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Pore water colloid properties in argillaceous sedimentary rocks

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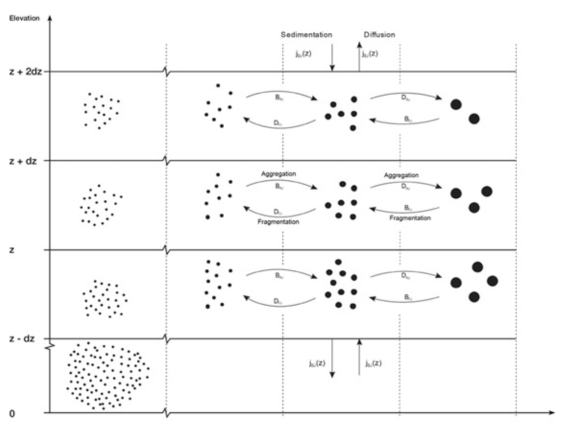
HIGHLIGHTS

- This study predicts pore water colloid properties from argillaceous sedimentary rocks.
- The study combines field, laboratory and model results.
- The high mineralization of the pore water limits the clayeous colloid concentration.
- The study predicts colloid concentrations below the ppb level for sizes from 10 to 100 nm.

GRAPHICAL ABSTRACT



Processes describing the colloid live to build up a colloid population.

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ABSTRACT

The focus of this work is to evaluate the colloid nature, concentration and size distribution in the pore water of Opalinus Clay and other sedimentary host rocks identified for a potential radioactive waste repository in Switzerland. Because colloids could not be measured in representative undisturbed porewater of these host rocks, predictive modelling based on data from field and laboratory studies is applied. This approach allowed estimating the nature, concentration and size distributions of the colloids in the pore water of these host rocks.
As a result of field campaigns, groundwater colloid concentrations are investigated on the basis of their size distribution quantified experimentally using single particle counting techniques. The colloid properties are estimated considering data gained from analogue hydrogeochemical systems ranging from mylonite features in crystalline fissures to sedimentary formations. The colloid concentrations were analysed as a function of the alkaline and alkaline earth element concentrations.
Laboratory batch results on clay colloid generation from compacted pellets in quasi-stagnant water are also reported. Experiments with colloids in batch containers indicate that the size distribution of a colloidal suspension evolves toward a common particle size distribution independently of initial conditions. The final suspension size distribution was found to be a function of the attachment factor of the colloids.
Finally, calculations were performed using a novel colloid distribution model based on colloid generation, aggregation and sedimentation rates to predict under in-situ conditions what makes colloid concentrations and size

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distributions batch- or fracture-size dependent. The data presented so far are compared with the field and laboratory data.
The colloid occurrence, stability and mobility have been evaluated for the water of the considered potential host rocks. In the pore water of the considered sedimentary host rocks, the clay colloid concentration is expected to be very low (<1 ppb, for 10–100 nm) which restricts their relevance for radionuclide transport.

1. Introduction

In environmental science, colloids are investigated for their role in transport of contaminant in aquifers (McCarthy and Degueldre, 1993). The colloid properties studied under in-situ conditions include not only their total concentration but most importantly their size distribution (Degueldre et al., 1996). The focus of this work is to evaluate the colloid properties in the pore water of Opalinus Clay and other potential host rocks identified for deep geological disposal of radioactive waste such as Effingen Member, "Brauner Dogger", and Helvetic Marl (Nagra, 2014; Mazurek et al., 2012; Pearson et al., 2003). In contrast to the homogeneous Opalinus Clay, some of these host rocks might form fracture networks where hydraulic flow locally governs fluid transport. In the safety assessment studies, colloid mobility in the Opalinus Clay has been evaluated and colloids were found to be of minor relevance (Voegelin and Kretzschmar, 2002). However this has to be assessed for the other potential host rocks. Based on the science of deep groundwater colloids and especially the experience gained at Wellenberg, Switzerland, the colloid mobility in fractured clay rocks with regard to colloid borne radionuclide transport is reconsidered. This work gives the expert's view on the colloid data required for the long-term safety assessment of a deep geological repository.

In this work colloid concentrations are estimated from the size distribution measured using single particle counting techniques or on the basis of a model. The colloid properties are investigated based on analogy to hydrogeochemical systems encountered in sedimentary formations described in previous studies. The present work combines field, laboratory and modelling studies.

The field studies concern geochemical formations ranging from various crystalline to sedimentary rocks (Degueldre et al., 2000). Section 2 summarizes the former field studies. The colloid concentrations are analysed as a function of the alkaline and alkaline earth element concentration in the water. Extrapolation for the salt concentration of the considered sedimentary formation water is suggested based on the properties of the attachment factor of these colloids. The attachment factor (a number between 0 and 1) is a function of the chemistry of the system and describes the fraction of collisions which actually lead to an aggregation event.

At the laboratory level, studies of colloid generation at a clay bed/water interface and colloid sedimentation from suspensions are performed under quasi-stagnant conditions. These quasi-stagnant conditions correlate to diffusive transport in the clay-rich host rocks that are of relevance to deep geological disposal. The generation of clay colloids from a compacted pellet in quasi-stagnant water is currently studied. Experiments with colloids in batch containers indicate that the size distribution of a colloidal suspension evolves toward a common particle size distribution for both extremes of initial conditions: starting from an almost colloid-free liquid phase on top of a pellet or starting from a very concentrated colloidal suspension (Bessho and Degueldre, 2009). The size distribution and the morphology of the colloids or aggregates at pseudo-equilibrium have been studied experimentally for example using scanning transmission X-ray microscopy (Degueldre et al., 2009a) and a model has been developed to understand their behaviour in batches (Degueldre et al., 2009b; García-García et al., 2009; Bessho and Degueldre, 2009).

Model calculations are performed, revisiting a recent original study (Degueldre et al., 2009b). The modelling results together with experimental tests, combine phenomena such as colloid generation, aggregation and sedimentation. This takes into account mechanisms in the water phase in order to identify the colloid properties under in-situ conditions. The data presented so far are compared with those of the hydrogeological systems studied earlier.

By combining for the first time field data with laboratory and modelling results, this paper yields a comprehensive understanding of the colloid properties in deep groundwaters. This integrative approach is needed to assess the importance of colloids in clay-rich host rocks for deep geological disposal, because field data from a fracture-free clay-rich host rock (such as Opalinus Clay) are not available and the combination of batch and modelling results alone provide limitations to the assessment of colloid generation. Thus the extrapolation of field results in combination with laboratory and modelling results provides the most reliable assessment of colloid properties needed for the performance assessment of a deep geological repository.

2. Colloid field studies

Field studies have been performed in formations ranging from crystalline (fractures filled with clay minerals) to sedimentary, with groundwaters going from weakly mineralised to saline and colloid going from the nanometre to the micrometre scale. The colloid data gained from these analogue hydrogeochemical systems are used for comparison.

2.1. Geological formations

The crystalline systems comprise recent granitic formations. The Central European Basement is first considered, with studies in the Black Forest of Southern Germany, in the Northern and in the Alpine area (Grimsel) of Switzerland. It also includes older granitic systems such as the Scandinavian and Canadian shields. The rock is mainly composed of quartz, feldspar and mica. Minerals currently found in the granitic fractures are: illite, biotite/muscovite, quartz and chlorite, calcite, iron oxy-hydroxides, with some pyrite. Their organic content is rather low (Degueldre et al., 1996).

The Swiss Crystalline studies (Degueldre et al., 1996) collect data of the Menzenschwand uranium mine, the Bad Säckingen spring, the Bad Zurzach thermal well, the Leuggern borehole and in the Alps both Grimsel Test Site and the Transitgas Tunnel. The depth ranges from about 100 to 1680 m in a 60 Ma old granite. The Experimental Underground Laboratory at Äspö (Sweden) lies in a 2 Ba old granite at a depth of 50–1562 m (Laaksoharju et al., 1995). In Manitoba, Canada (Vilks et al., 1991), waters were collected at depths down to 1150 m in the Whiteshell research facility that is underlain by the Lac du Bonnet granite batholith. The clay minerals in the water bearing fractures include: biotites, muscovites, chlorites and illites.

The Yucca Mountain (Nevada, USA) formation may also be described as crystalline with amorphous mineralizations and low organic concentrations. It consists of a thick accumulation of Miocene silica ash-flow tuffs of volcanic origin. The rock consists mainly of quartz, feldspar and zeolite with local gel layers (Levy, 1992).

Morro do Ferro, Minas Gerais, Brazil, is an altered (weathered) formation issued from volcanic activities in subtropical area containing some organic content. The rocks were predominantly carbonatite with layers deeply weathered to laterites. As its name suggests, Morro do