

Characterisation of fluid flow in aquitards using helium concentrations in quartz, Gunnedah Basin, NSW

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Executive summary

The project 'Research to improve treatment of faults and aquitards in Australian regional groundwater models to improve assessment of impacts of coal seam gas (CSG) extraction' focuses on method development to underpin the risk assessments associated with deep groundwater extraction and depressurisation from energy resource development. The project aims to develop methodologies and techniques that will improve the predictive capability of regional groundwater models used in this context, specifically with respect to the representation of faults and aquitards. The project has three components: (i) an examination of aquitards, (ii) an examination of faults, and (iii) an examination of the upscaling of aquitard and fault properties such that they can be adequately represented in regional groundwater flow models.

This report provides an overview of a novel approach using environmental tracers to derive formation-scale hydraulic conductivities of key aquitards in the Gunnedah Basin (New South Wales). Concentrations of helium in quartz were measured and used to calibrate a fluid flow model. Specifically this study:

- Measured a vertical profile of helium concentrations in the Watermark/Porcupine Formation aquitard sequence.
- Constrained rates of helium diffusion to ensure helium reached equilibrium between pore water and quartz.
- Simplified the modelling process to allow the quantitative assessment of helium equilibrium between pore water and quartz.
- Modelled the formation-scale transport, production and partitioning of helium in the aquitard sequence.
- Compared results to core-scale measurements of aquitard permeability.

Modelling suggests a vertical fluid velocity on the order of 0.002–0.02 mm/year, which is very slow, especially because the aquitard thickness exceeds 500 m. The shape of the helium profile suggests helium concentrations in the underlying Maules Creek Formation varied over recent geological time.

The future usage of this method depends on the depth and thickness of the aquitard of interest. Deeper aquitards are at higher temperatures, which enhance the diffusion of helium in quartz, resulting in faster equilibrium. Subsurface temperatures should be 40 °C at a minimum. In thicker aquitards (tens to hundreds of metres), internal helium concentrations are slow to adjust when helium concentrations change in adjacent formations. Therefore, equilibrium of helium between quartz and pore water can be assumed.

Furthermore, it is recommended that future utilisation of the method should be accompanied by the quantification of tortuosity and diffusion in the porous medium. Also, diffusion testing should be undertaken at lower temperatures to better constrain helium equilibrium between pore water and quartz. Environmental tracers, such as helium, are invaluable in the characterisation of aquitards at the formation-scale, aiding in the impact assessment of CSG development in groundwater basins.

1 Introduction

The subsurface containment or migration of hydrocarbons and other fluids is dependent on the integrity of the geological units that separate the reservoir from adjacent aquifers. In most conventional hydrocarbon reservoirs, the overlying cap rock or aquitard is quite impermeable (permeabilities in the mD to nD range). This, among other factors such as wettability, limits hydrocarbon migration from the reservoir throughout geological time. However, in unconventional hydrocarbon reservoirs, such as coal seams that have retained and accumulated natural gas, the integrity of the overlying aquitard is not a requirement for trapping the gas. This is because the sorption of hydrocarbons has often sufficiently limited their migration such that an exploitable resource exists, eventhough the overlying geological unit may not be impermeable. Indeed, coal seam gas reserves are retained in coal seams by reservoir pressure, which keeps gas molecules physically adsorbed onto the surface of the coal micropores (Dallegge and Barker, 2000; Moore, 2012). Groundwater extraction leads to a reduction in reservoir pressure, thereby desorbing gas from the coal cleats. Taking these conditions into account, there is a greater need to assess isolation properties of aquitards and other low permeability formations within coal seam gas (CSG) producing basins. In particular, there is a need to assess to what degree there are hydraulic connections between the target reservoirs and overlying aquifers. The latter may be utilised for irrigation, stock and municipal water supply. Hydraulic connections in this case refers to faults, fractures and changes in lithology (Cartwright et al. 2007). Other hydraulic connections may exist due to leaky wells, poorly installed wells, improperly decommissioned wells, or wells that have existed beyond the rated lifespan of their construction materials (Wu et al. 1998; Dusseault and Jackson 2014; Doble et al. 2018). The risks of impacts from leaky wells are significant but will not be considered here, as the investigation techniques discussed in this study are insensitive to these modern changes in fluid flow and mass transport.

While aquitards in deep basins are usually tens to hundreds of metres thick, their permeability is usually assessed on the scale of centimetres. This is the scale at which traditional permeability assessment techniques (i.e. diamond coring and triaxial permeametry) are undertaken (Yu *et al.* 2013). Such small-scale analyses are unable to characterise the influence of formation-scale features such as faults, fractures and heterogeneous lithology (Clauser 1992).

The effects of faults, fractures and changes in lithology have not been studied explicitly in this study. However, the helium data is likely to represent effects of changes in lithology and possibly small-scale fractures: these may have contributed to the variability in the observed helium concentrations (see further). Whether or not faults and fractures have played a critical role will be inferred from the modelling: in case a diffusion-based model describes the data reasonably well, there is sufficient evidence to believe that transport through faults and fractures has been negligible for the single core analysed.

An alternative approach is to assess fluid flow at the formation scale. Because rates of fluid flow in aquitards are typically very low (possibly significantly less than mm/year), the assessment of fluid flow at this scale requires the use of environmental tracers, which have been present in aquitards for millennia. If the sources, sinks and transport properties of the environmental tracers can be

constrained, it is possible to use them to infer fluid velocity at the formation scale (Mazurek *et al.* 2011). Figure 1.1 shows how the distribution of a tracer through an aquitard depends on the dominant transport method (i.e. advection versus diffusion) and the direction of advection.



Figure 1.1 Examples of tracer distribution in an aquitard and the dominant transport process for upward and downward water flow.

Numerous tracers have been used to assess fluid flow rates in aquitards (see Mazurek et al. (2011)). The present study examines the use of helium (He), which was demonstrated successfully in previous studies (Gardner *et al.* 2012; Smith 2015). Helium is completely inert, is produced in the subsurface as the result of the radioactive decay of naturally occurring uranium (99.3% ²³⁸U and 0.7% ²³⁵U) and thorium (²³²Th). Helium concentrations in groundwater can vary over several orders of magnitude. Helium has two stable isotopes: ³He and ⁴He. The former is very rare and comprises approximately one millionth or less of the total helium. Therefore the total helium is essentially equal to the ⁴He concentration. The present study did not analyse ³He, so it will not be considered further.

One of the greatest difficulties in measuring helium in aquitard pore water is that helium is sparingly soluble and is lost from core samples almost immediately after core retrieval. Therefore, core samples must be immediately preserved using specialised methods. In Australia, millions of metres of core have been collected for hydrocarbon and mineral exploration and are currently stored in public core libraries (e.g. the Londonderry Drillcore Library, NSW, contains 1,130 km of core (NSW Government 2016)). Unfortunately, only approximately 1 m of core has been specially preserved to capture pore water helium.

An alternative and novel method to assess helium concentrations in pore water is to use a proxy. The majority of sedimentary rocks contain the mineral quartz (SiO₂). Helium initially present in the pore water will readily diffuse into the intragranular porosity of quartz (Wood *et al.* 1990). Following an extended period of contact, the helium will equilibrate between the quartz and pore

water. Helium concentrations are believed to be retained in quartz grains for a period of at least several decades, provided that the samples are stored at ambient temperatures. By measuring helium concentrations in quartz from a vertically distributed set of samples, it is possible to constrain a solute transport model, as previously undertaken using pore water helium observations (Mazurek *et al.* 2011; Gardner *et al.* 2012).

The Gunnedah Basin, NSW, contains several coal measures that are prospective for coal seam gas extraction. The depressurisation process used to extract methane gas may decrease pressures in adjacent aquifers if the permeability in the intervening aquitards is significant. Groundwater flow modelling of the Gunnedah Basin has previously been undertaken as part of Environmental Impact Assessment for the Santos Ltd Narrabri Gas Project (CDM Smith 2014). Based on a literature review, limited information was presented with regards to the hydraulic properties of relevant aquitard units. In particular, the availability of hydraulic conductivity data for the deepest aquitards (i.e. Watermark and Porcupine Formations) was very limited. This lack of data motivated several studies, including the current study, to develop and apply methods that would increase data availability while making use, as much as practically possible, of existing information (e.g., reanalysis of wireline log data, see Turnadge et al. (2018)).

In this study, helium concentrations in quartz grains were measured in 13 samples from the Watermark and Porcupine Formations. These two aquitards are part of a thick aquitard sequence that directly overlies the Early Permian Maules Creek Formation (the primary coal seam gas target formation). Diffusion testing and numerical modelling were performed to determine whether helium had equilibrated between quartz and pore water. The vertical transport of helium through the aquitard sequence was simulated in order to estimate formation-scale hydraulic properties. This information could provide additional evidence of fluid flow, or absence thereof, across low permeability formations in the Gunnedah Basin.

2 Methods

2.1 Study area

Core samples for quartz-helium analysis come from the Slacksmith 1 corehole in the Gunnedah Basin, NSW (Figure 2.1). The corehole was drilled by Santos in 2009 and core was collected from 100.6–1410.0 m depth (Santos (QNT) Pty Ltd 2009).



Figure 2.1 Surface geology of the Gunnedah Basin and the location of the Slacksmith 1 corehole.

2.1.1 Geology and hydrogeology

The Gunnedah Basin is a structural trough located in NSW and contains Permian and Triassic sediments deposited in marine and non-marine environments (Tadros 1993). The primary CSG targets are the Hoskissons Coal and the coal members of the Maules Creek Formation. Aquifers include the Cainozoic alluvium and the Pilliga Sandstone.

Aquitards of interest to the present study are the Purlawaugh, Napperby, Watermark and Porcupine Formations (Table 2.1). The Purlawaugh and Napperby Formations separate the Pilliga Sandstone from the underlying Black Jack Group, which contains the Hoskissons Coal. The Watermark and Porcupine Formations separate the Hoskissons Coal from the underlying coals seams of the Upper Maules Creek Formation.

The Watermark Formation characteristically contains an upward-fining sequence of silty sandstone through to laminated claystone and siltstone, which is overlain by an upward-coarsening sequence of laminated claystone and siltstone through to laminated siltstone and sandstone (Tadros 1993). The Porcupine Formation characteristically contains an upward-fining sequence of conglomerate, sandstone, siltstone and mudstone (Tadros 1993).

Because the quartz-helium method relies on relatively high temperatures to expedite the diffusion of helium into the quartz, a focus was placed on cores available from deeper formations, which led to the analysis of the Watermark and Porcupine Formations. At Slacksmith 1, downhole temperatures were between approximately 46 to 64 °C at the depths of the Watermark and Porcupine Formations (700–1220 m). Here the Watermark Formation is described as an upward-coarsening sequence of silt grading to fine-grained sandstone (Santos (QNT) Pty Ltd 2009). The Porcupine Formation is described as an upward-fining conglomerate containing sandstones, siltstones and a few igneous intrusions (Santos (QNT) Pty Ltd 2009). The combined thickness of these two formations is over 520 m. The Watermark Formation is overlain by the Pamboola Formation and underlain by the Maules Creek Formation.

Table 2.1. Summary of geological units, geological model layers, hydrostratigraphic units and groundwater flowmodel layers represented in the CDM Smith Gunnedah Basin groundwater flow model (modified after CDM Smith(2014).

GEOLOGICAL UNIT	GEOLOGICAL MODEL LAYER	HYDROSTRATIGRAPHIC UNIT	GROUNDWATER MODEL LAYER(S)	
Cainozoic Alluvium	1	aquifer	1	
Liverpool Range Volcanics	- 2		2	
Wallumbilla Formation	2		3	
Bungil Formation		aquitard	4	
Mooga Sandstone	3		5	
Orallo Formation			5	
Pilliga Sandstone	4	aquifer	6	
Purlawaugh Formation	5		7	
Garrawilla Volcanics	6		8	
Deriah Formation	7		0	
Napperby Formation	/		9	
Digby Formation	8		5	
Trinkey Formation		aquitard	10	
Wallala Formation			10	
Breeza Coal Member	9		11	
Clare Sandstone	5		11	
Howes Hill Coal Member			12	
Benelabri Formation			12	
Hoskissons Coal	10 CSG reservoir		13	
Brigalow Formation			14	
Arkarula Formation	11		15	
Melvilles Coal Member	11	aquitard	16	
Pamboola Formation		αγυπαια	17	
Watermark Formation	- 12		18	
Porcupine Formation	12		19	
Maules Creek Formation (upper)		interburden	20, 21	
Maules Creek Formation (coal measures)	13	CSG reservoir	22	
Maules Creek Formation (lower)		interburden	23, 24	

2.2 Sample preparation and analysis

2.2.1 Porosity and permeability measurements

Ten core samples from the Slacksmith 1 core were analysed for porosity and gas permeability at the CSIRO Petrophysics laboratory (several additional samples were collected, but these samples were friable, breaking into small pieces, which prevented testing). Sub-coring was required in order to fit the samples into a fixed core holder of 1 inch diameter (for cores which had an original diameter of 45 mm) or 1.5 inch diameter (for cores which had an original diameter of 60 mm).

Samples were sub-cored with water using a 1.5 inch or 1.0 inch-diameter coring bit, depending on the original core size. Sub-core lengths varied from 20–150 mm, depending on the length of the original core. The top and bottom faces of sub-core samples were trimmed into flat parallel faces and heated in an oven at 105 °C for at least three days, in order to remove all moisture. After drying, the weight, length and diameter of each sample was measured and the bulk density calculated. Porosity and permeability measurements were then performed using nitrogen gas permeametry (for details, see Turnadge et al. (2018)). Sub-core permeability *k*, originally expressed in mD (milliDarcy) units, was converted to equivalent saturated hydraulic conductivity, K (m/s), using the conversion factor 1 mD = 9.6127 × 10⁻⁹ m/s (at 20 °C).

2.2.2 Mineral separation and helium analysis

Before the helium concentration in quartz grains can be measured, the quartz must be separated from other minerals. The helium concentrations in other minerals may not be in equilibrium with pore water helium concentrations. The methods used for a similar mineral separation scheme were described in detail by Smith (2015). The methods used for the present study are described briefly as follows. Sub-core samples weighing approximately 200 g were crushed to sub-millimetre size pieces using a jaw crusher. The material was repeatedly rinsed with deionised water to remove all clay-sized particles before being treated with 10% nitric acid to remove carbonates followed by 30% hydrogen peroxide to remove organic material. The use of hydrogen peroxide was found to be marginally effective so heavy liquid separation (described below) was used.

All samples were briefly dried at 40 °C before being sieved to 45–150 μ m. Select samples were treated with sodium hexametaphosphate (4 g/L) to remove additional clay particles that coated the mineral grains. Dry samples were magnetically separated using a Frantz isodynamic magnetic separator. This process separates non-magnetic minerals (such as quartz) from magnetic minerals (such as olivine, pyroxene, mica, iron oxides, etc.). However, minerals such as feldspars, pyrite, apatite and zircon, as well as organic materials, are only partially removed by magnetic separation, or may not be removed at all (Rosenblum 1958).

The non-magnetic separate was further separated by density (ρ) using heavy liquid (lithium metatungstate). The density of the liquid was mixed to 2.7 g/cm³ so that quartz (ρ = 2.63 g/cm³), organics ($\rho \approx 1.3$ g/cm³) and most feldspars (ρ = 2.55–2.76 g/cm³) would float and so that heavy minerals such as apatite (ρ = 3.16–3.22 g/cm³), zircon (ρ = 4.6–4.7 g/cm³) and pyrite (ρ = 4.95–5.10 g/cm³) would sink. The liquid density was then decreased to 2.5 g/cm³ so that quartz and any remaining feldspars would sink while organics would continue to float.

The resulting cleaned and dried quartz separates may have contained some feldspars, but additional separation techniques were avoided as they involve hydrofluoric acid. This would dissolve the feldspars, but would also partially dissolve the quartz, changing the grain size. As a result, the samples likely contain some feldspar minerals. Since the diffusivity of helium in feldspar is similar to that of quartz, and since feldspars generally do not contain U and Th, the effects of this impurity are assumed to be insignificant.

Approximately 2 g of each quartz separate sample was heated at 290 °C for 6 days in order to release the helium from the quartz grains. Helium (⁴He) concentrations were measured at the CSIRO Waite Campus using a mass spectrometer system designed for the measurement of noble

gases (Poole *et al.* 1997). Helium and any neon (neon-20; ²⁰Ne) in the samples were further purified using non-evaporable getters and cryogenics before being analysed using a residual gas analyser. While neon was not expected to be released from the quartz, it was measured to ensure that the helium was not contaminated with atmospheric gas during heating. In the case of contamination, the following correction was used:

$${}^{4}\text{He}_{q} = {}^{4}\text{He}_{m} - {}^{20}\text{Ne}_{m} ({}^{4}\text{He}/{}^{20}\text{Ne})_{atm}$$
(1)

where ${}^{4}\text{He}_{q}$ is the helium concentration attributed to quartz, ${}^{4}\text{He}_{m}$ is the total measured helium concentration, ${}^{20}\text{Ne}_{m}$ is the total measured neon concentration, and $({}^{4}\text{He}/{}^{20}\text{Ne})_{atm}$ is the atmospheric ${}^{4}\text{He}/{}^{20}\text{Ne}$ ratio (0.318). Samples with more than 5% of helium coming from leakage were reanalysed using a new subsample.

To determine the effective solubility of the helium in quartz, which varies between samples, the quartz was impregnated with pure helium. Samples were then heated to release the helium and then analysed as described above. This effective solubility is known as the helium accessible volume (*HAV*) and is calculated as:

$$HAV = {}^{4}\mathrm{He}_{imp}(p_{1}/p_{imp})(T_{imp}/T_{1})$$
⁽²⁾

where ⁴He_{*imp*} is the helium concentration in quartz (cm³ STP He/cm³_{quartz}) at the standard pressure p_1 (101,325 Pa) and standard temperature T_1 (273 K). T_{imp} and p_{imp} are the experimental temperature (563 K) and impregnation pressure, respectively. The helium pore water concentration (He_w) can then be calculated using (Lehmann *et al.* 2003):

$$He_w = S \times p_1 (He_{ini}/HAV) (T_{ini}/T_1)$$
(3)

where *S* is the air-water solubility of helium (cm³ STP He/g atm) while *T_{ini}* (K) and He_{ini} are the in situ formation temperature (Santos (QNT) Pty Ltd 2009) and the initial helium concentration measured in the quartz, respectively. In essence, Equation 3 uses the ideal gas law to calculate the partial pressure of helium in quartz and then converts that value to a pore water helium concentration by means of the solubility, which is dependent on temperature and salinity.

Helium diffusion rate in quartz

The reliability of quartz-derived pore water helium concentrations depends on equilibrium exchange of helium between the two domains (Figure 2.2). At ambient and moderately elevated temperatures (i.e. 20 to 60 °C), the diffusion of helium in quartz is very slow, requiring thousands to hundreds of thousands of years to achieve equilibrium conditions (Trull *et al.* 1991). Temperatures at our sampling depths were in the upper range of the 20-60 °C interval, which will be shown to be high enough to achieve equilibrium (see section 3.5.2). At elevated temperatures (i.e. 290 °C; as used above), the diffusion rate increases dramatically and equilibrium exchange occurs in hours to days. However, observed rates of diffusion have varied considerably between studies (Trull *et al.* 1991; Shuster and Farley 2005), which may be a function of grain damage and/or the presence of fluid inclusions (Smith *et al.* 2013). To quantify the diffusion rate of helium in quartz at the study site, step-heating diffusion experiments were conducted.

Quartz separates from three Slacksmith 1 core samples were sieved to 60–75 μ m and impregnated with helium at approximately 150 Torr (760 Torr = 1 atm). After impregnation, the samples were transferred to new sample containers and evacuated. The samples were then heated to 290 °C for increasingly longer time steps of 2–1438 minutes. Each time step corresponds to an expected helium release of 10% of the total helium. This was calculated using an analytical solution to the diffusion equation that assumes spherical quartz grains and a diffusion coefficient of 2.86 × 10⁻¹⁴ m²/s (Crank 1975; Trull *et al.* 1991).



Figure 2.2 Conceptual model of helium diffusion between aquitard pore water and quartz grains.

2.2.3 Uranium and thorium samples

The rate of helium production in the subsurface is the result of the radioactive decay of uranium and thorium and their subsequent isotopic daughters. Because uranium and thorium are the source elements of decay chains, these two elements can be measured in order to determine helium-4 production rates. To measure U and Th, whole rock samples were first crushed to <1 mm particles. The samples were then digested using hydrofluoric acid and the digested material was analysed using ion coupled plasma mass spectrometry.

2.3 Modelling methods

Helium transport was modelled for two reasons: (i) to estimate vertical fluid velocities at the formation-scale and (ii) to estimate equilibrium between quartz and pore water at the pore-scale. These transport mechanisms were then combined into a single model to account for formation-scale helium transport and the equilibrium of helium between quartz and pore water in order to

infer if the helium concentrations in quartz were representative of helium concentrations expected in the pore water. The transport of helium was simulated using analytical and numerical approaches. Modelled concentrations were calibrated to observed concentrations through the estimation of transport parameters, including vertical fluid velocity. The degree of model-tomeasurement misfit for each model was quantified using a normalised sum of least squares metric.

2.3.1 Flow through porous media

To estimate the formation-scale fluid velocity through an aquitard and thereby constrain an estimate of aquitard permeability, a solute transport model was used, which is a solution of the one-dimensional advection-diffusion equation with production:

$$D_p \frac{\partial^2 C}{\partial z^2} - v \frac{\partial C}{\partial z} = \frac{\partial C}{\partial t} - g_w \tag{4}$$

where D_p is the pore dispersion coefficient of helium in the porous medium, C is the helium concentration, z is the vertical position, v is the fluid velocity, t is time elapsed, g_w is the rate at which helium is released into the pore water following production in the rock. The pore dispersion coefficient is dependent on the mechanical dispersion D_m and the effective molecular diffusion coefficient D_e :

$$D_p = D_m + D_e \tag{5}$$

where D_m is calculated as $v \times \lambda$, with λ the dispersivity or dispersion length (m). For low advective velocities, D_p is equal to the effective diffusion coefficient D_e , as the contribution of dispersion to D_p becomes insignificant. It is assumed that the transport process is diffusion dominated (i.e., dispersion is negligible). Regardless, dispersion was included in the models for completeness. Dispersivity λ is often defined empirically and is a function of the scale of solute transport (Molz *et al.* 1983; Gelhar *et al.* 1992). A λ of 10 m was chosen to be appropriate to the 520 m thick aquitard sequence examined in the present study (Schwartz and Zhang 2003). For similarly low-permeability formations, much smaller λ -values have been used for transport simulations. For instance, based on tracer tests with small undisturbed clay cores Aertsens et al. (2003) derived dispersion length values in the order of 0.001 m. While our value is at the high end for such a low-porosity and low-permeability medium, we demonstrate that the transport is diffusion dominated, hence the magnitude of λ does not matter. In other words, the transport process will be shown to be insensitive to the magnitude of the dispersion length (see section 3.5).

The effective diffusion coefficient can be defined using the temperature-dependent free-water diffusion coefficient ($D_{0,w}$) (Jähne *et al.* 1987), which is multiplied by aquitard porosity (*n*) and flow path tortuosity (τ):

$$D_e = D_{0,w} n\tau \tag{6}$$

Because flow path tortuosity is typically not a well-constrained parameter, D_e is instead commonly approximated as:

$$D_e = D_{0,w} n^m \tag{7}$$

where m = 2 is the maximum expected porous media diffusion coefficient for helium (Mazurek *et al.* 2011). For deionised water at a temperature of 25 °C, the free-water diffusion coefficient $D_{0,w} = 7.22 \times 10^{-9} \text{ m}^2/\text{s}$ (Jähne *et al.* 1987).

The dominant transport process (i.e. advection or diffusion) can be assessed using the Péclet number:

$$Pe = \frac{vL}{D_e} \tag{8}$$

where L = aquitard thickness. Values of *Pe* below unity indicate that transport over distance *L* is dominated by diffusion, whereas values above ten are characteristic of advection-dominated systems.

The helium production rate is estimated from the concentrations of U and Th in the porous medium, which decay to produce a series of daughter isotopes and several alpha particles (helium-4 nuclei). It is assumed that over geological time, secular equilibrium is achieved (i.e., helium is released into pore water from the U- and Th-containing minerals at the same rate that it is produced). The helium production rate, g_r (cm³ STP/g_r/s), is calculated as:

$$g_r = 3.82 \times 10^{-21} [\text{U}] + 9.09 \times 10^{-22} [\text{Th}]$$
 (9)

where U and Th concentrations are in ppm. The parameter g_w (cm³ STP/g_w/s) is calculated from g_r by:

$$g_w = g_r \rho_b \left(\frac{1}{n} - 1\right) \tag{10}$$

where ρ_b is the dry bulk density of the porous medium.

Two different solutions to Equation 4 were computed: a steady-state analytical solution and a transient implicit numerical solution. Both models featured Dirichlet (i.e., specified concentration) boundary conditions. The numerical model featured time-varying Dirichlet conditions. The numerical model also allowed several parameters, including diffusivity, to be specified as functions of depth.

2.3.2 Helium diffusion in quartz

Model results were compared to the step heating helium release data in order to estimate the rate of helium diffusion from quartz grains to pore water. Assuming a constant diffusion coefficient within the quartz (D_q), the governing equation is (Crank 1975):

$$\frac{\partial C}{\partial t} = D_q \left(\frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right) \tag{11}$$

where *r* is the radial distance from the centre of the sphere. Assuming a constant Dirichlet boundary at the surface and a uniform initial helium concentration in the quartz, Equation 11 was solved with an analytical solution (Crank 1975). The model was calibrated for the step heating helium release data using the GRG (generalised reduced gradient) algorithm implemented with MS Solver to minimise the sum of squared normalised residuals, $\Sigma \chi^2$:

$$\sum \chi^2 = \sum_{t=1}^{N} \left(\frac{C_{o,t} - C_{m,t}}{\sigma_t} \right)^2 \tag{12}$$

where $C_{o,t}$ and $C_{m,t}$ are the observed and modelled helium concentrations at time t, respectively, and σ_t is the standard deviation of the observation at time t. The standard deviation is assumed to be the analytical uncertainty of the helium measurement. Because the helium concentration at each time step is a cumulative release, the uncertainty of model results increases with each time step according to standard error propagation.

Using a single diffusion coefficient was found to result in a poor model calibration (i.e., $\Sigma \chi^2 = 11-39$). Consequently, two diffusion coefficients (i.e., slow and fast) were implemented. This would suggest a slow diffusion path and a fast diffusion path due to a heterogeneous distribution of grain defects (Clay *et al.* 2010). The total helium release is then related to two diffusion coefficients, each contributing a fraction of the total helium. Model calibration was performed similarly to the single-rate diffusion model with the addition of the GRG algorithm implemented with MS Solver, which was used to minimise $\Sigma \chi^2$.

The diffusion coefficients determined from the release experiments at 290 °C (i.e., $D_{290,q}$) were adjusted to formation temperatures (Santos (QNT) Pty Ltd 2009) using the Arrhenius equation, by first solving for the maximum diffusion rate (D_0 ; m²/s) and then finding D_q at the required temperature (T; K):

$$D_q = D_{0,q} e^{-\frac{E_a}{RT}},$$
 (13)

where E_a is the activation energy (105 kJ/mol; Trull *et al.* 1991) and *R* is the gas constant (8.314 J/K mol). If the step heating experiment was performed at multiple temperatures, E_a could have been estimated independently. However, as this was not performed due to time constraint, it was instead assumed that literature activation energy values were accurate.

2.3.3 Simplifying the quartz-pore water equilibration process for the formationscale aquitard model

Ultimately, the equilibrium of helium between quartz and pore water needs to be assessed using a transient form of the advection-diffusion transport model because the boundary conditions on the quartz grains are likely to have been variable over geological time (i.e., changes in helium concentrations at the base of the aquitard due to climatic factors (e.g., variations in recharge rates) and/or geological factors (e.g., fracturing, faulting and diagenetic processes)). Furthermore, if changes in pore water helium concentrations and/or hydraulic conditions have occurred in adjacent aquifers then their effects on aquitard pore water helium and quartz helium concentrations need to be accounted for.

The analytical solutions to spherical diffusion generally require limited boundary conditions and initial conditions (Crank 1975). However, the scenario described above has more complex time-variable boundary conditions. Therefore, to model the diffusion of helium in quartz simultaneously with the advection-diffusion transport model, the diffusion calculations were simplified as follows.

Measured rates of helium diffusion were used to estimate a series of first-order rate constants $k_{i...N}$:

$$C(t) = \sum_{i=1}^{N} F_i e^{-k_i t}$$
(14)

where C(t) is the concentration of helium in quartz at time t and F is the volumetric fraction of the sample, where:

$$\sum_{i=1}^{N} F_i = 1 \tag{15}$$

Equation 14 was calibrated to the measured helium concentrations through least squares-based estimation of the set of first-order rate constants $k_{i...N}$. The number of rate constants N was selected to fit the data and its uncertainty. In an attempt to accurately represent three-dimensional diffusion, Equation 14 was also calibrated to the simulated concentrations obtained with the analytical solution to Equation 11; in this case, three rate constants were required to accurately represent these concentrations.

 D_q (m²/s) was converted into the same units as k (1/s) by dividing by the squared grain radius (a^2 ; m²). This conversion was necessary because the spherical diffusion model accounts for grain geometry when calculating the loss or gain of helium, whereas the first-order rate model does not. The first-order rate constants $k_{i...N}$ were then related to the two diffusion coefficients D by a linear relationship, allowing $k_{i...N}$ to be adjusted to represent the specified temperature and grain size.

2.3.4 Testing the first-order rate model

Analytical solutions for spherical diffusion include time-varying boundary conditions, but these are limited in their application (Crank 1975). Alternatively, a transient form of the partial differential equation (PDE) for advective-diffusive transport (including spherical diffusion) can be solved using a standard numerical solver (e.g., the pdepe solver for MATLAB[®]). The pdepe function solves various initial-boundary value problems for systems of parabolic and elliptic PDEs. The accuracy of the first-order rate model was tested through comparisons to the pdepe solver (Equation 14) for models featuring step-change boundary conditions with three frequencies.

2.3.5 Integrating quartz-helium partitioning in the advection-diffusion equation

Helium equilibrium between pore water and quartz as well as partitioning between these domains was modelled by relating Equation 14 to the numerical solution to Equation 4. Equation 14 was modified to be used as a step-wise function and the helium exchange between domains became an additional source/sink term in Equation 4. The helium partitioning factor between quartz and pore water was estimated at 20–60 %. These estimates were based on the helium accessible volume and the total volume of quartz. The latter was derived from x-ray diffraction (XRD) analysis of Eromanga Basin aquitards (unpublished data).

3 Results

3.1 Helium transport parameters

Porosity, bulk density and vertical hydraulic conductivity measurements are presented as functions of depth in Figure 3.1 (Turnadge *et al.* 2018). At each depth two porosity values are shown: values were obtained from gas-porosity measurements and bulk density measurements, which assumed a mineral density of 2.7 g/cm³. Bulk density-based porosity estimates were consistently higher as these represent total porosity, whereas gas-porosity estimates represent effective porosity (n_e). The minimum, mean, and maximum values for total porosity are 4.9, 10, and 13%. The minimum, mean, and maximum values for effective porosity are 0.4, 0.9, and 2.3%. Vertical hydraulic conductivity values have a minimum, harmonic mean, and maximum of 0.9×10^{-7} , 2.1×10^{-7} , and 30×10^{-7} m/d.



Figure 3.1 Physical analyses of aquitard core samples including (a) bulk density; (b) gas permeametry-based porosity (blue circles) and bulk density-based porosity (black circles); and (c) vertical hydraulic conductivity derived from air permeability measurements.

Calculated formation temperatures, helium solubility and helium free-water diffusivity are shown in Figure 3.2. Temperature increases with depth from approximately 48 °C at the top of the Watermark Formation to 62 °C at the bottom of the Porcupine Formation. Solubility increases by 4% over the same depth range with a mean of 8.8×10^{-3} cm³ STP/g/atm. The free-water diffusion coefficient D_0 increase by 22% over the same depth range with a mean of 1.1×10^{-8} m²/s.



Figure 3.2 Factors that affect helium transport and partitioning: (a) temperature from downhole wireline logs (Santos (QNT) Pty Ltd 2009), (b) temperature-corrected helium solubility for freshwater (Crovetto *et al.* 1982) and (c) free-water diffusion of helium (Jähne *et al.* 1987).

3.2 Helium production parameters

Uranium and thorium concentrations and associated helium production rate are shown in Figure 3.3. Mean concentrations of U and Th are 1.9 and 4.8 ppm, respectively and are similar between the Watermark and Porcupine Formations. Mean concentrations of U and Th give a helium production rate of 1.16×10^{-20} cm³ STP/g_r/s.



Figure 3.3 Helium release factors: (a) uranium (black circles) and thorium (blue circles) concentrations and (b) production rates calculated from Equation 9.

3.3 Helium concentrations

Helium concentrations in quartz, helium accessible volumes (*HAVs*) and helium concentrations in groundwater are given in Figure 3.4. Error bars represent analytical uncertainties. Standard error propagation was used where appropriate. Helium concentrations in quartz increase with depth by a factor of 3 and reach a maximum in the middle of the aquitard. Concentrations then decrease slightly before increasing near the base of the aquitard. *HAV* values have no trend with depth and have a mean of 1.6×10^{-3} cm³_{He}/cm³_q. Applying sample-specific *HAVs* partially remove the vertical trend in helium concentration. Therefore, the average *HAV* (dashed line in Figure 3.4b) was used to calculate the helium concentrations in pore water.



Figure 3.4 Helium results: (a) measured helium concentration in quartz, (b) measured (circles) and mean (dashed line) helium accessible volume of quartz and (c) calculated helium concentration in pore water.

3.4 Rates of helium diffusion in quartz

3.4.1 Spherical model

Helium release rates during diffusion experiments were higher than expected, with approximately 60% of total helium released during the first heating step (i.e., after two minutes). Therefore, there are no data to constrain diffusion rates at early times. While the total heating time was initially expected to exceed 24 hours, all helium was released in less than three hours.

Calculated diffusion coefficients at 290 °C for a median grain diameter of 68 μ m are given in Table 3.1 and show a poor fit when using a spherical model (Equations 6.18 and 6.19 from Crank (1975)) with a single diffusion rate (Figure 3.5). With spherical geometry and two diffusion rates, the fit is good ($\Sigma \chi^2 = 0.07-0.32$; Table 3.1; Figure 3.5). When two diffusion rates were included in the solution, the faster rate (D_1) varied by a factor of 1.9 across three samples whereas the slower rate (D_2) was more relatively consistent, varying by a factor of 1.3.

At 290 °C and for a median grain diameter of 150 μ m, the equilibration of helium between pore water and quartz occurs within 8–11 hours. At 40 °C, the equilibration time increases to 54–76 × 10⁴ years (Crank 1975).

3.4.2 First-order model

First-order diffusion modelling produced a good fit to the data when two or more rates were used ($\Sigma \chi^2 = 0.09-0.46$; Table 3.1; Figure 3.5). Figure 3.5 shows that the inclusion of two rates produces

an acceptable fit to observed data. However, this first-order model does not match the spherical model. This is because there are no data to constrain the model at early times (i.e., t < 2 minutes) and the first-order model does not adhere to the geometry of the quartz (i.e., spherical). Therefore, the first-order model was fit to synthetic data derived from the spherical two--parameter diffusion model, which required at least three first-order rates k to produce a good fit to the synthetic data ($\Sigma \chi^2 = 0.29-0.64$; Table 3.1; Figure 3.5).

To use the modelled first-order rates at different temperatures, they must be related to D_q . The units of the parameters were matched by dividing D_q by the grain radius squared (D_q/a^2). The parameters D/a^2 and k were related by ratios that are independent of temperature.



Figure 3.5 Estimated helium diffusion rates for sample SLK696. The first-order two-rate model was fitted to experimental data. The first-order three-rate model was fitted to synthetic data (i.e. generated by the spherical model).

Table 3.1 Experimental diffusion rates. F_{D1}, F_{D2}, F_{k1}, F_{k2}, and F_{k3} are optimised volume fractions (see Eq. 15)

	SPHERICAL				FIRST-ORDER										
	one rate		two rates				two rates (fi	t to experimer	ntal data)		three rates (fit to spherical	model)		
Sample	<i>D</i> ₂₉₀ /a ² (10 ⁻⁴ 1/s)	$\Sigma\chi^2$	D _{290,1} /a² (10 ⁻⁴ 1/s)	<i>D</i> _{290,2} /a ² (10 ⁻⁴ 1/s)	FD1:FD2	$\Sigma\chi^2$	<i>k</i> _{290,1} (10 ⁻⁴ 1/s)	<i>k</i> _{290,2} (10 ⁻⁴ 1/s)	Fk1:Fk2	$\Sigma\chi^2$	<i>k</i> _{290,1} (10 ⁻⁴ 1/s)	<i>k</i> _{290,2} (10 ⁻⁴ 1/s)	<i>k</i> ₂ _{90,3} (10⁻⁴ 1/s)	Fk1:Fk2:Fk3	$\Sigma\chi^2$
SLK696	4.13	29.1	12.2	0.9	1.3:1	0.14	148	9.46	2.5:1	0.16	4090	186	13.1	1.0:3.0:2.4	0.64
SLK1137	1.91	20.6	6.55	0.73	1:1.4	0.32	110	8.17	1.4:1	0.46	1820	97.1	9.07	1.0:2.7:3.1	0.29
SLK1213	2.97	11.4	6.79	0.97	1.2:1	0.07	114	10.6	2.1:1	0.09	2630	133	15.1	1.0:2.8:2.9	0.43

a = 34 µm (median grain radius)

Comparisons to numerical solutions

Comparisons between analytical and numerical solutions for the step change in boundary conditions case are shown in Figure 3.6. The first-order rate analytical model was found to be in good agreement with the numerical model. Slight discrepancies between the two solutions are discussed below.



Figure 3.6 Comparisons between two-parameter spherical diffusion numerical model (solid orange line) and threeparameter first-order rate approximation analytical model (dashed blue line). Also shown is the pore water helium concentration (solid black line). The quartz grain size was specified as median diameter = 70 µm.

3.5 Modelled pore water velocity

Using the measured mean effective porosity (n_e) of 0.9% for both the Watermark and Porcupine Formations. For this case, the effective diffusion coefficient D_e becomes over four orders of magnitude smaller than the free water diffusion coefficient:

$$D_e = D_{0,w} n_e^m = D_{0,w} 0.009^2 = D_{0,w} 8.1 \times 10^{-5}$$
(16)

For this porosity value, the helium release rate (Equation 10) is 268 g_w/g_r . This suggests that, for every mass unit of water there are 268 mass units of rock contributing helium to pore water. Low porosity coupled with a high release rate results in relatively high helium pore water concentrations, as internally produced helium has great difficulty diffusing from the formation given such low porosity values (i.e., < 1%; Figure 3.8).

Estimates of effective porosity derived from gas permeameter testing were expected to be less than total porosity estimates derived from bulk density values. The latter of which is the more appropriate value to use when calculating helium release rates. Total porosity is appropriate for modelling helium transport because helium's small size, neutral charge and high diffusivity, which allows it to access the total porosity. Therefore, the effective diffusion coefficient using the total rock porosity (mean = 10%) is calculated as:

$$D_e = D_{0,w} n^m = D_{0,w} 0.1^2 = D_{0,w} 0.01$$
⁽¹⁷⁾

While this increase in effective diffusivity results in greater helium loss from the aquitard, the modelled helium pore water concentrations are still too high to match observed data (Figure 3.7). This suggests that the effective diffusion coefficient is larger than Equation 17 suggests.

The factor *m* in Equation 17 is based on the assumption that flow path tortuosity can be calculated as a power function of rock porosity using the coefficient *m* ((Jury *et al.* 1991), rather than being directly estimated. When analysing core samples from the San Andreas Fault Zone, California, USA, Ali et al. (2011) calculated the effective diffusion coefficient of helium in pore water at the core-scale and determined that tortuosity was a factor of approximately 2.0–4.3 times higher than the porosity. Use of a tortuosity value four times greater than porosity resulted in an adequate model to measurement misfit for the Watermark Formation, but resulted in helium concentrations in excess of measured values for the Porcupine Formation. This discrepancy is discussed below.





3.5.1 Steady state modelling

Solutions of the steady state form of the advection-diffusion equation (Equation 4) can be used to produce steady state distributions of helium in the pore water and quartz of an aquitard. Figure 3.8 shows calculated steady state helium distributions in the pore water for a range of upward and downward flow velocities. The model results compare well to the observed helium concentrations in the Watermark Formation when the vertical flow velocity is 0.02 mm/year or less. Conversely, steady state model results compare poorly to observed helium concentrations for the Porcupine Formation. This suggests that the helium profile in the latter unit is not at equilibrium, which may be the result of transient helium concentrations in the underlying Maules Creek Formation. Possible causes of such transient helium concentrations include changes in groundwater flow velocities owing to climate change, bursts of helium associated with tectonic events, etc. At present this is an important uncertainty that needs further corroboration.

Modelled helium profiles were found to be insensitive to the direction of vertical groundwater flow, indicating that diffusion is the dominant mechanism of helium transport in this context.



Figure 3.8 Quartz-helium measured pore water concentrations (closed black circles) and calculated steady state helium distributions in pore water (solid blue lines) and derived from quartz-helium (dashed orange lines) with (a) downward advection and (b) upward advection.

3.5.2 Transient modelling

For the purposes of transient numerical modelling, the helium concentration at the base of the Porcupine Formation (i.e., the lower boundary condition) was assumed to vary in time according to the following sine function:

$$C = \left(2 + \sin\left(\frac{t}{2 \times 10^5}\right)\right) \times 10^{-5} \tag{18}$$

The initial parameters of this model were chosen rather arbitrarily, and subsequently adjusted manually until they allowed a better description of the non-uniform helium profile. This resulted in input concentrations that ranged from $1-3\times10^{-5}$ cm³ STP/g with a period of 1.26×10^{6} years. For this boundary condition and for both upward and downward groundwater flow velocities of 0.02 mm/year resulted in solutions that were in acceptable agreement with observed data. Decreasing the vertical flow velocity by one order of magnitude produced comparable results. Greater decreases in vertical flow velocity had little effect on the vertical profile of helium as transport was dominated by diffusion.

Increasing the vertical flow velocity by one order of magnitude produced a vertical profile that was in good agreement with observed data when upward advection was considered. Conversely, when downward advection was considered, a poor match was achieved. None of the numerical model solutions were found to fit the data perfectly. Calibration fits could potentially be improved by modelling additional combinations of vertical flow velocity and boundary condition variations. However, the boundary conditions required for transient modelling over geological time scales are arbitrary and cannot be independently estimated.

In summary, it is suggested that the most pertinent result of the present study is that the modelling presented, which was constrained by analyses of aquitard core samples, was used to constrain pore water velocities to be in the order of 0.02 mm/year. Modelling results suggest that velocities may be up to one order of magnitude higher or several orders of magnitude lower. Furthermore, transient transport modelling using time-varying source boundary conditions indicate that helium concentrations remain equilibrated between pore water and quartz grains.



Figure 3.9 Vertical distributions of helium in pore water (black circles) derived from quartz-helium measurements and helium distributions in pore water (blue solid) and derived from quartz-helium (orange dashed) with (a) downward advection and (b) upward advection.

4 Discussion

4.1 Comparison with independent estimates

Measured core-scale vertical hydraulic conductivities for the Watermark and Porcupine Formation ranged between $0.9-30.0 \times 10^{-7}$ m/day while the estimated modern hydraulic gradient ranged between 0.001-0.015 m/m (Turnadge *et al.* 2018). Using the measured mean effective porosity of 0.9%, the modern pore water velocity therefore ranged between 1×10^{-8} m/day (0.00365 mm/year) and 5×10^{-6} m/day (1.8 mm/year). The lower end of this range is comparable to those derived from helium transport modelling (i.e., 0.002-0.02 mm/year). Both methods suggest that the Watermark/Porcupine aquitard features very low permeability at this location and, given its thickness (i.e., exceeding 500 m in some locations), leakage across these formations is quite low at this location.

However, it should be noted that these hydraulic gradients are not constrained by any directly observed data and represent modern conditions for specific areas of the Gunnedah Basin. Because hydraulic conditions have likely varied over millennia, the modern hydraulic gradient is unlikely to be representative of that in which the vertical profile of helium initially developed. Therefore, it is likely that estimates of aquitard vertical hydraulic conductivity derived using the present day hydraulic gradient coupled with vertical profiles of helium may result in incorrect estimates of formation-scale properties.

Under the very low fluid velocities predicted, mass transport across the aquitard at the well location will be dominated by diffusion. This is confirmed by Péclet numbers of 0.07–0.7 (see Equation 8), given that Péclet numbers of less than one indicate diffusion-dominated transport (Mazurek *et al.* 2011).

Using the quartz-helium technique a reasonably well constrained vertical hydraulic conductivity was derived. The associated uncertainty of approximately one order of magnitude (or larger if *K* values lower than the current minimum of 0.9×10^{-7} m/day are considered) may seem large, but for such low K_v values this uncertainty is considered acceptable, because inter-aquifer leakage at the well location would be insignificant. We did not pursue deriving a quantitative uncertainty about K_v because transient helium distributions greatly complicates the modelling procedure such that Monte Carlo-type randomisation approaches similar to those of Smith (2015) has proven beyond the scope of this project.

Transience in vertical profiles of aquitard helium concentrations has previously been observed in other groundwater basins, including the Eromanga Basin (Gardner *et al.* 2012) and the Adelaide Plains Basin (Bresciani *et al.* 2015), both of which are located in Australia. Continental helium fluxes are known to be spatially and temporally variable (Torgersen 2010), which may explain the seemingly transient distribution of helium observed in the Gunnedah Basin. Heterogeneities in geological properties may also affect the helium distribution.

Application of the quartz-helium method to other parts of the Gunnedah Basin to derive estimates of aquitard K_v would require carefully selection of aquitard thickness, temperature and hydraulic properties to ensure that equilibria between quartz and pore water helium concentrations could

be expected. For example, in the present study, aquitard core samples from the Slacksmith 1 corehole were selected because the Watermark and Porcupine Formations were quite deep at this location; therefore subsurface temperatures were sufficiently high to enhance helium diffusion in quartz and thereby establish equilibrium conditions required for analysis.

In a similar study using cores samples from several different sites in a GAB aquitard in South Australia, Gardner et al. (2012) obtained best-fit formation-scale hydraulic conductivities that ranged from 2×10^{-9} m/s to $\leq 1 \times 10^{-12}$ m/s with corresponding fluid fluxes ranging from 2×10^{-10} to $\leq 1 \times 10^{-13}$ m/s. Additional variability was observed using the measured helium concentrations in shallow samples: this could be explained when the vertical helium flux through the aquitard could vary by three orders of magnitude across their study area. These results indicate that substantial preferential flow paths exist, with vertical fluid fluxes through the aquitard that, at some locations, can be up to 1000 times higher than that derived from the core profiles. Thus, extrapolating the results of a few core profiles to a large area cannot be justified without an associated regional sampling campaign.

4.2 Effects of linearisation

Modelling of quartz – pore water helium equilibrium is a valuable tool that may be used to identify aquitards for which the quartz-helium method may provide meaningful results. The linearisation of the helium diffusion process in quartz grains (i.e., the replacement of *D* with *k*) greatly simplifies the modelling process by avoiding the need to solve a transient partial differential equation. However, the linearisation of transient diffusion processes requires some consideration because of potential for loss of fine-scale detail. First-order model approximations accurately reproduce the experimental helium release data (Figure 3.5) when two or three rate coefficients are used. However, as shown in Figure 3.5, at early times (when experimental data are potentially unavailable), first-order model results differ from those produced by the spherical diffusion model. For this reason, the first-order model was calibrated to the results of the spherical model. Without this correction, the diffusion from a spherical object where the majority of the volume, and thus helium, is contained in the outer 16% of the sphere.

The omission of spherical grain geometry from transport modelling warrants consideration as a loss of fine-scale detail occurs when the distribution of helium within quartz grains is lost. Complex helium gradients within quartz grains can result in rates of diffusion that cannot be characterised using the first-order model.

To assess this loss of fine-scale detail, consider again the step-change model from Figure 3.6. Figure 4.1 shows a subset of Figure 3.6 including the helium concentration versus grain radius relationship derived from the numerical spherical transport model and compares it to the concentration derived from the three-parameter first-order analytical model. At all steps, the concentration at the centre of the sphere only changes slightly, whereas the concentrations in the outer third of the sphere change most significantly. For example, at approximately 3×10^9 seconds, the boundary concentration has decreased to zero (i.e. helium concentration in the pore water becomes zero). The helium that previously diffused into the sphere will now diffuse outwards towards the lower concentrations at the centre of the sphere. This multi-directional diffusion generates non-uniform concentration profiles within quartz grains; these are conditions that cannot be accurately modelled by the simple first-order model.

While these complex diffusion patterns could be important under certain circumstances (especially at microscopic scale), for the purpose of this modelling (and given that larger sources of uncertainty exist), the simplification resulting from use of a first-order model is considered minor and should not significantly impact the modelling of quartz – pore water partitioning at the formation scale.



Figure 4.1 Example of the loss of fine-scale detail caused by the linearisation of helium diffusion from quartz: temperature is 40 °C and boundary step change frequency is 5.0×10^9 seconds (i.e., 158 years); inset boxes show normalised helium concentrations (*C*/*C*₀; y-axis) versus quartz grain radii (*r*; x-axis) (grain centres are located at lower left corners of X-Y boxes; interfaces between grain surface and pore water are located at bottom right corners).

4.3 Future usage and limitations

This study shows that helium can be considered to be in equilibrium between quartz and pore water in the Watermark and Porcupine Formations at the Slacksmith 1 corehole. At this location the formations are considerably thick (i.e., >500 m) and occur at significant depths within the basin (i.e., ~700 m); therefore a sufficiently high temperature is present to enhance diffusion. To assess whether equilibrium conditions are present in other parts of the basin where these aquitards are thinner and occur shallower in the basin, hypothetical helium transport modelling was undertaken. Three temperature profiles and three thicknesses were considered. All temperature profiles featured gradients of 25 °C/km with maximum temperatures of 20, 40 and 60 °C located at the base of the profile. The three aquitard thicknesses tested were 5, 50 and 500 m. The time period of each model run was 10^2 , 10^4 and 10^6 years, with longer times selected to be

appropriate for thicker formations, in which more time is required to approach a steady state helium distribution in pore water. For each temperature and thickness combination, three different velocities were considered. Three groundwater flow velocities were specified as functions of aquitard thickness:

$$v_i = 10 \times \left(\frac{10^{i-1}}{b}\right)$$
, $i = 1, 2, 3$ (19)

The combinations of flow velocities and aquitard thicknesses specified ensured that a range of advection-dominant and diffusion-dominant conditions were evaluated.

The initial pore water and quartz helium concentration distribution across the formation is a linear profile between the following boundary concentrations: 4×10^{-5} cm³ STP/g at the base and 0.1×10^{-5} cm³ STP/g at the top. At t > 0, the boundary concentration at the base shifts to 2×10^{-5} cm³ STP/g; the boundary concentration at the top of the profile shifts to 1×10^{-5} cm³ STP/g. This allows the observation of equilibrium of helium between pore water and quartz via the numerical model.

For a 5 m thick aquitard, equilibrium between domains does not occur at any temperature (Figure 4.2). This is not surprising because helium can be transported quickly through a relatively thin aquitard. The pore water helium distribution can therefore reach an equilibrium state much faster than quartz grains can gain or lose helium. This suggests that the quartz-helium method is not suitable for applications to thin aquitards as it is unlikely that equilibrium conditions exists in such contexts. Alternatively, application of the method in thin aquitards requires an independent indication that helium boundary conditions have not varied significantly over recent geological time.

The results from the 50 m model show that quasi-equilibrium conditions can be expected only at high temperatures, with the closest to equilibrium being seen at 60 °C and 0.2 mm/year (Figure 4.3). This greater degree of equilibrium can be attributed to the greater thickness, which means helium transport is slower because helium has larger distances to diffuse, which also means the helium gradients are lower. The longer time required for pore water helium to approach steady state also results in more time for helium to diffuse in and out of the quartz.

For the 500 m thick aquitard, most scenarios show complete or nearly complete equilibration of helium concentrations. Similar to the results of the 50 m thick model, the increased thickness and longer transport time across the formation result in a system that is slow to react to changes in boundary conditions; therefore sufficient time elapses to enable the equilibration between domains.

However, although equilibrium conditions may be expected in thick aquitards, this does not guarantee that conditions will be suitable for the estimation of pore water velocities. This is because changes in boundary conditions have lasting effects within the aquitard, which may result in helium distributions that are not readily interpretable, at least without requiring some assumptions with regards to temporal changes in boundary conditions. As previously stated, there is no way to independently estimate initial helium concentrations (Mazurek *et al.* 2011), which in turn means there is no way to independently estimate transient boundary conditions. However, formation-scale pore water velocities, and possibly hydraulic conductivities, can be estimated by trialling various initial conditions and boundary conditions. Because hydraulic conductivity values vary over orders of magnitude, the ability to constrain *K*_v estimates to within a single order of



magnitude represents a significant scientific advance, particularly when no alternative analyses currently exist by which to estimate fluid flow at the formation-scale.

Figure 4.2 Results of hypothetical modelling of helium transport between pore water (solid blue lines) and quartz grains (dashed orange lines) for in 5 m thick aquitard; vertical fluid flow direction is upward; *D* uses $\tau = 4n$; quartz grain diameter is 46–150 µm and is weighted to equal mass per grain size; time step shown is 5 years; and total simulated time is 100 years.



Figure 4.3 Results of hypothetical modelling of helium transport between in pore water (solid blue lines) and quartz grains (dashed orange lines) for a 50 m thick aquitard; vertical fluid flow direction is upward; *D* uses $\tau = 4n$; quartz grain diameter is 46–150 µm and is weighted to equal mass per grain size; time step shown is 500 years; and total simulated time is 10,000 years.



Figure 4.4 Results of hypothetical modelling of helium transport between pore water (solid blue lines) and quartz grains (dashed orange lines) for a 500 m thick aquitard; vertical fluid flow direction is upward; *D* uses $\tau = 4n$; quartz grain diameter is 46-150 µm and is weighted to equal mass per grain size; time step shown is 50,000 years; and total simulated time is one million years.

5 Conclusions

The quartz-helium method was successfully applied to core samples of the Watermark-Porcupine aquitard sequence from the Slacksmith 1 corehole. Analytical and numerical helium transport models were used to estimate a pore water velocity of 0.02 mm/year or less, which generally agrees with core-scale gas permeametry measurements. A transient vertical distribution of helium in the Porcupine Formation suggests that helium concentrations have been variable in the underlying source unit (i.e., the Maules Creek Formation) over recent geological time. The existence of transient conditions means that it is difficult to estimate the uncertainty of the derived pore water velocities and associated vertical hydraulic conductivities.

Laboratory measurements of diffusion in quartz were undertaken and subsequently upscaled for inclusion in a formation-scale advection-diffusion helium transport model. Model results indicate that pore water and quartz grain helium concentrations should currently be in equilibrium at the Slacksmith 1 corehole. Furthermore, modelling results also indicate that helium concentrations in quartz grains are a suitable proxy for helium concentrations in pore water, provided that the aquitard of interest is adequately thick (i.e., tens to hundreds of metres) and subsurface temperatures are at least 40 °C. It is recommended that the suitability of the quartz-helium method be evaluated on a case-by-case basis.

Additional recommendations for future studies include:

- Collection and analysis of pore water helium concentrations when new coring occurs. These results should be compared with quartz-helium measurements to further assess the equilibrium of helium between pore water and quartz.
- Performing diffusion testing at lower temperatures to capture the initial rate of helium release.
- Independently assess diffusion in the porous medium (i.e., quantification of tortuosity and total porosity).
- Study the role of faults on the helium migration and profiles through numerical modelling.

Given that there is currently no other method capable of estimating aquitard fluid flow properties at the formation scale, it is suggested that these datasets and interpretations are of much relevance to the characterisation of low permeability formations.

Shortened forms

ABBREVIATION	DESCRIPTION
CSG	Coal Seam Gas
CSIRO	Commonwealth Scientific and Industrial Research Organisation (Australia)
HAV	Helium Accessible Volume
MC	Monte Carlo (sampling methodology)
mD	milliDarcy
NSW	New South Wales
PDE	Partial Differential Equation
STP	Standard Temperature and Pressure (0 °C and 1 atm)
XRD	X-Ray Diffraction

Symbols

SYMBOL	DESCRIPTION [UNIT]
д	partial differential (-)
С	concentration (cm ³ STP/g)
X	goodness of fit (-)
D _{0,q}	maximum diffusion coefficient for helium in quartz, (m ² /s)
D_q	diffusion coefficient of helium in quartz at a specified temperature (m ² /s)
Dp	porous media dispersion coefficient (m ² /s)
De	porous median diffusion coefficient (m ² /s)
D _{0,w}	free water diffusion coefficient (m ² /s)
Ea	activation energy (kJ/mol)
F	fraction of quartz sample with a given diffusion rate (-)
g _w , g _q	helium release rate into water (cm ³ STP/ g_w), helium release rate from quartz
k	first-order rate (1/s)
k	permeability (mD or m ²)
К	hydraulic conductivity (m/day)
λ	dispersivity (m)
т	empirical exponent (-)
n	porosity (%)
ne	effective porosity (%)
p1, p _{imp}	standard pressure, pressure of helium impregnation (Torr or Pa)
Pe	Péclet number: ratio of advective versus diffusive transport (-)
R	gas constant (J K/mol)
$ ho_b$	bulk density (g/cm³)
S	solubility (cc STP/g/atm)
σ	standard deviation
T ₁ , T _{ini} , T _{imp}	standard temperature, in situ temperature, temperature of helium impregnation (°C or K)
t	time (seconds or years)
τ	tortuosity (-)
v	fluid velocity (mm/year)
z	depth (m)

Glossary

TERM	DESCRIPTION
Aquifer	Rock or sediment in a formation, group of formations or part of a formation, which is saturated and sufficiently permeable to transmit quantities of water to wells and springs
Aquitard	A saturated geological unit that is less permeable than an aquifer and incapable of transmitting useful quantities of water. Aquitards often form a confining layer over aquifers
Coal measure	Geological strata of the Carboniferous or Permian periods usually containing sequences of coal seams
Coal seam	Individual layers containing mostly coal. Coal seams store both water and gas. Coal seams generally contain groundwater that is saltier than that in aquifers that are used for drinking water or agriculture
Coal seam gas	A form of natural gas (generally 95 to 97% pure methane, CH ₄) typically extracted from permeable coal seams at depths of 300 to 1000 m. Also called coal seam methane (CSM) or coalbed methane (CBM)
Darcy's law	A constitutive equation that describes the flow of a fluid through a porous medium such as rock or soil
Depressurisation	The lowering of static groundwater levels through the partial extraction of available groundwater, usually by means of pumping from one or several groundwater bores or gas wells
Dirichlet boundary condition	Also known as a first type boundary condition, involves specification of the value that the solution of a differential equation needs to produce along the boundary of a model domain. Applicable to both numerical and analytical models
Effective porosity	The fraction of pores that are connected to each other and contribute to flow. Materials with low or no primary porosity can become very permeable if a small number of highly connected fractures are present
Groundwater	Water occurring naturally below ground level (whether in an aquifer or other low-permeability material), or water occurring at a place below ground that has been pumped, diverted or released to that place for storage. This does not include water held in underground tanks, pipes or other works
Hydraulic conductivity	A coefficient of proportionality describing the rate at which a fluid can move through a permeable medium
Hydraulic gradient	The difference in hydraulic head between different locations within or between hydrostratigraphic units, as indicated by water levels observed in wells constructed in those units
Hydrostratigraphic unit	A formation, part of a formation, or group of formations of significant lateral extent that compose a unit of reasonably distinct (similar) hydrogeological parameters and responses
Monte Carlo simulation	The use of Monte Carlo analysis techniques to estimate the most probable outcomes from a model with uncertain input data
Permeability	The measure of the ability of a rock, soil or sediment to yield or transmit a fluid. The magnitude of permeability depends largely on the porosity and the interconnectivity of pores and spaces in the ground
Porosity	The proportion of the volume of rock consisting of pores, usually expressed as a percentage of the total rock or soil mass
Regional groundwater models	Models that encompass an entire groundwater system, geological basin or other significant area of interest that extends well beyond the measurable influence of individual bores or borefields
Reservoir (hydrocarbon)	Porous or fractured rock formations that contain significant reserves of hydrocarbons. Naturally- occurring hydrocarbons such as crude oil or natural gas are typically trapped in source or host rocks by overlying low permeability formations
Saturated flow	Flow through a porous medium (such as soil or rock) in which the void space within the porous medium is entirely occupied by water (as opposed to water and gas)
Stratigraphy	An arrangement of sedimentary, metamorphic and/or igneous rocks
Unconventional gas	Natural gas found in a very low permeability rock, such as coal seam gas, shale gas, and tight gas. Unconventional gas such as coal seam gas is trapped in coal beds by adsorption of the gas molecules to

TERM	DESCRIPTION
	the internal surfaces of coal. It cannot migrate to a trap and form a conventional gas deposit. This distinguishes it from conventional gas resources, which occur as discrete accumulations in traps formed by folds and other structures in sedimentary layers
Upscaling	Upscaling is the process of transforming the detailed description of hydraulic parameters in a grid constructed at measurement scale to a coarser grid with less detailed description. It replaces a heterogeneous domain with a homogeneous one in such a way that both domains produce the same response under some upscaled boundary conditions

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