysis applied so far to repository PA models can be grouped in the following categories:

- **Correlation or regression methods**. Such methods linearize the model input-output relation and calculate the parameter sensitivities from correlation or regression coefficients, which are normalised to a range between -1 and 1, negative values indicating inverse sensitivity (higher input -> lower output). For models with a close-to-linear behaviour, these methods produce clear and unique sensitivity statements. The degree of linearity can be assessed via the coefficient of model determination, r^2, which is always between 0 and 1. While 1 means true linearity, a low r^2 indicates that the model can only poorly be approximated by a linear approach. This does not necessarily mean, however, that the calculated sensitivities are meaningless or of low relevance, as they may represent just the interesting aspects of the model. Application of such methods to nonlinear models can be useful, but requires thorough model understanding
- **Rank-based correlation or regression methods**. For these methods, a rank transformation is performed on the model output, replacing each value by its rank in an ordered list. For nonlinear but monotonic models, this normally leads to significant increase of r^2 and, apparently, to more significant sensitivity results. It should be kept in mind, however, that rank transformation tends to undervalue high and overvalue low model output values, which might influence the sensitivity statements. Moreover, the rank transformation is non-reversable and causes loss of information.
- **Non-parametrical sensitivity tests**. Such tests work by dividing the input sample into two or more subsamples according to some criteria and comparing the corresponding model out-put. The resulting sensitivity statements are not easy to interpret. These methods should be applied with care and only for specific reasons.
- **Variance-based sensitivity analysis**. The variance of the model output that occurs under variation of the input parameters can – under certain circumstances – be uniquely decomposed in contributions caused by the individual parameters and their combinations. The relative contributions to the total variance, the Sobol indices, provide a measure for sensitivity that does not require model linearity or monotonicity. 1st-order indices quantify the influences of the individual parameters alone, higher-order indices describe parameter interactions. The sum of all indices of any order that contain a specific parameter is called the total-order index of that parameter and quantifies its total influence on the model output in interaction with all other parameters. A number of computational methods are available for calculating these indices.
- **Graphical methods of sensitivity analysis**. A number of methods have been invented to represent the sensitivities graphically instead of compressing them to a single numerical value. Such figures provide a clear optical impression of the sensitivities and are often more meaningful than a single value. A typical drawback is that many separate figures are needed to visualise the sensitivities of time-dependent models.
- **Density-based sensitivity analysis**. This kind of method is based on the idea to compare the conditional and the unconditional probability density of the model output. Such methods work with any model and can improve model understanding but are not easy to apply and interpret.

Since for probabilistic sensitivity analysis with several uncertain parameters it is necessary to perform a rather high number of model runs, such investigations can become very time-consuming and occupy considerable computational resources. This problem can possibly be mitigated by using metamodels. Metamodels are analytical functions of the parameters, such as polynomials, designed to simulate the behaviour of the actual

model under investigation, including its sensitivities. Where the original model might run several hours, a metamodel can be evaluated within milliseconds. So, it is possible to perform a high number of metamodel runs for calculating sensitivity measures. In exchange, however, it is necessary to adjust the metamodel accurately by performing a sufficient number of runs of the original model. Experiments showed that defining and adjusting a metamodel and using it for sensitivity analysis with standard methods in separate steps does not provide actual advantages compared to direct sensitivity analysis of the original model. Specific methods, however, have been developed that combine metamodeling with calculation of sensitivity indices. The main strength of such methods seems to be the ability to effectively compute higher-order sensitivity indices, which is a task difficult to perform with direct methods. This is a field of current and future research.

Already in the 1990s, but specifically in the past decade, some research has been done or initiated aiming at applying different sensitivity analysis methods to different repository PA models and investigating their specific strengths and weaknesses under specific consideration of the typical properties of such models (Sinclair, 1993; Becker, 2009; Kuhlmann, 2013; Spiessl, 2017). Currently, an exercise is being carried out with a number of integrated PA models and one process model from USA, Germany and Belgium. Teams from France, Switzerland, Finland and Russia recently joined the group either for observation or for active contribution (Röhlig, 2018).

4.3.2 Uncertainty treatment methods

To provide samples from the Bayesian posterior distribution, it is usual to use MCMC methods, typically based on the Metropolis-Hastings (MH) algorithm, which requires high number of evaluations of the underlying numerical model. The computational costs can be reduced by full replacement of the numerical model by a surrogate model; however, this would provide samples from a modified distribution. To preserve the exact posterior distribution, it is preferable to use the surrogate model within the DAMH algorithm, see Christen (2005). In recent literature, we can also encounter DAMH-based sampling processes that iteratively update the surrogate model: Zhang (2013) uses adaptive sparse grid interpolation with hierarchical polynomial basis; Cui (2011) adaptively constructs a stochastic model of the error of the approximated posterior distribution; Cui (2015) constructs a reduced-order model using forward model evaluations calculated during the sampling process.

The Bayesian approach had been applied to material parameter identification in model porous media flow problems, see Blaheta, Béreš, et al. (2018), Domesova (2017) and Domesová (2018). As the sampling procedure, the DAMH algorithm was used. Current research publication propose several possibilities of the surrogate model updates during the process of posterior sampling. According to our first results, the surrogate model can be constructed and iteratively updated during the sampling process, which reduces the precomputation cost, see Domesová (2018).

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