^{222}Rn Production in Geothermal Fluids and Its Application to Quantifying Fracture Attributes

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ABSTRACT

The precise and accurate characterization of fracture attributes, such as spacing and surface area, in geothermal systems is essential for increasing geothermal energy production. Fracture characterization is particularly important for enhanced geothermal systems (EGS) where fracture permeability must be increased and sustained compared to pre-development conditions to make EGS economical. Natural and synthetic geochemical tracers are a promising tool for fracture characterization; however, model validation remains an unmet challenge in many cases.

We present results of from batch radon emanation experiments designed to calibrate ^{222}Rn as a tracer of fracture aperture and a preliminary model that combines the calculated aperture data with tracers of fracture spacing (e.g. $^{87}Sr/^{86}Sr$ and $\delta^{18}O$) to calculate the available fracture surface area for heat exchange. Later experiments test the model with a custom-designed, flow-through hydrothermal column with an approximately 1-liter volume. The experiments are precisely controlled for temperature, pressure, and flow rate. Our preliminary results are applied to prior published data from the Long Valley geothermal system to assess the fracture attributes and compare to other tracers.

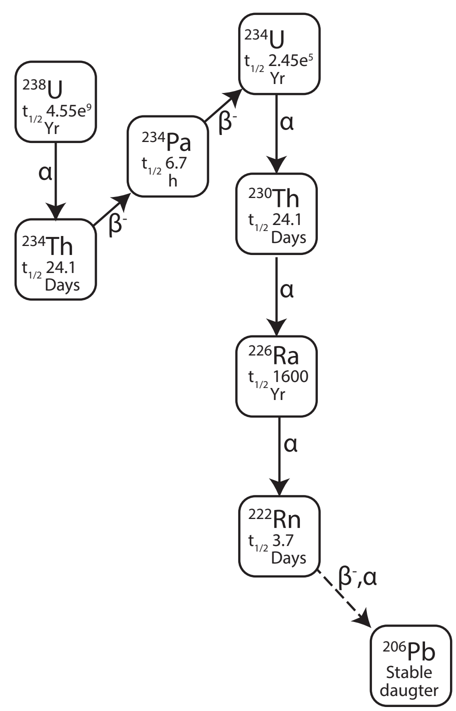

Figure 1. Schematic representation of the ^{238}U decay series. The long-lived isotopes of ^{234}U and ^{226}Ra are the most important parent isotopes that can lead to isotopic disequilibrium in the decay series. Daughter isotopes of ^{222}Rn are omitted except for the final, stable isotope ^{206}Pb for brevity.

1. Introduction

Sustainable energy production from enhanced geothermal systems (EGS) requires an understanding of fracture attributes such as spacing, aperture and surface area. Direct observation of fracture properties from drilling is impractical and difficult to extrapolate core scale observations to Km scale geothermal systems. The development of geochemical tracers for inferring fracture properties is well-established (Rose et al., 2011; 2001). While injected tracers are very promising for interrogating fracture attributes it is difficult to validate the results without complimentary measurements that are not dependent on the same underlying theory. We propose the use of natural variations in isotopic ratios such as ^{222}Rn, $^{87}Sr/^{86}Sr$, and $\delta^{18}O$ should be useful for characterizing the physical attributes of geothermal systems.

Variations in stable and radiogenic isotopes in geothermal systems offer a possibility to calculate fracture attributes from the natural water-rock reaction. Geothermal fluids are frequently out of isotopic equilibrium with the reservoir rock, however, chemical reactions between the fluid and reservoir rock will cause a measureable change in the fluid composition along a reactive flow path. The rate of change in the fluid composition for multiple isotopic ratios can be used to infer the spacing between the primary fluid-bearing fractures (Brown et al., 2013; DePaolo, 2006).

Calculating fracture surface properties such as surface area and aperture require tracers that do not sample the rock matrix between fractures. In the case of injected tracers this is commonly achieved with an adsorbing tracer such as Cs or Li, which have to be calibrated for their respective exchange behavior as a function of temperature and reservoir rock type. For natural isotopic tracers short-lived radioisotopes offer a compliment to adsorbing tracers. For example ^{222}Rn has a half-life of ~3.8 days, meaning that only atoms within ~1-2 cm of the fracture surface can possibly diffuse fast enough to be sampled in the fracture fluid. This means that the ^{222}Rn concentration in the fluid is related to the parent isotope abundance (^{226}Ra) and the fracture surface area to volume ratio(Nelson et al., 1983), also known as the fracture aperture.

Prior studies have utilized ^{222}Rn in groundwater to quantify fracture properties, however, they were not completely successful, due to uncertainties about the distribution of parent isotopes (^{226}Ra, ^{234}U and ^{238}U), possible effects of well pumping, and the general limits of extrapolating a conceptual theory to field-scale studies (Folger et al., 1997; Le Druillennec et al., 2010; Torgersen, 1980).

Our proposed study aims to bridge earlier theoretical and field studies of ^{222}Rn in geothermal systems with quantitative lab-based experiments and current generation reactive transport modelling to fully account for the sources of ^{222}Rn to geothermal fluids. The modelling results will produce calculated fracture apertures, and when coupled with traditional isotopic tracers such as $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ the models will calculate the fracture spacing and surface area as well.

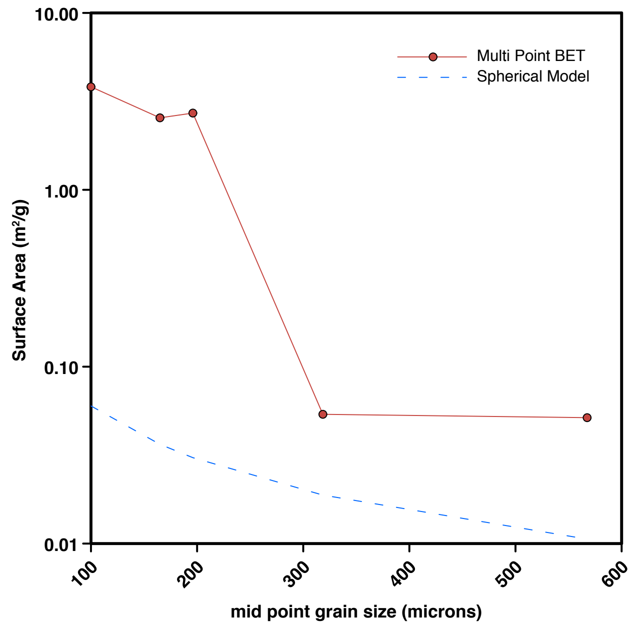

Figure 2. Surface area of the Bishop Tuff size fractions as determined by BET. The dashed blue curve gives the surface area of perfect spheres for reference. The steep drop off between the ~212 micron grain size is consistent with the visual shift from glass dominated mineralogy to lithic fragment and phenocryst dominated mineralogy.

2. Experimental Material Characterization

Samples of air fall Bishop Tuff were sieved into discrete size fractions including 63-150 µm, 150-180 µm. 180-212 µm, and 212-425 µm. The size fractions were then characterized for surface area, mineralogy, and U-series isotope activities. The surface area was determined by Brunauer–Emmett–Teller (BET) gas adsorption, and the results are reported in Fig 2. The <212 µm size fractions have surface areas >1 m^2g^{-1} while the larger size fractions have less than 10% of the surface area per gram (<0.1m^2g^{-1}). For reference, the surface area of perfect spheres as a function of grain size is also shown in Fig 2. The ~50x difference between the measured surface areas in the <212 fraction compared to spheres provides a quantitative assessment of the high surface area nature for the volcanic glass.

Additional SEM images of the selected size fractions were used to visually verify the presence of volcanic glass and confirm the high surface area nature of the sieved grains. Fig 3 shows representative images of volcanic glass fragments from the 150-180 µm size fraction.

Since ^{222}Rn is a daughter isotope of the ^{238}U decay series the abundance of the parent isotopes is important for interpreting any measured ^{222}Rn in fluid-rock systems. We measured the activities of ^{226}Ra, and the $^{234}U/^{238}U$ isotopic ratio. The $^{234}U/^{238}U$ is reported relative to secular equilibrium which is defined as $A^{234}U=A^{238}U$, where A is the isotope activity, which is the number of atoms (n) multiplied by the isotope decay constant (λ). The $^{234}U/^{238}U_{AR}$ for all analysed glasses are near secular equilibrium with the largest

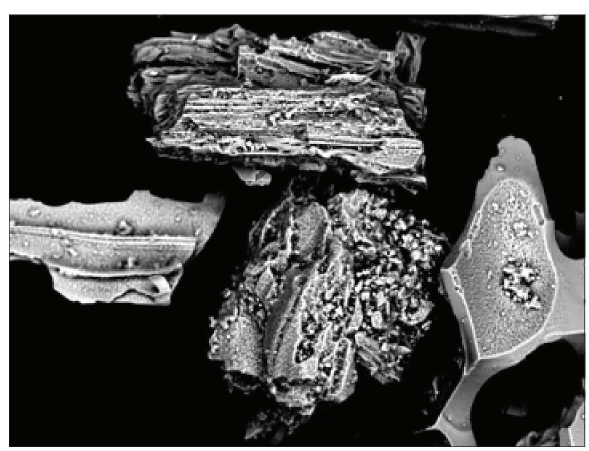

Figure 3. 150-180 um fragments of Bishop Tuff volcanic glass.

enrichment ~1%. The ^{226}Ra activities for the sieved tuff fractions range between 28 Bq/Kg in the >212 micron fractions to 87 Bq/Kg in the <63-150 micron fraction. Isolated Fe-Ti oxides have the highest ^{226}Ra activity (128-183 Bq/Kg).

^{222}Rn activities were measured for batch samples in both dry and water saturated emanation experiments. All ^{222}Rn activities were measured using a RAD7 radon detector (Durridge; Billerica, MA). The RAD7 measures short-lived polonium isotopes in equilibrium with ^{222}Rn to quantify ^{222}Rn activity. A dry emanation experiment resulted in 0.87 Bq/Kg, or 1.3% of the ^{226}Ra activity, while the water saturated emanation experiment yielded 4.70 Bq/Kg or 7% of the ^{226}Ra activity.

3. Model Development

The ^{222}Rn activity of fluids can be related to the fluid-rock ratio because the ^{222}Rn production is related to the rock or mineral surface area relative to the fluid volume. Depending on other quantifiable parameters this theoretically allows for calculating the pore size (or fracture aperture). Emanation from mineral grains to the surrounding pore space (air or water) can occur by alpha recoil and by diffusion. The percentage of radon that is emitted from minerals compared to the amount that decays inside minerals is the emanation factor (E). In the case of alpha recoil only, emanation (E_r) is a function of the recoil length relative to the mineral grain size. In most silicate minerals the recoil distance is ~30-40 nm, 95 nm in water and 65 µm in air (Semkow, 1990). Thus the E_r is a function of the grain size and shape. For spherical geometries E_r can be calculated based on the recoil length (L) and the grain radius (r)(DePaolo et al., 2006; Kigoshi, 1971; Semkow, 1990):

$$E_r = \frac{3}{4}\left(\frac{L}{r} - \frac{L^3}{12r^3}\right) \tag{1}$$

The spherical grain model can underestimate E_r because most mineral grains are not spheres and have oblate or similar geometries with greater surface area. Also grain size distribution is important, as smaller radius grains will have disproportionally high E_r compared to larger grains. An alternate method to estimating E_r is to use the BET-derived surface areas:

$$E_r = \frac{1}{4} LS\rho \tag{2}$$

where S is the surface area ($cm^2\ g^{-1}$), and p is the density ($g\ cm^{-3}$). Using the BET measured surface areas for the Bishop Tuff this yields E_r ~0.04-0.06, similar to our measured values of E_r.

Additionally diffusion of ^{226}Ra and ^{222}Rn from the interior of mineral grains to the mineral surface-fluid interface can also contribute to fluid ^{222}Rn budget. For ^{222}Rn the important parameter is the diffusivity relative to the isotope decay constant.

$$L_{Rn} = \frac{D_{Rn}\phi_m}{\lambda_{222Rn}} \tag{3}$$

Where L_{Rn} is the radioactive decay length, D_{Rn} is the diffusivity, ϕ_m is the matrix porosity and λ_{222Rn} is the isotope decay constant. Using EQ 3 we can Equation 3 allows us to calculate the contribution s

Unlike porous flow environments, fracture dominated systems have a somewhat simpler relationship between surface area and ^{222}Rn production. The water-saturated fractures with apertures >0.1 µm are not substantially affected by implantation (i.e. ejection of a 222Rn atom from one grain and incorporation into the adjacent grain), thus the fracture surface area to volume ratio largely dictate the fracture fluid ^{222}Rn activity. Nelson and co-workers (1983) developed a "thin crack" model that can be summarized as:

$$h = \frac{E}{\lambda C_{rn}} \tag{4}$$

Where h is the fracture aperture, E is the emanation factor, λ is the decay constant is C is the measured ^{222}Rn activity. The thin crack model is additionally summarized in Fig 4. For a fixed value of E the measured C_{rn} in a fracture fluid decreases as the fracture aperture increases.

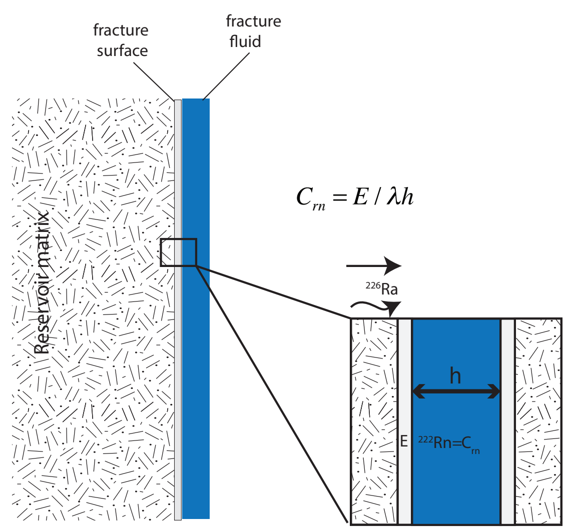

Figure 4. A schematic representation of the relationship between fracture aperture and the fluid radon activity.

4. Example Application to the Long Valley Geothermal System

The Long Valley geothermal system provides an opportunity to test the application of ^{222}Rn to fracture properties. There are a few published data from thermal springs and many additional geochemical data published over the last ~35 years(Brown et al., 2013; Evans et al., 2013; Sorey et al., 1991; Wollenberg et al., 1984). The ^{222}Rn activity at Casa Diablo is 23 pCi/L and nearby Hot Bubbling Pool is 43 pCi/L. Using the fracture model presented above this yields a fracture aperture of ~2.5 mm. In contrast, the ^{222}Rn activity at Little Hot Creek is 319 pCi/L, which yields a fracture aperture of ~0.25 mm.

The discrepancy in apparent fracture apertures from the Long Valley geothermal system could arise from variable amounts of fluid degassing before reaching the surface. Springs in Long Valley have experienced various amounts of degassing(Brown et al., 2013). Alternately, there could be a change in the concentrations of U in the host rock yielding differences in E_r. Order of magnitude changes in E_r seem unlikely given the relatively homogenous lithology in the caldera. Finally the large range in ^{222}Rn activity could be due to fluids sampled from fractures with very different apertures.

Brown and coworkers(2013) independently estimated the fracture aperture in Long Valley from the inferred fracture spacing and the volume of fluid discharge to be 0.05-0.25 mm, similar to the calculated values from the Little Hot Creek ^{222}Rn activity, suggesting that the Casa Diablo and Hot Bubbling Pool data maybe affected by fluid degassing. Sampling from production or monitoring wells where single-phase fluids can be sampled could resolve this apparent discrepancy.

Finally, the combined fracture spacing and fracture aperture data can be used to estimate the fracture surface area in the Long Valley geothermal reservoir. Using the reservoir model size from Brown et al (2013), the 0.25 mm fracture size, and the 10 m fracture spacing we calculate a total fracture surface area of 10^5 m^2/m.

References

Brown, S.T., Kennedy, B.M., DePaolo, D.J., Hurwitz, S., Evans, W.C., 2013. Ca, Sr, O and D isotope approach to defining the chemical evolution of hydrothermal fluids: Example from Long Valley, CA, USA. Geochimica et Cosmochimica Acta 122, 209–225.

DePaolo, D.J., 2006. Isotopic effects in fracture-dominated reactive fluid–rock systems. Geochimica et Cosmochimica Acta 70, 1077–1096. doi:10.1016/j.gca.2005.11.022

DePaolo, D.J., Maher, K., Christensen, J.N., McManus, J., 2006. Sediment transport time measured with U-series isotopes: Results from ODP North Atlantic drift site 984. Earth and Planetary Science Letters 248, 394–410. doi:10.1016/j.epsl.2006.06.004

Evans, W.C., Hurwitz, S., Bergfeld, D., Lewicki, J., Huebner, M.A., Williams, C.F., Brown, S.T., 2013. Water-rock interaction in the Long Valley Caldera (USA). Procedia Earth and Planetary Science 7, 252–255.

Folger, P.F., Poeter, E., Wantye, R.B., Day, W., Frishman, D., 1997. 222 Rn transport in a fractured crystalline rock aquifer: results from numerical simulations. Journal of Hydrology 195, 45–77. doi:10.1016/S0022-1694(96)03243-X

Kigoshi, K., 1971. Alpha-Recoil Thorium-234: Dissolution into Water and the Uranium-234/Uranium-238 Disequilibrium in Nature. Science 173, 47–48. doi:10.1126/science.173.3991.47

Le Druillennec, T., Ielsch, G., Bour, O., Tarits, C., 2010. Hydrogeological and geochemical control of the variations of 222 Rn concentrations in a hard rock aquifer: insights into the possible role of fracture-matrix exchanges. Applied …. doi:10.1016/j.apgeochem.2009.12.002

Nelson, P.H., Rachiele, R., Smith, A., 1983. Transport of radon in flowing boreholes at Stripa, Sweden. Journal of Geophysical Research: Solid Earth (1978–2012) 88, 2395–2405. doi:10.1029/JB088iB03p02395

Rose, P., Riasetto, D., Siy, J., Bartl, M., 2011. Quantum dots as tracers in geothermal and EGS reservoirs. … on Geothermal ….

Rose, P.E., Benoit, W.R., Kilbourn, P.M., 2001. The application of the polyaromatic sulfonates as tracers in geothermal reservoirs. Geothermics. doi:10.1016/S0375-6505(01)00024-4

Semkow, T.M., 1990. Recoil-emanation theory applied to radon release from mineral grains. Geochimica et Cosmochimica Acta 54, 425–440. doi:10.1016/0016-7037(90)90331-E

Sorey, M.L., Suemnicht, G.A., Sturchio, N.C., 1991. New evidence on the hydrothermal system in Long Valley caldera, California, from wells, fluid sampling, electrical geophysics, and age determinations of hot-spring …. Journal of Volcanology … 48, 229–263. doi:10.1016/0377-0273(91)90045-2

Torgersen, T., 1980. Controls on pore-fluid concentration of< sup> 4</sup> He and< sup> 222</sup> Rn and the calculation of< sup> 4</sup> He/< sup> 222</sup> Rn ages. Journal of Geochemical Exploration 13, 57–75. doi:10.1016/0375-6742(80)90021-7

Wollenberg, H.A., Smith, A.R., Mosier, D.F., Flexser, S., Clark, M., 1984. Radon-222 in groundwater of the Long Valley caldera, California. PAGEOPH 122, 327–339. doi:10.1007/BF00874602