



Contribution ID: 113

Type: Verbal

Tritium (HTO) as a conservative tracer used for characterization of contaminant migration in porous rock environment

Chemical leaching of uranium in Northern Bohemia (Straz pod Ralskem) took place in between 1972 to 1996. Acid technology solutions, mainly consisting of sulphure acid (more than 4 mil tons), nitric acid, ammonia and fluoric acid, were pumped into uranium bearing layers and caused contamination of Cenomanian and Turonian water reservoirs. Cenomanian aquifer containing U was contaminated by more than 5 mil tons of dissolved species, including mainly H₂SO₄, sulfates, Fe, Al, NH₄. However, trace metals are also presented in a broad range of mobile species –e.g. Th, As, V, Mn, Tl and Be both released and migrate within contaminated aquifer and neighbouring layers.

Contaminant migration within different layers must be considered during evaluation of measures and methodologies to be used during remediation process. Target rock layers include both porous „friable“ sandstones with permeability coefficient up to 10⁻⁵ m/s (majority of contamination) and upper „fucoid“ sandstones, containing more impermeable clay and organic fraction with permeability coefficient from 10⁻⁸ to 10⁻⁷ m/s. Diffusion transport of potential contaminants is expected to dominate mainly within fucoid sandstone layers.

In presented study ³H (HTO) was used as a conservative tracer in order to study diffusion of non-sorbing, i.e. the most mobile contaminants, within porous environment saturated with groundwater. The through-diffusion technique was used for different samples, including both friable and fucoid sandstones. Break-through curves were evaluated using GOLDSIM diffusion module (NRI Rez/CTU), enabling to take into account the unsteady boundary condition in the inlet reservoir.

Although the determined values of effective diffusion coefficient D_e fell into relatively narrow interval of (2.0•10⁻¹⁰ –6.34•10⁻¹⁰) m².s⁻¹, the dependence of tracer diffusion through sandstone sample on rock properties could be distinguished. It was found that ³H diffusion rate was dependent on pore size distribution, even though the total porosity of different samples did not differ significantly (22-27 % range), and furthermore on mineral content. Kaolinite was finally identified as the main influencing factor for species diffusion rate within sandstone as its content in pores results in different pore size distribution. In samples with higher content of kaolinite (max. 16 %) mainly pores with small size were observed in which tracer diffusion movement was slowed down, and vice versa. Moreover, this phenomenon influences also out-diffusion process of species/contaminants, retained in sandstone samples: faster out-leaching was observed for samples with lower kaolinite content and larger pores.

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Track Classification: Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management