Comparison of novel synthetic DNA nano-colloid tracer and classic solute tracer behaviour

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ABSTRACT

Tracer experiments are commonly used in hydrogeological and geothermal reservoir studies to determine attributes of fluid flow paths. In complex fractured media, it is often necessary to use several different tracers, injected at different locations, to determine hydraulic connections and preferential flow paths. A relatively recent development is the use of tracers in tomography studies, requiring a large number of tracers. However, the number of suitable tracers is reduced when their combined effects and interferences are taken into account and particularly when temperatures in the studied media are high, as elevated temperatures can degrade the artificial tracers. Consequently, the scientific community is continuously attempting to increase the number of available artificial tracers that are environmentally benign, can be easily detected in dilute concentrations, are typically conservative, and do not degrade (or degrade at known rates) at elevated temperatures. A new class of tracers are DNA nanotracers, which are environmentally friendly, sub-micron scaled silica particles encapsulating small fragments of synthetic DNA.

Using synthetic DNA in tracer tests enables the development of a virtually unlimited number of distinct tracers, and consequently, “contamination” of the studied system by one specific environmentally benign tracer type is not a limitation anymore. In addition, these DNA nanotracers can potentially be used to determine the approximate maximum temperature the tracer has encountered, which is of particular interest in geothermal systems. However, as these tracers are new and constitute colloids, rather than solutes, their transport behaviour within the fluid flow path is not well understood. As colloids tend to move preferentially within the main fluid flow paths, they typically move faster through the porous or fractured medium than the mean fluid velocity, emphasizing preferential flow paths. Thus, classic tracer test analyses, developed for solute tracers, may not be completely applicable to the DNA colloid tracers.

In this study, we test these novel DNA nanotracers in both a sedimentary porous medium setting, where a classic tracer test has been performed before, and in a fractured rock system, where heat and solute tracer tests are also conducted. Additionally, comprehensive column experiments will complement the study. This allows us to compare the new DNA nanotracers to classic solute tracer behaviour within these two fundamentally different settings. The fracture-based system investigated here is the Deep Underground Geothermal (DUG) Laboratory at the Grimsel Test Site in the Swiss Alps.

1. INTRODUCTION

Tracers in hydrogeology are used to understand transport processes, to quantify their parameters, and to investigate hydraulic connections in the subsurface. The characterization of a groundwater system is important in several areas of research, industry and resource management. It has applications, for example, in geothermal reservoir studies, groundwater protection and remediation, and in underground infrastructure management (e.g. Käss 1998, Leibundgut et al. 2009).

Dissolving salts and fluorescent dyes are common types of tracers used in hydrogeology. In fact, they are considered as ideal tracers, as they represent the groundwater flow (Leibundgut et al. 2009). These tracers are readily available, and monitoring the development of their breakthrough is relatively straight-forward. However, a natural input or residue concentrations in the subsurface from previous tracer tests can interfere with subsequent tracer data analyses. Additionally, interferences and combined effects impede the use of multiple tracers simultaneously. There is also a special consideration for geothermal reservoir characterization, as the tracers must be thermally stable at reservoir conditions. Several, although a limited number of tracers have been shown to be suitable for use in geothermal reservoirs (Rose et al. 2001, Axelsson 2013).

The novel DNA nanotracers tested in this study overcome the problem of interferences and combined effects. They are sub-micron sized silica particles encapsulating small fragments of synthetic DNA (Paunescu et al. 2013b). The encapsulation increases the chemical and thermal stability of the DNA (Paunescu et al. 2013a), making it also a potential tracer candidate for geothermal reservoirs. DNA has a significant information storage capability, and thus using it as a tracer has an advantage of producing theoretically unlimited number of distinct tracers, all of which have the same transport properties. This property follows from the encapsulation of the DNA between a support silica particle and silica coating. As a consequence, the DNA nanotracers are transported based on the properties of the silica particle, but identified based on the encapsulated DNA with ultra-low detection limits (Paunescu et al. 2013a, b). These features allow the use of multiple distinct DNA nanotracers simultaneously, for example in tomography studies, or repeat tracer tests to take place.

2. EXPERIMENTAL SETTINGS

Two different field settings have been chosen for the evaluation of the novel DNA nanotracers as tracers in hydrogeological applications, and for the comparison of their behaviour in the subsurface to classic solute tracers. The field settings are sedimentary porous medium and fractured crystalline rock. These field experiments are pioneering cases for a field-scale application of the DNA nanotracers. They have a high potential of becoming an established tool in hydrogeological, oil and gas, as well as geothermal investigations, to name only a few. Thus, the field tests provide valuable input for studying the feasibility of these new tracers. In addition to the field experiments, we conduct packed-sand column experiments with both DNA nanotracers and solute tracers.

2.1 Widen test site

A well-studied sedimentary porous medium site is located in Widen in north-eastern Switzerland, next to the river Thur (Diem et al. 2010). The site hosts several wells, including four multichamber wells that allow isolated injection or production at different depths of the well. The array of densely located wells at the site is ideal for conducting tracer tests with the novel DNA nanotracers and to compare their transport properties to classic solute tracers.

2.2 Deep Underground Geothermal (DUG) Laboratory

The DUG Laboratory is located near the Grimsel Pass in the Swiss Alps in granitic rock at a depth of about 450 m. The laboratory is located within the Grimsel Test Site operated by NAGRA at a location where three major shear zones intersect each other between two tunnels (Fig. 1). A comprehensive in-situ stimulation and circulation experiment is taking place in the DUG Laboratory, and this is accompanied by pre- and post-stimulation characterization of the rock mass. Utilization of various methods for site characterization (e.g., seismic and GPR surveys, pumping tests, and tracer tests) will provide abundant amount of data to quantify the transport properties of the fracture-dominated system. This, in turn, allows us to test the new DNA nanotracers in concert with solute tracer tests and to compare the migration behaviour of DNA nanotracers with classic solute tracers.

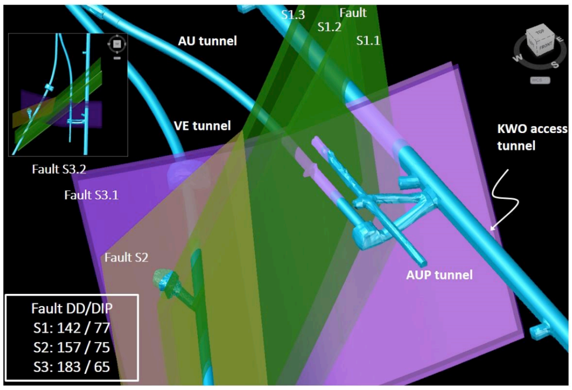

Figure 1: Illustration of the DUG Laboratory, locating between VE and AU tunnels, and characterized by three major shear zones (S1, S2 and S3) intersecting within the DUG Laboratory test volume.

2.3 Column experiments

A well-controlled setting with fewer uncertainties compared to the field experiments is provided by column experiments. Here, a column, filled with sand, allows detailed investigation of the transport properties of the DNA nanotracers. Also, this closed system provides insights into possible accumulation or retardation properties of the colloid DNA nanotracers.

3. ON-GOING RESEARCH AND FUTURE PROJECTS

The study described here is ongoing at the moment of submitting this paper. Unfortunately, due to delays in conducting the tracer field experiments, results and interpretation have to be omitted from this paper. However, preliminary results indicate some fundamental differences in transport properties between DNA nanotracers and solute (dye) tracers as briefly outlined in the next paragraph. By the time of presentation in three months, we hope to be able to present these results.

Previous studies suggest that colloidal transport is characterized by shorter mean transport times, smaller fractional mass recoveries, and smaller dispersion coefficients, compared to solute tracers (e.g., Becker et al. 1999, McKay et al. 2000, Zvikelsky and Weisbrod 2006, Baumann et al. 2010). The preliminary results obtained from the DUG Laboratory experiments agree with these findings.

They indicate that the DNA nanotracers were transported through the fractured medium with faster mean transport velocities than the solute dye tracers, and that they experienced less dispersion. We also plan to evaluate the potential applications of the novel DNA nanotracers in hydrogeological, oil and gas, and geothermal settings.

REFERENCES

Axelsson, G.: Tracer tests in geothermal resource management, *EPJ Web of Conferences*, 50, (2013), 02001.

Baumann, T., Toops, L. and Niessner, R.: Colloid dispersion on the pore scale, *Water Resources Research*, 44, (2010), 1246-1254.

Becker, M.W., Reimus, P.W. and Vilks, P.: Transport and attenuation of carboxylate-modified latex microspheres in fractured rock laboratory and field tracer tests, *Groundwater*, 37, (1999), 387-395.

Diem, S., Vogt, T., Hoehn, E.: Räumliche Charakterisierung der hydraulischen Leitfähigkeit in alluvialen Schotter-Grundwasserleitern: Ein Methodenvergleich, *Grundwasser – Zeitschrift der Fachsektion Hydrogeologie*, 15, (2010), 241-251.

Käss, W.: Tracing technique in geohydrology, *A.A.Balkema*, Rotterdam, (1998).

Leibundgut, C., Maloszewski, P. and Külls, C.: Tracers in hydrology, *John Wiley & Sons*, Chichester, (2009).

MacKay, L., Sanford, W. and Strong, J.: Field-scale migration of colloidal tracers in a fractured shale saprolite, *Groundwater*, 38, (2000), 139-147.

Paunescu, D., Fuhrer, R. and Grass, R.N.: Protection and deprotection of DNA – High-temperature stability of nucleic acid barcodes for polymer labelling, *Angewandte Chemie International Edition*, 52, (2013a), 4269-4272.

Paunescu, D., Puddu, M., Soellner, J.O.B., Stoessel, P.R. and Grass, R.N.: Reversible DNA encapsulation in silica to produce ROS-resistant and heat-resistant synthetic DNA 'fossils', *Nature Protocols*, 8, (2013b), 2440-2448.

Rose, P.E., Benoit, W.R. and Kilbourn, P.M.: The application of the polyaromatic sulfonates as tracers in geothermal reservoirs, *Geothermics*, 30, (2001), 617-640.

Zvikelsky, O. and Weisbrod, N.: Impact of particle size on colloid transport in discrete fractures, *Water Resources Research*, 42, (2006).