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The design and fabrication of beta sensor system for in situ diffusion tests in mudstone in France

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ABSTRACT

Long term in situ diffusion experiments in the Callovian-Oxfordian mudstone (France) are designed in the context of nuclear waste management. β-emitters HTO and ^{36}Cl radiotracers are representative of neutral and anionic species in these experiments. Studies have been carried out to design an in situ beta monitoring system in order to quantify tracer migration in the rock pore water over time. The development, design and calibration of β-emitter radiation sensors were performed. An optimised geometry was calculated for the detection chamber of ^{36}Cl in solution (Ø × L = 30 × 2 mm^2) via Monte-Carlo N-Particle transport simulation tools. A better SNR (Signal to Noise Ratio) was obtained with YSO cylindrical crystal (Ø × L = 6.5 × 0.5 mm^2). A sensitivity of 0.21 cpm/Bq/mL was measured using ^{36}Cl standards. The ^{36}Cl detection limits were 18 Bq/mL and 14.2 Bq/mL after 1 h and 24 h of accumulation time respectively. Such beta sensors, placed 350 mm from the radiotracer injection borehole, could successfully discriminate ^{36}Cl anisotropic diffusion in various directions within 4 years. It will be tested in situ and can be adapted to other in situ experience.

1. Context and objectives

1.1. The Callovian-Oxfordian mudstone (COX)

Argillaceous media are being considered in numerous countries as potential host rocks for the geological disposal of radioactive waste (Norris, 2014). In France, an underground research laboratory (URL) has been built by Andra (The French National Radioactive Waste Management Agency) to investigate the feasibility of a radioactive waste disposal facility at a depth of 490 m within the Callovian-Oxfordian (COX) mudstone (Delay et al., 2014). The research objective includes the characterization of the confining properties of this argillaceous rock through in situ hydrogeological tests, chemical measurements and diffusion experiments (Delay et al., 2006).

1.2. Previous in situ diffusion tests (DIR) in the Andra's URL

To investigate and quantify the radionuclide diffusion properties of the Callovian-Oxfordian rock, in situ tests (DIR) have been carried out in boreholes (Dewonck et al., 2007). The design of these tests was based on previous experiments performed in the Swiss Mont Terri URL in Opalinus Clay rock (Palut et al., 2003; Wersin et al., 2004). At the bottom of vertical downward boreholes, synthetic water circulated in contact with the rock. Tracers were added to the synthetic water when stable chemical and hydrostatic state had been reached. The tracers injected were: Tritiated water: HTO, ^{125}I, ^{36}Cl, ^{22}Na, ^{85}Sr, ^{134}Cs. A circulation pump was maintaining a homogeneous concentration of the tracers in the test interval. Tracer contents in the circulating fluid were regularly measured by water sampling and analysing, allowing to monitor the activity decrease over time. At the end of the experiment, the rock around the injection chamber in which tracers had diffused, was over-cored in order to analyse tracer concentration profiles within the rock.

Since 2004, similar in situ experiments were also performed at Mont Terri and modelled by several teams (Wersin et al., 2008; Yi et al., 2012; Gimmi et al., 2014).

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1.3. Development of a new in situ diffusion test

A new in situ experiment is considered by Andra regarding the study of radionuclide migration (Diffusion of RadioNuclide: DRN). The aim of this experiment is to collect data on the migration of radionuclides, by means of experimental set-up as contrasting as possible with previous studies. This includes larger space scale (hundreds of mm), longer time-scale (over 8–10 years) and if possible in situ monitoring of radionuclides migration through the argillaceous rock during the experiment. HTO and ^{36}Cl radiotracers have been selected to monitor neutral and anionic species mobility in the COX mudstone. ^{36}Cl is one of the anionic radionuclides considered in the case of impact assessment studies of disposal units due to its mobility and long half-life (Fréchou et al., 2004). HTO is a neutral tracer which would be used as a reference compared to previous experiments. Both tracers are injected simultaneously in the synthetic water circulating in the packed-of test interval of a borehole.

The purpose of the present work was to develop a beta detection system based on the system patented under the reference "INPI 2.933.777 – (A1) – [08 54695]" and to evaluate the possibility to monitor in situ ^{36}Cl diffusion thanks to this system. This included optimization of the design and characterization of sensitivity and limits of detection (L.D).

2. Design of DRN in situ beta monitoring system

The general geometrical design of the in situ ^{36}Cl diffusion experiment was estimated under a 2D modelling of ^{36}Cl diffusion. As showed in Fig. 1, the modelling scenario considered a beta monitoring sensor placed in one borehole inside the mudstone at a distance of 500 mm or 300 mm from the injection borehole. This injection borehole contains the solution with radiotracers in contact with the mudstone. A parietal zone (green) was included to account for the drilling induced disturbed zone surrounding the boreholes. A void between the parietal zone and the sensor is filled by water. Water is in contact with detection crystal via a stainless steel filter plate. An initial activity (A_0) of 2000 Bq/mL was chosen, which represent an equivalent activity for ^{36}Cl as in DIR experiment (~20MBq). Standard diffusion parameters were taken for ^{36}Cl as measured in COX mudstone by Descostes et al. (2008). Results are illustrated by a cartography of ^{36}Cl activity after 5 years (Fig. 1, middle) and the temporal evolution of ^{36}Cl activity at injection boreholes (x = y = 0, blue curve) and then at 350 and 500 mm (red curves) from injection borehole (Fig. 1, bottom). The L.D at 14 Bq/mL (black dotted line) was determined from beta sensor performance as reported in the following section. These results show that no significant activity could be detected even in 10 years ^{36}Cl plume diffusion when the sensor is placed 500 mm away from the injection borehole (Fig. 1, bottom: dotted red line x = 500 mm). In contrast, after around 4 year radiotracer diffusion, ^{36}Cl could be detected by the sensor placed at 350 mm away from the injection borehole.

These modelling results led us considering the final geometrical design of in situ beta monitoring system illustrated in Fig. 2. Beta sensors could be placed at 350 mm around the injection borehole. This would guarantee the detection of ^{36}Cl diffusion in various directions within 4 years. Considering the possible axial deviation during drilling that can induce anisotropic diffusion along X (perpendicular to the bedding of the mudstone) versus Y (Parallel to the bedding of the mudstone), it has also been studied that with an anisotropy coefficient of 1.5, the sensors placed along two axes (S1 and S2 for example) could differentiate anisotropic diffusion after around 4 years.

In terms of beta particles detection sensor system, the well-known beta detection methodology is liquid scintillation counting which is an indirect fluorescence counting system used to quantify the activity of samples emitting beta and alpha particles (Staff, 2004). Such a method cannot be applied for direct continuous and in situ detection, as required for this project. Fiber-optic sensors (FOSs), consisting of a

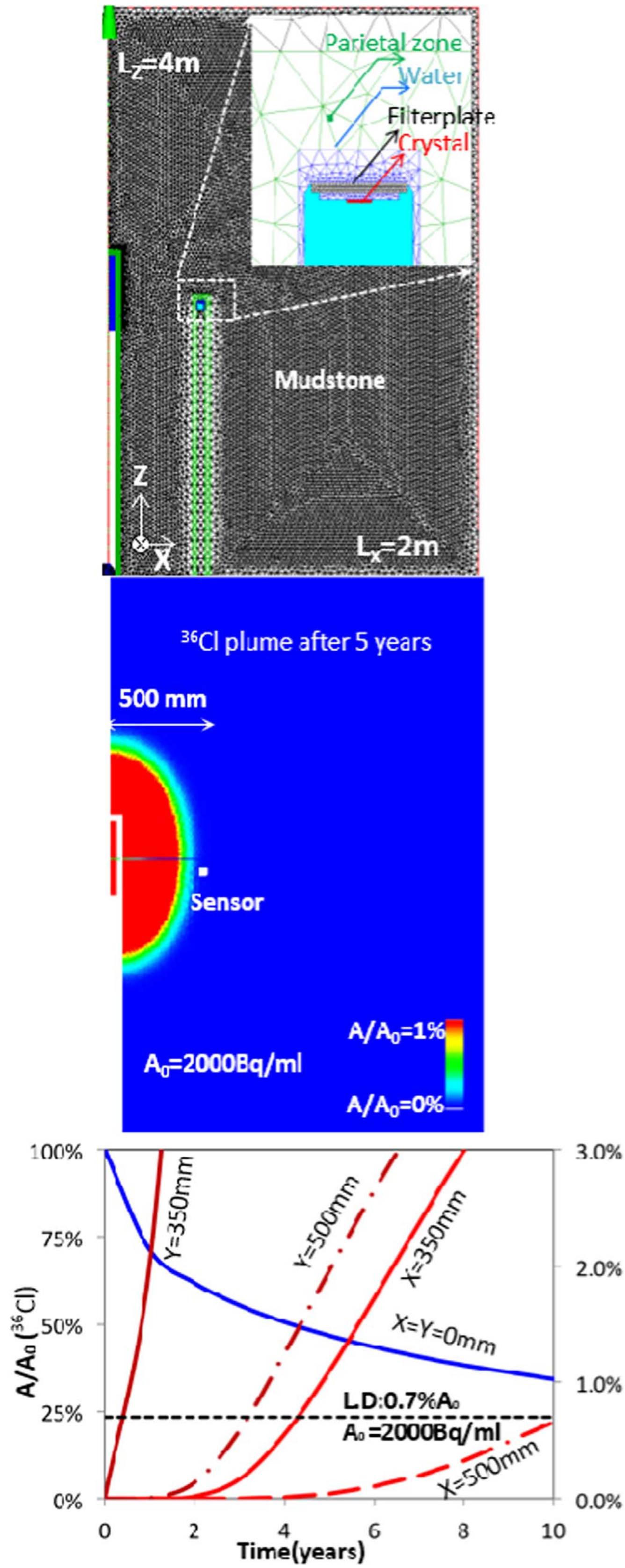

Fig. 1. (top): Mesh used for diffusion modelling. The cross section (y = 0) of beta monitoring system includes the injection borehole and the probe inside its borehole; (middle): 2D distribution of radiotracers modelled 5 years after injection; (bottom): Evolution of ^{36}Cl activity with time in sensor boreholes at various positions (350 mm and 500 mm away from injection borehole along X or Y axis). Vertical axis left scale holds for injection solution (blue curve, x = y = 0) whereas right scale holds for red curves.