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Novel application of β/γ autoradiography and collimated γ-spectrometry to study in situ radionuclide migration paths in fractured rock

A. Möri [a,*], C. Biggin [b], U. Mäder [a], J. Eikenberg [c], M. Rüthi [c]

[a] Rock–Water Interaction Group, Institute of Geological Sciences, University of Bern, CH-3012 Bern, Switzerland
[b] National Cooperative for the Disposal of Radioactive Waste (Nagra), CH-5430 Wettingen, Switzerland
[c] Division for Radiation Protection, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland

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Abstract

β/γ-Autoradiography and collimated γ-spectrometry were applied on in situ impregnated core samples containing various radionuclides such as ^{137}Cs and ^{60}Co to study diffusion paths in fractured granitic rocks from the Grimsel test site (GTS). The radionuclide tracers were injected into a water conducting feature (WCF) during years (^{137}Cs) or months (^{60}Co) before the flow field and the adjacent rock matrix were overcored for analysis.

The retardation sites of the radionuclides in and around the flow paths were first determined by means of a state of the art β/γ-autoradiography scanner. Increased activity above natural background was observed both in the WCF and in the adjacent rock matrix showing structural control of radionuclide transport into the rock matrix as activity in the matrix was mainly bound to grain-boundary pores.

In a second step, the areas of increased activity within the WCF and in the neighbouring rock matrix were investigated using a HPGe γ-spectrometer. A new setup for collimated γ-spectrometry was developed, which revealed spatial and nuclide specific information about tracer concentrations in the samples. Although this technique is hampered by much reduced counting efficiencies, it allows non-invasive determination of spatial distribution of radionuclides at the centimetre scale.

A preliminary data set produced to show the application possibilities of these techniques indicated maximum ^{137}Cs diffusion into the rock matrix adjacent to a WCF to a depth of 45 mm within 3 years and for ^{60}Co to about 3 mm in 2 months.

Keywords: Radionuclide transport; Matrix diffusion; β/γ-Autoradiography; γ-Spectrometry

1. Introduction

This study is part of the long-term diffusion (LTD) project in Nagra's underground rock laboratory, the Grimsel test site (http://www.grimsel.com), which is located in a granitic host rock in the Swiss Alps (Kickmaier et al., 2005). The present work applies state of the art β/γ-autoradiography in combination with γ-spectrometry to determine retardation sites of radionuclides in a water conducting feature (WCF) and, in particular, in the adjacent rock matrix to investigate structural and/or mineralogical control on the migration paths of radionuclides. This paper describes the approach and analytical methods and includes preliminary data to show the applicability of these techniques in contaminant transport studies including radionuclides.

The investigated rock material originates from a shear zone located in the granodioritic host rock at the GTS. The drillcore material was excavated and sampled in the frame of the Excavation Project in 1996 (Alexander et al., 2003; Möri et al., 2005). The shear zone consists of mica-rich mylonite bands which underwent brittle reactivation which is characterised by an array of cleavage parallel

* Corresponding author. Tel.: +41 31 381 31 15; fax: +41 31 389 34 31.
E-mail address: andreas.moeri@geo-online.com (A. Möri).

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fracture planes, mainly filled with fault breccia and/or fine-grained, cohesionless fault gouge (see Bossart and Mazurek, 1991; Möri et al., 2005 for details).

Within this WCF, numerous in situ dipole tracer tests were performed between 1988 and 1996 (Alexander et al., 1992; Frick et al., 1992; Smith et al., 2001). Three tracer tests with ^{137}Cs were carried out in 1993, injecting each time between 6.0E+06 and 6.5E+06 Bq. ^{137}Cs recoveries at the extraction borehole varied between 70% and 100% within the first 1000 h. In 1996, a cocktail of sorbing radionuclides (^{60}Co, ^{75}Se, ^{99}Tc, ^{113}Sn, ^{152}Eu, $^{234/235}$U and ^{237}Np) was injected under reducing groundwater conditions into the 1.7 m long flow field (Eikenberg et al., 1998) and the radionuclides remaining in the WCF were immobilised 2 months later by in situ impregnation with a Na-fluorescein doped epoxy resin (Frieg et al., 1998). Due to the relatively low viscosity[1] of the injected resin and the high hydraulic gradients during in situ impregnation, it is suggested that the resin-impregnated volumes roughly correspond to those parts of the WCF where radionuclides could be transported advectively. At this time, ^{137}Cs from the earlier experiments had already been in contact with the WCF and the surrounding rock matrix for 3 years.

2. Analytical methods

2.1. FUJI BAS-1800 II scanner for β/γ-autoradiography

β/γ-Autoradiography is a well known technique to visualise the spatial distribution of β/γ emitting radionuclides in rock samples and thin sections with limited discrimination between different radionuclides. In the past, the technique was based on conventional film technology whereas the new BAS-1800 II system consists of a scanner unit with re-usable imaging plates (IP's). The background radiation[2] during exposure of the IP's was minimised by means of a 5 cm thick lead shielded box combined with an internal shield of wood and copper minimising the effects of the secondary Pb X-rays (generated by particle matter interaction in the lead shielding) on the IP's.

IP's are flexible image sensors with dimensions of 23×25 cm and consist of a 250 μm thick PVC foil uniformly coated with a 150 μm thick layer of small crystals (grain size about 5 μm) of light-sensitive barium fluorobromide that contain trace amounts of bivalent europium. The excited Eu-electrons are stimulated in the scanner by a He–Ne laser light source and the emitted photons are collected by an optical guide and quantified in a detector. The maximum spatial resolution of the scanner is 50 μm. However, a certain amount of γ-rays emitted from lower levels in the samples may reach the rock surface and thus produce a scatter effect on the IP. This must be taken into account when interpreting the autoradiographs.

The sensitivity of the solid state imaging plates is 10–100 times higher than that of conventional X-ray films. The effective dynamic linear range is 5 orders of magnitude with a gradation of 65,536 grey levels in 16 bit mode.

2.2. γ-Spectrometry

Radionuclide activities were measured using an intrinsic HPGe γ-spectrometer equipped with a cryo-compressor for cooling. These detectors have a peak resolution of less than 2.5 keV at 1332 keV, which is sufficiently precise to determine photon emissions from mixtures of radionuclides simultaneously (e.g. ^{137}Cs and ^{60}Co). The efficiency curves for bulk rock analysis were determined using a software package called *WinnerTrack*™ taking into account the relative activity distribution within the core slabs (available from β/γ-autoradiography images), the sample geometry and sample self-absorption.

2.2.1. Collimated γ-spectrometry

Spot analyses of the activity of selected radionuclides were required in order to obtain information on the radionuclide distribution within the active areas without disturbing the rock by sub-sampling. γ-Rays cannot be collimated in a strict sense but only shielded to varying degrees, depending on the photon emission energy of the radionuclide and the thickness of the lead shield. Such spot measurements required developing an appropriate measurement setup with an optimised collimation aperture and adequate shielding.

The effectiveness of the collimation was tested with a mock-up WCF in form of an artificial ^{137}Cs/^{60}Co radionuclide source made of a 200×40 mm filter paper strip placed between a pair of 4 cm thick granite plates with straight edges cut. The activity of the artificial source was determined to be 41.5 kBq for ^{137}Cs and 34.6 kBq for ^{60}Co (±5% 2σ standard deviation). The test setup consisted of 10 cm of lead shielding between the sample and the detector with variable collimator holes.

The efficiency curve for this complex measurement geometry with a collimator hole of 20 mm was modelled for quantification of the activity at the measurement spots. The measured activities by collimation are shown in Table 1 and were 50.0 kBq for ^{137}Cs and 43.5 kBq for ^{60}Co (±20% 2σ standard deviation[3]). The deviations from the known activities of the source were 22% for ^{137}Cs and 26% for ^{60}Co, respectively and thus are almost within the error. This suggests that the geometric physical model provides a reasonable approximation of the nuclide specific activities determined with the collimated setup.

[1] Resin density is 0.987 kg dm^{-3} and viscosity after 30 min of injection at 13 °C is 173 mPa s (Frieg et al., 1998).

[2] The scanning facility was installed at the GTS in a radiation controlled (IAEA Level C) area.

[3] The statistical uncertainty resulting from collimated measurements was calculated using the law of error propagation for uncorrelated parameters and is mainly due to counting statistics and uncertainties concerning the calculated efficiency (i.e. modelled complex measurement geometry).