

## Tracer tests in fractured rocks with a new fluorescent dye—pyrene-1,3,6,8-tetra sulphonic acid (PTS)

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**Abstract** Two multi-tracer tests were performed in fissured rocks accessible in underground laboratories to examine a new fluorescent dye: pyrene-1,3,6,8-tetra sulphonic acid (PTS). The first test was carried out at the Lindau Rock Laboratory (LRL), Germany, in a highly permeable ore dike, and the second, at the Grimsel Test Site (GTS), Switzerland, in a heterogeneous granite fault zone (AU 126). At the LRL new tracer was injected together with uranine in a convergent flow field (monopole test), and slightly different tracer breakthrough curves were observed according to different diffusion coefficients of both tracers. The matrix porosity calculated with the aid of the one-dimensional (1-D) single-fissure dispersion model (SFDM) agrees well with that found in earlier tracer tests and with measurements performed on core samples. At the GTS, the PTS tracer was applied together with pyranine in two-well injection–withdrawal (dipole) tests. Both tracers yielded identical tracer concentration curves, which confirm their conservative behaviour. Mathematical simulations performed with the aid of a 3-D numerical model (FRAC3DVS) yielded equally good fits for different sets of parameters, independent of whether matrix porosity was included or neglected. That lack of unique solution and the difficulty in observing the influence of matrix diffusion result from a wide distribution of the transit times of particular streamlines, which is characteristic for injection–withdrawal tests. However, both tracer tests clearly indicated that the new tracer (PTS) behaves conservatively at high pH values and can be successfully used for groundwater labelling.

**Key words** fractured rock; monopole and dipole tracer tests; new fluorescent dye; transport modelling; matrix diffusion

### Tests de traçage en roches fracturées avec un nouveau produit fluorescent—l'acide pyrène 1.3.6.8 tétra sulfonique (PTS)

**Résumé** Deux tests multi-traceurs ont été effectués en milieux de roches fracturées, dans le but de tester un nouveau produit fluorescent: l'acide pyrène 1.3.6.8 tétra sulfonique (PTS). Le premier test a été opéré au laboratoire Lindau Rock (LRL), en Allemagne, dans un filon minéralisé très perméable, tandis que le deuxième test a été effectué sur le site de Grimsel (STS), en Suisse, dans une zone de failles de granites hétérogènes (AU 126). Au LRL, le nouveau traceur a été injecté, mélangé à de l'uranine, dans les conditions d'un régime d'écoulement convergent (test monopôle), et des courbes d'arrivée légèrement différentes ont été identifiées, en fonction des différents coefficients de diffusion des deux traceurs. La porosité de matrice calculée à l'aide d'un modèle 1D de dispersion à travers une seule fracture (SFDM) est en conformité avec les résultats obtenus par des tests de traçage préalables et par des mesures réalisées sur des carottes. Au GTS, le traceur PTS a été injecté, mélangé à de la pyranine, au niveau de deux forages avec injection et pompage (tests dipôles). Les deux traceurs révèlent des courbes de concentrations identiques, confirmant ainsi leur comportement conservatif. Les simulations mathématiques réalisées avec un modèle numérique 3D (FRAC3DVS) donnent des résultats de qualités égales avec différents jeux de paramètres, indépendamment de la prise en compte ou non de la porosité de matrice. Cette non-unicité de la solution et la difficulté d'observer l'influence de la matrice de diffusion proviennent d'une grande distribution des temps de transit selon des directions d'écoulement particulières, ce qui est caractéristique des tests par injection-pompage. Cependant les deux tests de traçage indiquent clairement que le nouveau traceur (PTS) se comporte de manière conservative pour de fortes valeurs de pH et peut être utilisé avec succès pour la caractérisation des eaux souterraines.

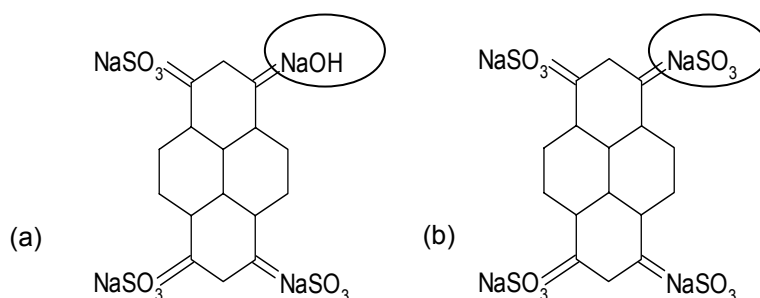
**Mots clefs** roches fracturées; tests monopôle et dipôle de traçage; nouveau produit fluorescent; modélisation de transport; matrice de diffusion

## INTRODUCTION

A number of fluorescent tracers, such as uranine, eosine, pyranine and rhodamines, have been used to estimate transport parameters (velocity, dispersivity, water storage capacity, etc.) in porous media and karst channels (e.g. Seiler *et al.*, 1989; Abelin *et al.*, 1991; Behrens *et al.*, 1997; Ptak, 1997), or in fissured rocks (e.g. Neretnieks *et al.*, 1982; Novakowski *et al.*, 1985; Maloszewski & Zuber, 1993; Himmelsbach *et al.*, 1998; Hoehn *et al.*, 1998; Reimus *et al.*, 1999; Meigs *et al.*, 2001; Sanford *et al.*, 2002; Witthüser *et al.*, 2003; Mazurek *et al.*, 2003).

For the application of fluorescent tracers, several conditions should be satisfied: low detection limit and high solubility of the tracer, no toxicity, a simple analysis method, no or low sorption behaviour under different pH values, no microbial degradation, and different wavelength spectra (EMW) for multi-tracer experiments. These conditions cannot be well satisfied simultaneously for known tracers. Therefore, it is always of interest to search for new tracers, which could better satisfy the above requirements. In particular, there is a need for a tracer, which could be applied under different pH conditions and which has different fluorescence emission spectra from those of other commonly used dye tracers (e.g. uranine, eosine, pyranine), so that more dye tracers can be detected simultaneously. For instance, Leibundgut & Wernli (1988) proposed naphthionate for groundwater studies while Hadi *et al.* (1996) have tested succinyluranine disodium salt and 5(6)-carboxyuranine trisodium salt. In the present study the pyrene-1,3,6,8-tetra sulphonic acid (PTS) was tested as a new groundwater tracer. In comparison to pyranine, PTS has four sulphonic acid functional groups (Fig. 1) and is not toxic. Its fluorescence spectrum is stable for pH values between 2 and 11, and can be detected online in the presence of pyranine or uranine without pH separation (EMW<sub>PTS</sub>: 403 [nm], EMW<sub>uranine</sub>: 512 [nm], EMW<sub>pyranine</sub>: 512 [nm]). In comparison to the commonly used uranine, the PTS tracer has a very low degradation in the presence of light and in column experiments in tertiary sand and quaternary gravel it shows significantly lower sorption at lower pH values (<pH 7) than uranine (Einsiedl *et al.*, 1999). Generally, fluorescent tracers show different transport behaviour depending on the pH value (Smart & Laidlaw, 1977; Einsiedl, 1999). However, at pH values above 9, all used fluorescent dyes should have an anionic character and in consequence low or non-reacting transport behaviour. At the Lindau Rock Laboratory (LRL), the fracture water shows a high pH solution of 10.2 and at the Grimsel Test Site (GTS) in a heterogeneous granite fault zone (AU126), pH values of 8.5–9.3 were measured.

The detection limit for PTS is 0.05 µg l<sup>-1</sup> under laboratory conditions and about 1 µg l<sup>-1</sup> for an online system in the field. After successful tests of PTS in flow-through laboratory columns (Einsiedl, 1999), the present study focuses on the examination of PTS transport behaviour at the field scale at two test sites. The first study was performed at the Lindau Rock Laboratory (LRL), southern Black Forest, Germany, where a 600-m long observation tunnel was excavated to investigate the hydraulic effect of a highly permeable fault zone (ore dike) at a planned dam site (Himmelsbach *et al.*, 1998). The second test was carried out at the Grimsel Test Site (GTS), which is the NAGRA (National Cooperative for the Disposal of Radioactive Waste) underground rock laboratory in the Swiss Alps (Haslital, Berner Oberland). NAGRA has been operating the GTS laboratory over the last 20 years to develop concepts of secure radioactive waste disposal (Frick *et al.*, 1992; Smith *et al.*, 2001).



**Fig. 1** Chemical structure of (a) pyranine and (b) the new fluorescent tracer pyrene-1,3,6,8-tetra sulphonic acid (PTS).

The field tests had two main goals: (a) the examination of the PTS-transport behaviour in the fissured aquifers in comparison to pyranine and uranine, which are regarded by Smart & Laidlaw (1977) as non-reacting dyes under favourable conditions in porous aquifers; and (b) the estimation of rock parameters at the LRL ore dike and the fault-zone AU 126 of the GTS. Rock parameters found at the LRL were compared to those previously obtained by Himmelsbach *et al.* (1998) who used deuterium, pyranine and uranine.

### Experimental site and test design at the LRL

The Lindau Rock Laboratory (LRL) is situated in the Albtal granite pluton with a transmissivity of  $2.5 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$  and matrix porosity of more than 6%, which represents a dense fracture system. The ore dike is nearly vertical with a thickness of 0.3–3.0 m. The tunnel system of the rock laboratory is about 90 m below the ground surface. Details on the test site and the experimental set up for the LRL can be found in Himmelsbach *et al.* (1998). In the LRL a tracer experiment was performed under a radial convergent flow field (monopole test). The distance between the injection (BL8) and detection boreholes (BL10) is 11.2 m. An instantaneous injection of the tracer cocktail containing uranine and PTS was performed. According to the results of Himmelsbach *et al.* (1998), the thickness of the ore dike is approximately 2 m, while the hydraulic conductivity is  $2.4 \times 10^{-5} \text{ m s}^{-1}$  ( $2.07 \text{ m day}^{-1}$ ). A constant hydraulic head was adjusted by opening borehole BL10, which led to a constant flow rate of  $0.23 \text{ l s}^{-1}$ . In comparison to previous tracer tests, the flow rate was two times larger than that of the monopole tests ( $0.1 \text{ l s}^{-1}$ ) but close to the dipole experiments ( $0.2\text{--}0.25 \text{ l s}^{-1}$ ) of Himmelsbach *et al.* (1998). A pH value of approximately 10.2 and an electrical conductivity of  $77 \text{ } \mu\text{S cm}^{-1}$  were observed during the present tracer test. It is unclear if the dissolution of concrete at the tunnel wall, and/or mineral reactions, led to that high pH value. For such a pH value, uranine has an anionic character (Behrens, 1988), and should exhibit a non-reactive transport behaviour.

### Experimental site and test design at the GTS

The GTS is located at a depth of 450 m in the crystalline rock of the Aare Massif. The rock massif consists mainly of pre-Hercynian gneisses and Hercynian granites. The Hyperalkaline Plume in Fractured Rocks (HPF) experiment site is set up in a conducting

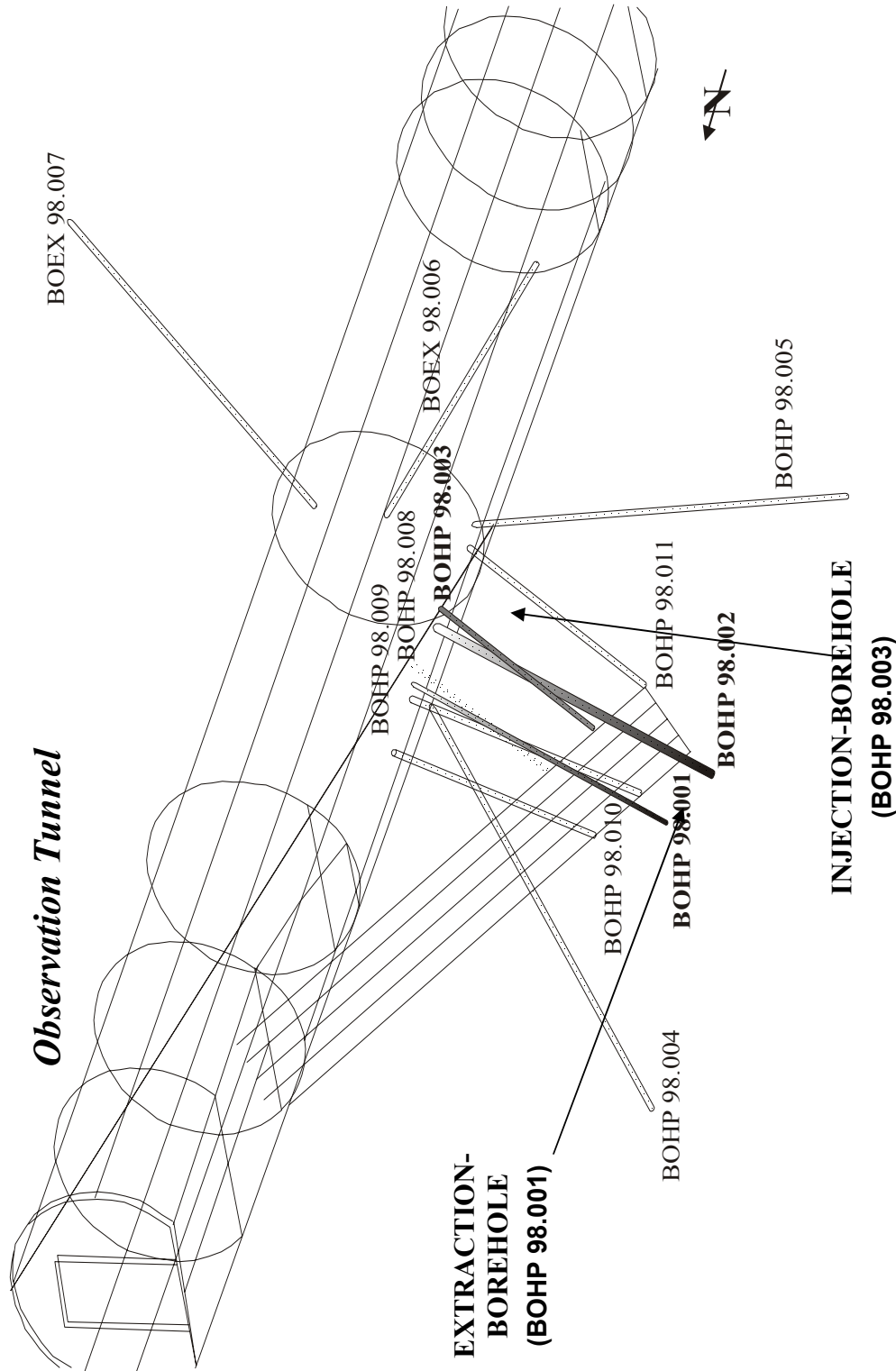


Fig. 2 Observation tunnel at the GTS test site (adapted from NAGRA).

conducting shear zone (AU 126) of the granodiorite of the Aare Massive. The major fault zones (AU 96, AU 126) have a regional dip of 70–90° and a principal NE–SW strike (Frick *et al.*, 1992). Ten boreholes were drilled into the HPF shear zone AU 126 and the granite rock to investigate their hydraulic properties (Bossart *et al.*, 1991; Meyer *et al.*, 1989). Figure 2 presents the GTS tunnel with all observation wells. Due to the low permeability in the HPF shear zone, a withdrawal experiment could not be performed by pumping, but only by free outflow (Pfingsten & Soler, 2003) or by water injection–withdrawal test with the same flow rates (about 60 ml h<sup>-1</sup>). In such a case a symmetrical dipole flow field can be assumed in a homogeneous flow system when the natural gradient is close to zero. Generally, dipole tracer tests should not be recommended for double-porosity systems because they yield wide distributions of streamlines with flow times going theoretically to infinity. Then, even for an instantaneous injection of tracer, the resulting breakthrough curve is characterized by a strong tailing effect (“hydraulic tail”) and significantly lower tracer recovery (Himmelsbach *et al.*, 1998). In addition, other studies, for example by Kunstmann *et al.* (1997) or Becker & Shapiro (2000), have shown that the hydrodynamic dispersion in a single fracture and/or the transverse dispersivity may also generate strong tailing effects. As a consequence, both the “hydraulic tail” and “dispersive tail”, may mask the tailing effect resulting from the matrix diffusion and make the interpretation of tracer curves very ambiguous.

A triple packer system, recommended by Novakowski (1992) and Bäumlé *et al.* (2001) was used for the injection (BOHP 98.003)–extraction (BOHP 98.001) system. At the test site, the fault zone has a thickness of 0.45–0.55 m, a fracture permeability of about  $1.5 \times 10^{-8}$  m s<sup>-1</sup> and a hydraulic conductivity of the rock matrix of about  $9.7 \times 10^{-11}$  m s<sup>-1</sup> (Fisch, 1999). The matrix porosity of the granite and mylonite has an average value of about 0.1–2%, whereas the dominant openings (filled with fault gouge, i.e. coarse rock components) have porosities even larger than 20% (Pfingsten & Soler 2003). Borehole imaging performed in the shear zone AU 126 showed fractures with apertures of 1 mm or more (Pfingsten & Soler, 2003).

The tracer test was performed over a distance of 0.77 m. The groundwater from the fault zone AU 126 is relatively poorly mineralised with an ionic strength of about 1 mM. During the present experiment the pH value ranged from 8.5 to 9.3. More details on pH values in the shear zone AU 126 can be found in Pfingsten & Soler (2003).

The injection system consisted of two tanks, one with the tracer cocktail and the second with local fault-zone water from AU 96 having identical chemical composition to water in the shear zone AU 126. To obtain steady-state hydraulic conditions, local freshwater was injected for four days into the injection borehole with a constant flow rate ( $Q_{inj}$ ) of 66 ml h<sup>-1</sup>. The withdrawal was obtained by opening the borehole BOHP 98.001 and adjusting a free outflow ( $Q_{out}$ ) of 58 ml h<sup>-1</sup>. After reaching steady-state flow conditions, the tracer cocktail, consisting of pyranine and PTS was injected as a step-pulse (top hat) during 27 h with the same injection flow rate. After the end of tracer injection, the local groundwater was injected further with the same  $Q_{inj}$  to keep hydraulic conditions at a steady state. During the tracer test, quasi-constant pressure of about 195 kPa and 98 kPa were observed at the injection and observation boreholes, respectively.

## RESULTS AND DISCUSSION

### LRL experiment

The tracer experiment performed at the LRL in a radial-convergent flow field was interpreted using the Single Fissure Dispersion Model (SFDM). This model, developed by Maloszewski & Zuber (1985), combines the convective-dispersive equation for tracer flow in a single fracture with diffusion processes of the tracer into an adjacent infinitely extended rock matrix. The assumption of the infinite matrix is well satisfied for short-term tests in densely fractured media because then the diffusion of tracer is not influenced by the presence of adjacent fractures. Then, the solution to the transport equation for a fixed distance reads (Maloszewski & Zuber, 1985, 1990):

$$C_f(t) = \frac{aM}{2\pi Q} \int_0^t \sqrt{\frac{t_0}{P_D}} \exp\left[-\frac{(t_0-u)^2}{4uP_D t_0} - \frac{a^2 u^2}{t-u}\right] \frac{du}{\sqrt{u(t-u)^3}} \quad (1)$$

where  $C_f$  is the flux-averaged tracer concentration observed in the outflow;  $M$  is the injected mass of the tracer;  $Q$  is the volumetric flow rate through the system (pumping rate);  $t_0$  is the mean transit time of water;  $P_D$  is the dispersion parameter;  $a$  is the diffusion parameter describing the matrix diffusion process; and  $u$  is the integration variable (varies between 0 and  $t$ ).

This model has three fitting parameters:

$$t_0 = x/v \text{ or } V/Q \quad (2a)$$

$$P_D = D/(vx) = \alpha_L/x \quad (2b)$$

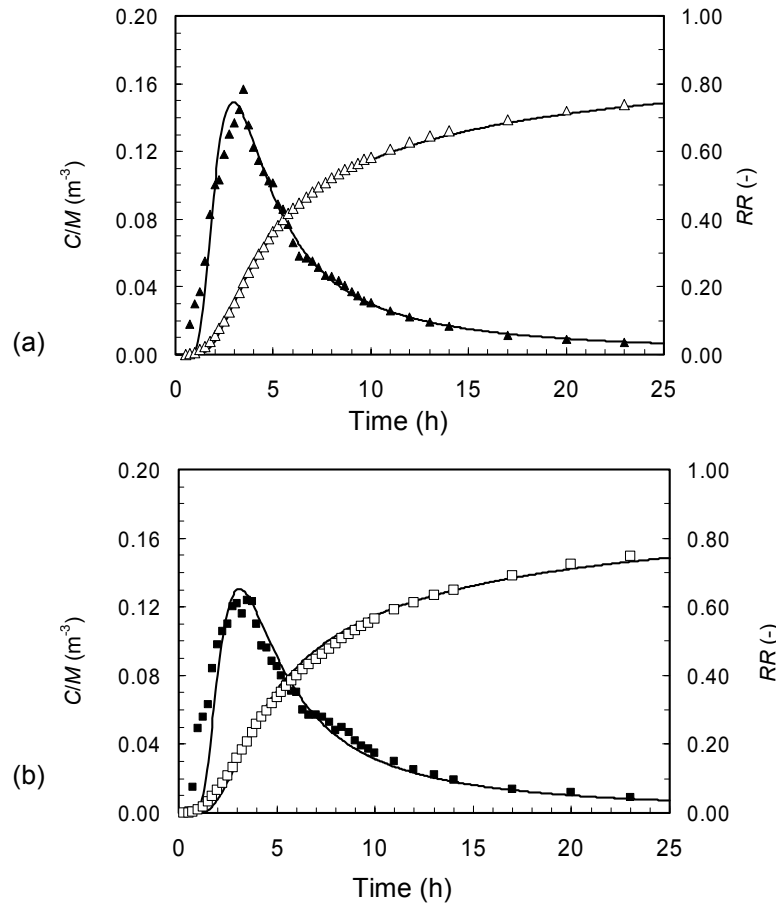
$$a = n_p(D_p)^{1/2}/(2b) \quad (2c)$$

where  $x$  is the distance between the wells;  $v$  is the average water velocity;  $V$  is the volume of mobile water in the system;  $D$  is the dispersion coefficient;  $\alpha_L$  is the dispersivity ( $\alpha_L = D/v$ );  $n_p$  is the matrix porosity;  $2b$  is the fracture aperture; and  $D_p$  is the effective diffusion coefficient of the tracer in water in the matrix ( $D_p = D_m/\tau_p$ ), where  $D_m$  is the molecular diffusion coefficient of the tracer in water and  $\tau_p$  is the tortuosity factor of the matrix (assumed as equal to 1.5).

In the case of monopole or dipole tests, it is important to include in the calibration procedure the concept of a relative mass recovery and to fit tracer concentration and recovery curves simultaneously (Klotz *et al.*, 1988; Maloszewski & Zuber, 1990). The relative tracer recovery rate ( $RR$ ) is calculated from the experimental and theoretical tracer concentration curves as a function of time:

$$RR(t) = Q \int_0^t C_f(t) dt / M \quad (3)$$

The experimental data and the best-fit curves for both concentration and the recovery of uranine and PTS are shown in Fig. 3. The shape of both curves clearly suggests the existence of tracer diffusion into porous rock matrix. The concentration curve of PTS has a slightly lower peak with  $(C/M)_{\max} = 0.125/0.155$  ( $1 \text{ m}^{-3}$ ), and a slightly longer tail in comparison to uranine, due to a higher molecular diffusion coefficient than that of uranine. Therefore, it diffuses faster and deeper into the porous rock matrix than



**Fig. 3** Modelled curves (solid lines) obtained as a best-fit to (a) observed uranine normalized concentrations (▲) and relative recoveries (Δ) and (b) PTS normalized concentrations (■) and relative recoveries (□).

uranine. The values of fitting parameters support the existence of matrix diffusion. Both tracers yield the same values of transit time and dispersivity:  $t_0 = 2.4$  h and  $\alpha_L = 0.56$  m, while the values of  $a$  parameter are  $7.6 \times 10^{-3} \text{ s}^{-1/2}$  and  $8.5 \times 10^{-3} \text{ s}^{-1/2}$  for uranine and PTS, respectively.

For a radial-convergent flow field, the fracture porosity (defined as  $n_f = 2b/L$ , where  $L$  is the fissure spacing) can be calculated from (e.g. Zuber, 1974):

$$n_f = (Q \cdot t_0) / (\pi \cdot m \cdot x^2) \quad (4)$$

where  $m$  is here the thickness of the ore dike, whereas the fracture aperture,  $2b$  ( $\mu\text{m}$ ) based on the cubic law is:

$$2b = 4.29 \tau_f (K/n_f)^{0.5} \quad (5)$$

where  $K$  is the hydraulic conductivity expressed in  $\text{m day}^{-1}$  and  $\tau_f$  is the fracture tortuosity.

Using a thickness of the ore dike of 2 m, the fracture porosity of 0.24% was obtained from equation (4). The obtained value of  $n_f$ , together with the hydraulic conductivity of  $2.07 \text{ m day}^{-1}$ , and the fracture tortuosity of 1.5, were used in equation (5) to calculate the fracture aperture of  $188 \mu\text{m}$ . The molecular diffusion coefficient for uranine is equal to  $4.5 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$  (Skagius & Neretnieks, 1986). For PTS, the

**Table 1** Model and rock parameters found from the tracer test performed at the LRL.

Tracer	Model parameters:			Rock parameters:			
	$t_0$ (h)	$P_D$ (-)	$a$ (s <sup>-0.5</sup> )	$\alpha_L$ (m)	$2b$ ( $\mu$ m)	$n_p$ (%)	$n_f$ (%)
Uranine	2.4	0.05	$7.6 \times 10^{-3}$	0.56	188	8.1	0.24
PTS	2.4	0.05	$8.5 \times 10^{-3}$	0.56	188	5.0	0.24

$t_0$ : mean transit time of water;  $P_D$ : dispersion parameter;  $a$ : diffusion parameter;  $\alpha_L$ : longitudinal dispersivity;  $2b$ : fracture aperture;  $n_p$ : matrix porosity;  $n_f$ : fracture porosity

molecular diffusion coefficient of pyranine  $D_m = 1.5 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$  (Himmelsbach *et al.*, 1998) was assumed, because both tracers have nearly the same chemical structure (see Fig. 1). Finally, using the values of  $a$ , fracture aperture ( $2b$ ) and diffusion coefficients ( $D_p = D_m/1.5$ ), the matrix porosity ( $n_p$ ) was calculated by applying equation (2c). The matrix porosity found from the PTS and uranine data was equal to 5.0 and 8.1%, respectively. The modelling results of LRL test are summarized in Table 1.

The present values of fracture porosity (0.24%) and dispersivity (0.56 m) are approximately two times larger than those obtained in previous monopole experiments performed between the wells (Himmelsbach *et al.*, 1998), whereas the fracture aperture shows a similar value. The pumping rate ( $Q$ ) applied in the recent study was two times higher than that in the previous monopole tests. The higher pumping rate mobilized the larger water volume (more fractures with mobile water are activated) which perhaps results in greater fracture porosity and dispersivity. This finding is supported by similarly higher fracture porosity obtained by Himmelsbach *et al.* (1998) in dipole tests which were carried out with flow rates close to those used in the present test.

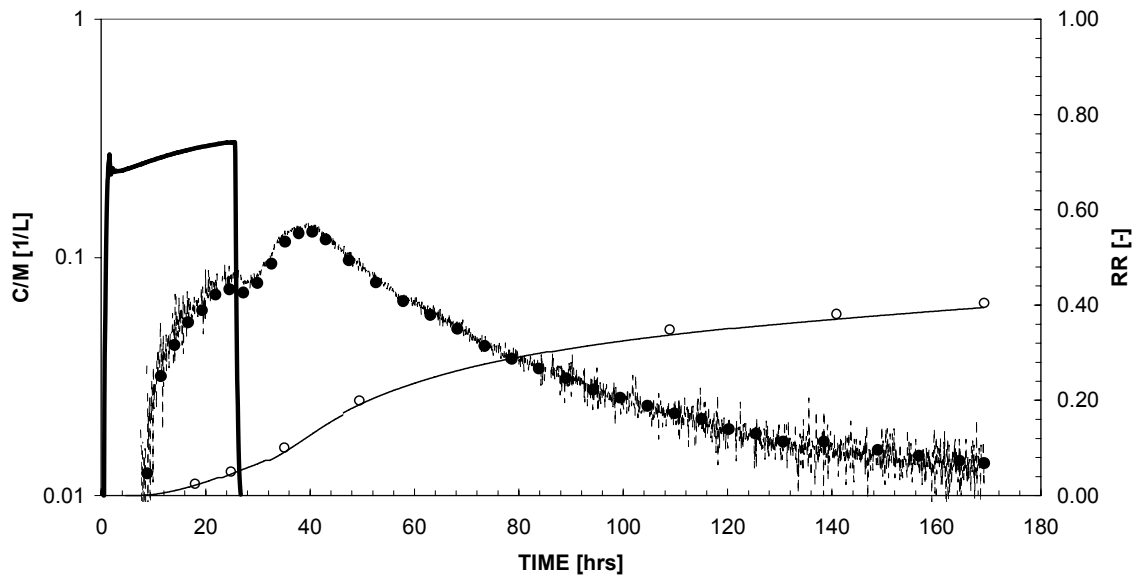
The matrix porosity found from the present test with PTS agrees with the results obtained earlier using eosine, uranine, pyranine and deuterium, and with the mercury porosimetry data (less than 3% for the granitic to more than 6% for the hydrothermally altered rocks) reported for the LRL test site (Himmelsbach *et al.*, 1998). However, matrix porosity obtained from the uranine breakthrough curve within this study is about 1.5 times higher than that measured on the core samples. Uranine seems to be reactive in the micro-porous matrix, which leads to a larger value of  $a$  parameter and to the overestimation of the matrix porosity, if the matrix sorption is not taken into account (see e.g. Maloszewski *et al.*, 1999).

## GTS experiment

The concentrations of pyranine and PTS were measured online in the injection (BOHP 98.003) and extraction (BOHP 98.001) boreholes as a result of a step-pulse injection (see Fig. 4). The largest concentrations of 0.46 and 4.6 mg l<sup>-1</sup> for pyranine and PTS, respectively, were reached in the injection borehole after 1.5 h, and next the fluorescence signal was constant for the injection period of 24 h. After that, the tracer injection was stopped, and the original background fluorescence in the injection borehole was reached after 1.5 h. The complete duration of tracer injection was approximately 27 h (Fig. 4). The injected masses ( $M$ ) of pyranine and PTS were calculated from:

$$M = Q_{\text{inj}} \int_0^{t_{\text{inj}}} C_{\text{inj}}(t) dt \quad (6)$$





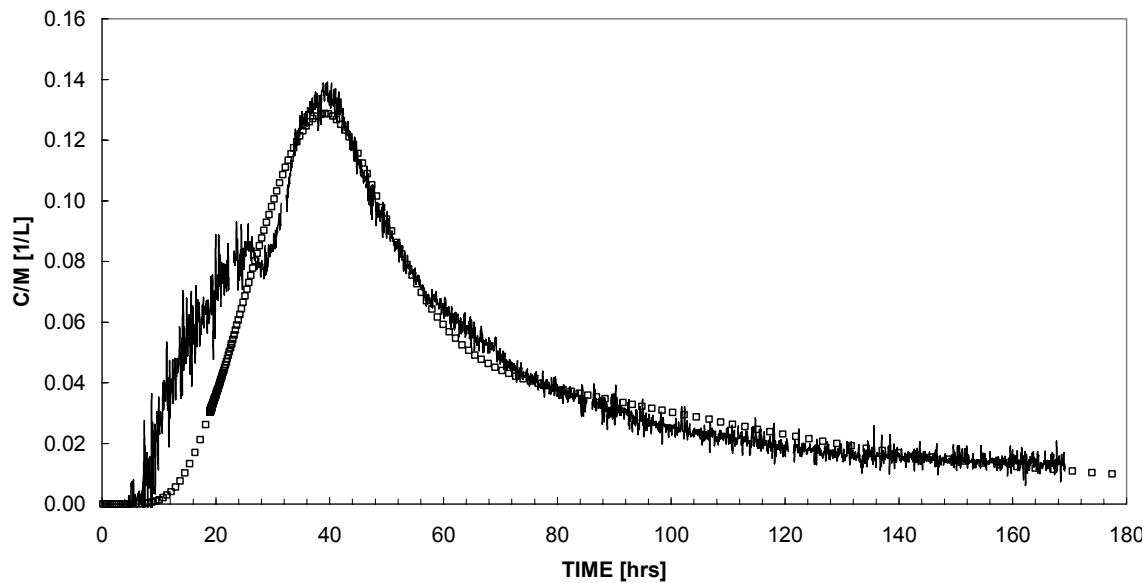
**Fig. 4** Normalized tracer concentration curves ( $C/M$ ) for pyranine (●) and PTS (dashed line), recovery rates  $RR$  for pyranine (○) and PTS (line) observed in withdrawal well and the injection function of both tracers (heavy line).

where  $Q_{inj}$  is the flow rate ( $66 \text{ ml h}^{-1}$ ) and  $C_{inj}(t)$  is the input tracer concentration (see Fig. 4) in the injection borehole. The injected masses of pyranine and PTS were equal to 0.65 and 6.5 mg, respectively. PTS is detectable at higher concentration and therefore a larger injected mass of that tracer was required.

The concentrations of pyranine and PTS observed in the withdrawal borehole (BOHP 98.001) normalized to the total injected mass ( $C/M$ ) are shown in Fig. 4. Both tracers show nearly the same concentration ( $C/M$ ) and recovery ( $RR$ ) curves. Taking into account that pyranine and PTS have the same molecular diffusion coefficients, the same shape of those curves clearly demonstrates the conservative behaviour of PTS at the GTS.

The existence of two peaks in the observed tracer breakthrough curve (Fig. 4) results either from a depth stratification (e.g. Zuber, 1974) or from channelling (Moreno *et al.*, 1993; Smith *et al.*, 2001). Within the present study too few data exist to perform a quantitative interpretation of two flow paths.

Under an assumption of a single flow path, a quantitative interpretation of the breakthrough curves was performed using the FRAC3DVS numerical model developed by Therrien & Sudicky (1995), which considers the 3-D tracer transport in discretely-fractured, saturated–unsaturated media. It includes advection, longitudinal and transverse dispersion and the tracer diffusion into the rock matrix. For the numerical calculations, the following dimensions of the fracture plane were used:  $x = 3.0 \text{ m}$  (horizontal) and  $z = 3.0 \text{ m}$  (vertical), while in the direction perpendicular to the plane a matrix domain with  $y = 0.01 \text{ m}$  was assumed. Variable spacing was used for discretization with a finer nodal spacing being equal to  $0.01 \text{ m}$  near the injection and extraction boreholes in the  $x$ - and  $z$ -directions, and to  $0.0001 \text{ m}$  in the rock matrix ( $y$ -direction). The flow distance was  $0.77 \text{ m}$ , while the volumetric flow rates in the injection and withdrawal wells were  $66$  and  $58 \text{ ml h}^{-1}$ , respectively. Some additional rock parameters required for the numerical calculation (matrix density of  $2.67 \text{ g cm}^{-3}$ , matrix hydraulic conductivity of  $9.65 \times 10^{-11} \text{ m s}^{-1}$ , and the fracture conductivity of



**Fig. 5** Best fit obtained with the FRAC3DVS model (□) to the experimental tracer curve of PTS (line).

**Table 2** Model parameters chosen for the simulation of the tracer test performed at the GTS with the aid of the FRAC3DVS program.

Tracer	$\alpha_L$ (m)	$\alpha_T$ (m)	$2b$ ( $\mu\text{m}$ )	$n_p$ (%)
Pyranine	$2 \times 10^{-2}$	$5 \times 10^{-5}$	900	1.5
PTS	$2 \times 10^{-2}$	$5 \times 10^{-5}$	900	1.5

$1.5 \times 10^{-8} \text{ m s}^{-1}$ ), were taken from Fisch (1999). The diffusion coefficient in the matrix ( $D_p = D_m/\tau_p = 1.0 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ ) was also taken as being known for both tracers. Under these assumptions four parameters were sought in FRAC3DVS: longitudinal and transverse dispersivities ( $\alpha_L$  and  $\alpha_T$ , respectively), matrix porosity ( $n_p$ ) and fracture aperture ( $2b$ ). Performed simulations showed that a number of different parameter sets yielded nearly the same good fit shown in Fig. 5. The set of the finally selected parameters based on the results of earlier investigations is shown in Table 2. The borehole imaging performed in the shear zone AU126 has shown fracture apertures of  $1000 \mu\text{m}$ . The Peclet number ( $\text{Pe} = 1/P_D$ ) of  $\text{Pe} = 35$  agrees reasonably well with the values of 20 and 30 found within the shear zones AU 96 and AU 126, respectively by Heer & Hadermann (1996) and Pfingsten & Soler (2003). The matrix porosity at the location of the packer intervals of the boreholes reported by Pfingsten & Soler (2003) vary from less than 0.1% to more than 20%, with the result of the present study being within that range.

## CONCLUSIONS

Two tracer tests were performed in radial-convergent and injection-withdrawal flow fields at the Lindau (LRL) and Grimsel (GTS) underground rock laboratories. The

results of both tests clearly document the conservative behaviour of a new fluorescent tracer (PTS) and its applicability for groundwater investigations.

In the test performed at the LRL, the fitted and resulting rock parameters found using the PTS concentration and recovery curves (fracture aperture, matrix and fracture porosities) are in a very good agreement with earlier findings of Himmelsbach *et al.* (1998).

The dipole tracer test performed in the GTS using PTS and pyranine (tracers of equal diffusion coefficients) showed identical transport behaviour confirming a conservative behaviour of the PTS in these bedrock environments. The modelling of tracer curves has shown that the unique inverse solution is not available. An acceptable set of fit parameters was selected on the basis of knowledge obtained in earlier studies. The present experiment has shown that the influence of the matrix porosity to radionuclide transport for the shear zone AU 126 cannot be well determined by applying injection–withdrawal tests. Practically, all tracer experiments performed until the present at the shear zone AU 126 (Pfingsten & Soler, 2003) have shown that the dipole tests in fissured rocks supply little information.

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