GEOCHEMICAL TRACERS: CAPABILITIES AND POTENTIAL FOR GEOTHERMAL RESERVOIR CHARACTERISATION

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ABSTRACT

Geochemical tracers have been used for many years to improve the understanding of reservoir dynamics in geothermal systems. Tracers can be classified as either conservative or reactive, and can be used in liquid-phase, vapour-phase or two-phase reservoirs at temperatures up to and above 300°C. They are commonly used to map flow pathways between injection and production wells in a geothermal field, to monitor the effects of reinjection and identify wells that might experience premature thermal breakthrough if left unmanaged. Tracer tests also provide information about reservoir fluid residence time, fluid recharge location or direction, swept pore volumes, inter-well connectivity, temperatures, fracture surface area, flow-storage capacity relationships and volumetric fluid sweep efficiencies. In addition, tracer data can be used with numerical transport codes to help validate 2D or 3D reservoir models. Thus, tracer tests can provide powerful insight into geothermal reservoir characteristics, and they can be performed at many stages of project development, from small-scale demonstration projects (e.g. an injection-production well doublet) through to large-scale commercial fields (e.g. Wairakei, New Zealand). New 'smart' tracers have the potential to be used with a single well to evaluate changes in fracture surface area following reservoir stimulation, and thus have applications to both conventional and unconventional (engineered) geothermal projects.

1. INTRODUCTION

Geothermal reservoirs in conventional geothermal systems are typically dominated by fracture permeability with high fracture permeabilities and low matrix permeabilities, and with the exception of porous hot sedimentary aquifer systems, this will likely be the case for most enhanced or engineered geothermal systems that are stimulated by hydraulic, chemical or thermal means. Understanding the distribution, aperture, orientation, and connectivity of new and existing fractures in a geothermal reservoir is important to both assess the effectiveness of a stimulation effort, or to manage the field over the longer term and minimise thermal breakthrough or too-rapid thermal drawdown (Bodvarsson and Stefansson, 1989). Down-hole tools such as borehole televiewers can provide some of this information proximal to the wellbore, such as fracture aperture and orientation, stress regime and lithologic-associations (e.g. Davatzes and Hickman, 2009), and micro-seismic monitoring during hydraulic stimulation can map out the location of fractures more distal to the well (e.g. Asanuma et al., 2005). However ultimately, fracture connectivity (and thus inter-well-connectivity) can only be assessed by direct measurements.

The two key methods for determining whether hydraulic connectivity exists between wells include pressure-transient testing, and tracer testing. Pressure transient testing includes both interference and pulse testing: both provide an indication of whether a connection exists between wells. With some assumptions of flow models between the wells (e.g. radial flow or linear flow), transmissivity and storativity of the formation in the interval between the wells can be estimated (Leaver, 1986; Fan et al., 2005). It typically involves initiating a pressure change at the active well (either by shutting-in or opening the well) and monitoring the pressure response in the shut-in observation well. Tracer testing of flow between geothermal (or groundwater) wells is an effective tool to map fluid flow pathways in a reservoir, and break-through curves (BTC) can provide a variety of useful, quantitative data depending on the type of tracer used (conservative or reactive). In many instances, this data can be used to provide insight into projected thermal drawdown in the reservoir or thermal breakthrough of re-injected fluids (e.g. Pruess and Bodvarsson, 1984; Robinson et al., 1984; Axelsson et al., 2001). A benefit of tracer testing (compared to pressure-transient testing) is that wells do not need to be shut in to assess connectivity – shutting-in wells is likely to be less desirable for commercially-producing fields.

2. BACKGROUND

2.1 Tracer testing basics

Geochemical tracer testing has been performed at many geothermal fields around the world for several decades. In a geothermal tracer test, a chemical tracer is injected into the reservoir to track the movement of fluid, and monitored either in one or more production wells or during flow-back from the same injection well (single-well injection-backflow testing). Geochemical tracers in either groundwater or geothermal applications should not affect the water flow regime (by changing fluid density or viscosity), and they are ideally not present in the reservoir or aquifer of interest (i.e. have a low background level). In addition, they should be environmentally benign, have low toxicity, be detectable at low concentrations and be affordable (100's of kilograms may need to be injected) (Davis et al., 1980). A key requirement for geothermal tracers is that they are stable at reservoir temperatures, or have well-characterised thermal-decay kinetics (e.g. Adams and Davis, 1991).

Although radioactive tracers have been used in the past (e.g. ^{82}Br, ^{125}I, ^{131}I and HTO (tritiated water) (McCabe et al., 1983; Robinson and Tester, 1984; Bixley et al., 1995; IAEA, 2004; Axelsson et al., 2005)), many tracer tests now use chemical tracers that are less hazardous to handle, including organic compounds such as fluorescent dyes (e.g. fluorescein, rhodamine-WT), other fluorescent compounds (e.g. naphthalene disulfonate), and dissolved inorganic solutes (e.g. potassium iodide (KI) and potassium bromide (KBr)). Most tracer tests performed to date in geothermal reservoirs have used these solute tracers, however current research is also investigating the use of nano-colloids as 'smart' geothermal tracers (discussed later).

In the reservoir, various transport processes affect the return of the dissolved tracer: solutes are affected by advection, dispersion and diffusion as they move through the reservoir, and these processes are reflected in the character of the tracer break-through curve that is observed in the production well(s).

A method of estimating inter-well volumes and flow geometries in liquid-phase geothermal reservoirs using conservative tracer tests was presented by Shook (2003). This approach is centred on moment analysis and analysis of tracer residence times (Levenspiel, 1972; Robinson and Tester, 1984): assuming that steady-state conditions are met and the tracer behaves conservatively, it enables quantitative information to be derived from tracer break-through curves including mean residence time, volume-averaged flow geometry, swept pore volume, and volumetric-fluid sweep efficiency (Nalla et al., 2005; Shook and Forsmann, 2005).

Without discrete monitoring of tracer transport through individual fractures (e.g. as in Robinson and Tester (1984)), it is challenging to translate a tracer breakthrough curve (and its corresponding residence time distribution (RTD)) from a fractured reservoir into interpreted discrete reservoir geometries or flow paths. This is partly due to the difficulties in separating the effects and contribution of diffusion vs. dispersion processes to the resulting RTD, but also the complexity of the flow pathways themselves. The RTD reflects the integrated tracer response from one or many flow pathways, with varying contributions of diffusion and advective processes, depending on the reservoir and flow regime. Nonetheless, many researchers have investigated different physical and numerical models to explore this further (e.g. Horne and Rodriguez, 1983; Jensen and Horne, 1983; Robinson and Tester, 1984; Hull et al., 1987; Bullivant, 1988; Moreno et al., 1988; Bullivant and O'Sullivan, 1989; Niibori et al., 1995; Neretnieks, 2002; Juliusson, 2012; Radilla et al., 2012).

2.2 Types of tracers

Geochemical tracers (geothermal or groundwater) can be divided into those that are believed to behave in a conservative manner (i.e. all tracer that is injected is eventually returned), and those that are not conserved, either through interactions with the reservoir rock (e.g. adsorption/desorption), the reservoir fluid (e.g. hydrolosis), or natural decay (e.g. radioactive decay). These reactive or 'smart' tracers have the potential to provide additional information about the reservoir, and are best used in conjunction with a conservative tracer to provide constraints on reservoir hydraulic properties.

Different tracers are used depending on the fluid phase in the reservoir (i.e. liquid phase, two-phase or vapour-dominated).

Commonly-used conservative liquid-phase tracers include the UV-fluorescent polyaromatic-sulfonate family, which have demonstrated thermal stability at temperatures of 300°C and above (Rose et al., 2001). There are 9 compounds in this family that have been identified as suitable geothermal tracers, and these have been used successfully to map fluid flow pathways and provide insight into reservoir properties at many geothermal fields around the world over the last 10 years (e.g. Philippines (Nogara and Sambrano, 2005), Japan (Watanabe et al., 2005), New Zealand (Rose,1998; Rose et al., 2000), Mexico (Iglesias et al., 2011), Germany (Behrens et al., 2006), France (Sanjuan et al., 2006), and the USA (Rose et al., 1998, 1999, 2000, 2002)). In late 2008, they were used at Geodynamics Ltd's Habanero project in central Australia to map flow properties of the reservoir between the Habanero #1 and #3 wells (Yanagisawa et al., 2009). The naphthalene sulfonates are detectable at very low levels in a reservoir (100 parts per trillion), and they are mutually compatible, meaning that any number of the family can be used simultaneously in a tracer test. They are analysed using conventional high-performance liquid chromatography (HPLC) with fluorescence detection (Rose et al., 2001).

Tracers for vapour and two-phase reservoirs have also been explored and tested. Injection of tracers into a vapour-dominated or two-phase system requires that the tracer be dissolvable in the liquid injectate, but will partition into the steam phase once it is in the reservoir (Adams et al., 2001). Gas-phase tracers will enter the steam phase at different rates depending on their volatility and the boiling parameters in the reservoir. Gas-phase tracers that have significant solubility in both the gas and liquid phase have potential for use as two-phase tracers: currently these include alcohols such as methanol, ethanol, n-propanol and isopropanol, n-butanol, 2-butanol, and isobutanol that all possess excellent detection limits in the low-ppb range (Fukuda et al., 2005; Mella et al., 2006a; Mella et al., 2006b). Tracers that are less soluble in water (i.e. have higher volatility) are better candidates for tracing fluid pathways in vapour-phase reservoirs. Tracers with these properties include: sulfur hexafluoride (SF6), hydrofluorocarbons (HFC's e.g. R-23 and R-134a), hydrochlorofluorocarbons (HCFC's, e.g. R-12 and R-13), and perfluorocyclic hydrocarbons (PFTs, e.g. perfluorocyclohexane) (Adams, 2001; Hirtz et al., 2010). Depending on the reservoir (two-phase or vapour-phase), two or more tracers are commonly injected into the reservoir during a tracer test, to distinguish different reservoir boiling zones and boiling conditions (e.g. Bixley et al., 1995; Moore et al., 2000; Adams, 2001; Fukuda et al., 2005; Hirtz et al., 2010). The interpretation of such tracer results can be more complicated than for liquid-phase reservoirs, owing to the variable partitioning of the tracer between the gas and liquid phase depending on the tracer volatility, and the specific boiling characteristics in the reservoir (i.e. single or multi-step boiling, and the degree of superheat if present).

The idea of using tracers that are temperature-sensitive (and thus reactive) has been explored for a couple of decades (e.g. Robinson et al., 1984; Tester et al., 1986; Robinson and Birdsell, 1987; Rose and Adams, 1994; Adams, 1995; Plummer et al., 2011). Popular groundwater tracers (e.g. rhodamine-WT and fluorescein) are usable in low-temperature geothermal reservoirs as conservative tracers, but at higher temperatures (>160°C for rhodamine-WT and 250°C for fluorescein), they are thermally unstable (Adams and Davis, 1991; Adams et al., 1992; Rose and Adams, 1994; Behrens et al., 2006). When used in conjunction with a conservative tracer, this reactive behaviour can be used to provide information about flow-path temperatures in the reservoir (e.g. Adams et al., 1989; Rose and Adams, 1994).

Similarly, the idea of using tracers that undergo adsorption/desorption reactions in the reservoir has been proposed in earlier studies (e.g. Robinson and Birdsell, 1987; Fox and Horne, 1988; Shan and Pruess, 2005). Developing such tracers is no easy feat and this is the focus of several current research projects in the USA (discussed in Section 4.0).

3. WHEN TO USE TRACERS: OPPORTUNITIES FOR GEOTHERMAL PROJECTS

Tracer tests can provide different information depending at what phase of a project they are utilised. Although they are often used to map flow pathways in established geothermal fields, tracer tests can provide useful information at many stages of project development and new 'smart' tracers may be able to be used with a single well (via an injection-backflow test) to evaluate changes in fracture surface area following reservoir stimulation (e.g. Fayer et al., 2009; Ghergut et al., 2007, 2010; Reimus et al., 2012). Currently, tracer tests can be used to:

- determine whether any connections exist between a newly-drilled injection-production well doublet, and if so, the hydraulic properties of the connected reservoir (fluid residence time, velocity, how 'open' the system is, flow capacity: storage capacity relationships, thermal sweep efficiencies);
- evaluate the success of a hydraulic or chemical stimulation effort – either identifying whether new hydraulic connections exist between an injector and producer or between an existing field and a previously 'dry' or isolated well, or, whether fracture surface area in the reservoir has changed (heat exchange area); or
- map flow pathways in a larger field between multiple injectors and producers, to manage injection returns and identify areas where thermal breakthrough might occur in the future if left unmanaged.

Future capabilities are expected to involve the use of 'smart' tracers in geothermal tracer testing.

4. SMART TRACERS: NEW RESEARCH DIRECTIONS IN THE USA AND EUROPE

4.1 Overview

Through the US Department of Energy's (DOE) Geothermal Technologies Program, there are several research projects in progress in the USA that are aiming to develop new technologies and techniques in tracer science. These research projects are typically focusing on the development and validation of 'smart' tracers: tracers that have the ability to provide additional information about the reservoir beyond hydraulic properties. Reservoir characteristics of interest include (1) fracture surface area, (2) matrix-fracture relationships, and (3) distributed temperature regime along a flow pathway. The ability to measure changes in fracture surface area (and thus the area available for heat exchange) is particularly important for engineered geothermal systems (EGS), and creating and maintaining connectivity of an engineered fracture network is one of the major challenges in EGS development. Thermally-sensitive and sorbing tracers may both have the capability to assess changes in fracture surface area.

4.2 Reactive tracers (solute tracers)

Solute tracers that undergo sorption/desorption reactions are currently being explored for their potential to measure changes in fracture surface areas in enhanced geothermal reservoirs (Shan and Pruess, 2005). These include cation-exchange tracers such as lithium ion (Reimus et al., 2012; Dean et al., 2013) and fluorescent compounds that exhibit both sorption behaviour and thermal decay (such as safranin-T) (Leecaster et al., 2012; Rose et al., 2012). Sorption of a reactive tracer onto geothermal reservoir surfaces results in the delay (or retardation) of the reactive tracer relative to the bulk water velocity as measured by a conservative tracer. Laboratory tests with flowing column studies have demonstrated that safranin-T reversibly sorbs at high temperatures and that its retardation is positively correlated with amount of surface area that it interrogates relative to the conservative tracer 1,5 naphthalene sulfonate (Leecaster et al., 2012). The retardation (and thus inferred-sorption) decreases as temperatures increases. Safranin-T is also thermally unstable at geothermal reservoir temperatures, and its thermal degradation has been characterised in auto-clave reactor and column flow-through experiments (Leecaster et al., 2012). A field test of the safranin-T tracer at the Soda Lake geothermal field (Nevada, USA) between two wells, indicated that the safranin-T breakthrough curve was retarded relative to the conservative naphthalene sulfonate tracer, and did show thermal decay (Rose et al., 2012).

Other solute tracers that are thermo-sensitive are being investigated by researchers in Germany. Reaction kinetics of esters derived from naphthol sulfonates are being explored and they appear to show promise as thermally-sensitive tracers under a range of reservoir conditions (temperature and pH) and tracer test durations (Nottebohm et al., 2010; 2012).

4.3 Nano-tracers (colloidal geothermal tracers)

Colloidal tracers may be able to provide additional information about geothermal reservoir properties. Particle transport in physically heterogeneous and fractured systems deviates significantly from the transport of solute species, due to the effects of particle interactions (flocculation), mechanical clogging effects, and surface reactions (e.g. sorption) (James and Chrysikopoulos, 1999). In addition, colloids may be less affected by diffusion into the matrix compared to solute tracers, and thus breakthrough of colloids can occur ahead of the breakthrough of solute tracers (McKay et al., 1993; Vilks and Bachinski, 1996, Redden et al., 2010).

A team at the University of Utah is developing a method for the fabrication of quantum dots that have potential for use in conventional and engineered geothermal systems (Bartl et al. 2009a; 2009b; Rose et al., 2010). Quantum dots are small crystallites of semiconductors in the size range of 1 to 20 nano-metres and composed of a few hundred to several thousands of atoms. Importantly, quantum dots may have the capability to perform as either conservative tracers (with customisable diffusivities) when fabricated a particular way, or as reactive tracers, either through temperature-sensing capability or sorption capabilities. As a result of quantum size effects and strongly confined excitons, quantum dots display unique size and shape-related electronic and optical properties. In particular, they can be made to fluoresce over a wide range, including the visible and near infrared (NIR) regions of light – regions where geothermal and EGS reservoir waters possess very little interference. The inorganic semi-conducting nano-crystal core of each quantum dot can be tuned to deliver various emission colours (ranging from the visible to the NIR). The surface chemistry of colloidal quantum dots can be adjusted independently, by varying the choice of ligands to optimise their interaction with the sensing environment (e.g., hydrophilic/hydrophobic, functional chemical groups, positively/negatively charged surface, etc.). Thus, it is hoped that quantum dot tracers can be designed to possess all of the qualities of the conventional solute conservative tracers (i.e. the naphthalene sulfonates), or be converted to reactive tracers depending on the surface treatment (Rose et al., 2010). Currently, the quantum dots being fabricated and tested at the University of Utah are using a cadmium selenide (CdSe) core (Siy et al., 2011) and this core is being protected by either a cadmium sulphide (CdS) or a silica shell, or combination of both, to which ligands can be attached to modify the surface chemistry (Riassetto et al., 2011). The thermal stability of these candidate quantum dots is being evaluated using autoclave batch reactor experiments at temperatures up to 300°C, and timescales of up to a week, and preliminary column flow-through experiments with comparisons to a conservative, solute tracer have already been conducted (Brauser et al., 2013). Work continues on evaluating their potential as geothermal tracers.

Researchers at Stanford University have investigated other temperature-sensitive nano-tracers including tin-bismuth alloy nano-particles and silica nano-particles with covalently linked dye (Alaskar et al., 2011; Ames, 2011). The tin-bismuth alloy has a melting temperature that varies between 139°C and 271°C depending on its composition: this is the premise for the temperature-sensing capability. Other temperature-sensitive nano-tracers are being investigated at Idaho National Laboratory, incorporating temperature-dependent processes such as mineral thermo-luminescence, or racemisation in polymers of organic compounds. Researchers are investigating the possibilities of encapsulating such temperature-sensitive minerals/ compounds in a protective silica shell to protect them from alteration in the geothermal environment and enable them to be transported through the reservoir recording its thermal signature (Redden et al., 2010).

5. SUMMARY

Geothermal tracers continue to be a useful tool for exploring and characterising geothermal reservoirs in active geothermal fields worldwide. Tracer testing should be considered during the exploration/resource characterisation stage of project developments: the benefits are significant (qualitative data on flow pathways and quantitative data on reservoir hydraulic properties) and the tests are easy to perform. New research directions in 'smart' tracers show promise in providing further insight into engineered and natural geothermal reservoirs in both conventional and unconventional geothermal settings.

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