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Field tracer experiment in a low permeability fractured medium: results from El Berrocal site

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Abstract

A modelling experimental activity was developed to characterise the hydraulic behaviour of water-bearing fractures in crystalline rocks.

Three conservative tracers were injected into two packed-off sections of the same well, 69 m deep in a granite formation, and recovered by pumping from an isolated section of a second borehole, 46 m deep, 14 m apart. The concept of this design is to characterise separately an isolated fracture zone intersecting the lower parts of both wells from the fracture network intersecting the bulk of the rock.

Before using the tracers in the field, their behaviour was studied in the laboratory under controlled conditions. Fluorescein, eosin and iodide were finally chosen as the best spikes.

The field experiments were developed under strictly controlled conditions, such as (1) checking the hydraulic pressure in the packed-off sections and in other parts of circuits; (2) mixing the tracer solutions during the injection and checking their homogeneity; (3) performing a continuous and automatic monitoring of tracer concentration in the arrival well.

Iodide and fluorescein were injected in one section, and eosin in the other section. The pumping rate was maintained at $2 \ 1 \ \text{min}^{-1}$. The test lasted 27 d after which from 40 to 60% of the injected masses were recovered.

Breakthrough curve analysis considered two conceptual models: radial advective-dispersive transport with and without matrix diffusion: the first model returns thickness-porosity values around 0.2×10^{-1} m and dispersivity around 4 m. Parameters are remarkably consistent for

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iodide and fluorescein, although fittings can be improved. The matrix diffusion model provides much better fittings by decreasing thickness porosity to 0.8×10^{-2} m. Dispersivities range from 0.5 to 0.9 m and the molecular diffusion term differentiates the behaviour of conservative tracers such as fluorescein and iodide. © 1997 Elsevier Science B.V.

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1. Introduction

Solute transport in fractured aquifers is controlled by preferential pathways or channelling having different hydraulic properties and by diffusion in the rock matrix.

The understanding of the behaviour of contaminants in fractured aquifers requires validated procedures and techniques for the determination of model parameters. This may be achieved through the combination of hydrogeological investigations in field and model applications.

This paper describes the results of field tests carried out in the framework of 'El Berrocal Project', an international study aiming to understand and model solute transport processes in a fractured granite formation (Rivas et al., 1995). In this framework, a number of boreholes of different depths were drilled in an abandoned uranium mine. Radially converging tracer tests (using non-adsorbed spikes) were planned in order to improve the hydrogeological knowledge of the site (D'Alessandro et al., 1995). The characterization of the hydraulic behaviour of different fracture systems intersecting two boreholes at different depth is described in this paper. Previous cross-hole pumping tests showed that the hydraulic transmissivity of the fracture set connecting the bottom of the two boreholes was greater than that of the fractures intersecting the same wells in shallower sections. The tracer test hereafter described allowed a characterization of both water pathways.

The experiment was divided into different phases: (1) laboratory experiments aiming to choose the most suitable tracers; (2) hydraulic characterization of the rock system between the two wells; (3) development of an experimental design for the tracer test, defining pumping rate, time duration and tracer mass to be used; (4) cross-hole migration test with fluorescent tracers and iodide ions; (5) analytical model development and interpretation of the data from the tracer experiment.

2. Field tests

2.1. Site description

El Berrocal Project was developed within an experimental site located at the eastern end of Sierra de Gredos, Central Spain (Fig. 1) in an abandoned uranium mine. The ore body consists of a sub-vertical pitchblende-rich quartz vein intruded into a Hercynian granite during an alpine reactivation of a late-Hercynian fault. Preliminary tracer tests were performed in wells S1 and S7, 60 m deep, drilled inside the mine. The described



Fig. 1. Cross-section through the experimental site, with the boreholes drilled in an early phase.

experiment was performed in boreholes S11 and S12, (64 and 46 m deep, respectively) drilled outside the mine near the bottom of the Tarica Valley (Fig. 1).

2.2. Tracer selection

Before performing the cross-hole migration test in the field, the conservative behaviour of the fluorescent tracers employed in the present study was checked using granite samples and ground water collected from the site (Gutiérrez et al., 1991).

Both batch K_d measurements and column migration experiments were performed. These experiments showed that fluorescent dyes behave as tritiated water (Fig. 2) and may be considered as fully conservative tracers.

2.3. Borehole hydraulic characterization

Preliminary hydraulic tests were performed on S11 and S12. The results of these tests, together with previous information from structural study of the cores (Campos Egea and Marin Benavente, 1994) provided the guidelines for the tracer test design.

Single borehole constant head tests were performed on S11 using a conventional straddle packer system, with monitoring intervals of 3 to 5 m. These tests gave transmissivity values ranging from 10^{-6} to 10^{-9} m² s⁻¹. A cross-hole pumping test was then performed aiming to find hydraulic responses between the boreholes. The monitored borehole section in S11 extended from 49 m depth to the hole bottom (-64 m). The section was isolated using a single packer at that depth. S12 was used as an observation well. Head measurements were performed in a borehole section extending from -24 to -49 m (hole bottom).



Fig. 2. Breakthrough curves from miscible displacement tests of eosin and tritium flowing through a column of crushed and weathered granite.

2.4. Tracer test experimental design

One pumping borehole (S12) and injections of tracers into the other borehole (S11) at two different depths were planned. Since anisotropy and heterogeneity effects were neglected, the symmetry of the flow field was assumed to be radial. For test planning it is necessary to know: (1) how long the experiment will last; (2) the mass of the tracer to be used.

The time is governed by the following expression:

$$t_R = \frac{\pi R^2 \phi_c b}{Q_{\text{max}}} \tag{1}$$

where R = distance between injection and pumping wells (m), ϕ_c = flow porosity (dimensionless), b = thickness of the tested rock mass section (m), Q_{max} = pumping rate (m³/sec).

The tracer travel time is inversely proportional to the pumping rate. Then Q must be maximised in order to optimise the time required and costs for the experiment, with an additional restriction based on the allowable drawdown. Under steady-state conditions, such drawdown can be computed using Thiem's equation

$$s = \frac{Q}{2\pi T} \ln \frac{R'}{r} \tag{2}$$

where s = drawdown (m), Q = pumping rate (m³/sec), T = transmissivity (m²/sec), R = radius of influence of the pumping well (m), r = distance from the observation point to the pumping well (m).

The experiment was performed at a depth where the initial hydraulic head was some

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5-6 m below ground level. A 15 m drawdown was considered acceptable. Considering $T = 0.3 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ (resulting from cross-hole tests) the optimum pumping rate was 2.0 l min⁻¹, which is consistent with the pumping rate used during hydraulic characterization. The estimated travel time with this flow rate ranged between 0.2 and 20 d, depending on the $b\phi_c$ value considered in Eq. (1).

Previous hydraulic characterisation showed that the maximum pumping rate to be imposed at the extraction well was around $2 \, l \, min^{-1}$. This lowered the piezometric head of the pumping section from 5 to 15 m, which is the maximum value allowing the steady-state conditions to be maintained.

The mass of tracer to be used (Davis et al., 1986) depends directly on the detection limits of the analytical techniques, and on the volume of water in which the tracer solution will be diluted:

$$g = CpV\beta \tag{3}$$

where Cp = lower detection limit (g/m³), V = volume of water (m³) accessible to the tracer, $\beta =$ enlarging factor (dimensionless) accounting for dispersion, dead-end pores and other unquantifiable dilution factors, around the ratio R/2r (distance between boreholes/borehole diameter).

2.5. Tracer test

The experiment consisted of the injection of the tracers into two packed-off sections of S11, and of the recovering of the spiked water by pumping from an isolated section of borehole S12, 14 m apart (Fig. 3). An important aim of this test was to prove that the planned set-up and the operating procedures might guarantee this simultaneous injection and the tracer migration under strictly controlled conditions (Molz et al., 1987; Abelin et al., 1991). The purpose of this design was to characterise two different water pathways: the fracture zone connecting the lower parts of the two wells and the fracture network intersecting the bulk of the rock. More details about the equipment and the set-up are shown in Fig. 3.

2.5.1. Set-up description

In the injection well two sections were separated by a double-packer. The upper section (hereafter called S11-A) was 24 m long, and isolated a volume of about 140 l of water between -25 and -49 m. The lower one (S11-B) was 20 m long, between -49 and -69 m, with a volume of 118 l.

A system of external circulation of water for homogenisation of the tracers in the wells was adopted. The system consisted of equipping each section with a recirculation circuit, connected to the surface, and allowing access to the borehole sections.

Each circuit, connected to a pump (1,3 in Fig. 3), had one valve for tracer injection (b) and one sampling point for checking the tracer concentration (f). In order to allow the injection of flushing water, both loops were connected to a tank of clean water (7) to push the tracer solution into the rock formation. Water from the tank was pushed into the circuits by nitrogen pressure (a). The water flow through each loop was checked by both a flow meter (e) and a pressure indicator (not shown). The piezometric head



Fig. 3. Set-up for in-situ tracer test.

reached in each section in different phases of the test was measured by a pressure transducer (2,4) connected to both a display panel (6) and a data logger (16).

All the checking instruments and the input–output points of the external circuits were gathered in a control panel near the well casing.

The recovery section S12 is located between the well bottom at -44 m and the single packer location at -27 m. The different elements are also shown in Fig. 3. This section was also equipped with a recirculation loop having different purposes:

- 1. to homogenise the tracer solution
- 2. to bring to the surface the ground waters to be sampled and analysed
- 3. to maintain a pumping rate as low as 2 1 min⁻¹ from the withdrawal well, by subtracting part of the recirculating water by a peristaltic pump.

The driving force for the tracers was provided by the peristaltic pump (12). The more powerful pump downhole (9) only had the task of recirculating the solution through the loop and bringing it to the surface.

An automatic sampler (11) was also connected to the circuit. It consisted of a computer-controlled carousel of 24 bottles whose sampling frequency and volume can

S11 - Injection - Upper section: (from -25 to -49 metres)

- 1) Grundfoss pump (3-7 l/min)
- 2) Pressure transducer

S11 - Injection - Lower section:

- (from -50 to 64 metres)
- 3) Grundfoss pump (3-7 l/min)
- 4) Transducer
- 5) Two 70 mm. packers

S11 - Surface

- 6) Display for hydrostatic pressures + data recorder (16)
- 7) Tank for flushing water (300 I.) Circuit equipped with:
 - a) Device for nitrogen pressure
 - b) Inlets for tracers injection
 - c) Injection valve
 - d) One-way valves
 - e) Flowmetres
 - f) Sampling points

S12 - Pumping - One section: (from -27 to - 46 metres bottom)

- 8) 70 mm Packer
- 9) 3 " Grundfoss pump
- 10) Transducer

S12 - Surface:

- 11) Automatic sampler (ISCO)
- 12) Peristaltic pump (1-3 l/min)
- 13) lodide automatic analyzer
- 14) Manual sampling
- 15) Hydrostatic pressure display
- 16) Data recorder
- 17) Paper recorder

be programmed. An ion-selective electrode was also mounted in the external part of the circuit, allowing a continuous check of the tracer concentration in the arrival well (13). The electrode was connected to a paper recorder (17). Finally, a pressure transducer (10) allowed the control of the hydraulic head to be maintained in the pumping well. It was connected to the display panel (15) and to the data logger (16).

As indicated previously, two fluorescent dyes, eosin and fluorescein, were used. An ionic spike, which is easy to detect by selective electrodes, was then added. This gave the opportunity to check the conservative behaviour of fluorescent dyes. Iodide was selected as ionic spike. Since it was measured automatically, it was used as a 'breakthrough alarm bell' and it was injected in the most conductive section (S11-B) together with fluorescein. Eosin and fluorescein were measured manually in the field; further checks on all the tracers were successively made in the laboratory.

During some preliminary hydraulic manoeuvres the fracture zone intersecting the lower section was more conductive than the upper one (by a factor of two). Iodide and fluorescein were then injected into section B.

Taking the previous calculations (Eq. (3), Section 2.4) into account, the following type and quantity of tracers were injected into the two sections of S11:

Upper section S11-A	Eosin (5.022 g)
(accounting for 133 l)	
Lower section S11-B	Fluorescein (5.054 g)
(accounting for 110 l)	KI (4.578 g, corresponding to 3.5 g of I^-)

2.5.2. Operating procedures

After setting up the equipment, some tests were performed in order to check the operating conditions of the test, i.e. the seal of the packers, the drawdown rate and the general working conditions of the system.

The packer sealing was checked first in S11. Pumping from the lower section (S11-B) for a total lowering of 8 m in the piezometric head resulted in an observed drawdown in the upper section (S11-A) of 8 cm only. Reversibly, a lowering of 11 cm of the phreatic level was induced by a drawdown of 5.60 m in S11-A.

The sealing was less effective in S12, where the phreatic level lowered 71 cm for a drawdown of 5.65 m of the packed-off section. The seal was in any case considered acceptable.

Later, the hydraulic manoeuvres planned for the experiment were tested. An extraction rate of $2 \ lmin^{-1}$ was applied to the pumping section in S12: after 5 h this resulted in a 9.3 m drawdown. Responses of 2.73 m and 1.16 m were observed in S11-B and S11-A respectively. Then, the lower section of S11 revealed a higher sensitivity. This was confirmed during flushing: injecting water at 2.4 atm resulted in a rate of flushing of 1.8 l min⁻¹ in S11-B, and 0.6 l min⁻¹ in S11-A.

Pumping from S12 started at a rate of $1.5 \ lmin^{-1}$. After 48 h it was increased to 2 lmin⁻¹. One day later hydraulic equilibrium was achieved. Then, iodide and fluorescein were injected sequentially into S11-B. After 40 min recirculation, the concentration was normalised; water was flushed for 3 h, with a total injection of 225 l. Eosin was injected



Fig. 4. Disappearance curve of fluorescein from S11-B injection section. Arrows indicate the flushing periods. Note the increase of concentration when the flushing pressure releases.

into S11-A 4 h 21 min later than fluorescing. Flushing continued for 9 h, during which 300 l were introduced.

It is worth noting the behaviour of the tracer after flushing. An increase of the concentration in the injection well is evident along the disappearance curves (Fig. 4). Once the pressure of flushing water relaxes, the tracer apparently comes back from the rock matrix to the well section. The same behaviour is less evident but still visible in the eosin disappearance. Then a second flushing step was decided on: additional 160 l were added to S11-B, and 200 l to S11-A.

Eosin and fluorescein were detected in the pumping well about 3 d after injection. Because of the lower sensitivity of the analytical technique, iodide could be detected later.

The test continued normally for 9 d when a failure in the recirculation loop inside the pumping well caused a slowdown of the experiment, followed by an interruption. This irregular trend, although visible in the breakthrough tracer curves, did not affect the test results. After repairs, the experiment continued regularly for a further 15 d. It was finally stopped 27 d after injection because of a malfunction of the main pump in S12. At this moment, however, a reasonable recovery of the tracers was achieved.

3. Modelling

Three breakthrough curves were modelled independently using the computer code TRAZADOR (Benet and Carrera, 1992) which analytically solves transport equations for conservative tracers, following the inverse problem approach. Fitting is achieved by

Tracer	Model ^a	L (m) ^b	Q (1 min ⁻¹) ^c	Recovery %	$b\phi_{c}$ (m) ^d	$\begin{array}{c} \alpha_{\rm L} \\ ({\rm m})^{\rm e} \end{array}$	ϕ'_{m} (-) ^f	D _m (-) ^g
Fluorescein	AD	22.0	1.375	62.54	0.0187	3.400	_	
Iodide	AD	22.0	1.375	61.3	0.0224	4.000	_	_
Eosin	AD	14.5	0.625	41.16	0.0201	4.400	_	_
Fluorescein	Dm	_			0.0088	0.600	2.000	0.5479
Iodide	Dm		_	_	0.0081	0.570	2.000	0.9000
Eosin	Dm	_	_	_	0.0078	0.904	2.000	0.6039

Table 1 Tracer recoveries and parameter values returned by different conceptual models

^a AD = advection dispersion, Dm = matrix diffusion.

^b L = distance.

Q = flow rate.

^d $\tilde{b}\phi_c$ = thickness porosity.

^e $\alpha_{\rm L}$ = longitudinal dispersivity.

matrix thickness).

the maximum likelihood method. The code can simulate several models. From these models, radial advection-dispersion and radial advection-dispersion with matrix diffusion were chosen.

3.1. Implementation of the conceptual model

The flow field was assumed to be radial in spite of the test geometry and the fracture distribution. The pumping rate applied at S12 affected the isolated intervals S11-A and S11-B in different ways. Differences in the measured drawdowns reveal such an effect,



Fig. 5. Cumulative recovery curves for dyes and iodide.



Fig. 6. Fitting of experimental data by radial-advection and radial-advection/matrix-diffusion models

which is due to the hydraulic conductivity distribution through the rock mass. The pumping flow rate was distributed according to the measured drawdowns and distances between intervals, in order to reproduce flow conditions for each tracer breakthrough. As a result, flow rates were 0.625 ml min⁻¹ for eosin and 1.375 ml min⁻¹ for fluorescein and iodide (see Table 1).

Fig. 5, where the cumulative recoveries are shown, reveals that iodide and fluorescein display different behaviour. The parts of the curves from day 0 to day 13 run relatively parallel. From day 13 on, the curves tend to intersect each other. That means that two solutes, considered as conservative, did not behave in the same way, even though they were injected at the same time. Therefore, it is expected that models will return different parameters. Transport of eosin takes place over a different part of the formation, and so the cumulative curve displays a completely different picture.

Several failures occurred during the experiment which affected the quality of the breakthroughs. The data most influenced by such failures were not used during the calibration, but are represented by different symbols in Fig. 6.

3.2. Results for radial advection-dispersion

Calibration of eosin resulted in a reasonably well-fitted curve, specially in the first part. The model needs relatively high dispersivity to reproduce the tail of the curve ($\alpha_L = 4.4$ m, compared to the distance between wells, 14.5 m). The model returns a slightly high thickness porosity value as well (0.02 m, Table 1). As a result, data belonging to the first two days are not properly fitted. This is probably the counterpart of keeping the tail of the curve reasonably well reproduced. A more complex model might provide more accurate results.

Iodide and fluorescein calibration return similar parameter values: thickness porosity around 0.02 m and dispersivity 4.0 m and 3.4 m respectively. Given the quality of data, fittings are acceptable (Fig. 6). The rising part of the fluorescein breakthrough is specially well fitted, compared to eosin as well as iodide. The two tracers were supposed to be conservative, therefore we attribute such differences to retardation processes which affect fluorescein and iodide selectively. Such differences are also shown in Fig. 5.

After the performance and analysis of the tests, some questions remain open, such as the definition of the first arrival time (t_0) . While the peak concentration time and the 'mean' time corresponding to 50% of the recovery are quite well defined, the first arrival lacks precision. This is mainly due to analytical restrictions, which selectively affect the tracers used. It is important to have good control over t_0 because it is least influenced by chemical processes; its value depends only upon advective-dispersive processes.

In the tests described, the detection limit of analytical techniques for iodide and fluorescein differed by about one order of magnitude. This difference means that first arrivals cannot be considered as the first detected. Therefore, another definition is needed. Use of a 'first arrival time' derived from the breakthrough curve obtained is recommended. In this way, t_0 should be considered as the time for which a small percentage of recovery is achieved (i.e. 1% or so).

However, in this way, retardation processes affecting the recovery are inherited by t_0 .

In summary it is not clear how to deal with the restrictions carried by the t_0 definition. To compare flow processes affecting solute transport, one will continue to consider part of the early arrivals of the breakthrough.

3.3. Results from advection-dispersion and matrix diffusion

Regarding eosin calibration, tail fitting benefits largely from the new model. However, improving the fitting of the tail results in a worse reproduction of the rising part of the curve. Thickness porosity and dispersivity are reduced by a factor of four (Table 1), which results in a different slope on the first part of the curve (Fig. 6).

As in the previous case, the fitting of the tail of fluorescein improves with the matrix diffusion model. Thickness porosity is reduced by a factor of about three, while dispersivity decreases by a factor of eight. The parameters returned by the fluorescein model were used to calibrate the iodide breakthrough. In this case, the model displays a significant improvement of the fitting curve, even though the fit using advection dispersion was remarkably good. Matrix diffusion fitting displays a curve of lower computed values than measured tail data—as for fluorescein—while those for advection—dispersion were consistently higher. Matrix diffusion parameter values returned for iodide do not differ from those for fluorescein, apart from dimensionless molecular diffusion (Table 1). However, parameters returned for eosin (matrix diffusion) showed thickness porosity values similar to iodide and fluorescein, but longitudinal dispersivity is twofold. Eosin also displays for molecular diffusion higher values than fluorescein and lower than iodide.

4. Conclusion

The field test showed that the planned operating procedures and the designed set-up were suitable to control any step of the experiment.

Concerning the results, an important aspect is the behaviour of tracers after flushing with a slug of fresh water. After pressure release, the spiked water comes back from the fractures to the well: this slows the access of tracer into the fractures, making the assumption of a 'pulse injection' more approximate.

Comparing advection-dispersion models poses problems because there were difficulties in the proper definition of that part of the breakthrough curve affected only by porosity and dispersivity. This part is the first arrival which is also affected by the performance of the injection.

The three tracers displayed noticeable differences in spite of their conservative nature. Eosin breakthrough also differs from the rest because it affects a different part of the rock mass. However, iodide and fluorescein were injected in the same borehole section and, surprisingly, display different tail effects. Retardation selectively concerns iodide and fluorescein. As a result, when advection–dispersion parameters are comparable, fittings display noticeable differences. One can question the convenience of using the concept 'conservative' in field experiments, which is more probably a myth rather than a scientific concept.

The matrix diffusion model calibration largely improved fitting for all tracers. None of the tracers used showed a "true conservative" behaviour.

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