Numerical modeling of temperature-reporting nanoparticle tracer for fractured geothermal reservoir characterization

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1/1	Highlights							
17								
15	• Temperature-reporting nanoparticle tracer tests are numerically implemented in a three-							
16	dimensional fractured geothermal reservoir.							
17	• The working mechanisms of the temperature-reporting nanoparticle tracers are illustrated							
18	through field simulations using a proposed modeling approach.							
19	• Our proposed analysis curves can provide responses to the reservoir temperature							
20	distribution as well as geological and thermal heterogeneities based on the tracer							
21	breakthrough data.							

Abstract 22

Information on the temperature distribution of subsurface reservoirs is essential for geothermal 23 24 energy development. One of the promising tools to detect the reservoir temperature distribution is temperature-reporting nanoparticle tracers whose functionality has been extensively investigated 25 26 in both theoretical and experimental ways in the last decade. However, most related studies were limited to simplified geometries and ignored the dynamic interplays of fluid flow, heat transfer, 27 28 transport and reaction of the temperature-reporting nanoparticle tracer. The response behavior and working mechanisms of such nanotracers in a realistic three-dimensional system still have not been 29 fully revealed through a systematic study. In this work, we develop a numerical modeling approach 30 to simulate field implementation of these nanotracers in a fractured geothermal reservoir. This 31 32 study aims to evaluate whether the injection of multiple temperature-reporting nanoparticle tracers with different thresholds can be used to estimate the temperature distribution and provide 33 information on the thermal and geological heterogeneities. Several scenarios have been 34 investigated for the geothermal reservoir including homogeneous and non-homogeneous cases 35 (e.g., thermal and geological heterogeneities). Our obtained results from the nanotracer 36 breakthrough curves show that the deviation temperatures in peak concentration values provide an 37 upper limit of the lowest temperature and precise highest temperature for the reservoir temperature 38 range. The deviation temperature of the peak arrival time curve accurately estimates the highest 39 temperature along the main streamlines between the wells. The proposed analysis curves based on 40 the nanotracer breakthrough data were visibly affected by geological heterogeneities including 41 their conductivities and orientations as well as thermal heterogeneities in the geothermal reservoir. 42 Keywords: nanoparticle; tracer; temperature-reporting; geothermal; reservoir characterization; 43 tracer breakthrough curve

1 Introduction 45

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46 Geothermal energy is a clean, renewable and sustainable alternative to traditional energy sources of fossil fuels for direct heat utilization and electricity generation (Moeck, 2014). 47 48 According to the World Geothermal Congress (2023), a total of 1,476 PJ (410 TWh) of geothermal energy was used globally in 2022, a 44% increase from 2020, with geothermal heating and cooling 49 50 of buildings accounting for about 79% of the total. While major geothermal systems are restricted to structurally dynamic or volcanically active regions (Lund et al., 2008; J. W. Tester et al., 1989), 51

enhanced geothermal system (EGS) technology radically expands the global geothermal potential 52 through hydraulic/thermal/chemical stimulation on target reservoirs to enhance permeability and 53 fluid flow rate, in impermeable and low heat flow regions. Nonetheless, to improve the economic 54 benefits and reduce investment risk in geothermal projects, reservoir characterization is 55 particularly needed to evaluate subsurface heat/energy utilization capabilities in terms of 56 geothermal power generation, as well as the management, maintenance and sustainability of 57 operational plants (Domra Kana et al., 2015; S.-M. Lu, 2018; Olasolo et al., 2016). In general, 58 59 geothermal reservoir characterization includes assessing reservoir conditions such as estimating temperature profiles (i.e., temperature range and distribution), surveying well-to-well or inter-well 60 connectivity (Dashti et al., 2023) and extrapolating the volume of the fractured zones (B. Sanjuan 61 et al., 2006). Among all reservoir characteristics, temperature profiles are critical for geothermal 62 63 energy exploration and assessment in the geothermal reservoir (re-)siting step, as well as for evaluating reservoir thermal performance (e.g., thermal breakthrough prediction due to reinjection 64 65 of cooled geothermal fluid) and adjusting production strategy in the production step. Nevertheless, the reliable and accurate measurement or estimation of temperature profiles throughout the 66 67 lifecycle of geothermal energy development has always been a major, complex and difficult challenge for geothermal reservoirs (Frey et al., 2022). 68

69 Common methods to measure or estimate temperature profiles include wellbore 70 measurements, geothermometers, analytical/numerical models and tracer tests. Temperature 71 measurements at or near the wellbore do not provide temperature distributions between injection and production wells due to the limited space of the wellbore in the geothermal reservoir. Since 72 the introduction of geothermometers in the 1960s, geothermometer technology has continued to 73 advance in-situ temperature measurements by evaluating sensitive parameters with respect to the 74 75 chemical equilibrium of fluids and reservoir rocks (e.g., aluminum concentration, pH, vapor loss, 76 etc.) (Fournier & Rowe, 1966; Nitschke et al., 2017; B. Sanjuan et al., 2014; Ystroem et al., 2020). 77 However, the spatial distribution of reservoir temperature is still unknown using geothermometers. Gringarten et al. (1975) presented an analytical solution for temperature determination, based on 78 pure fluid flow and heat transfer in a hot-dry rock reservoir with infinite, equidistant and parallel 79 80 fractures. Following that, many researchers (P. Cheng, 1979; A. H.-D. Cheng et al., 2001; Fox et al., 2016; Tang et al., 2020; Wilkins et al., 2021) continued to develop analytical solutions and 81 numerical simulators to predict the temperature distribution in fractured geothermal reservoirs. 82

Nevertheless, with the large uncertainties in the geometry of fractures (e.g. aperture, scale, spacing 83 and network) and heat transfer of unfractured zones, their results can only serve as references in 84 practice, although these results are sometimes informative. To compensate for such shortcomings, 85 conservative solute tracers (Erol et al., 2022; Pollack et al., 2021; Robinson, 1985; Robinson & 86 Tester, 1984; Williams et al., 2010; Xu et al., 2022) and adsorbing solute tracers (C. Dean et al., 87 2012; Hawkins et al., 2018; Leecaster et al., 2012; Williams et al., 2013) which are able to adhere 88 to the fracture surface were used in field tests to identify key geometry properties (i.e., heat transfer 89 surface area, fracture aperture, etc.) of geothermal reservoirs, assisted by fitting analytical 90 solutions of tracer transportation. Utilizing the obtained geometry properties, the reservoir 91 temperature distribution is roughly estimated by using analytical or numerical models for heat 92 transfer (Robinson, 1985; Williams et al., 2013). In addition, temperature-dependent degrading 93 94 (Arrhenius reaction kinetics) solute tracers are proposed to infer the average reservoir temperature in fields by analytically fitting the tracers' degradation characteristics (Plummer et al., 2010; 95 96 Plummer et al., 2011; P. E. Rose, 1994; J. W. Tester et al., 1987). It should be noted that the reaction rates of such kind of tracers vary with the environmental temperature and this method 97 98 only provides the average reservoir temperatures without spatial temperature information in a single test. Although solute tracers have been used in many fields for temperature estimation in 99 100 fractured geothermal reservoirs, the combined effects of their diffusion and interaction with 101 reservoir rocks as well as highly mineralized reservoir fluids give rise to less reliable tracer tests 102 due to the high mass loss, low detectability and collectability of the solute tracers (Aydin et al., 2022; Rudolph et al., 2020; Vitorge et al., 2014). 103

To eliminate deficiencies in solute tracer tests, nanoparticles with controllable size, structure 104 and physical and chemical properties are utilized to measure or estimate temperature profiles in 105 106 geothermal reservoirs by different transport (e.g., low diffusion) and working mechanisms from 107 solute tracer (Divine & McDonnell, 2005; X.-Z. Kong et al., 2018; Redden et al., 2010). One kind of tracer named 'temperature-sensitive nanotracer' is attractive and utilized to detect thermal 108 drawdown and average temperature by quantifying the extent of tracer degradation. The 109 degradation starts from a certain temperature threshold and its rate is influenced by the 110 environmental temperature (Axelsson et al., 2001; Nottebohm et al., 2012; Robinson et al., 1988)). 111 Many theoretical and experimental studies have been done to investigate the performance behavior 112 of temperature-sensitive nanotracers. For example, theoretically, Ames et al. (2015) used the 113

analytical solution of tracer distribution in the one-dimensional model to inversely predict the 114 thermal drawdown. However, their work does not consider the dynamic interplays of heat transfer, 115 fluid flow, transport and reaction of the nanotracer. In addition, Alaskar et al. (2015) analytically 116 and experimentally exhibited the prospects of temperature-sensitive nanotracers for forecasting 117 the thermal drawdown. In 2021, a field demonstration of temperature-sensitive nanotracers by 118 Hawkins et al. (2021) showed that the estimated effective inter-well reservoir temperatures have 119 an error of less than 5°C from the true values. However, the above studies have not demonstrated 120 121 how inter-well test results from temperature-reporting nanotracers are related to geological heterogeneities, temperature distributions and well positioning. 122

Different from the degradation principle of a temperature-sensitive nanotracer after reaching 123 the temperature threshold, a novel tracer called a 'temperature-reporting nanoparticle tracer' (also 124 125 'temperature-reporting nanotracers' for simplicity) can be quickly, fully and irreversibly converted when the environmental temperature reaches a certain threshold, are being studied to characterize 126 127 the temperature distribution of geothermal reservoirs (Puddu et al., 2016; Rudolph et al., 2020). It was Williams et al. (2010) and Alaskar et al. (2011) who first introduced a dye-release mechanism, 128 129 wherein encapsulated dyes are released from the nanotracer upon reaching a specific temperature threshold. France et al. studied polymer microcapsules encapsulating dyes that release dyes at a 130 131 certain temperature threshold. Alaskar et al. (2015) experimentally developed irreversible thermochromic microspheres and dye-attached silica nanoparticles and exhibited their prospect as 132 133 temperature sensors for forecasting the thermal drawdown analytically and experimentally. Puddu 134 et al. (2016) invented the submicrometer-sized particle, demonstrating for the first time the feasibility of using nucleic acid damage quantitatively to measure temperature. Rudolph et al. 135 (2020) conducted experiments to develop temperature-reporting nanotracers by silica particles 136 synthesized with the core-shell-hull layers. The outer dye in the nanotracer is released irreversibly 137 138 once the environmental temperature is above its thresholds, giving rise to changes in the structure of the developed nanoparticle tracers. Nevertheless, these works primarily focused on concept 139 development, laboratory research and analytical analysis. 140

The motivation of this work is to investigate whether temperature-reporting nanotracers can be used for the characterization of 3D geothermal reservoirs related to reservoir temperature distribution prediction, and provide methodologies that when applied enable the development and exploitation of geothermal energy resources. Presently, there have not been theoretical studies to

reveal the working behavior of the temperature-reporting nanotracers, nor have these nanotracers been tested in realistic 3D geothermal reservoirs. The mutual interplay among fluid flow, heat transfer, transport and reaction of the temperature-reporting nanotracers and reservoir heterogeneity necessitates a detailed investigation. In addition, how to design and implement a temperature-reporting nanotracer test in geothermal reservoirs is also questionable. Therefore, numerical simulation can be utilized as a useful approach to shed light on these issues and help us gain insight into the potential of implementing temperature-reporting nanotracers in the field.

152 The goal of the current work is to study the working mechanisms (i.e., transport, reaction, distribution and resulting concentration breakthrough curves) of temperature-reporting nanotracers 153 in synthetic but typical 3D fractured geothermal reservoirs as well as their performance in the 154 detection of reservoir temperature distributions through analyzing its breakthrough data. To 155 156 achieve that, a numerical modeling approach is developed for the reaction of temperature-reporting nanotracers. The novelty of this work is the application of a new analysis method based on the 157 158 peak information of nanotracer breakthrough curves proposed to estimate the temperature along the tested injection-production positions and reservoir temperature range of the fractured 159 160 geothermal reservoir.

The paper is organized as follows. We first present the employed methodology for modeling 161 temperature-reporting nanotracer transport in fractured geothermal reservoirs, including the 162 reaction mechanism of temperature-reporting nanotracers and numerical modeling approaches. 163 164 Secondly, the potential of temperature-reporting nanotracers in fractured geothermal reservoirs and the effect of well configuration on the temperature-reporting nanotracers' responses are 165 evaluated in a homogeneous model. Finally, the thermal distributions (i.e., different temperature 166 gradients and regional thermal anomalies) and effects of reservoir heterogeneity (i.e., embedded 167 by inclined zones) within the geothermal reservoir are studied. 168

169 2 Materials and Methods

A liquid solution is injected into the fractured geothermal reservoir through the injection well for a short period, followed by pure water injection the rest of the time. The solution is a mixture of water and temperature-reporting nanotracers with different temperature thresholds. The temperature-reporting nanoparticle nanotracers being simulated are representative of silica particles synthesized with core-shell-hull layers developed to characterize temperature distribution

when the environmental temperature reaches its threshold (Rudolph et al. (2020)). The nanotracer
breakthrough data are monitored in the production well during the injection process.

The modeled physical and chemical processes consist of fluid flow, heat transfer, transport 177 and the reaction of temperature-reporting nanotracers in the reservoir. The numerical models for 178 simulating fully coupled processes of fluid flow, heat transfer, transport and reaction for 179 temperature-reporting nanotracer are developed for the first time and implemented in the finite 180 element simulator-PorousFlow module (Wilkins et al., 2021) within the MOOSE framework 181 (Permann et al., 2020). The relevant equations are described in detail below. The numerical 182 algorithm and validation of fully coupled processes among fluid flow, heat transfer, transport and 183 reaction for temperature-reporting nanoparticle tracers are given in Appendices I and II, 184 respectively. 185

186 **2.1 Governing Equations**

187 **2.1.1 Fluid Flow and Heat Transfer**

Firstly, the fluid mass balance equation (Cacace & Jacquey, 2017) for compressible and liquid-phase water flow in porous media is written as:

190
$$\frac{\partial(\phi\rho_w)}{\partial t} + \nabla \cdot (\rho_w \boldsymbol{u}_w) - Q_w = 0$$
(1)

where ϕ is the porosity (-) of the porous medium, *t* represents time (s), the subscript *w* refers to water, p_w is the pressure (Pa), *T* is the temperature (K), $\rho_w = \rho_w(p_w, T)$ is the water density (kg·m⁻³) as a function of pressure and temperature, u_w is the Darcy velocity (m·s⁻¹) and Q_w is the water mass source (kg·m⁻³·s⁻¹).

195 The Darcy velocity $\boldsymbol{u}_{\boldsymbol{w}}$ (Qiao et al., 2018) is given as:

196
$$\boldsymbol{u}_{\boldsymbol{w}} = \frac{k}{\mu_{\boldsymbol{w}}} (-\nabla p_{\boldsymbol{w}} + \rho_{\boldsymbol{w}} \boldsymbol{g})$$
(2)

where *k* is the reservoir permeability (m²), $\mu_w = \mu_w(p_w, T)$ refers to the water viscosity (Pa s) as a function of pressure and temperature, and *g* is the gravitational acceleration (m·s⁻²).

Secondly, the heat transfer equation (T. Kohl & Rybach, 1996) for both solid and water in theporous media is written as:

201
$$\left[\phi c_{p,w} \rho_w + (1-\phi) c_{p,s} \rho_s\right] \frac{\partial T}{\partial t} - \left[\phi \lambda_w + (1-\phi) \lambda_s\right] \nabla^2 T + \rho_w c_{p,w} \boldsymbol{u}_w \nabla T - Q_T = 0$$
(3)

where the four terms on the left side individually represent a transient variation of temperature, heat conduction, heat convection and heat source. The subscript *s* represents the solid phase, $c_{p,w}$

and $c_{p,s}$ are separately the specific heat capacity of water and solid (J·m⁻³·K⁻¹), ρ_s denotes the solid density (kg·m⁻³), *T* is the temperature (K), λ_w and λ_s refer to the heat conductivity of the water and solid (W·m⁻¹·K⁻¹) and Q_T is the heat source (W·m⁻³).

207 **2.1.2 Reaction and Transport of Temperature-reporting Nanoparticle Tracers**

208 (a) Reaction Process

Figure 1 illustrates the working mechanism for reporting temperature information of the 209 temperature-reporting nanotracer which are silica particles synthesized with the core-shell-hull 210 layers. Specifically, the hull melts/degrades, dyes are released and the structure of the developed 211 212 tracer (reactant) changes when the environment temperature exceeds the temperature threshold of the temperature-reporting nanotracer. The product resulting from this temperature-dependent 213 reaction is our research focus. The released dyes only act as indicators of the reaction completion 214 degree in the lab, without directly reflecting temperature changes (Rudolph et al., 2020). In 215 216 addition, dyes may interact with highly mineralized reservoir fluids and adsorb onto the reservoir 217 rock. Therefore, in this study released dyes are not involved in the temperature reporting mechanism. 218





In the following, we present our numerical approach for simulating the temperature-reporting

nanotracers reaction. Referring to Figure 1, the relevant reaction can be described as follows:

225
$$C_{T_{thre.}}^{reac.} \xrightarrow{T > T_{thre.}} C_{T_{thre.}}^{prod.}$$
 (4)

where $C_{T_{thre.}}^{reac.}$ denotes the concentration of reactant and $C_{T_{thre.}}^{prod.}$ is the corresponding product's concentration after the reaction. $T_{thre.}$ refers to the temperature threshold of the reactant.

228 (b) Transport Process

mmod

Each temperature-reporting nanotracer with a certain temperature threshold has a group of the two advection-diffusion equations (Shan & Pruess, 2005) for both reactant and product:

231
$$\frac{\partial C_{T_{thre.}}^{reac.}}{\partial t} - \nabla \cdot (\boldsymbol{D} \nabla C_{T_{thre.}}^{reac.}) + \nabla \cdot (\boldsymbol{u}_{\boldsymbol{w}} C_{T_{thre.}}^{reac.}) - Q_{C_{T_{thre.}}^{reac.}} = 0$$
(5)

232
$$\frac{\partial c_{T_{thre.}}^{prod.}}{\partial t} - \nabla \cdot (\boldsymbol{D} \nabla C_{T_{thre.}}^{prod.}) + \nabla \cdot (\boldsymbol{u}_{\boldsymbol{w}} C_{T_{thre.}}^{prod.}) - Q_{C_{T_{thre.}}^{prod.}} = 0$$
(6)

where **D** refers to the diffusion coefficient (m²·s⁻¹), \boldsymbol{u}_{w} is the Darcy velocity (m·s⁻¹), $Q_{C_{T_{thre.}}^{reac.}}$ and $Q_{C_{T_{thre.}}^{prod.}}$ represent the mass source of reactant and product (kg·m⁻³·s⁻¹), respectively. Here we assume that the nanotracers are well mixed with water as components of the liquid solution and the gravity segregation between water and nanotracer is ignored due to the low mass fraction (<10⁻ ³) of nanotracers in the liquid. In addition, for simplicity we do not consider the deposition and aggregation of nanotracers during the flow process.

The detailed discretization and algorithm for solving the fully coupled processes of fluid flow, heat transfer, transport and reaction of temperature-reporting nanoparticle tracers are given in **Appendix I**.

242 **2.2 Model Description and Input Data**

Our model is inspired by a typical fractured reservoir setting such as Soultz-sous-Forêts EGS 243 (Egert et al., 2020) which contains several irregularly distributed fractures. In this study, a highly 244 permeable and thin reservoir is used to mimic a fracture ($\phi = 1$). We use the model settings 245 illustrated in Figure 2 (a). The reservoir model consists of two types of 600 m thick rocks: an inner 246 stimulated (called 'inner reservoir') and an outer non-stimulated reservoir (called 'outer 247 reservoir'). Both are covered by low permeable caprock and underlain by a low permeable 248 bedrock. The ground surface temperature is assumed to be 20°C and the initial geothermal gradient 249 is 0.05°C·m⁻¹. The initial pressure distribution is based on the hydrostatic gradient. For the model 250 boundary conditions, we use a constrained (initial) pressure and (initial) temperature at the top (2 251 252 km depth) and a constant (initial) temperature at the bottom (3.6 km depth). The other facies of the model are set with closed boundaries. 253

We assume two wells into the inner reservoir, each having three possible injection/production points, thus leading to nine model configurations with different fluid/tracer schemes. The reservoir

depth is between -2.5 km to -3.1 km and extending -0.5 km to 0.5 km horizontally. The unstructured mesh consisting of tetrahedral elements was created by the GMSH software (Geuzaine & Remacle, 2009). The element size differs between 1 m (around the wells) and 400 m (close to the boundaries) with a typical element size of 25 m inside the inner reservoir. Mesh sensitivity analysis is shown in **Appendix III**. The physical properties of rock and fluid are summarized in **Table 1** and **Table 2**, respectively.



reservoir reservoir
Figure 2. The reservoir model was used in this study. (a): A thin and homogeneous reservoir
(permeability=5·10⁻¹¹ m²) located within a 3D model. (b): Three injection (I1, I2, I3) and three production (P1,
P2, P3) well positions embedded in the simplified reservoir model. The dimensions of the inner reservoir are (1
km·0.6 km·1 m) and the initial reservoir temperature distribution ranges from 145°C to 175°C.

Properties	Symbols	Units	Caprock	Outer reservoir	Inner reservoir	Bedrock
Porosity	φ		0.01	0.1	1	0.01
Permeability	Ŕ	m^2	10-18	5 10-16	5 10-11	10-20
Density	ρ^s	kg m ⁻³	2600	2600	2600	2600
Specific heat capacity	c_p^s	J kg ⁻¹ °K ⁻¹	850	850	850	850
Heat conductivity	λ^{s}	W m ⁻¹ °K ⁻¹	2	2	2	2

267 **Table 1.** Properties of the rock (Bächler et al., 2003; Baillieux et al., 2013)

268	Table 2.	Fluid	proj	perties (Ľ.	Smith	&	Cha	pman,	1983)).
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Properties	Symbols	Units	Value
Water			
Bulk modulus	K_w	Pa	$2 \cdot 10^{10}$
Density	$ ho_w$	kg m ⁻³	$\rho_w = 1000 \cdot e^{\frac{p_w}{K_w}}$
Viscosity	μ_w	Pa s	10-3
Specific heat capacity	$C_{p,w}$	J kg ⁻¹ K ⁻¹	4000
Heat conductivity	λ_w	W m ⁻¹ K ⁻¹	0.6
Nanotracer			
Diffusion coefficient	D	$m^2 \cdot s^{-1}$	$4 \cdot 10^{-12}$

Tracers (conservative and temperature-reporting nanotracers with different temperature thresholds) are injected only on the first day at a mass rate of 6 g·s⁻¹ for each tracer, whereas the injection and production flow rates of water are constant at 40 L·s⁻¹ for five years. The water is

injected at a constant temperature of 70°C. The inner reservoir has a thickness of 1 m in the X direction and has a permeability of $5 \cdot 10^{-11}$ m². As shown in Figure 2 (b), the initial temperature range is 145°C to 175°C for a reservoir depth interval of 0.6 km. Three alternative injection points I1, I2 and I3 as well as production points P1, P2 and P3 have depth positions of -2.7 km, -2.8 km and -2.9 km, respectively see Figure 2 (b).

277 **3 Results and Discussions**

Several numerical simulations are conducted to illustrate the working mechanisms of 278 temperature-reporting nanotracers by analyzing the response behavior from the data collected at 279 280 the production well. Firstly, a case with homogeneous permeability of the inner reservoir is tested. The main purpose is to understand how the velocity field obtained from different well positions 281 282 affects the breakthrough curve response of the temperature-reporting nanotracers for a given initial temperature distribution. The breakthrough concentrations of these nanotracers are analyzed by 283 284 extracting the peak information (i.e., peak concentration values and peak arrival time) from their breakthrough concentration curves. Secondly, more complex features are added to the model to 285 286 investigate the effect of inclined zones (with different permeabilities) embedded within the reservoir and different thermal distributions. 287

288 **3.1 Reference Case: Homogeneous System**

The temperature-reporting nanotracers we use in this example have different temperature 289 thresholds from 145°C to 180°C with variations of 5°C. For specific and detailed investigations, 290 the temperature threshold differences were refined to 1.25° C in part of the temperature range. It 291 292 should be noted that the selected temperature threshold range of the nanotracers is valid only for our numerical model setting. In future nanotracer field tests, the temperature threshold range can 293 be estimated from a simplified linear relationship between reservoir thickness and temperature 294 data at or near the wellbore from the exploration and well-drilling stages. Conserved nanotracers 295 are co-injected for comparison. Nine scenarios of injection-production position setup are simulated 296 with the same well operating conditions, injected materials and volumetric rates. 297

3.1.1 Interdependency among Fluid Flow, Heat Transfer, Transport and Reaction of Temperature-reporting Nanoparticle Tracers

To illustrate the temperature distribution and associated flow regimes related to the tracer test, consider two well positions: I1P1 and I1P3. The results are depicted in Figure 3 when the

302 conservative nanotracer concentration reaches its peak concentration at the production well (80 303 days for I1P1 and 90 days for I1P3). The nanotracer is transported toward the production well, 304 following the displayed fluid flow directions. The fluid (i.e., water well-mixed with nanotracers) 305 flow direction is essentially based on the fluid velocity which is computed from Equation 2. It does 306 not indicate its magnitude. The fluid flow direction together with the velocity field (subplots a2 307 and b2) reflect the streamlines of fluid flow.



Figure 3. Typical thermal and hydraulic states of the reservoir (depth range 2.5~3.1 km): (1) Temperature and
fluid flow direction distribution and (2) velocity fields at 80 days and 90 days which correspond to concentration
peaks of the conservative nanotracer collected at the production well when the injection-production points (a)
I1P1 and (b) I1P3 are chosen, respectively. The black lines in (a) and (b) are the reservoir temperature contour
and Darcy velocity distributions, respectively.

308

The volume around the injection point cools down (Figures 3 (a1) and 3 (b1)) since the injected water has a lower temperature. The temperature field is asymmetrical due to the flow field not being aligned with the initial temperature distribution. The injected fluid sweeps the reservoir symmetrically around the main streamline region towards the production point but is affected by the reservoir boundaries. The overall magnitude of fluid velocity for both I1P1 and I1P3 (Figures

3 (a2) and 3 (b2), respectively) is similar along the main streamlines since the injection and
 production rates are the same.

Figure 4 illustrates the nanotracer concentration distribution for three types of nanotracers: 321 one conservative ($C_{cons.}$) and two temperature-reporting nanotracers ($C_{T_{160^\circ C}}^{prod.}$ and $C_{T_{170^\circ C}}^{prod.}$) with 322 temperature threshold 160°C and 170°C, respectively. We track only the converted temperature-323 reporting nanotracer concentrations ($C_{T_{thre.}}^{prod.}$) as the sum with unconverted nanotracer concentration 324 is preserved (acting as a conservative nanotracer). Injection-production position setups are still 325 I1P1 (Figures 4 (a) and 4 (b)) and I1P3 (Figures 4 (c) and 4 (d)). In each case, two groups of times 326 were selected for plotting: the former are 80 days and 300 days and the latter are 90 days and 160 327 days, which individually correspond to the peak arrival time of conservative nanotracer and 328 329 temperature-reporting nanotracer with T_{thre}=170°C in the I1P1 case and the I1P3 case, respectively.



Figure 4. Distributions of conservative nanotracer and temperature-reporting nanotracers with $T_{thre.}=160^{\circ}C$, 170°C. (a, b): I1P1 setup at 80 and 300 days (peak arrival time of conservative nanotracer and nanotracer with $T_{thre.}=170^{\circ}C$); (c, d): I1P3 setup at 90 days and 160 days (peak arrival time of conservative nanotracer and nanotracer with $T_{thre.}=170^{\circ}C$). The black contours with their magnitudes on the right clearly show the reservoir temperature distribution.

336 In the case of I1P1, conservative nanotracer flows from the injection point to the production 337 point, covering both shallow and deep parts of the reservoir (Figures 4 (a1) and (b1)). The highconcentration part of the conservative nanotracer is best maintained along the main streamline of 338 fluid, while the concentrations are more diffuse in the weak-current region. This is associated with 339 the nanotracer concentrations traveling at different speeds and mixing with low fluid 340 341 concentrations at neighbouring streamlines. The temperature-reporting nanotracers can react if they reach temperatures above their thresholds thus being converted. As a result, shallow and deep 342 formations with comparatively low and high temperatures have different abilities to convert the 343 temperature-reporting nanotracers with their corresponding temperature thresholds. From Figures 344 4 (a2) and 4 (a3), there are large proportions of converted temperature-reporting nanotracers with 345 a threshold of 160°C in the lower half reservoir and only small amounts of converted temperature-346 reporting nanotracers with a threshold of 170°C appear in the lower quarter reservoir. Above the 347 middle reservoir, there is zero concentration of $C_{T_{160^\circ C}}^{prod.}$ and $C_{T_{170^\circ C}}^{prod.}$ meaning that the corresponding 348 349 nanotracers are not converted in the upper part of the reservoir. Compared to the conservative nanotracer, the two temperature-reporting nanotracers are just partly converted. Compared to the 350 results in 80 days, the concentrations of the three nanotracers become weakened after 300 days 351 due to continuous production. In addition, the distributions of $C_{T_{160^\circ C}}^{prod.}$ and $C_{T_{170^\circ C}}^{prod.}$ are similar to 352 each other after 300 days, which reflects the slow tail production. 353

From case I1P1 to I1P3, the fluid flow direction has changed (see Figures 4 (a1) and (c1)). The conservative nanotracer in Figures 4 (a1) and 4 (c1), flows through a wide region and a slightly longer distance to the production point. The peak arrival time is thus longer with I1P3 than with I1P1. However, when comparing Figures 4 (b3) and 4 (d3), the peak arrival time of the nanotracer with a 170°C threshold is less with I1P3 (160 days) than with I1P1 (300 days) due to the shorter flow path to cross regions where temperatures reach above 170°C.

360 3.1.2 Well Configuration Impact on Temperature-reporting Nanotracer Breakthrough 361 Curve

As mentioned earlier, there are a total of nine injection-production well positions. Figure 5 summarizes the nanotracer breakthrough concentrations at the production well, including both conservative nanotracer and nine converted temperature-reporting nanotracers with temperature thresholds varying from 150°C to 172.5°C.

The nanotracers are injected with the same concentration and can be directly compared. The conservative nanotracer profile is always above the other nanotracer profiles. The temperaturereporting nanotracers are converted only when their thresholds are met in certain reservoir regions.



369
 370 Figure 5. Nanotracer breakthrough curves in comparison to the conservative nanotracer (blue points) with a
 371 total of nine injection-production well configurations.

Referring to Equation 4, the sum of reactant and product concentrations equals the conservative 372 nanotracer concentration. When a nanotracer is not fully converted, the product concentration is 373 less than the conservative nanotracer concentration. Therefore, in all subplots of Figure 5, the 374 curves produced by the temperature-reporting nanotracers with low-temperature thresholds, such 375 as 150°C and 155°C, are very close to the conserved one. In addition, high peak concentration 376 values in these nanotracer breakthrough curves normally correspond to short travel time. On the 377 contrary, the temperature-reporting nanotracers with high-temperature thresholds such as 172.5°C 378 379 are less converted and have lower peak values of concentration. They also take a longer time to reach the peak concentration than those with low-temperature thresholds such as 160°C. 380 Nanotracers with higher thresholds have to be transported further to be converted. 381

The effects of injection-production positions are visible on the nanotracer breakthrough 382 383 curves in Figure 5. Subplots (a1) I1P1, (b2) I2P2 to (c3) I3P3 show that the magnitude of temperature-reporting nanotracer breakthrough curves increases towards the conservative curve 384 385 with deeper injection-production position. More nanotracer is converted at a greater depth with higher temperatures. The process to reach their corresponding peaks is also accelerated because 386 387 the conversion happens along (or closer to) the fastest streamline. The same trends apply to the cases where well position I or P is constrained but the paired well position P or I moves towards 388 389 the deep formation, referring to subplots (a1)-(b1)-(c1) in Figure 5.

The results from injection-production positions I1P3 (Figure 5 (a3)) and I3P1 (Figure 5 (c1)) differ, although the geometric settings are symmetrical. The 162.5°C nanotracer curve in subplot (c1) with I3P1 has a higher magnitude than in subplot (a3) with I1P3. This difference is due to the nanotracer starting at a high temperature in the first case, and not meeting that high temperature along all flow lines in the second case.

395 3.1.3 Analysis of Temperature-reporting Nanotracer Breakthrough Curves for Reservoir 396 Characterization

A key factor that can be used to maximize the production of geothermal energy is the knowledge of temperature which is the main streamline of injected water experiences. Since the nanotracers follow the water, the temperature-sensitive tracer information can, ideally, reflect the temperature characteristics of the streamlines. Based on that, a new analysis method is proposed to quantify reservoir temperature information from temperature-reporting nanotracer breakthrough curves (such as Figure 5). Peak concentration values and peak arrival time are plotted versus the

403 nanotracer temperature thresholds, see Figure 6. Temperature-reporting nanotracers with 404 thresholds 175°C and 180°C were not converted due to the highest reservoir temperature being 405 175°C. Therefore, their peak concentrations in subplot (a) are zero and the peak arrival time in 406 subplot (b) does not exist. Here we need to mention that it is important to use a broad range of 407 nanotracer temperature thresholds to cover the reservoir temperatures since the exact temperature 408 distribution is unknown in realistic field tests.





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The curves of both peak concentration value and peak arrival time versus threshold temperature have flat sections at low-temperature thresholds, which correspond with the

415 conservative nanotracer, indicating full conversion. The temperature encountered on the flow paths
416 between the wells must therefore be above these thresholds.

When the peak concentration curves deviate from the flat sections, for a nanotracer with a 417 sufficiently high threshold temperature, some of that nanotracer has traveled along a flow path 418 with a temperature below the threshold where it was not converted. In other words, the threshold 419 temperature at the transition from fully converted nanotracers to less converted nanotracers 420 indicates that there are flow paths not exceeding this indicated temperature. This provides an upper 421 422 limit of the lowest temperature in the flow region. Similarly, when the threshold temperature of the nanotracers becomes sufficiently high, they are not converted (zero peak concentration) 423 indicating no flow paths reach that high temperature. Thus, the threshold temperature where the 424 nanotracers firstly stop being converted is an indication and approximation of the highest 425 426 temperature in the flow region. As the sum concentration of nanotracer reactant and product acts like a conservative nanotracer, this holds regardless of reservoir properties (this could change if 427 the nanotracer interacted chemically different within the reservoir). 428

As indicated, there is an important precaution regarding the history-dependent behavior of the 429 430 nanotracers. If they have passed through a region above the threshold temperature, they are activated regardless of what happens later. Consider the difference between injection from a deep 431 432 towards a shallow producer, with injection from a shallow position towards a deep position. (i) In the former case the high temperature is at the injector and the different nanotracers are exposed to 433 434 high temperature from the start. They might not see much higher temperatures and although there are lower temperatures downstream, they are already activated, yielding a narrow range for the 435 deviation thresholds. Such a case mainly provides reliable information about the highest 436 temperature. (ii) In the latter case the nanotracers are exposed to a low temperature at the injector 437 438 and take multiple directions having different temperatures towards the producer. The initial high-439 temperature fluid at the producer is produced before the nanotracers encounter it, meaning the maximum interpreted temperature may be less than the initial temperature at the producer. Some 440 flow paths can however go deeper to reach even higher temperatures. 441

In the following, the nanotracer temperature threshold at the turning point of each nanotracer curve is discussed and compared to the temperature along the injection-production positions. Consider first the injector-producer pairs positioned at the same depth. In Figure 2 (b), the initial temperatures of I1P1, I2P2 and I3P3 are 155°C, 160°C and 165°C, separately. In Figure 6 (a1) the

peak concentration values deviate at 151.25°C, 156.25°C and 161.25°C, respectively. This means 446 not all the injected nanotracer was converted by the initial temperature surrounding the injector. 447 Keep in mind that the nanotracer mixture (and then water) is injected at 70°C and thus the 448 nanotracer needs some residence time to reach a higher temperature. As seen in Figure 3 (a1) there 449 is a low-temperature region near the injector, while the temperature distribution in the rest of the 450 reservoir is less affected. Some nanotracers, particularly the end of the slug, can follow paths that 451 do not reach the high initial temperatures. Concerning the peak arrival time in Figure 6, we observe 452 a deviation from the horizontal section at 155°C, 160°C and 166°C, which corresponds more 453 closely with the initial temperatures at the injectors and producers. In Figure 6 (a1), nanotracer 454 peak concentrations become zero when the temperature threshold approaches 175°C. This means 455 the maximum temperature of the overall reservoir is below 175°C (as confirmed in Figure 4). 456 457 While this high temperature was detected in all three cases, the lowest temperature of the reservoir of 145°C (see Figure 4) was not detected by any. Flow lines were passing through temperatures 458 459 below 150°C for the case I1P1 (Figure 4), thus 145°C might have been expected to be the highest stable point. Likely, each flow line contained a region (close to the well) with a higher temperature 460 461 that activated the nanotracers, as suggested before. From the above analysis, we conclude that it is generally difficult to estimate the lowest temperatures in the reservoir. However, when the wells, 462 especially the injector, are placed shallower, the threshold temperature where the peak 463 concentrations start to decrease, becomes better, but not reliable, estimates of the minimum 464 temperature. 465

The results of inclined wellbore positions I1P3 and I3P1 are shown in Figures 6 (a2) and (b2). 466 As seen in Figure 4 for I1P3, flow lines pass through both regions below 150°C and above 170°C 467 and the same is true for I3P1 by symmetry. For both cases, the concentration curves flatten 468 precisely at 175°C, as for the three previous cases, indicating the maximum temperature of 175°C. 469 470 The peak concentrations of I1P3 and I3P1 cases deviate at 157.5°C and 160°C, respectively, close to the initial temperature of 160°C centrally between the two wells (Figure 2). This reflects that 471 nanotracer needs to flow through regions with this temperature whether it flows from the deep or 472 shallow configuration. Especially in Figure 4, we see that for I1P3 all flow lines cross a 473 474 temperature of 160°C at the indicated time. The deviation threshold temperature is higher for I3P1, related to the nanotracer encountering higher temperatures from the start. In Figure 6 (b2), the peak 475

arrival time deviates for both I1P3 and I3P1 at a temperature threshold of 165°C. This is closer to
the deeper wellbore's initial temperature.

It is noticeable that the peak concentrations deviate at lower temperature thresholds than the 478 arrival time. As seen in Figures 6 (a) and 6 (b), the peak concentrations reduce while their arrival 479 time stays the same. This is related to where the nanotracers are flowing. The peak of the 480 conservative nanotracer represents the flow from the fastest flow lines between the wells. When 481 482 the nanotracer is converted along the same lines, we also see the arrival of that nanotracer with the 483 same peak, however, when the nanotracer needs to take a longer path to be converted, the arrival time is increased. Several of the colder streamlines at the top of the reservoir may supply an 484 485 unconverted nanotracer, yielding less production and a lower peak concentration of the converted nanotracer. But as long as the main flow line between the wells has sufficient temperature the 486 487 arrival time is similar to the conservative nanotracer. Similarly, we can suggest that the higher peak arrival time represents the time needed for nanotracers with the indicated thresholds to pass 488 489 through flow lines with those temperatures. In Figure 6 (b1) the arrival time of a nanotracer with a given threshold decreases when the horizontally positioned well pair is deeper, because the main 490 491 flow line has a higher temperature and activates more nanotracers there, and because the nearby flow lines also have a higher temperature. At shallow locations, the nanotracer needs to follow a 492 493 long flow path to be activated. Considering the inclined positioned wells (Figure 6 (b2)), the arrival 494 time profiles are almost identical. They deviate from the flat section when the threshold 495 temperature exceeds the highest temperature of 165°C along the main flow line. Only flowlines 496 deeper than the well pair reach higher temperatures. With the flow pattern being symmetrical and most of the temperature distribution remaining as the initial, the nanotracers are activated in the 497 same flow lines for the two cases and get the same arrival time. 498

To summarize the main points, temperature-reporting nanotracer breakthrough curves can be interpreted as follows:

The nanotracer peak concentrations as a function of threshold temperature:

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 Deviate from full conversion at a threshold temperature which is an upper limit of the lowest temperature encountered along all the flow lines. This is not a reliable estimate of the minimum temperature.

First reaches a zero value at a threshold temperature which is a precise estimate of 505 0 the maximum temperature in the flow region. The estimate is improved by using 506 multiple tracers with small differences in the temperature thresholds. 507 The nanotracer arrival time as a function of threshold temperature: 508 _ • Deviates from the arrival time of the conservative nanotracer at a threshold 509 temperature equal to the highest temperature along the fastest/main flow line 510 between the wells. This deviation temperature is usually higher than that from the 511 512 nanotracer peak concentration curve. • Higher arrival time on the curve indicates the time needed to flow along lines 513 reaching the corresponding threshold temperatures. 514 515 In the following sections, we utilize the presented analysis method on more complicated

reservoir conditions, including **Section 3.2** different thermal distributions (varied temperature ranges at certain depths and regional thermal anomalies) within the geothermal reservoir and **Section 3.3** inclined zones embedded within the reservoir. The purpose of these investigations is to determine the impact of these thermal and geological uncertainties on the analysis of performance evaluation of temperature-reporting nanotracers in geothermal reservoirs. Schematic illustrations of varied temperature ranges, regional thermal anomalies and inclined zones are shown in Figure 7 (a-c), respectively.



523 524 Figure 7. Schematic representation of geological heterogeneities and temperature anomalies in a reservoir (a): 525 Different initial temperature ranges with gradients $\nabla T = 0.05^{\circ}$ C·m⁻¹, 0.0375°C·m⁻¹ and 0.025°C·m⁻¹ along the inner reservoir depth from -2500 m to -3100 m; (b) Lower-left or lower-right located high-temperature regions 526 527 within the inner reservoir; (c): Left-inclined (LIZ) or right-inclined zones (RIZ) with higher, same, or lower permeabilities compared to the inner reservoir. Note that the inclined zone with dimensions (1000 m·600 m·1 528 529 m) is a plane perpendicular to the thin reservoir.

3.2 Impact of Different Thermal Distributions in the Fractured Geothermal Reservoir 530

Temperature distributions and heat flux densities in the subsurface can vary greatly depending 531 on location. Values above average are referred to as positive anomalies. Conversely, a negative 532 anomaly indicates a decrease in temperature or heat flux relative to the surrounding mean. Thermal 533 anomaly in subsurface formations is a common geological phenomenon that can be caused by 534 variations of thermal conductivities around structures such as salt domes, geological and tectonic 535 activity, geochemical reactions, or hydrothermal activities in faults and fractures (Cherubini et al., 536 2013; Emry et al., 2020; Yan et al., 2023). Subsurface formations with positive thermal anomalies 537 are targeted as areas of geothermal development for heat and electricity production (Moeck, 2014). 538 In the following, we investigate two scenarios impacted by different local temperature gradients 539 or a positive regional thermal anomaly and whether they are detectable by the nanotracer analysis. 540

541 **3.2.1 Different Temperature Ranges**

We consider three different initial temperature gradients as shown in Figure 7 (a) where the middle of the reservoir is constrained to 160° C. Here, cases 1, 2 and 3 respectively correspond to temperature gradients of 0.05° C·m⁻¹, 0.0375° C·m⁻¹and 0.025° C·m⁻¹, with initial reservoir temperature ranges of 145° C~175°C, 148.25° C~170.75°C and 152.5° C~167.5°C. Note that only the initial reservoir temperature range is varied in the numerical model; the remaining parameters are the same as in the reference case. Case 1 is essentially the reference case studied in **Section 3.1**.

The resulting concentration peak and arrival time trends from the nanotracer breakthrough curves are shown in Figures 8 (a) and 8 (b) for injection-production position setups I1P1 and I1P3.

551 There is an obvious distinction among the curves of the three cases with different initial temperature gradients. In Figure 8 (c) the curve deviation temperatures are compared with initial 552 well temperatures (red markers) and initial reservoir temperature range (grey bars) for additional 553 well setups. Each curve in Figures 8 (a1) and 8 (a2) flattens at zero concentration at a high 554 555 temperature. As discussed, this point indicates the maximum temperature, which is confirmed to be accurate by the comparison (top of the grey bar and top green point) in Figures 8 (c1) and 8(c2). 556 557 The lower temperature deviation on the concentration curve, however, does not reliably estimate the minimum reservoir temperature and is in some cases above the lowest initial well temperature 558 559 and, and some cases below. The deviation temperature of the arrival time curve (blue marker) has been noted to reflect the maximum temperature along the streamline between the wells. As the 560 reservoir is homogenous, this flow line goes directly between the wells and the maximum 561 temperature of this flow line is likely to be approximately the highest initial well temperature. This 562 563 comparison (blue point and highest red point in Figure 8 (c) is very accurate (0 to 2°C difference) for all 10 cases. We have thus demonstrated accurate prediction of the highest reservoir 564 temperature and main streamline temperature. 565

When the initial temperature range (i.e. local gradient) increases the estimated max temperature increases accordingly. For the horizontally positioned wells, a higher gradient (thus lower min temperature) is reflected in a lower min temperature estimate for I1P1, but when the wells are positioned deeper (I2P2 or I3P3), there is little to no difference with the gradient. The flow lines then start at hotter temperatures (slightly below the initial well temperature) and activate according to higher temperatures than found at the top of the reservoir. Changing the gradient for

- 572 I1P3 also has little effect on the lower limit temperature estimate, as the flow lines all pass 160°C
- 573 in the center.



Figure 8. Comparison of nanotracer performance for peak concentration (a), peak time (b) and estimated reservoir temperature data (c) for initial temperature gradients $\nabla T = 0.025$ or 0.05° C·m⁻¹. Well configurations are indicated. T_{inje}. and T_{prod}. are initial temperatures at injection and production positions, while grey bars indicate the reservoir's initial temperature range. $T_{C_{peak}}^{rese}$ show estimated reservoir temperatures based on peak concentration curve deviation. $T_{t_{peak}}^{mstr.max}$ is the temperature where the arrival time curve deviates (estimating the highest temperature on the main streamline between the wells).

The temperatures between the concentration turning points indicate a possible reservoir 581 temperature range. For a given well positioning the jump of each curve in Figure 8 (a1) or (a2) 582 will become sharp when the temperature gradient decreases. Regardless of the initial temperature 583 gradients of the reservoir, subplot (c1) shows that the low reservoir temperature obtained is 584 dependent on the tested positions, the maximum temperature can always be detected, and shallow 585 tested locations such as horizontal I1P1 exhibit more effectiveness in measuring reservoir 586 temperature intervals. Looking at the inclined cases I1P3 and I3P1 in subplot (c2), the estimation 587 588 performance for reservoir temperature ranges looks pretty generic.

589 **3.2.2 Regional Thermal Anomalies**

High-temperature regions are artificially added to the reservoir in the lower-left or lower-right 590 positions to mimic regional positive thermal anomalies of up to 200°C from deep radiant heat 591 592 sources, see Figure 7 (b). Note that only the high-temperature regions are added to the numerical model, the remaining parameters are the same as in the reference case. The temperature thresholds 593 of the nanotracers have extended from 145°C~180°C to 145°C~205°C with variations of 5°C. For 594 specific and detailed investigation, the temperature threshold difference is refined to 2.5°C in part 595 596 of temperature ranges. We compare tracer results in Figure 9 (a, b) for a homogeneous case without thermal anomaly (HOM), a case with left thermal anomaly (LTA), and a case with right thermal 597 anomaly (RTA). 598

The high-temperature region enhances the conversion of temperature-reporting nanotracers 599 600 with high thresholds in the deep reservoir, see subplots (a1) and (a2). The cases with LTA and RTA produce nearly the same results for I1P1 in subplot (a1), probably because the streamlines 601 are symmetric in the left-right direction and the cooled-down region due to injection has not 602 affected the high-temperature distribution in the deep reservoir during the given period. 603 Nevertheless, when the injection-production position is changed to the inclined I1P3, the 604 difference between LTA and RTA is clearly distinguishable in subplot (a2) where the case with 605 RTA results in higher peak concentration values than with LTA. The streamlines from I1 to P3 606 flow through the high-temperature region on the right side (RTA) close to P3 rather than the left 607



Figure 9. Comparison of nanotracer performance for peak concentration (a), peak time (b) and estimated reservoir temperature data (c) in models with no (HOM), left-located (LTA) or right-located (RTA) different thermal anomalies. Well configurations are indicated. $T_{inje.}$ and $T_{prod.}$ are initial temperatures at injection and production positions, while grey bars indicate the reservoir's initial temperature range. $T_{cpeak}^{rese.}$ show estimated reservoir temperatures based on peak concentration curve deviation. $T_{tpeak}^{mstr.max.}$ is the temperature where the arrival time curve deviates (estimating the highest temperature on the main streamline between the wells).

one (LTA) which is not close to either well. For I3P1 the resulting behavior between RTA andLTA is opposite.

In subplot (b), the trends of the curve's relative position are similar to those plotted based on 617 peak concentration values in subplot (a). It should be mentioned that a high-temperature region in 618 the reservoir could behave similarly to the one with large temperature gradients when comparing 619 Figure 9 (a, b) to Figure 8 (a, b). Specifically, the low-temperature gradient case $(0.025^{\circ}\text{C}\cdot\text{m}^{-1})$ in 620 Figure 8 (a, b) always gives a narrow threshold temperature range which is also seen in the 621 622 behavior of the HOM case without high temperature anomaly in Figure 9 (a, b). Moreover, based on the results presented in Figure 9 (a2, b2), we are able to distinguish the differences caused by 623 the existence of thermal anomalies as well as the locations of thermal anomalies in the reservoir, 624 when the injection-production well configuration is inclined. 625

A performance evaluation of the nanotracers in the reservoir with thermal anomalies is given 626 according to the breakthrough curve peak analysis (Figure 9 (a, b)) for selected cases. The 627 maximum temperature within the reservoir is estimated accurately, which means the thermal 628 anomaly is detected. Although a shallow injection-production position can help to more accurately 629 630 estimate the reservoir temperature range as seen in Figure 9 (c1), this effect is reduced when a thermal anomaly with high temperature is located in the deep reservoir, including both RTA and 631 632 LTA. This observation is also valid for inclined well positions, see subplot (c2), when comparing the case HOM with the cases LTA and RTA. The temperature where the peak arrival time curve 633 634 deviates indicates the highest temperature along the main streamlines between the wells and in most cases corresponds well with their highest initial temperature. 635

636 **3.3 Impact of Geological Heterogeneities Resulting from Inclined Zones**

Reservoirs are usually highly heterogeneous and some distinguishing features are large spatial 637 differences in reservoir permeability. In some cases, low permeable zones are encountered, such 638 as faults developed by tectonic movement; high permeability layers exist in other cases, such as 639 thief zones due to possible clay erosion or sand production after a long period of water injection 640 (C. Lu et al., 2021). Tracer testing can identify the properties of fractures or inclined zones, 641 dependent on the shapes of tracer breakthrough curves (J. Li et al., 2016; L. Li et al., 2017). Those 642 studies mainly focused on the conservative tracer, while here we will explore how such geological 643 features affect the performance of temperature-reporting nanotracers in fractured geothermal 644 reservoirs. 645



X [km]X [km]647Figure 10. Tracer concentration distributions after 70 days for a geological heterogeneous model, i.e. an648inclined zone (white line) and configuration IIP3. The difference between a conservative tracer (left column)649and a temperature-reporting nanotracer (T_{thre.}=160°C, right column) is clearly demonstrated in dependence of650the hydraulic setting. Subplot a/b: low-permeable left-/ right-inclined zone (i.e., $5 \cdot 10^{-16} m^2$); Subplots c and d:

high-permeable left and right-inclined zone (i.e., $5 \cdot 10^{-10} \text{ m}^2$). Note that the permeability of the inner reservoir is $5 \cdot 10^{-11} \text{ m}^2$. The black contours with their magnitudes on the right show the reservoir temperature.

653 Figure 7 (c) illustrates a thin inclined zone centrally placed between the wells. The effects of left-inclined and right-inclined zones are studied with permeabilities higher (i.e., $5 \cdot 10^{-10} \text{ m}^2$) or 654 much lower (i.e., $5 \cdot 10^{-16} \text{ m}^2$) than the inner reservoir permeability (i.e., $5 \cdot 10^{-11} \text{ m}^2$). Note that only 655 the inclined zones with different directions and permeabilities are added to the numerical model, 656 657 the remaining parameters are the same as in the reference case. As illustrated in Figure 10, concentration distributions of a conservative tracer and a temperature-reporting nanotracer with a 658 659 160°C threshold are shown with injection-production setup I1P3 at 70 days (when tracers flow through the inclined zone). The results with a low-permeable inclined zone are given in Figure 10 660 (a, b), and those with a high-permeable inclined zone are shown in Figure 10 (c, d). As expected, 661 the temperature-reporting nanotracer with a 160°C threshold mainly exists in the lower part of the 662 reservoir where it is converted whereas the conservative tracer has a large area of distribution in 663 the reservoir. 664

In the cases with a low-permeable inclined zone, the inclined zone behaves as a tight barrier for tracer transport and tracer concentrations are separated. In subplot (a2) there is a region of highconcentration converted tracer above the inclined zone. Temperature-reporting tracer has been transported around the upper region of the reservoir (above the inclined zone) and converted after passing the 160°C isotherm.

In the cases with a high-permeable inclined zone in Figure 10 (c, d), we notice in subplots (c1) 670 and (c2) that the fluid flow is attracted up towards the high permeability zone giving a more 671 horizontal flow in the reservoir. This flow diversion is less clearly seen in subplots (d1) and (d2) 672 since the direction of I1P3 is perpendicular to the right-inclined zone. The concentration of 673 674 conservative tracer is less in subplot (c1) than in subplot (d1). The conservative tracer meets the high-permeable zone earlier in subplot (c1) and more tracer transports to the high-permeable zone 675 where the tracer can be accumulated, compared to subplot (d1). Moreover, since the fluid flow is 676 attracted towards a horizontal direction in the upper part of the reservoir in subplot (c2), less 677 678 temperature-reporting nanotracer transports downward being converted than in subplot (d2).

A comparison of the tracer peak concentration and peak arrival times is shown in Figure 11 for two configurations: I1P1 and I1P3. As seen in Figures 11 (a1) and (b1) the results for I1P1 are very similar when comparing whether the inclined zone is left or right-oriented for a given permeability, due to symmetry. For I1P3, left- or right-inclined orientations do not give

symmetrical flow and thus the orientation of the inclined zone impacts the tracer profiles, see 683 curves in Figures 11 (a2) and (b2). Referring to Figure 10 (c1), a high-permeable inclined zone 684 can divert flow towards it and attract tracers to flow in its plane from the reservoir, resulting in 685 low peak concentration values. The separation effect on the tracer transport due to the low-686 permeable inclined zone can decrease the peak concentration collected in the production point, 687 referring to Figure 10 (b1). In Figure 11 (a1) the (red) curves of peak concentrations for low-688 permeable inclined zone take an abrupt turn at 155°C. This is the result of a low permeability zone 689 forcing tracers to flow in the lower part of the reservoir, as we can see in Figure 10 (a2). In the 690



Figure 11. Comparison of tracer performance for peak concentration (a) and peak arrival time (b) in
homogeneous (HOM) and geological heterogeneous models (LIZ or RIZ) with well configurations of (1) I1P1
and (2) I1I3. The permeability in the left- or right-inclined zone (LIZ or RIZ) is 5·10⁻¹⁶ m² or 5·10⁻¹⁰ m² while
the reservoir permeability is 5·10⁻¹¹ m².

I1P1 setup, the fastest streamline will be at the top and stay above 155°C (the initial well 696 temperatures) and not deliver converted tracers with higher thresholds. Streamlines that take the 697 opposite side of the barrier will all have reached 170°C which explains the jump in arrival times 698 at 150°C to a higher stable value until 170°C, where only the outermost streamlines can convert 699 the tracers. As the threshold increases gradually less tracer is converted but following a different 700 trend than at first since the tracer is primarily converted at the bottom of the reservoir. Note that 701 the discontinuity in arrival time and the trend of peak concentration happen at the same temperature 702 703 and are clear indicators of two separate flow line groups. While there were two separate streamlines groups also for I1P3, they arrived at similar times, while the reactive tracer mainly converted in 704 the lower one. 705

Figure 11 (2) gives the results when the injection-production position changes to I1P3 which 706 707 is exactly the setup shown in Figure 10. When the inclined zone is low permeable and the orientation is similar to the injection-production direction, the tracer transport will be efficient and 708 709 give rise to high peak concentration values of tracer concentration in the production point. That is why it is seen in Figure 11 (a2) that the dashed red line (right-inclined) has a lower magnitude than 710 711 the solid red line (left-inclined). On the other hand, the green dashed line in Figure 11 (a2) has higher values than the green solid one since the attraction of the high permeable zone plays a more 712 713 significant role in reducing the tracer concentration in the latter case (solid line, left-inclined), which has been discussed in the above for comparisons between Figure 10 (c) and (d). Therefore, 714 715 inclined injection-production position I1P3 is useful for the test to differentiate the right- or leftinclined zones. In addition, it is noticeable in Figure 11 (a) that the curve trends within the high 716 tracer threshold range of 165°C~175°C are similar: top (low permeable zone), middle 717 (homogeneous) and bottom (high permeable zone). 718

719 Regarding reservoir temperature estimation using the deviation points of the peak 720 concentration and peak arrival time curves in Figure 11, we find that the maximum temperature is correctly estimated to be 175°C in all cases. For a given well setup a fairly consistent minimum 721 temperature is estimated as 152°C~153°C for I1P1 and 157°C~158°C for I1P3 although the actual 722 minimum for all cases is 145°C. The highest temperature of the coldest streamline is determined 723 724 by how deep they go, i.e. the depth of the wells, as explained earlier. The deviation threshold temperature of the arrival time curve shows different trends compared to the homogeneous case. 725 When there is a barrier, the flow is concentrated and it can be diverted from or towards the well, 726

⁷²⁷but also from or towards the higher temperatures. In the example of I1P3 with a low permeability ⁷²⁸RIZ (see Figure 10 (b2)) the flow is diverted straight down (increasing the arrival time of the ⁷²⁹conservative tracer), exceeding 170°C and then straight towards the producer. The deviation of the ⁷³⁰arrival time (red curve in Figure 11 (b2)) correctly indicates that temperatures have exceeded ⁷³¹170°C on the main streamlines.

732 **4 Conclusions**

Temperature-reporting nanoparticle tracers, which quickly, fully and irreversibly convert 733 734 when the environmental temperature reaches its threshold value, were studied to characterize the 735 temperature information of geothermal reservoirs. We developed a numerical modeling approach to illustrate their working mechanisms in a thin 3D reservoir. A mixture of tracers with different 736 737 temperature thresholds was injected. First, a homogeneous and fractured geothermal reservoir was studied with nine injection-production well configurations. Furthermore, adding heterogeneities 738 739 such as thermal anomalies and inclined zones were investigated. The following conclusions can be made: 740

- Reservoir temperature can be changed due to cold water injection and fluid intrusion from different depths. The injected temperature-reporting nanotracers travel along all streamlines but are converted on a streamline only if they reach the temperature threshold of that specific tracer. This happens if the streamline goes sufficiently deep or sufficiently close to a thermal anomaly.
- Injection-production positions and inclined high- or low-permeable zones embedded
 within the reservoir influence the streamlines, thus impacting the collected tracer
 breakthrough data. Deeper well positions steer all the streamlines through higher
 temperatures, while the mentioned zones can spread or deviate streamlines towards or away
 from the high temperatures.
- The peak concentration of a given temperature-reporting nanotracer is determined by the proportion of the swept reservoir area above its temperature threshold. The changes in tracer peak arrival time result from alterations in the flow paths of its main streamlines. A long flow path generally corresponds to a long peak arrival time of the tracer.
- A new analysis method was proposed, based on plotting the peak concentration and arrival
 time of each tracer against their temperature threshold.

- At sufficiently low temperatures, all tracers are fully converted, giving the same
 peak concentration as the conservative tracer. The temperature where the tracers
 start to yield lower peak concentrations is an upper limit of the minimum reservoir
 temperature.
- At sufficiently high temperatures, no tracer is converted, giving zero peak
 concentration for higher temperatures. The temperature threshold where the peak
 concentrations first become zero is identical to the maximum reservoir temperature.
- The arrival time of the tracer peak is the same as the conservative tracer as long as
 it is converted on the main (fastest) flowline between the wells. The threshold
 temperature when the time starts to increase indicates the highest temperature on
 the main flowline.
- The range in temperature thresholds between the deviations on the peak concentration 768 curve reflects only a part of the reservoir temperature range, but the highest temperature 769 deviation corresponds to the highest reservoir temperature, while the lowest deviation 770 temperature can be near or far away from the lowest reservoir temperature. Changing the 771 reservoir temperature conditions (with other conditions the same), consistently changes the 772 773 max temperature (actual and estimated) but may not necessarily impact the lower threshold deviation temperature. While a reduced temperature range may be expected to reduce the 774 difference between the thresholds (a sharper peak concentration curve), a sharp curve does 775 not directly imply a narrow reservoir temperature range. 776
- At a reduced temperature range (and other conditions the same), the flow paths next to the main streamline now have a temperature closer to that of the main streamline. The same streamlines have the same arrival time, but tracers with lower thresholds, corresponding to the streamline max temperature, will dominate. In total, the streamlines with the same arrival times obtain threshold temperatures closer to the main streamline, i.e. a sharper curve.
- The effects of thermal anomalies (existence and locations) and inclined zones (conductivity and orientation) in the reservoir can be more easily observed when the injection-production position is non-horizontal as the tracer data become more sensitive to the orientation and location of such features.

In practice, forward simulations are required to match the breakthrough data from the field tests. Although it provides useful insights into the effects of geological and thermal heterogeneities on the tracer breakthrough data, a precise estimation of these unknowns relies on inverse modelling which uses model sensitivities to update parameters to be more consistent with observations. Our results show clear responses to geological and thermal heterogeneities which are clearly detectable when choosing a non-horizontal injection-production location.

The conclusions of this work are not limited to fractured reservoirs but can also applied to 793 794 other types of geothermal reservoirs. During the entire lifecycle of geothermal reservoir exploitation, temperature-reporting tracer tests can be conducted to detect temperature range and 795 reservoir thermal drawdown in the reservoir as well as other geological and thermal 796 797 heterogeneities. At the exploration stage, the initial in-situ reservoir temperature information can 798 be characterized quickly by the tracer curve using high flow rates, especially if shallow injectionproduction positions are chosen and the tracer can sweep the entire reservoir. For the production 799 800 stage, the thermal drawdown status can be periodically quantified by comparison of the present temperatures to the initial ones. Potential challenges or limitations that one might be faced with 801 802 when implementing our proposed approach in a realistic fractured geothermal reservoir scenario include injection-production configurations for inferring the temperature range, and uncertainties 803 804 in the properties of liquid solution.

In addition to the investigations discussed in this work, the robustness of our proposed analysis approach has also been verified by simulations that consider the variations of communication between the inner reservoir and outer reservoir, injection and production rate and conversion degree of nanotracers. Nevertheless, the following aspects still deserve further studies:

- The impact of complex geometries which may include additional fractures and randomly
 distributed geological heterogeneities on the temperature-reporting nanotracer's behaviour
 in the reservoir and analysis curves.
- Use field test data to validate simulation results to ensure the practical applicability of this research work and its findings.
- Consider the impact of nanotracer properties and mechanisms such as gravity segregation,
 deposition and aggregation on the results predicted by the proposed numerical model.
- Incorporate the impact of potential fluid source (supply) and/or sink (leakage) from
 neighbouring geological strata.

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Appendix I: Fully Coupled Processes of Fluid Flow, Heat Transfer, Transport and Reaction for Temperature-reporting Nanoparticle Tracers

1016 (a) Detailed Equations of Fully Coupled Processes

Equation 4 indicates a sharp conversion process of the injected nanotracers / reactants into products. To release this constraint, we include a conversion factor *Z* in the following reaction expression for temperature-reporting nanotracers:

1020
$$(C_{T_{thre.}}^{reac.\,n})^{updated} = Z^n \cdot C_{T_{thre.}}^{reac.\,n}$$
(A1)

1021 where $(C_{T_{thre.}}^{reac.\,n})^{updated}$ represents the newly updated concentration of the reactant after reaction 1022 (at time step *n*) and *Z* is defined as:

1023
$$Z^{n} = \begin{cases} 1, & T^{n} < (T_{thre.} - a); \\ \frac{1}{1 + e^{b \cdot (T^{n} - T_{thre.})}}, & (T_{thre.} - a) \le T^{n} \le (T_{thre.} + a); \\ 0, & T^{n} > (T_{thre.} + a). \end{cases}$$
(A2)

where a (°C) and b (=6/a, °C⁻¹) are conversion constants that can be controlled such that a smooth conversion between 100% and 0% nanotracer conversion can be achieved when environment temperature *T* is near the nanotracer temperature threshold T_{thre} .

1027 The product concentration after the reaction at the time step n is updated with:

1028
$$(C_{T_{thre.}}^{prod.^{n}})^{updated} = (1 - Z^{n}) \cdot C_{T_{thre.}}^{reac.^{n}} + C_{T_{thre.}}^{prod.^{n}}$$
 (A3)

1029 where the two parts on the right side separately refer to the concentration increase of the product 1030 after the reaction and product concentration at time step n.

1031 The updated results $(C_{T_{thre.}}^{reac. n})^{updated}$ and $(C_{T_{thre.}}^{prod. n})^{updated}$ at time step *n* are separately used 1032 explicitly in the following tracer transport Equations A4 and A5 for the implicit computation of 1033 the reactant and product concentrations at time step *n*+1.

1034
$$\frac{C_{T_{thre.}}^{reac.\ n+1} - (C_{T_{thre.}}^{reac.\ n})^{updated}}{\Delta t} - \nabla \cdot \left(\boldsymbol{D} \nabla C_{T_{thre.}}^{reac.\ n+1} \right) + \nabla \cdot \left(\boldsymbol{u}_{w}^{n+1} C_{T_{thre.}}^{reac.\ n+1} \right) - Q_{C_{T_{thre.}}^{reac.\ n+1}} = 0 \quad (A4)$$

1035
$$\frac{C_{T_{thre.}}^{prod.\,^{n+1}} - (C_{T_{thre.}}^{prod.\,^{n}})^{updated}}{\Delta t} - \nabla \cdot \left(\boldsymbol{D} \nabla C_{T_{thre.}}^{prod.\,^{n+1}} \right) + \nabla \cdot \left(\boldsymbol{u}_{\boldsymbol{w}}^{\boldsymbol{n+1}} C_{T_{thre.}}^{prod.\,^{n+1}} \right) - Q_{C_{T_{thre.}}^{prod.\,^{n+1}}} = 0 \quad (A5)$$

1036 After obtaining $C_{T_{thre.}}^{reac.\,n+1}$ and $C_{T_{thre.}}^{prod.\,n+1}$, we need to update these two variables at time step n+11037 according to the new conversion factor Z^{n+1} using Equations A1 and A3.

1039 The numerical algorithms of the fully coupled processes of fluid flow, heat transfer, transport 1040 and reaction for temperature-reporting nanoparticle tracers are shown in Figure A1. At time step 1041 n, the fluid flow model, nanotracer transport model together with heat transfer model are solved 1042 implicitly. After obtaining the results, the reactant and product concentrations of tracers are 1043 updated using the nanotracer reaction model, which provides the inputs for the tracer computation 1044 at next time step n+1.



1046 Figure A1. Schematic of the fully coupled fluid flow, heat transfer, transport and reaction models of 1047 temperature-reporting nanoparticle tracers.

1048 Appendix II: Numerical Model Validation

A one-dimensional (1D) simulation is run to validate the numerical model with coupled fluid flow, heat transfer, transport and reaction for temperature-reporting nanoparticle tracers, shown in Figure A2. The numerical results produced by the PorousFlow module (Wilkins et al., 2021) within the MOOSE framework (Permann et al., 2020) are validated against an analytical solution. The analytical solution is simply a stepwise function, which satisfies the mass conservation law and chemical conversion.





Figure A2. Validation on the fully coupled processes of fluid flow, heat transfer, transport and reaction for temperature-reporting nanoparticle tracers: (a1) 1D model with boundary conditions; (a2) temperature and conversion factor profiles; (a3, a4) comparison of numerical and analytical results for reactant $C_{T_{2.5^{\circ}C}}^{reac.}$ and product $C_{T_{2.5^{\circ}C}}^{prod.}$ at 5·10⁴ s and 1.5·10⁵ s.

As shown in Figure A2 (a1), incompressible fluid is injected with a nanotracer reactant (threshold 2.5°C) into a 100-meter horizontal system. The Darcy velocity is $5 \cdot 10^{-4} \text{ m} \cdot \text{s}^{-1}$ and the injected nanotracer reactant concentration is 0.1 mg·L⁻¹. The porosity is assumed 1 and fluid properties are set constant (density 1000 kg·m⁻³, viscosity 10⁻³ Pa·s and thermal conductivity 0.6

- 1064 $W \cdot m^{-1} \cdot k^{-1}$). The system temperature (0°C~5°C) and pressure (1.1 MPa~0.1 MPa) distributions are 1065 constrained following a linear relation from inlet to outlet, during the whole process.
- 1066 The temperature distribution and the corresponding conversion factor *Z* along the model are 1067 shown in Figure A2 (a2). The comparisons between the analytical and numerical solutions show
- agreement for both reactant and product at the two selected times $5 \cdot 10^4$ s and $1.5 \cdot 10^5$ s.

Journal Pre-proof

1069 Appendix III: Mesh Sensitivity Analysis

1070 To illustrate the convergence of the chosen discretization, a comparison of the conservative 1071 tracer breakthrough curves at the production well is provided in Figure A3 for three different 1072 meshes. The curves are practically indistinguishable when the element number reaches 331'799. 1073 Consequently, the simulations reported in this study are performed with 331'799 elements.



1074Time [years]1075Figure A3. Comparison of conservative tracer breakthrough curves for different element numbers.

Highlights

- Temperature-reporting nanoparticle tracer tests are numerically implemented in a threedimensional fractured geothermal reservoir.
- ٠ The working mechanisms of the temperature-reporting nanoparticle tracers are illustrated through field simulations using a proposed modeling approach.
- Our proposed analysis curves can provide responses to the reservoir temperature distribution as well as geological and thermal heterogeneities based on the tracer breakthrough data.

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Declaration of interests

 The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☑ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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