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Advances in the Past 20 Years: Geochemistry in Geothermal Exploration, Resource Evaluation and Reservoir Management

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ABSTRACT

Most theoretical fundamentals of geothermal geochemistry were established by the mid-1980s, as were numerous practical applications of these fundamentals to geothermal resource evaluation and management. Since that time, these geochemical tools have been refined to various degrees. Advances are discussed in the categories of sampling and analysis, exploration and resource evaluation, and resource management. Noted developments include: widespread use of high-performance liquid chromatography (HPLC); advances in spectral analysis; new and refined chemical geothermometers (especially using non-condensable gas species); analysis tools that enable fluid inclusion stratigraphy; ground surface CO₂ flux measurement; integration of geochemical reaction models into numerical reservoir simulation; scaling and wellflow chemistry modification; new reservoir tracers and flow-line tracer enthalpy technology. Emphasis is placed on commercially applied technology, but academic developments are included.

Introduction

Most of the theoretical and practical fundamentals of geothermal geochemistry, its engineering applications and related chemical applications to geothermal technology had been established by the mid-1980s, and the enduring works of pioneers such as A.J. Ellis, Tony Mahon, Don White, Robert Fournier, Harold Helgeson, Alfred Truesdell, Stefán Arnórsson, Franco D'Amore and Werner Giggenbach quickly come to mind, among others. All of these scientists understood the value of practical fundamentals and how to apply them, and many of their contributions are an essential part of the

established science and technology (*e.g.* Ellis and Mahon, 1977; Giggenbach, 1980; Giggenbach, 1988; Henley and others, 1984; D'Amore, 1991; Nicholson, 1993; Arnórsson, 2000).

These contributions include chemical geothermometry, mixing models that use conservative solutes in combination with enthalpy, understanding the fundamentals of reaction kinetics and thermodynamics, relationships among geological processes, petrology, mineralogy, temperature and fluids chemistry, stable isotope behavior in geothermal systems, partitioning of dissolved species between co-existing liquid and vapor phases, the basic processes that underlie mineral scale deposition and how to control scaling and corrosion.

This document provides a brief, "post-1980s" overview of developments in geochemical theory and technology and applications of chemical tools to geothermal resource evaluation and management. The emphasis is on commercial technology, but some developments in academic research are included. It is organized in terms of several basic categories, for example fluids sampling and analysis technology, and exploration and resource evaluation tools, but of course there is overlap between them. Not included are chemical tools used in drilling, in power plants, for environmental monitoring and emissions abatement, and minerals extraction studies.

Of course it must be said that many new developments are underlain by the astounding advances in computation hardware and software of the last two decades and for field work there is that other new and priceless tool, the portable global positioning survey (GPS) meter.

Many recent developments are relatively complex but a few are closer to nuts and bolts. And some things needing progress seem to never change - the literature review for this work revealed that many "abstracts" still fail to abstract, and a few too many data maps still include automated contouring of hopelessly non-uniform data point distributions.

The references provided are examples only. Many are deliberately recent, and chosen because they include references to yet earlier developments. Accordingly, the first or early appearance of a new idea or development may not be cited, even if its origin is mentioned.

Sampling and Analysis Technology

The basics of collecting solids for chemical analysis (rocks, cuttings, cores, scales) were of course established long ago. Methods of collecting liquids, steam and gases from hot springs and wells were also established before the mid-1980s, but the equipment and methods used have become somewhat more refined and uniform. For example, the ASTM International Historical Standard E1675-95a, "Standard Practice for Sampling Two-Phase Geothermal Fluid for Purposes of Chemical Analysis," was not fully adopted until 1995, and this was superseded in 2004 by Active Standard E1675-04e1 of the same name (ASTM, 2007). The ASTM standard equipment and methods essentially come from the United States, and differ in some details from equipment and methods in New Zealand, Japan and elsewhere. Most of these differences are inconsequential, with occasional exceptions in poorer countries where old (and sometimes very well-worn) equipment would happily be replaced.

Two nuts and bolts improvements: the general adoption of Teflon stop-cock sealed steam and gas sample bottles in place of the older retractable stem design, and the PVC sewer pipe canisters in which to store and ship these bottles; these offset (just barely) the contemporary headache of shipping under IATA Dangerous Goods regulations

Downhole sampling equipment existed before the mid-1980s and occasionally was used to sample at temperatures exceeding 300°C. Under these conditions failure rates were high and sample quality was often poor. Some sampler chambers were pre-evacuated, allowing boiling; others used a flow-through design that would perform best in flowing wells and more questionably in a stagnant or low-flow condition. The current commercial choice is the Kuster Flow Through Sampler (FTS), which is specified to 230°C. During the past decade the Japanese government funded Calidus Engineering (United Kingdom) to research and design a 300°C controlled-piston-displacement tool that looked promising but was never built, and Calidus claims that a 500°C tool could be constructed given more research.

Recent significant improvements in solution analysis technology have included the increasingly wide-spread use of high-performance (or -pressure) liquid chromatography (HPLC), which yields much better data for sulfate, in particular, than previously available. The number of labs that analyze silica (SiO₂) using atomic absorption (AA) has increased (*e.g.* Lim, 2005). Mroczek and Graham (2005) have made a recent and valuable contribution to sampling and analysis for mercury in steam.

Infrared spectroscopy has been used to identify rocks and minerals for over thirty years, and portable, battery-powered instruments are now available. These promise to prove useful in the identification of alteration zones, degrees of alteration and stratigraphic correlations among cuttings samples from drill holes. Proof-of concept studies have been reported (Calvin and others, 2005). Infrared spectrometry is also gaining a role in remote (airborne and satellite) detection of soil mineralogy that is affected by geothermal leakages (*e.g.* Nash and others,

2004; and recent work by Coolbaugh and others at University of Nevada, Reno).

Equipment to measure CO₂ flux from the ground surface has recently evolved into portable systems that use an accumulation chamber and infrared gas analyzer, allowing the measurement of a site to be made within a few minutes (Lewicki and Oldenburg, 2005). This is enabling high-density surveys to detect and evaluate leakage zones, such as reported by Werner and Cardellini (2006).

The recent development of extremely sensitive metals analysis by inductively-coupled plasma mass-spectroscopy (ICPMS) has enabled investigations of trace element anomalies in soils that may (or may not) prove to be useful indicators of geothermal leakage (Hill and others, 2006).

Exploration and Resource Evaluation Tools

Chemical Geothermometry

The well-established silica (SiO₂) and cation geothermometers that use sodium, calcium and potassium to estimate temperatures of rock-water equilibration were in place by the mid-1980s, and additions that include magnesium and lithium were in place by about 1990 [Arnórsson (2000) provides a summary]. Since that time the principal geothermometry development has been a larger database and experience with their (hopefully) best use. This applies also to the sulfate-water oxygen isotope geothermometer. Silica geothermometry has benefited from increasing recognition of the dominant roles of chalcedony and quartz at lower and higher temperatures, respectively.

Occasional variants of the original defining equations continue to be published, based on selected data sets and/or refined mathematical approaches (*e.g.* Can, 2002). Some of these refinements may be of use in particular situations even though it often is changes and relative values of the calculated temperatures that are of more interest than the absolute accuracy of each estimate.

Most geothermometers based on non-condensable gases dissolved in water and/or the vapor phase have been developed more recently than the forms based on dissolved solids, and a number of contributions between about 1985 and 1998 are summarized by Arnórsson (2000). A recent addition is the H₂S geothermometer (liquid phase) of Blamey (2006).

Isotope Studies

Isotope studies have benefited from database growth, development and application experience since the mid-1980s, but only a few basic developments. The amount of research into helium isotopes, their significance as an indicator of magmatic origins, and effects of reservoir processes on the ratio ³He/⁴He is notable (*e.g.* Dobson and others, 2006), as is the application of isotopes to trace the origin of acidic fluids in geothermal systems (IAEA, 2005). Research studies into a number of other isotopes (of sulfur, carbon, argon, radon, radium, chloride, boron, strontium and potassium) have also appeared [see a brief summary in Arnórsson (2000)].

Fluid Inclusion Studies

Fluid inclusions are microscopic bubbles of water, solutes (dissolved and crystallized) and gases that are trapped in minerals and retain evidence of conditions at the time of formation (especially temperature and fluid composition), which may or may not match contemporary conditions. Traditional methods of inclusion analysis are somewhat slow, and an interesting new development is Fluid Inclusion Stratigraphy (FIS), which is made possible by automated methods of sample crushing and analysis using mass spectrometry. This method does not determine the temperature of inclusion formation, but it is hoped that the chemistry of trapped volatiles can be used to identify fractures and fluid types (Dilley and others, 2005), and during drilling to identify potentially productive wells from non-productive wells (Dilley and Norman, 2005). Up to 10,000 analyses may be returned by FIS measurements on 20 ft-interval samples in a 10,000 ft deep well (Norman and others, 2005).

Trace Element Studies

Dissolved trace elements (usually present at < 1 mg/l) have never played a major role in geothermal studies, particularly in the commercial realm. Few trace elements show patterns and/or temperature-controlled behaviors that are more useful than information provided by the more abundant solutes, particularly within commercial timelines and budgets. However, some trace metal sulfides (*e.g.* antimony, zinc) are sources of scale deposition at a few projects, and studies of arsenic have long been of interest due to issues of environmental contamination. Recent examples of trace elements applied to fluid origins and behavior are rare earth elements and yttrium in thermal waters of Jordan (Möller and others, 2006; Möller, P. in Savaşçin and Mertoğlu, 2005), and studies of trace elements in Icelandic geothermal waters (Stefánsson and Arnórsson, 2005).

Resource Management Tools

Geochemical Reaction Modeling

The chemical reaction modeling codes that appear most often in geothermal studies, such as WATCH, SOLVEQ-CHILLER, and EQ3/6 were all developed by about 1985. Subsequent improvements to these and other codes that model high-temperature processes (*e.g.* SOLMNEQ.88, TEQUIL and GEOFLUIDS) have included code enhancements, more and better thermodynamic data (some of the codes also model reaction kinetics), addition of the Pitzer method to model highly saline solutions, and for some an MS-Windows interface. The theoretical basis of reaction modeling has been summarized by Bethke (1996). Arnórsson and others (1982) and Reed and Spycher (1984) provide good summaries of the boiling problem in WATCH and SOLVEQ-CHILLER, respectively. Information about the most recent versions of the codes mentioned above is best obtained by searching the internet.

A larger development is the integration of reaction modeling with numerical reservoir simulation, the foremost example of which appears to be TOUGHREACT (Xu and others, 2005). Most published applications of TOUGHREACT so

far have been simplified case studies, but examples of more directed studies are beginning to appear. Examples include modeling the effects of injection at the Salton Sea, CA (McLin and others, 2006) and Coso, CA (Kovac and others, 2006), and the incomplete neutralization of acidic reservoir fluid at Onikobe, Japan (Todaka and others, 2005). FRACHEM is a similar code that has been developed in association with the European Enhanced Geothermal Systems (EGS) project (André and others, 2006).

These codes must be used advisedly, especially if investigating more than silica and calcite or at elevated salinity. André and others (2006) compared model predictions using six different codes and concluded that “differences in thermodynamic equilibrium constants, activity coefficients and kinetics models can result in significant differences in predicted mineral precipitation behavior and reservoir-porosity evolution. Differences in calculation schemes typically produce less difference in model outputs than differences in input thermodynamic and kinetic data, with model results being particularly sensitive to differences in ion-interaction parameters for high-salinity systems.” Xu and others (2005) have compared results of equivalent models that use different activity models, finding differences that increase with salinity and temperature.

A better understanding of the thermodynamic data is thus critical, and new applications of these data may arise. Pruess and Azaoual (2006), for example, have recently studied the feasibility of using supercritical CO₂ as a heat transmission fluid in engineered hot dry rock systems, concluding that the idea merits further consideration. Poparov (2005) has provided new experimental data on the pressure dependence of vapor/liquid distribution for Cl, Na and SiO₂. Villafañila García and others (2005) have studied the data and methods needed to better predict sulfate mineral scaling. Moller and others (2006) report progress in further understanding the chemical behavior of aluminum species in solution.

Scaling and Wellflow Chemistry Modification

Calcium carbonate scaling is now routinely controlled using inhibitors, and some applications of geothermal anhydrite scale inhibition are also appearing (*e.g.* Yanagisawa and others, 2006).

Silica over-saturation in heat exchangers, separated brines and injection wells is now commonly managed by adding acid to pH ~5.5 or occasionally using silica scale inhibitors, but formation scaling still occurs and silica deposits in high-temperature production wells remain a costly issue. Research into understanding silica deposition (*e.g.* Mroczek and others, 2000; Gunnarsson and Arnórsson, 2005; Molina Padilla and others, 2005; Potapov and others, 2005) and into improving inhibition (*e.g.* Gallup and Barcelon, 2005) is still being carried out.

The reaction transport models discussed above are increasingly being used to better understand these silica issues. For example, Kato and others (2005) have determined that pH modified injected brines may be depositing anhydrite (CaSO₄). Park and others (2006) recently evaluated amorphous silica precipitation near hypothetical injection wells under three scenarios, “acid injection,” “alternating acid and alkali injection,” and “multiple small diameter injection wells.” Their results

suggest that alternating acid and alkali injection (twenty days each was modeled) merits further evaluation.

Downhole injection of sodium hydroxide into production wells was first developed at vapor-dominated geothermal fields (Lardarello, Geysers) to neutralize small amounts of hydrochloric acid in steam (*e.g.* Bell, 1989). At Miravalles, Costa Rica, the principle has been extended to successfully and economically neutralize liquid-dominated wellflows that have pH values in the range of 2.3 to 3.2 (Moya and others, 2005). A by-product of the process is formation of anhydrite and amorphous silica scales in the wellbore which requires mechanical clean-out twice a year. It has been discovered that fine-tuning the pH can reduce this, and silica scale inhibitors are also being studied.

Tracer Developments

A reservoir tracer is a chemical compound that can be injected in some quantity into one well, and detected at others to discern pathways of permeability and reservoir fluid circulation. Until the mid-1980s the number of tracers suitable for injection into high-temperature reservoirs was very limited, and rarely was more than one injection well tested at one time. Most tracer tests used 50~100 kg of sodium fluorescein (also known as uranine), which has problems with decay caused by heat (see below), light, or oxygen, but is easily and cheaply detected at extremely low levels (0.1 ppb or less). A few tests used a larger amount (200~500 kg) of a stable salt such as potassium iodide, which is relatively expensive to analyze yet with a higher detection limit (~10 ppb). Other fluorescent tracers (*e.g.* rhodamine) were too unstable to use at higher temperatures (>~185°C). Tritium as a liquid or vapor phase tracer was being abandoned due to issues of radioactivity.

More recently, research funded by the U.S. Department of Energy has discovered that a number of organic aromatic alcohols are stable at temperatures as high as 300~340°C and suitable for use as liquid phase tracers (*e.g.* Rose and others, 2001). These compounds can be detected using HPLC, enabling as many as eight tracers to be measured simultaneously at concentrations as low as ~0.2 ppb. This allows simultaneous tests of multiple injection wells.

Examples of the aromatic tracers are toluenesulfonate, 1-naphthalene sulfonate, and several different naphthalene disulfonates (1,5-, 2,5-, 2,6- *etc.*), which cost about the same as fluorescein (~US\$5,000 or less for a single injection dose). Commercial analysis services are now available for ~\$50/analyte/sample or less, which is an order of magnitude higher than analyses of fluorescein. A typical response to the analysis cost is to reduce the number of measurements and sacrifice some detail in the tracer response curve, and the aromatics are now being used more and more widely (*e.g.* Watanabe and others, 2005).

Adams and Davis (1991) measured the thermal decay constant of fluorescein, which can now be used to estimate a correction for the amount lost between the point of injection and the point of production. The amount of decay is small below ~180°C yet becomes severe above ~250°C. Side-by-side tests using fluorescein and aromatic tracer have documented the fact that large amounts of fluorescein may disappear from

reservoir waters apart from the fraction affected by thermal degradation, at least in some locations (Gundersen and others, 2002).

Fluorescent tracers remain interesting because Rose and others (2002) have developed a highly sensitive laser fluorometer coupled with HPLC separation. This equipment has detected fluorescein and a fluorescein derivative (6-carboxy-fluorescein, also unstable at high temperatures) at concentrations as low as 40 parts per quadrillion, which enables injecting only a few kg.

Vapor phase tracers such as sulfur hexafluoride (SF₆), hydrofluorocarbons (HFCs) and hydrochlorofluorocarbons (since abandoned due to ozone depletion) have also been developed (*e.g.* Adams and others, 2001).

An interesting new alternative for vapor phase tracing is chain alcohols such as ethanol, n-butanol, i- and n-propanol and methanol, recently reported by Fukuda and others (2005), Fukuda and others (2006) and Mella and others (2006b). Compared to SF₆ and HFCs these are disadvantaged by lower thermal stability and higher detection limits. Advantages include water solubility, easy handling for injection, easier sampling, vapor pressures similar to water and a range of vapor pressures. Mella and others (2006a) have reported a 30-fold improvement in the detection limit for n-propanol, down to 1 ppb.

As reservoir tracer tests have become more and more common, interest in quantitative analysis of the results has also grown, to the extent that tracer matching is appearing in numerical reservoir simulation models (GeothermEx, proprietary).

Tracer flow testing is another new and valuable development which appeared in the mid-1990s [Hertz and others (2001), Lovelock (2006)]. This technique measures the respective flow rates of liquid and vapor in a two phase flow line to determine total flow enthalpy, by measuring dilution of liquid phase tracers (*e.g.* sodium benzoate) and vapor phase tracers (*e.g.* SF₆) between points of injection and points of sample collection.

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