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Contribution of natural tracer profiles for the characterization of transport properties of argillaceous formations

F. Bensenouci (1,2), J.L. Michelot (2), S. Savoye (3), J.M Matray (1), and H.N. Waber (4)

(1) IRSN, Institut de Radioprotection et de Sûreté Nucléaire, Av. du Gen. Leclerc BP No. 17, 92262 Fontenay-aux-Roses, France, (2) UMR IDES- CNRS, Université de Paris-Sud, Bât 504, 91405 Orsay, France, (3) CEA/Saclay, L3MR, 91191 Gif sur Yvette, France., (4) Gestein-Wasser-Interaktion, Institut für Geologie, CH-3012 Bern

Consolidated argillaceous formations are considered as potential host rocks for the disposal of high level radioactive wastes, because of their very low permeability and favourable sorption properties. Diffusion is assumed to be the dominant transport process in argillites like those studied in various underground research laboratories at Bure, Tournemire (France) and Mont Terri (Switzerland). In such media, investigations of the porewater natural tracers (^{2}H, 18O, Cl-) appear as a pertinent method for the understanding of the palaeohydrogeological evolution and the solute transport properties of the rock formation.

Two methods based on equilibration techniques between pore and synthetic waters (radial diffusion and vapour exchange) were used to determine the water stable isotope and chloride concentrations in argillite porewaters. The radial diffusion method was also used to estimate the diffusion coefficients of deuterium and chloride and their related geochemical porosities.

In the case of Tournemire, the curved profiles given by the water stable isotope compositions and obtained by the two techniques are very consistent and show a net positive shift compared to that obtained by Patriarche et al. (2004) who used the vacuum distillation technique at 55°C. The diffusion coefficients calculated in this study by equilibration techniques are in order of 10-11 m^2s-1 and 10-12 m^2s-1 for the water stable isotopes and chloride, respectively. The values of water contents obtained from chlorides by diffusion experiments are lower than those obtained from water stable isotopes. This result confirms the occurrence of an anionic exclusion in this claystone. Several simulations of diffusional exchange between the argillite and the aquifers above and below were performed for assessing the large-scale transport properties of the argillites. Modelling the transport of chloride and deuterium with HYTEC (a 2D coupled reactive-transport model from the Paris School of Mines) indicates that the diffusive process likely started between 13 Ma (deuterium) and 20 Ma (chloride) ago. This result is in quite good agreement with the known palaeo-hydrogeology of the Causses area, which indicates that karstification and the valleys' incision started during Miocene times.

References

Patriarche, D., Ledoux, E., Michelot, J.L., Simon-Coincon, R. & Savoye, S. (2004): Diffusion as the main process for mass transport in very low water content argillites Part 2. Fluid flow and mass transport modeling. Water Resources Res., 40, W01517, 2004.