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CROCK

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Dissemination Level		
PU	Public	X
RE	Restricted to a group specified by the partners of the CROCK project	
CO	Confidential, only for partners of the CROCK project	

Objective:

The objective of this WP 5.1 is to apply the outcome of other WPs to the application in the Safety Assessment, thereby decreasing the PA uncertainty and providing improvements for future site characterizations.

Status of work being performed by KEMAKTA:

The present work being carried out consists of two major subtasks:

The first subtask involves the creation of a reactive transport template using the well-known reactive transport simulation code, CrunchFlow (e.g. Steefel, 2009). This template will eventually incorporate the same sorption chemistry description to be developed in WP4 for interpretation of laboratory experiments although with different boundary conditions and deployed to model advective flow along a single transport path with matrix retention processes including matrix diffusion and sorption on internal micro-surfaces of the rock matrix. In conjunction with this, a number of different scenarios for groundwater chemical evolution are to be defined with the intention of exploring the parameter space of the sorption model(s) produced in WP4 and its (their) interaction with physical transport processes. Here, three scenarios are preliminarily proposed by Kemakta as a benchmarking exercise:

1. Intrusion of a meteoric water into a fracture flow system initially equilibrated with saline groundwater (no redox gradients);
2. Intrusion of a decaying alkaline plume into a fracture flow system with downstream pH neutralisation achieved by groundwater mixing;
3. Intrusion of an oxygenated water of glacial origin into a fracture flow system initially equilibrated with reducing saline groundwater;

The intention of these calculation cases is to impose sufficiently strong geochemical gradients on the system that non-linear effects of groundwater chemistry are readily apparent in the residence time distribution curves calculated for transported radionuclides. The three cases outlined above are intended to test model response to (1) ionic strength gradients, (2) pH and carbonate concentration gradients, and (3) redox gradients. To some extent, case (1) also probes pH and carbonate parameter space by way of mutual correlations between these variables in natural groundwaters. For solutes such as Ni that appear to exhibit both ionic strength and pH/carbonate sensitivity it may be useful to additionally simulate a simplified ionic strength case to evaluate differences between different sorption mechanisms with regard to transport retardation. Dissolution and precipitation processes involving rock matrix primary and secondary minerals will also be included to the extent deemed necessary to capture additional features of pH and redox buffering which might be expected when geochemical gradients are imposed on the system. At the present time, however, the above scenarios should be considered merely as suggestions and the final choice of test scenarios will be made at a later time in cooperation with other WP5 partners.

The second subtask involves the deployment of a simplified transport model to simulate the reactive transport of radionuclides where the description of sorption is decoupled from the temporal and spatial evolution of the major chemical components in the groundwater. In this case, instead of simulating the full reactive chemistry problem directly in the transport model, a parametric K_d surface calculated from the sorption model produced in WP4 is applied at run time in a numerical calculation where non-linearities of the sorption process are accounted for

by use of appropriate non-linear accumulation and pseudo-reaction corrections to the mass balance. Here the temporal changes in groundwater chemistry are modelled as a separate problem using mixing models or coupled reactive transport as deemed appropriate for the particular case being examined. The radionuclide transport model will be tested using the same benchmarking cases outlined above. The aim here is to identify situations in which such an approximation is valid and when it is not appropriate. This may be particularly important for solutes such as uranium which may exert a non-negligible influence on groundwater redox potential if present in sufficiently high concentration. At the present time, the basic methodology of the modelling concept has been tested using some simplified cases and the results suggest that it is possible to simulate long timespans (0.1-10 Ma) significantly faster than is achievable using a full reactive transport simulation.

References:

Steefel C, (2009) CrunchFlow. Software for modeling multicomponent reactive flow and transport. User's Manual, Lawrence Berkely National Laboratory, CA, USA.

Status of work being performed by Amphos 21:

When dealing with reactive transport models used in the framework of PA studies for deep geological repositories of nuclear waste, modellers have always faced the enormous computational challenges posed by the large spatial and temporal scales of the problems as well as the burden of solving thousands of coupled non-linear equations. It turns out that the use of K_d -based simulations offers an appealing alternative since it allows the system to be fully linearized. Yet, the definition of reliable K_d -values is a hard task since they depend on the site-specific geochemical conditions, namely on the pH and redox conditions. These conditions, in turn evolve over the considered time-frame. In the framework of WP4 of the CROCK project, we have developed a prototype for the calculation of "intelligent" K_d values. The prototype is based on the evidence that pH and redox conditions, which govern the K_d values of the considered radionuclides, are controlled by few minerals (e.g. calcite, pyrite, FeS), which in turn can be often described under equilibrium assumption. Thus, the prototype consists of two companion numerical models: (a) the "classical" K_d -based numerical model and (b) a fully explicit reactive transport model where the (equilibrium) mineral reactions are described using the efficient procedure proposed by De Simoni et al. (2008). After each simulation step, new pH and redox conditions are inferred from the "supporting" model. This geochemical information and a companion data-base (i.e. pH and redox vs K_d), feedbacks the K_d -based model by providing new K_d -values in each node of the model domain. The functioning of the prototype is illustrated in Figure 1. In order to test the underlying simplification assumption (i.e. K_d values controlled by pH and redox conditions in turn controlled by mineral equilibrium), a numerical tool, denoted as MC-PHREEQC, has been developed. This framework uses Monte Carlo simulations to automatically generate parameter values to be used as input for reactive transport codes (currently Phreeqc), based on provided probability distributions. The results are treated in a probabilistic manner to increase our knowledge on K_d sensitivity to specific parameters at different scales, thereby decreasing uncertainty in PA.

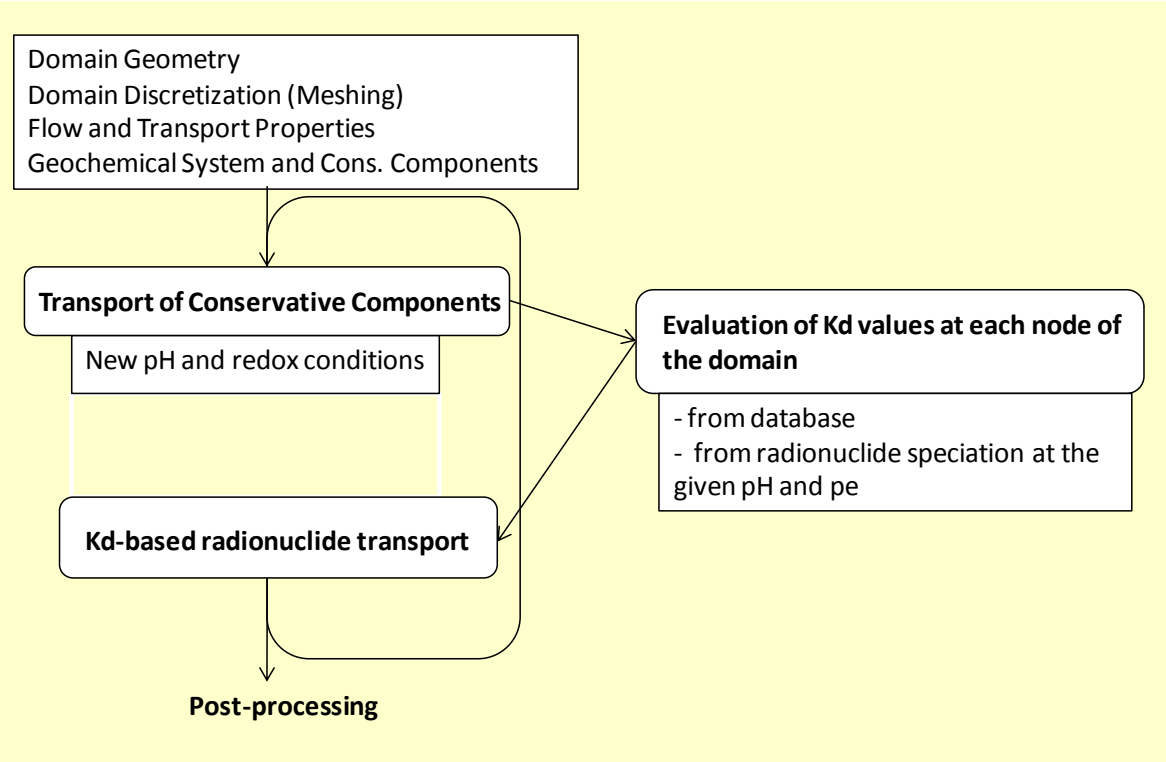

Figure 1. Overview of the method to incorporate "intelligent" Kd values.

References:

De Simoni M., Sanchez-Vila X., Carrera J., Saaltink M. W. (2007). A mixing ratios-based formulation for multicomponent reactive transport. WRR vol. 43.

Status of work being performed by VTT in WP5:

The present work being carried out involves the conceptualisation and planning of the test system to be applied in testing PA based models (WP5) against more detailed science based models to be developed in WP4.

The goals of PA level modeling at VTT are

- to evaluate uncertainties of K_d values
- to evaluate uncertainties of whole K_d based sorption model approach used in PA

The starting point is that the uncertainties are connected to scaling and extrapolation of the Kd values from the experimental conditions to the PA conditions.

VTT aims to develop a surface complexation model, e.g., nickel and europium on biotite. In addition to the biotite system VTT is also interested in extending the work to some other surfaces. VTT will also search open literature to find suitable sorption data for some other radionuclide on biotite.

A reactive sorption model for some interesting system is the first major goal of VTT's work, and the major goal is the conclusion derived from comparison between the results of this

model and the PA-based model in WP5. The modelling will be done in defined geometrical setups (matrix diffusion and fracture water), although the setup is still being planned and VTT will request suitable and realistic data from other CROCK partners. Sorption will be modelled by surface complexation and ion-exchange reactions, and these models will be implemented within the COMSOL Multiphysics platform. The results of the COMSOL model and some PA level radionuclide transport models will be compared.

In addition of transport model comparison, K_d-values and SCM-models in the literature will be evaluated and modelled in different scales if possible (includes uncertainties) to enable data comparison instead of comparing transport modelling results, but this work has not yet started effectively (see Figure 2, where the two approaches has been sketched).

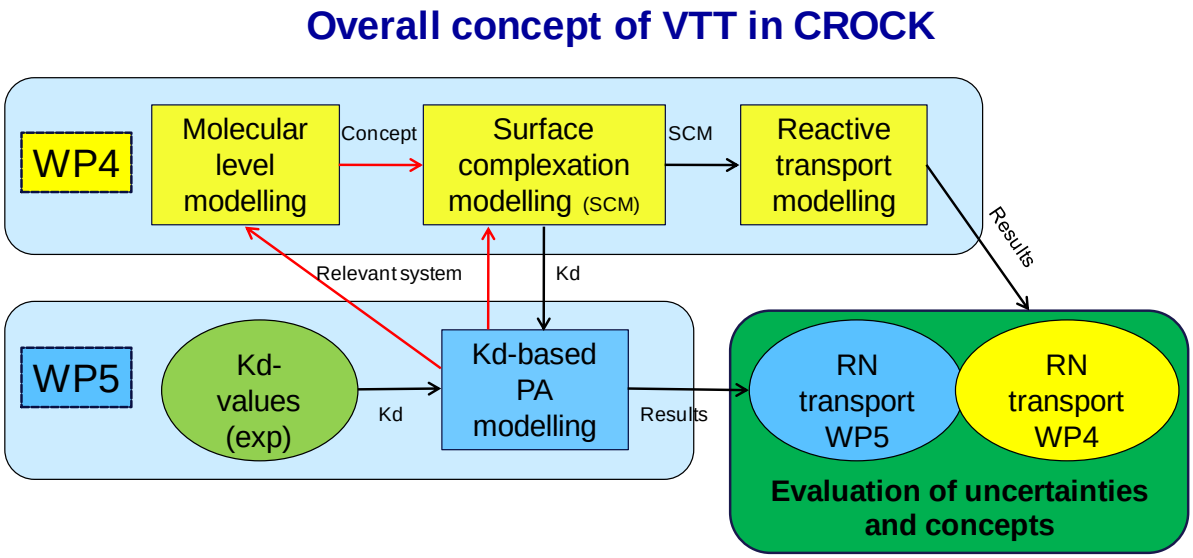

Figure 2. How to apply SCM and reactive transport modelling to enable comparison of both Kd-values and transport modelling.