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Radionuclide diffusion into undisturbed and altered crystalline rocks

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ABSTRACT

An extensive set of porosity, ε, effective diffusion coefficient, D_e, and hydraulic conductivity, K, data were obtained from 45 granitic samples from the Bohemian Massif, Czech Republic. The measured dataset can be used to define parameter ranges for data to be used in safety assessment calculations for a deep (>400 m) radioactive waste repository, even though the samples originated from shallower depths (<108 m). The dataset can also be used for other purposes, such as evaluating the migration of contaminants in granitic rock (e.g. from shallow intermediate-level radioactive waste repositories and chemical waste repositories).

Sample relaxation and ageing processes should be taken into account in research otherwise migration parameters might be overestimated in comparisons between lab results and those determined *in situ*.

Keywords: disposal, granite, radionuclide diffusion, sampling protocols.

Introduction

Crystalline rocks are being considered as potential host rocks for deep geological repositories (DGR) for radioactive waste in many European countries. The disposal method is usually based on three protective barriers. The radioactive waste has to be encapsulated in a metal canister made of either steel or copper. The metal canisters are then placed in crystalline basement rock at a depth of about 500 m and enclosed in bentonite clay. After disposal the tunnels are sealed.

Any deep geological repository or storage facility, which might contain radioactive waste, CO_2 or natural gas, has to prove its safe performance in order to be licensed by regulators. The safe performance is evaluated using safety assessment methodologies. Migration risks to humans and the environment are necessarily included. In considering the safe performance of DGRs in crystalline rock, a dataset of rock migration parameters, such as porosity, permeability, diffusivity and transmissivity, has to be available. As DGRs are typically planned to be constructed at a depth of 400–600 m below the ground surface, there is a lack of data about the deeper horizons. Moreover, it is not wholly clear whether data from samples from shallow horizons can substitute for them.

Even though the main transport mechanism in crystalline rock is advection, migration processes from fractures into their linings and unaltered rocks must also be studied. The conceptual model is based on the presumption that non-advective migration is driven by diffusion into altered mineral layers and the undisturbed rock matrix adjacent to water-bearing fissures. The diffusion process depends on many features including molecule size and charge, sorption onto mineral surfaces, effective porosity, pore constrictivity,

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tortuosity and groundwater composition. Moreover, alteration by metasomatic recrystallization, which would change the rock properties, must also be taken into account. Laboratory experiments commonly use samples that were removed from deeply buried high-pressure environments that are presumed to have undergone pore space relaxation. These samples are typically stored for future work at room temperature and in oxic conditions. Such conditions have the potential to lead to mineralogical changes and to further pore space relaxation.

To evaluate the importance of such effects, a set of relatively old archive samples of granitic rocks from different depths up to 100 m was gathered, together with samples from four new boreholes drilled in 2010–2011 up to 100 m depth as part of Czech project FR-TI1/367. The samples were then characterized to determine selected migration parameters relevant to diffusive transport (porosity, ε, diffusion coefficient, D_e, formation factor, F_f). In order to interrelate rock non-advective transport properties to other rock migration characteristics, hydraulic conductivity, K, was also studied. The sample composition was determined, however it is not discussed in detail herein.

Theory

The diffusion process is governed by Fick's laws, which are summarized elsewhere (e.g. Eriksen and Locklund, 1989; Ohlssons and Neretnieks, 1995; Bradbury and Green, 1986; Vilks *et al.*, 2004; Löfgren and Neretnieks 2003*a*,*b*, 2006). Changes in concentration over time are determined by the equation:

$$\frac{\delta C_p}{\delta t} = \frac{D_p}{R_p}\frac{\delta^2 C_p}{\delta z^2} \qquad (1)$$

where C_p is the concentration in pore water (mol m^{-3}), D_p is the pore diffusion coefficient (diffusivity, m^2 s^{-1}), R the retardation factor of the rock matrix (m^{-3} kg^{-1}), t is time (s) and z is coordinate of movement.

The effective diffusion coefficient D_e (m^2 s^{-1}) is defined as:

$$D_e = \varepsilon \frac{\delta_D}{\tau^2} D_w = \varepsilon D_p = F_f D_w \qquad (2)$$

where ε is the porosity, δ_D is the constrictivity, τ^2 the tortuosity, D_w the diffusivity in free water (m^2 s^{-1}) and D_p the pore diffusivity in pores (m^2 s^{-1}), and F_f is the formation factor.

Methodology

Granitic rocks are being considered for deep geological repositories. Therefore, samples of granitic rocks from different part of the Bohemian Massif were examined from relatively old archived drill cores (archive samples) and cores from boreholes drilled during 2010 and 2011 (fresh samples). The archived samples had been stored for between 5 and 40 years. The maximum sampling depth was 108 m. The samples were taken regularly along the cores. Different granitic rock types were selected in the effort to look at the different properties that can influence migration, such as mineralogical composition, porosity, grain size and alteration. A series of metasomatically altered samples from Cínovec were included in order to determine the extent to which rock alteration influences the rock migration properties. However, samples that had undergone high P–T metamorphic alteration were excluded. Sampling localities, details and rock types are listed in Table 1, and the locations are shown in Fig. 1. A simplified chemical composition of the rock samples is plotted on SiO_2 *vs.* K_2O plots in Fig. 2. For each sample, the porosity, ε, effective diffusion coefficient, D_e, and hydraulic conductivity, K, were determined.

Water accessible porosity, ε, was measured using the water saturation method of Melnyk and Skeet (1986). The method was also used for saturation of the samples for the diffusion experiments.

The hydraulic conductivity, K, was determined at various confining pressures. Rock samples were installed in standard pressure cells manufactured by Wykeham Farrance for triaxial tests. Core samples 50 mm in length were subjected to a confining pressure similar to the *in situ* pressure. A constant pressure gradient between upper and lower faces (Δ = 50 kPa) produced a flow of water through the cores. The volume of water that passed through the sample, and the pressure difference between the inlet and the outlet, were measured by pressure controllers and recorded at regular intervals. The hydraulic conductivity was then calculated according to equation 3:

$$K = \frac{Ql}{Ah} \qquad (3)$$

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TABLE 1. Locality, origin and petrographic characteristics of the samples.

	Symbol	Origin	Rock type
Cinovec	CS1	archive	Medium- to fine-grained albite-topaz-zinnwaldite granite, partly altered (kaolinization)
Krásno	Kž25	archive	Leucocratic mica-free granite to feldspatite with alternating albite/K-feldspar ratio
Melechov 2	Mel2	archive	Coarse-grained two-mica granite
Melechov 4	Mel4	archive	Fine-grained two-mica granite (biotite > muscovite)
Podlesi-Potůčky	PTP5a	archive	Medium-grained albite-topaz-Li mica granite
Pozdatky	VP5	archive	Porphyritic amphibole-biotite melasyenite (durbachite) and medium-grained biotite melagranite
Příbram	MV4	archive	Fine- to medium-grained biotite granodiorite.
Panské Dubénky	PDV1	2010	Fine-grained two-mica granite (Bi > Mu)
Pozďátky	PZV1	2010	Porphyritic amphibole-biotite melasyenite (durbachite) and medium-grained biotite melagranite
Melechov 2	MEV1	2010	Coarse-grained two-mica granite
Ctětín	CTV1	2011	Fine-grained amphibole-biotite granodiorite and migmatized biotite gneiss

where K is the hydraulic conductivity (m s^{-1}), Q is the volumetric flow rate (m^3 s^{-1}), l is the sample length (m), A is the cross section area of the sample (m^2) and h is the pressure difference between the top and bottom of the core (expressed as water head difference in m).

The through-diffusion methodology involves placing a rock sample disc (radius 25 mm,

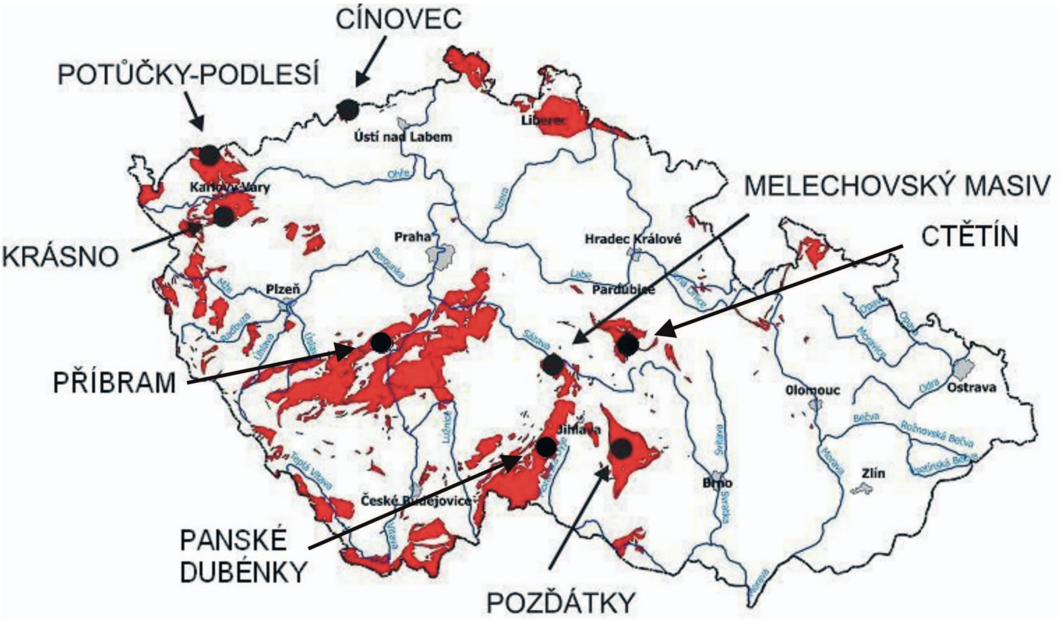

FIG. 1. The location of the sites in the Bohemian Massif where the samples originated. Red identifies granitic bodies (L. Rukavičková, Czech Geological Survey).

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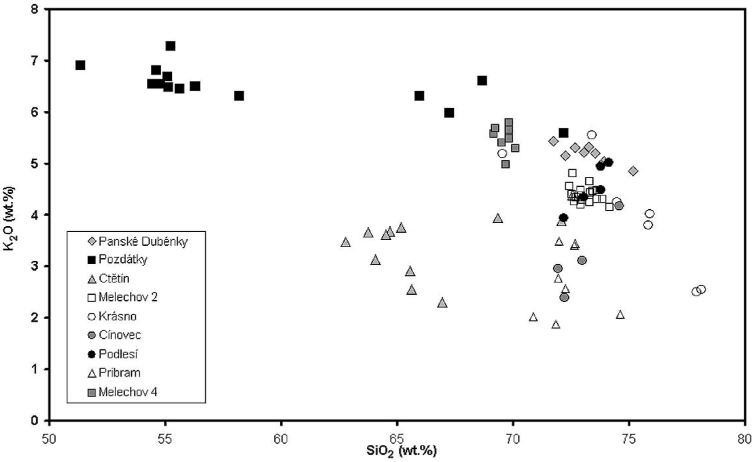

FIG. 2. The composition range of the studied granitoids on a SiO_2 *vs*. K_2O plot. The Si-poor and K-rich points are melagranitoids (durbachites) from the Pozdátky boreholes. The granodiorite from Ctětín is characterized by moderate SiO_2 and K_2O contents (62–66 wt.% and 2.0–3.5 wt.%, respectively). All the other rocks are true granites (68–78 wt.% SiO_2) with different alkali contents.

thickness 10 mm) between a spiked reservoir (a radioactive tracer in synthetic granitic groundwater) and a tracer-free reservoir (synthetic granitic groundwater). Synthetic groundwater was prepared by taking the average groundwater composition of granitic bodies in the Bohemian Massif (for composition see Table 2). The tracer that was used was ^3H ($T_{½}$ = 12.4 years, 1300 Bq l^{-1}). The activities in both input and output reservoirs were regularly measured using liquid scintillation spectrometry (HIDEX 300 SL, Hidex Oy, Finland).

The experimental breakthrough curve was then compared with modelled curves which were calculated using a compartmental diffusion module, based on the *GoldSim* transport code

TABLE 2. Composition of synthetic granitic groundwater (mg l^{-1}), prepared on the basis of the average composition in the Bohemian Massif down to 100 m depth (based on Rukavičková *et al.*, 2009).

	— Concentration (mg l^{-1}) —		
Na	1.8	Cl	42.4
K	10.6	SO_4	27.7
Ca	27	NO_3	6.3
Mg	6.4	HCO_3	30.4
		F	0.2

and a contaminant transport module (Vopálka *et al*., 2006; Havlová and Vopálka, 2010). Examples of diffusion breakthrough curves and the fitting procedure, using the *Goldsim* diffusion module, are shown in Fig. 3. Finally, the effective diffusion coefficient, D_e, values were calculated according to equation 2.

Results

A large set of migration parameters for 45 samples, namely porosity ε, effective diffusion coefficient, D_e, and hydraulic conductivity, K, was collected.

The variation of porosity ε with sample depth in the context of sample origin is shown in Fig. 4. Archive samples, altered archive samples and recent fresh samples are distinguished. Two general trends can be seen: (1) the porosity decreases slightly with sampling depth; (2) the porosity values are generally lower in fresh samples compared to archive material. A statistical analysis of the dataset clearly shows that the fresh samples have lower porosity values and less variance (Table 3). The data for samples from relatively shallow depths (<50 m) is more scattered, most probably due to of weathering. No important mineralogical changes or grain size irregularities are apparent within the first 50 m. The porosity rarely exceeds 1% for both types of samples at depths of more than 50 m (Fig. 4). The

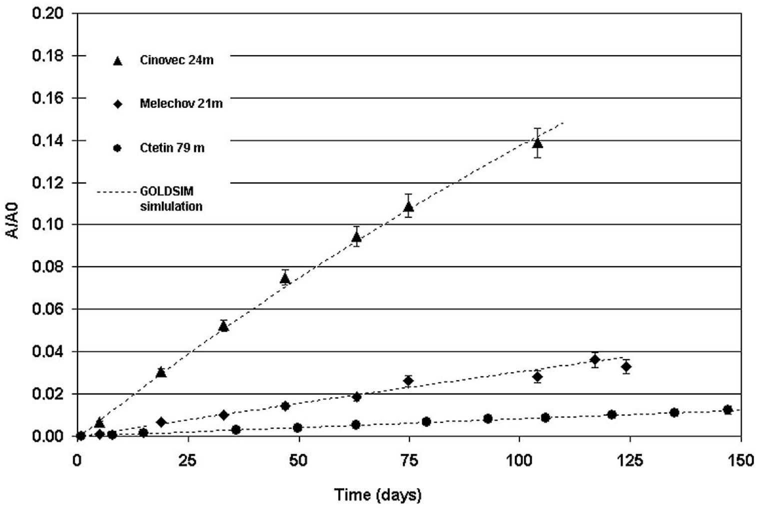

FIG. 3. Examples of diffusion breakthrough curves (points) and fits using the *Goldsim* diffusion module (dashed lines) for 3 samples: ▲ Cinovec site, 24 m depth, altered archive sample; ◆ Melechov, 21 m depth, archive sample, ● Ctětín, 79 m depth, recent (fresh) sample (2011).

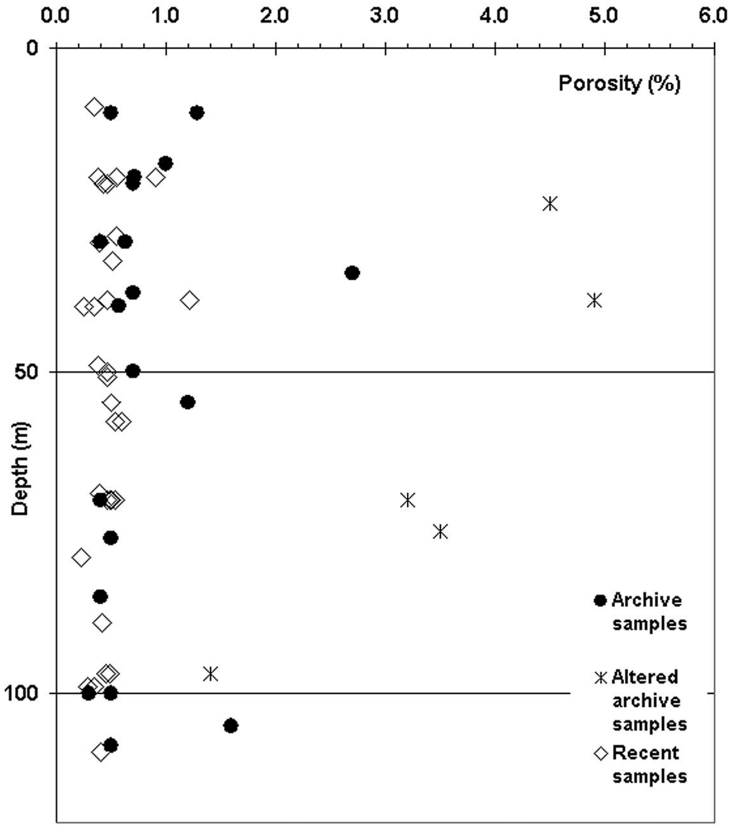

FIG. 4. Dependence of porosity, ε (%) on sampling depth (m) and sample origin.

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TABLE 3. Statistical evaluation of the datasets for porosity ε (%) and effective diffusion coefficient D_e ($m^2\ s^{-1}$) for the archive and fresh (recent) samples.

	Archive samples			Fresh (recent) samples		
	Porosity (%)	$D_e \times 10^{-12}$ ($m^2\ s^{-1}$)	$K \times 10^{-12}$ ($m\ s^{-1}$)	Porosity	$D_e \times 10^{-12}$ ($m^2\ s^{-1}$)	$K \times 10^{-12}$ ($m\ s^{-1}$)
Number of analyses	24	24	23	21	21	21
Minimum	0.3	0.65	0.07	0.23	0.4	0.01
Maximum	2.7	7.41	160	1.22	3.64	17
Mean	0.73	2.3	13	0.49	1.59	4
Median	0.73	1.62	2.4	0.49	1.36	1.8
Standard Deviation	0.53	1.68	33	0.22	0.89	5.4

altered samples of greisenized granite were excluded from the statistical analysis as they had porosities that were almost an order of magnitude greater than the unaltered samples.

A similar trend can be seen in the effective diffusivity, D_e, (Fig. 5). The altered samples have D_e values that are an order of magnitude higher than the unaltered ones. The D_e values show a

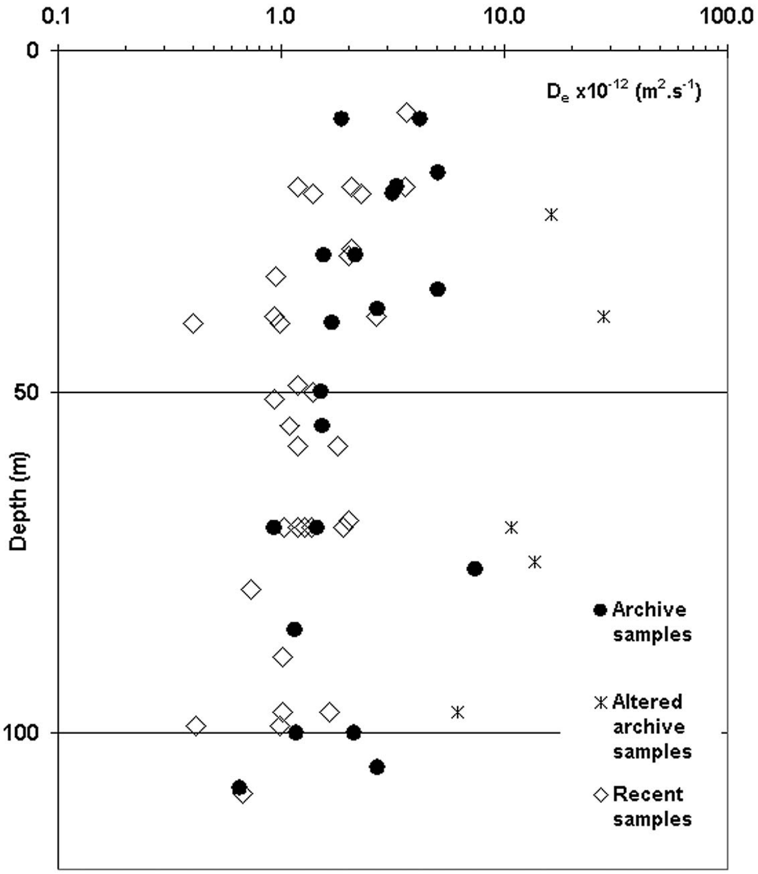

FIG. 5. Dependence of effective diffusion coefficient, D_e ($m^2\ s^{-1}$) on sampling depth (m) and sample origin.

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slight general reduction with increasing depth for both sample groups and the D_e values for the fresh samples have a lower mean value (Table 3). The D_e values of the archive samples are more scattered at shallow depths (<50 m), but otherwise they have comparable D_e values (Table 3).

Hydraulic conductivity, K, is usually defined as a measure how easily water can move through a porous space in the presence of a hydraulic gradient. It is a parameter usually applied to advective transport, however, in common with diffusion, it is related to pore space parameters (porosity, tortuosity, constrictivity) in porous media. Even though it might seem that diffusivity and hydraulic conductivity should be directly related, there is no clear relationship in the reported data. Figure 6 shows a generally decreasing trend with larger scatter for the archive samples (see also Table 3). However, extraordinary samples with high D_e and low K values and samples in which these parameters are reversed are present in the dataset (Fig. 6). No dependency of K on sampling depth was found.

Discussion

Comparing the chemical compositions and sample migration parameters (porosity, ε, effective diffusion coefficient, D_e and hydraulic conductivity, K), no straight dependency for any of the elements and any of the parameters was found. The porosity, ε, of deep crystalline rock is typically between 0.5–1.0%, although values <0.1% have been reported (Ohlssons, 2000; Vilks *et al.*, 2003). The rock in the vicinity of water-bearing fissures may be altered, resulting

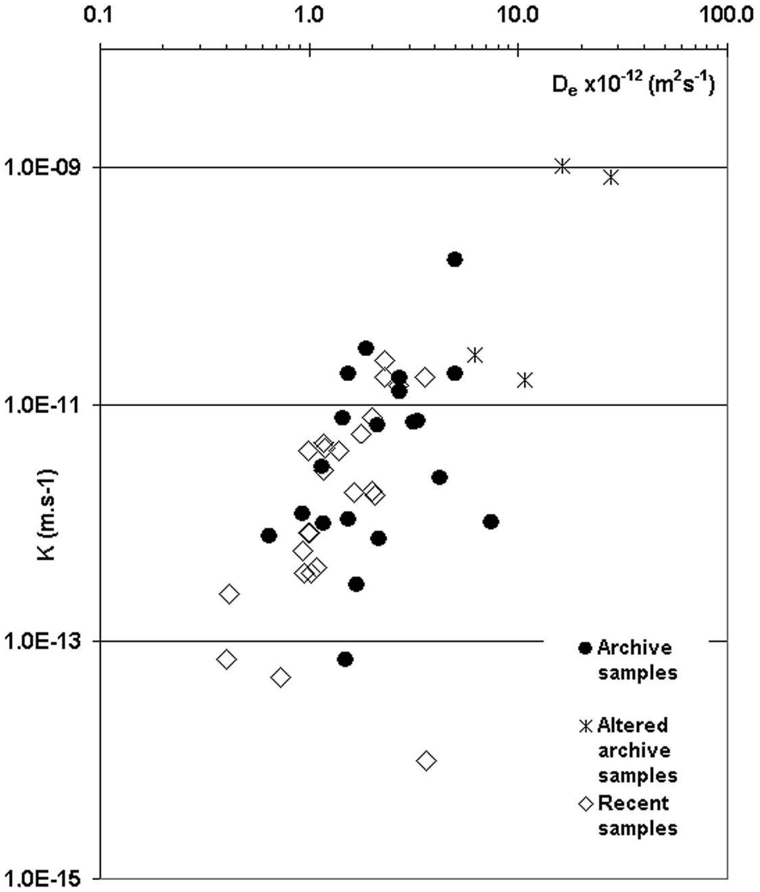

FIG. 6. Relation between the effective diffusion coefficient, D_e (m^2 s^{-1}) and hydraulic conductivity, K (m s^{-1}) and sample origin.

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both in increased porosity and mineralogical changes (cementation or new pore network formation).

It is clear that the determined porosity and diffusivity values for the granitic samples lie within certain value ranges. Based on statistical evaluation, the ranges are comparable with values from the depths that are being considered for a deep geological repository (400–600 m, Bradbury and Green, 1986; Selnert *et al.*, 2008; Valkiainen *et al.*, 1996; Vilks *et al.*, 2003). In this dataset, 91% of the porosity values for the granitic samples fall into the range 0.3–1.0%. Moreover, 80% of D_e values fall into the range $0.4–3.0 \times 10^{-12}$ m^2 s^{-1}.

As there is a lack of samples from deep horizons in the Czech Republic, the values can be compared with granitic samples from Grimsel test site in Switzerland, (400 m below surface, borehole LTD 06.10). These core samples were characterized using the same procedures as ours in the same laboratory. At the time of the experiment the samples had been stored for three years. The samples of Grimsel granodiorite had porosity values in the range 0.45–0.65% and D_e values in the range $3.5–10 \times 10^{-12}$ m^2 s^{-1} (V. Havlová, unpublished data). Moreover, even though of different origin, samples of Äspö diorite and fine-grained granite from 220 m depth revealed similar values on the lower value limit (porosities, ε, 0.5%, 0.3%, respectively; D_e in the range of $10^{-14}–10^{-13}$ m^2 s^{-1}; Byegard *et al.*, 1998).

Porosity and other rock properties can also change with depth: increasing geostatic pressure can result in a reduction in pore space and change in geometry, which also results in a decreasing diffusivity, D_e (Bradbury and Green, 1986). One might expect the same relation for hydraulic conductivity, K. After removal the rock cores are prone to stress release and an increase in pore space due to the reduction in the surrounding hydrostatic pressure (Bradbury and Green, 1986; Vilks *et al.*, 2003). The increase in pore space may continue with time due to alteration processes (Vilks *et al.*, 2003).

Both of the processes described above seem to be reflected in the results. The porosity, ε, and diffusivity, D_e, generally decreased with sample depth. The statistical analyses (Table 3), show that the fresh sample group has lower value ranges in comparison with the archive sample group and this could be due to the increase in pore space with time. As the time interval between sampling and the laboratory investigation of new boreholes was not long (several months), further relaxation and rock alteration can still occur. Vilks *et al.* (2003) determined a 50% increase of sample porosity after two years of storage.

The metasomatically altered samples from Cínovec had properties very different to those of the unaltered granitic samples. They are more comparable with sedimentary rock (porosity up to 7%, diffusivity 2.7×10^{-11} m^2 s^{-1}).

Although there was some limited correlation between hydraulic conductivity K and diffusivity D_e (Fig. 6), some samples had high D_e and low K values whereas others had high K and low D_e values. Possible explanations for this are as follows: (1) there may be an experimental artefact due to the different experimental set up for the two types of experiments; (2) there may be different types of pore that influence the diffusion and permeability in a different way. The first type, through-transport pores (Lever *et al.*, 1985), being interconnected, would influence mostly flow through the sample, i.e. hydraulic conductivity values. Moreover, the pore size may play an important role as well: hydraulic conductivity should logically be more influenced by the larger aperture pores. Small aperture pores and dead-end pores, i.e. no flow-through pores (Lever *et al.*, 1985), can influence tracer diffusive migration. If the dead-end pores are long, they may open during sample preparation and this can contribute to the flow-through porosity. If they are short and have space for tracer entrance, steady state diffusion would establish more slowly than expected on the basis of the K values. The different sample lengths for the through-diffusion experiments and permeability measurements (10 mm and 50 mm, respectively) may provide a potential explanation for the discrepancies. However, the characterization of pore apertures and the dead-end pore length distribution in samples is complicated (Lever *et al.*, 1985).

Sample relaxation following withdrawal of the cores should increase pore space and this in turn should lead to an increase in hydraulic conductivity. Therefore, the K values should be higher for the archive samples due to the increase in available porosity. This can be shown to be true by a statistical analysis of the K values (Table 3). The archive samples have higher K values in comparison with the fresh samples from recently drilled boreholes. Further studies to clarify the precise mechanisms are continuing.

The formation factor, F_f, can be also used as an operational geometrical parameter that is only

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dependent on the geometry of the micropore network and is independent of diffusing species properties (Löfgren, 2004). It can be determined using experimental effective diffusivity, D_e, data as follows:

$$F_f = \varepsilon \frac{\delta_D}{\tau^2} = \frac{D_e}{D_w} \quad (4)$$

where F_f is the formation factor, ε is the porosity, d_D is the constrictivity, τ^2 the tortuosity, D_w is the diffusivity in unconfined water (tabulated, $D_w(^3H)$ = 2.42×10^{-9} m^2 s^{-1}, Byegard *et al*., 1998) and D_e is the effective diffusivity, determined in through-diffusion experiments.

The formation factor is a function of porosity $F_f = k(\varepsilon)$. In Fig. 7, formation factors, F_f, for all granites studied are plotted *vs*. porosity, ε. Most of the data lie near a line defined by Archie's law (Parkhomenko, 1967):

$$F_f = a\varepsilon^m \quad (5)$$

where ε is the porosity, and *a* and *m* are empirical parameters. Values of *a* = 0.71 and *m* = 1.58 are found to be suitable for crystalline rock with less than 4% porosity.

The sample data reported herein are consistent with the results of Löfgren (2004) for granite rock samples from the Äspo underground repository. Neither the increased feldspar content of feldspars in the Pozďátky samples, nor the moderate feldspar content in the Ctětín samples (Fig. 2) had any influence on the sample position with respect to Archie's line. This contrasts with analyses reported by Ohlssons (2000) in which low feldspar content samples were separate from

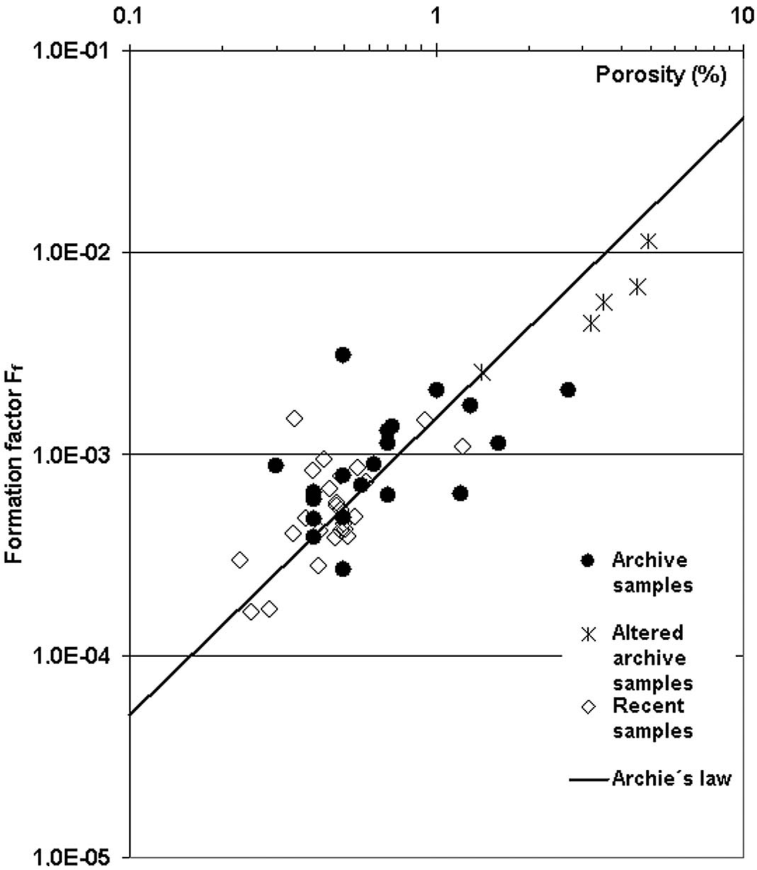

FIG. 7. Relation between the formation factor, F_f and porosity, ε in the context of sample origin. The line represents the empirical Archie's law.

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the main granitic rock group. An examination of Archie's plot shows that the samples from depths between 10 and 108 m exhibit similar pattern as samples from deeper intervals as in Ohlsson (2000) and Löfgren (2004). That forms the last argument enabling us to recommend that the obtained dataset (ε, D_e, F_f, K) can be used as an approximation of the ranges for crystalline rock migration parameters from deeper rock horizons. These data can therefore be used in a safety assessment calculations if there is a lack of data from deeper intervals.

Conclusions

An extensive porosity, ε, effective diffusivity, D_e, formation factor, F_f and hydraulic conductivity, K, dataset for 45 samples of different granitic rocks from the Bohemian Massif was collected. Our analyses show that samples originating from shallow depths (up to 108 m) can be used to define the data ranges for safety assessment of a deep (>400 m) geological repository. However, alteration can dramatically change sample properties.

Differences in the data ranges for archive and fresh samples is probably a result of sample ageing processes which include stress relaxation after bore core removal, pore space opening and possible alteration of samples during storage. Such processes can lead to an overestimation of migration parameters in comparison with *in situ* conditions if samples have been stored for long period. Further research is required to clarify the precise mechanisms.

Further research activities will continue within the frame of the project, and will focus on more detailed characterization of the parameters influencing tracer migration in granitic rock matrix (such as chemical and mineral analyses and pore space visualization).

Acknowledgements

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