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Method development for electronic autoradiography with geological samples

INTRODUCTION

The final disposal of spent nuclear fuel will take place in crystalline granitic rock in a multi-barrier repository system in Finland. The transport properties of radionuclides in the geosphere need to be considered when assessing the overall safety of the repository. To this end, in situ diffusion experiments have been conducted in Olkiluoto and Grimsel. In this study, autoradiography methods were used to determine the spatial distribution of ^{133}Ba and ^{75}Se activity in granitic rocks from sorption experiments.

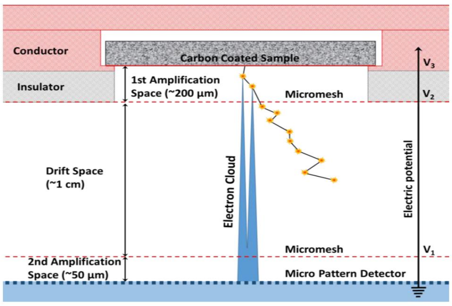

Fig 1. A schematic image of the BeaverTM where the primary electrons created during ionization by emitted electrons are multiplied in the amplification spaces. The resulting electron cloud is then localized on a micro pattern detector (Voutilainen et al., 2018).

EXPERIMENTAL

Sorption of ^{133}Ba and ^{75}Se on thin sections of veined gneiss and pegmatitic granite from the Olkiluoto site and granodiorite from the Grimsel Test Site was studied using autoradiography and gamma spectrometry. The sorption experiments on thin sections were carried out in the presence of groundwater simulants made to resemble the fracture groundwater of the in situ sites.

The spatial distribution of activity in the rock cubes and thin sections was studied using two autoradiography methods; digital autoradiography using phosphor imaging plate technique (Fuji 5100) and the Beaver^{TM} (Figure 2), which is based on coupling a micro patterned gaseous detector (MPGD) and parallel ionization multiplier device (PIM).

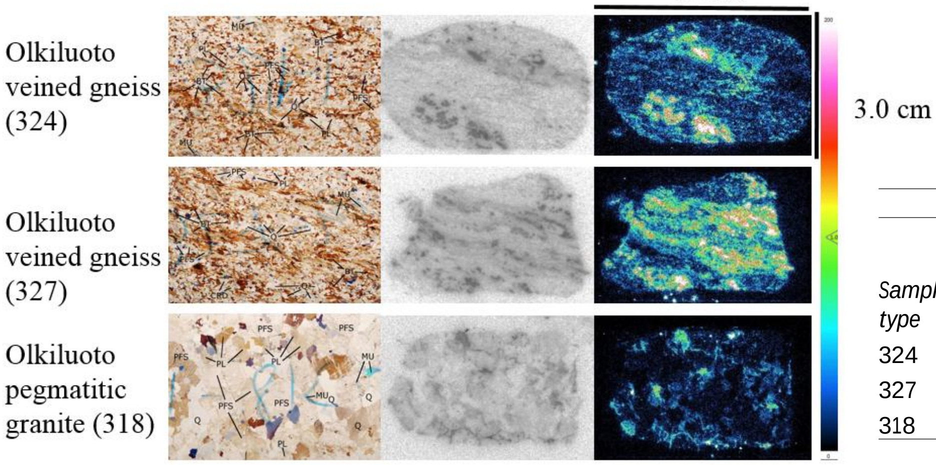

Figure 2. The photograph (left), digital autoradiograph (middle) and electronic autoradiograph with Beaver^{TM} (right) of veined gneiss (324 and 327) and pegmatitic granite (318) with ^{133}Ba.

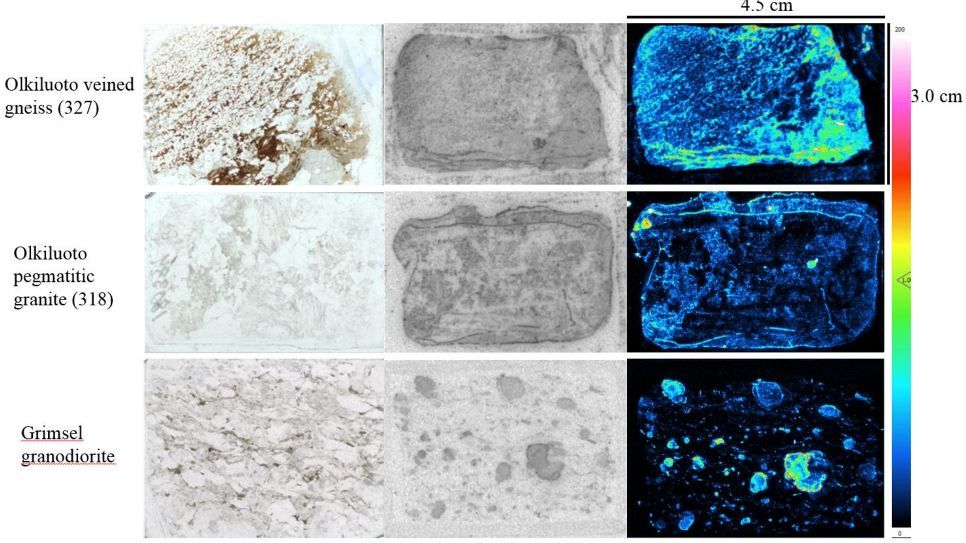

Figure 3. The scanned thin section (left), digital autoradiograph (middle) and electronic autoradiograph with Beaver^{TM} (right) of veined gneiss (327), pegmatitic granite (318) and Grimsel granodiorite with ^{75}Se.

RESULTS

A better resolution was obtained for 75Se than for 133Ba with the BeaverTM. 133Ba has more of higher-energy electrons emitting from its decay (Table 1), which decreases the resolution obtained with autoradiography methods.

The sorption of barium and selenium was more prominent in the mica-rich veined gneiss than in pegmatitic granite (Fig 2 and Fig 3). Distribution coefficients were calculated from the sorption results with gamma spectrometry and autoradiography through a developed polymethyl methacrylate PMMA standard sample. It was discovered that the values obtained in this study were in good agreement with previous results (Table 2).

Table 1. Energies and intensities of the electrons emitted by ^{133}Ba and ^{75}Se (NuDat).

^{133}Ba		^{75}Se	
Energy (keV)	*Intensity (%)*	*Energy (keV)*	*Intensity (%)*
4.05	138.0	1.24	130.2
17.18	10.6	9.11	41.5
30.20	14.20	12.51	4.19
43.63	4.0		
45.01	48.1		
75.54	7.3		

Table 2. Distribution coefficients of Ba obtained in this study compared with results from (Muuri et al., 2018a) and (Muuri et al., 2018b).

Distribution coefficient of Ba [m³/kg]			
	This study	Muuri et al. (2018a)	Muuri et al. (2018b)
Sample type	*Thin sections*	*Crushed rock*	*Rock pieces*
324	0.0023±0.0001	0.111±0.001	0.030±0.002
327	0.0023±0.0001		0.030±0.002
318	0.00046±0.0001	0.0109±0.001	0.007±0.001

CONCLUSIONS

Electronic autoradiography with the Beaver^{TM} has proven to be a useful tool in quantifying the spatial distribution of activity in granitic rock samples. Using image analysis, mineral specific distribution coefficients can be derived from the electronic autoradiographs obtained with Beaver^{TM}. This offers one step in upscaling sorption results into in situ conditions. The standardization of the method will be continued.

REFERENCES

[1].NuDat. http://www.nucleide.org/NucData.htm

[2].Voutilainen et al. (2018) Digital autoradiography on C-14-labelled PMMA impregnated rock samples using the Beaver^{TM}. MRS Advances. 3. 1161-1166.

[3]. Muuri et al. (2018a) The sorption and diffusion of 133Ba in crushed and intact granitic rocks from the Olkiluoto and Grimsel in-situ test sites. Applied Geochemistry. 89, 138-149

[4]. Muuri et al. (2018b) The in-diffusion of 133Ba in granitic rock cubes from the Olkiluoto and Grimsel in-situ test sites. Applied Geochemistry. 92, 188-195.

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