The principles of helium exploration

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Abstract: Commercial helium systems have been found to date as a serendipitous by-product of petroleum exploration. There are nevertheless significant differences in the source and migration properties of helium compared to petroleum. An understanding of these differences enables prospects for helium gas accumulations to be identified in regions where petroleum exploration would not be tenable. Here we show how the basic petroleum exploration playbook (source, primary migration from the source rock, secondary longer distance migration, trapping) can be modified to identify helium plays. Plays are the areas occupied by a prospective reservoir and overlying seal associated with a mature helium source. This is the first step in identifying the detail of helium prospects (discrete pools of trapped helium). We show how these principles, adapted for helium, can be applied using the Rukwa Basin in the Tanzanian section of the East African Rift as a case study. A thermal hiatus caused by rifting of the continental basement has resulted in a surface expression of deep crustal gas release in the form of high-nitrogen gas seeps containing up to 10% 4He. We calculate the total likely regional source-rock helium generative capacity, identify the role of the Rungwe volcanic province in releasing the accumulated crustal helium and show the spatial control of helium concentration dilution by the associated volcanic CO2. Nitrogen, both dissolved and as a free-gas phase, plays a key role in the primary and secondary migration of crustal helium and its accumulation into what might become a commercially viable gas pool. This too is examined. We identify and discuss evidence that structures and seals suitable for trapping hydrocarbon and CO2 gases will likely also be efficient for helium accumulation on the timescale of the Rukwa Basin activity. The Rukwa Basin prospective recoverable P50 resources of helium have been independently estimated to be about 138 BSCF (billion standard cubic ft: 2.78 × 109 m3 at STP). If this volume is confirmed it would represent about 25% of the current global helium reserve.

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The global helium provinces today, the US Mid-Continent, Canada, Algeria, Qatar/Iran, central Europe, Russia and Australia, are all areas of petroleum production. The history of helium exploration (Gluyas 2019a, b) is one of serendipity, helium only being discovered as a small fraction of the produced natural gas (methane). Many petroleum companies vent helium alongside other waste gases either because they do not know there is helium in their gas mixture or they fail to recognize its value (Clarke et al. 2012).

The USA held a near monopoly on helium supply throughout the twentieth century. However, new discoveries have not delivered the volume required to meet growing demand, particularly those associated with the use of helium in medical cryogenics. Currently, growth in the helium market is around 11% per annum (Research and Markets 2020). In the last decade both Algeria and Qatar have begun to sell helium extracted from hydrocarbon gas produced for liquefied natural gas exports. The concentration of helium in both Algerian and Qatari petroleum gas is typically below 0.1%. Should demand for liquefied petroleum gas decline, then extraction and sale of helium from this source would not remain economical.

The failure of petroleum exploration to deliver additional reserve volumes of associated helium has led to three periods of global shortage of helium since 2010. This caused interruption of some hospital services in the UK and elsewhere, and universities were informed that demand would not be met (Connor 2013; Stokes 2013; Kalin and Finn 2017; Murphy 2019). Given the importance of helium to modern society and its irreplaceable use in medical cryogenics, leak detection, as a lift gas and in breathing mixtures, it came as a surprise to the authors that back in 2011, when we began this work, we could find no exploration strategies specifically developed for helium. A tested and successful helium exploration strategy was clearly needed (American Physical Society et al. 2016; Ballentine 2017).

Here, established petroleum exploration questions have been adapted to create a helium exploration strategy to enable regions globally to be assessed for their helium prospectivity. The strategy was developed by addressing the following questions (Table 1):

1. What are the source rocks for helium?
2. How do the source rocks mature for helium generation?
3. What causes helium to escape from its source rock (primary migration)?
4. How is helium transported throughout the subsurface (secondary migration)?
5. How is helium concentrated in subsurface reservoirs to form a helium accumulation?
Earth’s cratonic basement rocks are both very old and commonly of granitic composition – rich in thorium at 20–30 ppm (René 2017), but containing only modest quantities of uranium (typically around 4 ppm: Adams et al. 1959). For example, basement rocks and cratons of Archean–Proterozoic age (3.8–0.54 Ga) in the Canadian Shield or the Yavapai-Mazatzal province in the southern USA are predominantly metamorphic or granitic, and contain on average 2.8 ppm U and 10.7 ppm Th (Burwash and Cumming 1976). Mafic basement rocks tend to have lower uranium and thorium contents (Rudnick and Fountain 1995; Heikal et al. 2013).

### Defining the helium system

**Helium source rocks and helium generation**

The dominant stable isotope of helium, $^4$He, is sourced radiogenically from the alpha decay of $^{238}$U, $^{235}$U and $^{232}$Th isotopes in Earth’s mantle and crust. Thus, a helium source rock will be one that has the combination of U and Th concentrations and age to provide the helium. Radioactive decay is a first-order physical process in which the rate is dependent upon the absolute amount of the isotope and is unaffected by temperature or pressure. It is measured in terms of radioactive ‘parent’ half-life: that is, the time taken for half of the parent element to ‘decay’ to the daughter element. It follows that the older the rock is, the more the isotopes will have decayed, with the corresponding production of helium. The half-lives of the three relevant isotopes are: $^{238}$U = 4.468 Gyr (billion years), $^{235}$U = 0.7 Gyr and $^{232}$Th = 14.05 Gyr.

### Primary migration

The primary migration of helium is a two-stage process that requires initial migration (diffusion) out of helium-producing minerals into the interstitial porosity, and migration out of the source rock.

Radiogenic $^4$He produced from the alpha decay of $^{238}$U, $^{235}$U and $^{232}$Th will be found within 10–20 μm of the parent radionuclide (e.g. Martel et al. 1990). This is due to the penetration distance of the original alpha particle (Meunier et al. 1990). This distance is within the same length scale as most mineral crystals and grains. Thus, helium can become trapped both within the mineral matrix and on mineral grain boundaries, depending on the distance of the radionuclide from the grain boundary (Martel et al. 1990; Ballentine and Burnard 2002; Barry et al. 2015).

### Maturation

During tectonically quiescent periods, subsurface concentrations of $^4$He naturally increase over time as uranium and thorium decay. The helium generated remains in both the mineral matrices and interstitial pore fluids (Reimer 1976; Bottomley et al. 1984; Zadnik and Jeffery 1985; Ballentine and Burnard 2002). This has been observed in the Canadian and Fennoscandian shields, Kaapvaal Craton and Greenland Craton (Zadnik and Jeffery 1985; Lippmann-Pipke et al. 2011; Holland et al. 2013; Neretnieks 2013; Warr et al. 2018).

The observation that shallow (<2 km) waters and gases in continental sedimentary systems contain low helium concentrations (ppm level) is mostly due to the ability of helium-retentive minerals, such as apatite, zircon, uraninite or monazite, to retain and accumulate helium below their closure temperatures (Table 2) (e.g. Ballentine and Burnard 2002), as well as the relatively young age of shallow sedimentary systems and associated fluids. For apatite, closure temperature is c. 70°C, indicating that under an average crustal temperature gradient of 30°C km$^{-1}$, assuming a surface temperature of 10°C, helium would only start being released from these minerals at 2 km or deeper. Since most helium-rich gas reservoirs generally occur at shallower depths (Table 3), helium atoms and thus helium accumulations both require a mechanism for their release from deeper regional geological systems, transport and a focusing mechanism.

### Table 1. Synthesis of components required for a viable helium province

| Stage                      | Helium system                                                                 |
|----------------------------|------------------------------------------------------------------------------|
| Source                     | $^{238}$U, $^{235}$U and $^{232}$Th decay in the crust, producing alpha particles that acquire electrons and become helium. |
| Maturation                 | Time to accumulate (stability of crust) v. volume of crust.                  |
| Primary migration          | Heat to above the closure temperatures of minerals with respect to helium retention v. diffusion. Release of nitrogen from associated minerals and clays. Tectonism and rock fracturing to release fluids. |
| Secondary migration        | Gas buoyancy and/or solution in water and hydrostatic drive.                 |
| Accumulation in reservoir  | Direct input into trap of a buoyant free-gas phase; or degassing of oversaturated groundwater; or gas stripping via hydrocarbon (CH4) or magmatic (CO2) gas phase. |
| Trap integrity and longevity | Risk of: microseepage; capillary failure; fracture failure; tectonic destruction of trap. |

### Table 2. Helium closure temperatures for helium-retentive minerals

| Mineral  | Closure temperature range (°C) | References |
|----------|-------------------------------|------------|
| Apatite  | 55–100                        | Lippolt et al. (1994), Wolf et al. (1996), Farley (2000), Farley (2002), Shuster et al. (2006) |
| Hematite | 90–250                        | Bähr et al. (1994), Farley (2002) |
| Zircon   | 180–200                       | Farley (2002), Reimers (2005), Reich et al. (2007), Cherniak et al. (2009) |
| Garnet   | 590–630                       | Dunai and Roselieb (1996), Farley (2002) |
| Monazite | 182–299                       | Boyce et al. (2005) |
| Titanite | 150–200                       | Reimers and Farley (1999), Farley (2002) |
| Uraninite| c. 200                        | Martel et al. (1990), Stuart et al. (1994) |

Ranges in closure temperature for individual minerals are due to combinations of differing grain sizes and cooling rates.
Table 3. Average depths of a selection of helium-producing reservoirs in the USA

| Field and location          | Producing reservoir | Helium concentration (%o) | Depth of producing reservoir (km) | References |
|-----------------------------|---------------------|---------------------------|-----------------------------------|------------|
| Hugoton, - Panhandle, Kansas | Chase Group Permian | 0.60 (average)            | 0.90                              | Lollar et al. (2002), Gage and Driskill (2005) |
| Oklahoma – Texas, USA       | Dolomite (Permian)  | 0.67 ± 2.18               | 1.31 (3 average)                  | Ballentine and Lollar (1992), Chidsey (1991) |
| Woodside, Utah, USA         | Entrada (Jurassic)  | 0.60 ± 2.34               | 0.95                              | Morgan and Chidsey (1991), Harris (1993) |
| Huntley Dome, Utah, USA      | Entrada (Jurassic)  | 0.62 ± 2.70               | 0.31                              | Chidsey et al. (2005), Chidsey and Harris (2005) |
| Model Dome, Colorado, USA    | Topeka (Pennsylvanian) and Wabaunsee (Pennsylvanian) | 0.52 ≤ | 0.73 | Dobbin (1968), Danabalan (2017) |
| McElmo Dome, Colorado, USA   | Leadville (Mississippian) | 0.38 ± 2.50 ± 70          | 0.94                              | Dobbin (1968) |
| LaBarge, Wyoming, USA        | Madison (Mississippian) | 0.50 ± 0.73               | 0.39                              | Hamak (1989), Stewart and Street (1992), De Bruin (1995), Martin et al. (2008) |

The principles of helium exploration

The first stage of primary migration, the diffusion of helium from producing minerals into the surrounding pore or fracture space, enables high concentrations of helium to accumulate in tectonically quiescent areas of the crust (Lippmann-Pipke et al. 2011; Holland et al. 2013; Neretnieks 2013; Warr et al. 2018). The bulk diffusion of $^4$He in the crust is limited in terms of length scale. Diffusion and buoyancy-driven fluid advection can both contribute to the migration of helium from the crust into the sedimentary cover, with advection likely to be the dominant process in regions of recent tectonic activity and diffusion controlling helium release in more quiescent systems (Torgersen 1989, 2010; Hussain 1997; Ballentine and Burnard 2002; Neretnieks 2013; Cheng et al. 2018).

In the case where advection is responsible for helium transport from the deep continental crust to shallow regions, then two conditions are required. First, a thermal event, one which is significant enough to overcome the closure temperature associated with the various minerals within which helium is trapped and to generate, via fluid-overpressure-induced fracturing, an interconnected fracture fluid migration pathway (Warr et al. 2018). Secondly, a carrier fluid is required to facilitate advective movement out of the source rock. The carrier phase may be groundwater with the gases held in solution, or a gas phase such as N$_2$, CO$_2$ or CH$_4$. Both conditions require a significant increase in the regional thermal gradient, which is likely to be caused by tectonism such as extensional rifting, orogeny or volcanic activity. Moreover, the timing needs to be correct insofar as the source rock needs to be mature and replete with helium.

An example of the primary migration of $^4$He is currently occurring within Yellowstone National Park in the USA (Lowenstern et al. 2015). Calculations show that the supervolcano, which is heating the Archean-aged Wyoming Craton, is releasing the $^4$He accumulated within the craton source alongside its magmatic CO$_2$ carrier gas.

In subsurface reservoirs, helium is always found with nitrogen, although nitrogen can also occur without significant helium. $^4$He/N$_2$ in natural gas fields typically ranges between 0.02 and 0.20 (Pierce et al. 1964; Poreda et al. 1986; Gold and Held 1987; Jenden et al. 1988a; Jenden and Kaplan 1989; Stilwell 1989; Hyagon and Kennedy 1992; Hutcheon 1999) and can be a mixture of helium-associated nitrogen and other sources of nitrogen (Ballentine and Sherwood Lollar 2002). The isotopic ($\delta^{15}$N) composition of the N$_2$ end member associated with the economic $^4$He in the Kansas–Texas Hugoton–Panhandle gas field falls within a very narrow range (~3.00 to +2.45‰). This compares with the range seen from both low-temperature metamorphism of the crust and the release of ammonia from clays (~5.00 to +4.00‰) (Zhu et al. 2000; Ballentine and Lollar 2002; Danabalan 2017). That radiogenically produced helium is associated with non-radiogenic nitrogen suggests they share a process in common, although their source rocks could be different; nitrogen, as the major gas, may be acting as an advective carrier gas phase for the helium or providing the primary mechanism of gas phase formation from solution.

The link between $^4$He and N$_2$ during primary migration is discussed in a study of the Eger Rift by Weinlich et al. (1999) with an added caveat: while this region exemplifies primary migration for the crustal-sourced helium system, it is also shows the potential dilution of $^4$He- and N$_2$-rich gases by the addition of magmatically sourced CO$_2$ and associated gases in a tectonically active region, also seen in the Yellowstone example discussed earlier (Weinlich et al. 1999; Lowenstern et al. 2015).

Gases from the Eger Rift in central Europe show a CO$_2$ dilution trend for N$_2$ and $^4$He concentrations close to current geothermal centres in the area (Fig. 1). This is an indicator that tectonism such as rifting and associated volcanism and high heat flows can facilitate the primary migration of helium out of previously quiescent source rocks. If a trap is already in place when tectonism occurs, there is a...
possibility that the upward migration of $^4$He and N$_2$ as a free-gas phase may be all that is needed to generate a helium-rich reservoir. If the primary fluid contains no gas phase, or mixing with shallower groundwater causes any gas phase to be fully dissolved into solution, the salinity and pressure–temperature regime controlling the gas saturation point of the groundwater will need to be exceeded for a helium-rich gas reservoir to form.

However, primary vertical migration alone cannot explain the presence of large-volume helium-rich gas in areas that have not experienced any recent major tectonic activity, such as in Kansas, USA (Jenden et al. 1988b) or the Williston Basin, USA–Canada (Cheng et al. 2018). Such cases instead may require long-distance secondary migration (i.e. lateral migration of helium) or diffusional control.

**Secondary migration**

Secondary migration of helium is the lateral and vertical movement of helium and other associated gases after primary migration has occurred. Secondary migration can occur as free-gas migration or movement of groundwater containing dissolved gases (Table 4), or, be it very slowly, by diffusion through static fluids (Torgersen 1989, 2010; Cheng et al. 2018).

Observed correlations between air-saturated-water (ASW)-derived noble gas isotopes such as $^{20}$Ne and $^{40}$Ar, and admixtures of $^4$He and $^4$He-associated N$_2$ in fields from Kansas, Oklahoma, Texas and Arizona show strong evidence of groundwater involvement in helium-rich systems (Ballentine and Lollar 2002; Gilfillan et al. 2008; Danabalan 2017). In addition to groundwater movement, regional uplift or other mechanisms resulting in a pressure drop could also cause the carrier gas, and hence helium, to exsolve from solution to generate a free-gas phase (e.g. Sorenson 2005).

**Accumulation in reservoirs**

When groundwater containing dissolved $^4$He and N$_2$ equilibrates with a CO$_2$ or CH$_4$ gas phase, insoluble $^4$He and N$_2$ will preferentially exsolve from the groundwater and partition into the free-gas phase (gas stripping). In this way hydrocarbon (or CO$_2$) generation and buoyant migration as a gas phase through a groundwater containing accumulated but dissolved helium and nitrogen will result in a helium-rich hydrocarbon (CO$_2$) phase, with the resulting helium concentration dependant on both the amount of helium accumulated in the groundwater and the hydrocarbon gas/water ratio. Similarly, a pre-existing hydrocarbon or CO$_2$ gas phase within a geological trapping structure promotes the degassing of groundwater containing accumulated $^4$He and N$_2$ (e.g. Barry et al. 2016, 2017). This process operated to form most of the helium deposits currently exploited. Gas stripping by its nature dilutes any $^4$He, and commercial gas fields containing admixtures of CH$_4$ or CO$_2$ contain at most concentrations of up to a few per cent of $^4$He.

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**Table 4. Possible mechanisms of secondary migration and gas phase formation for the helium system**

| Secondary migration mechanism | Description |
|------------------------------|-------------|
| Free–gas phase – gas buoyancy | • The $^4$He–N$_2$ gas phase released from minerals into proximal fracture porosity migrates surfacewards by gas buoyancy along faults or lithologies with enhanced porosity and permeability |
| Groundwater flow – hydrostatic | • The $^4$He–N$_2$ gas phase from primary migration or after limited secondary migration contacts groundwater and at high pressure is dissolved within the groundwater |
| The role of secondary CH$_4$ or CO$_2$ | • $^4$He–N$_2$-rich groundwater migrates due to hydrostatic drive. The formation of a $^4$He–N$_2$ gas phase may be subsequently caused by changes in water temperature, salinity or pressure that cause $^4$He–N$_2$ saturation within the migrating water |
|                             | • The addition of CO$_2$ and/or CH$_4$ to any $^4$He–N$_2$-rich groundwater phase will enable gas saturation to be reached at a deeper level, but will also result in $^4$He–N$_2$ being diluted in any exsolution gas |
|                             | • Gas-buoyancy-driven migration to trap |
|                             | • Migration of a $^4$He–N$_2$-rich groundwater that contacts an existing gas phase (or vice versa) will result in dissolved $^4$He–N$_2$ partitioning into the gas phase (so called ‘gas stripping’)

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**Fig. 1.** $^3$He/$^4$He ratio ($R/R_a$) in natural gas seeps plotted as a function of distance from the geothermal system in the Egger Graben, Czech Republic (inset) (modified from Weinlich et al. 1999). Samples close to the gas-escape centre contain only trace levels of $^4$He (percentage shown in boxes), are CO$_2$ dominated and have $^3$He/$^4$He ratios > 1. This is consistent with a significant contribution of crustal-derived gas in the distal fluids. This pattern is observed in other systems (e.g. Barry et al. 2015).
helium. There are, however, several gas accumulations that contain as much as 10% helium and 90% nitrogen. Examples include the Permian strata of the USA Kansas Basin, Central Kansas Uplift, as well as in N₂–⁴He-rich wells in Utah, Montana (USA) and Saskatchewan (Canada). In these occurrences, the role of diluant gases has been minimal and the groundwater must have been oversaturated with ⁴He-associated N₂. Depressurization of groundwater through uplift or upward migration of nascent fluids from deeper source rocks are mechanisms that would ensure the bubble point is exceeded for any dissolved gases and thereby the preservation or formation of a primary (generated in the source rock) ⁴He and ⁴He-associated N₂ free-gas phase.

**Trap efficacy, leakage and destruction**

Once ⁴He and associated gases have migrated into a gas trap, the preservation of ⁴He in that trap is a function of the rate at which ⁴He is supplied to the gas pool less the rate at which helium leaks out. The capillary entry pressure required to force helium gas through the seal is similar to that of CO₂ (Wollenweber et al. 2009). Thus, seals that are suitable for CO₂ and hydrocarbon gases would similarly trap helium with this mechanism. Diffusive loss, however, is a function of atomic or molecular mass, and helium has a higher diffusion coefficient than other gases. Low-permeability seals such as evaporite minerals, or a cap rock with similar sealing efficiency, have the physical characteristics that would allow helium to be stored for long periods (100 Myr) of time without significant diffusive leakage (Broadhead and Gillard 2004; Broadhead 2005; Cheng et al. 2018). There is also evidence that less efficient (higher-permeability) cap rocks could suffice. For example, several gas fields in the Pannonian Basin, a Miocene extensional basin with a high thermal gradient, contain helium concentrations at relatively high, although subcommercial, concentrations (Ballentine et al. 1991). Dominated by shale and mudrock seals, these gases contain radiogenic ⁴He/⁴⁰Ar* at a similar ratio to that predicted for production in the crust from regional estimates of the basement (U + Th)/K (where ⁴⁰Ar* is radiogenic ⁴⁰Ar produced from the decay of ⁴⁰K). The preservation of the predicted radiogenic ⁴He/⁴⁰Ar* in these gases suggests that ⁴He has not been preferentially lost, relative to the ⁴⁰Ar*, from the gas phase via diffusion on the timescale of trap filling. However, older reservoirs carry a greater risk of diffusional loss and loss due to tectonic events (similar to hydrocarbon plays).

**Tanzania: a helium exploration case study**

The Tanzanian section of the East African Rift System (EARS) is an exemplar region where all of the characteristics for development of a helium resource are met. In this sense, Tanzania represents a unique opportunity to develop a ‘play fairway’ approach for helium exploration. A play fairway is defined as the area occupied by a horizon with access to migrated helium from its mature source rock. Within the area occupied by the play fairway one would expect to find prospects, configurations of seal and reservoir rock that could trap helium and other buoyant gases.

**Geological background**

The East African Rift (EAR) is a classic example of a continental rift. Crustal extension typically occurred through normal faulting that thinned the crust and resulted in upwelling of dense and hot lithospheric mantle rocks. The lithospheric mantle in turn deformed and was replaced by hotter asthenosphere. The process of heat transfer beneath the rift to the lithosphere reduced rock density and caused time-dependant regional uplift on the timescale of tens of millions of years (McKenzie 1978; Sengör and Burke 1978). The location of crustal extension was often influenced by pre-existing and large-scale pre-rift tectonic boundaries (Nyblade and Brazier 2002).

The EARS has evolved broadly from north to south and can be divided into several discrete diachronous rift sectors. The northern end of the rift system in Ethiopia and Afar is the most mature where it is transitioning from continental rifting to seafloor spreading. In Kenya the rift is well advanced, whilst the southern end of the eastern branch of the EAR, in northern Tanzania, is still at a juvenile rifting stage. The earliest basaltic volcanism occurred between 45 and 39 Ma in southwestern Ethiopia, with a major phase of flood basalt volcanism by 31–30 Ma (Ebinger et al. 1993). Regional heating and widespread mantle metasomatism along the asthenospheric–lithospheric boundary, inferred from kimberlites in the Archean cratons around the rift, is likely to have occurred before any surface expression of rifting across the East African Plateau (Ebinger et al. 2013).

The most juvenile rift sector marks the southern termination of the eastern branch and is expressed by a pronounced splay in the Tanzanian Divergence Zone (TDZ). Volcanic activity began at 8–4 Ma in the centre of the TDZ, with volcanism extending to the outer graben after 4.5 Ma. The Manyara Rift has been affected by rifting over a larger area than other young rifting zones, and has retained a thick lithosphere and a lower crust that has not viscously deformed (Uwe 2014). Volcanism migrated southwards, with ages of 4.9–1.5 Ma in the northern Manyara Rift and 1.5–0.7 Ma in the southern branch (Bagdasaryan et al. 1973; Foster et al. 1997).

The western branch of the EAR extends over 2000 km, from Lake Albert in the north to Lake Malawi. In parts of the western branch, subsidence is at its greatest in the entire rift, while rift-flank uplift is also pronounced (Uwe 2014). In the Albertine Rift of western Uganda, the lithosphere has thinly thinned at all and the lower crust has not viscously deformed; the chemistry of the magmatic rocks is primitive (mantle derived) and the volume of magmatism is limited (Uwe 2014). Volcanism throughout the western branch occurs in four distinct centres (Toro-Ankole, Vrunga, South Kivu and Rungwe), but there is no clear trend in when the volcanism started, with estimates from three systems at c. 10 Ma (Rogers 2006; Ebinger et al. 2013).

The c. 350 000 km² Archean-aged Tanzanian Craton lies in the centre of Tanzania between two branches of the EARS (Fig. 2) (Weeraratne et al. 2003). It was emplaced at c. 2.7 Ga, and is primarily composed of greenstone belts and granite with ages upwards of 2.4 Ga (Pinna et al. 1994; Dawson 2008). The craton experienced several collisional events early in its history, resulting in peripheral mobile belts. The Usagaran (2.0–1.8 Ga) and Ubendian (2.1–1.8 Ga) mobile belts formed via subduction-related accretion, whereas the Mozambique Belt was formed by a later oblique collisional event (<1.3 Ga) and was then reworked by the multistage Pan African Orogeny (950–550 Ma) (Quennell 1956; Leoni et al. 1994; Muhongo and Leoni 1994; Mruma 1995; Fritz et al. 2005; Vogt et al. 2006; Dawson 2008; Macheyeky et al. 2008; Boniface et al. 2012).
the presence of trapping structures in proximity to helium migration pathways and which formed prior to helium migration.

**Developing a play fairway for Tanzania**

The \(^4\)He generation potential is an estimate of the volume of \(^4\)He. We separate the Tanzanian system into five distinct regions of interest: (1) the Tanzanian Craton (350 000 km\(^2\)); (2) the Ubendian Belt (75 000 km\(^2\)); (3) the Usagara Belt (57 600 km\(^2\)); (4) the North Tanzanian Divergence (NTD), which consists of parts of the reworked Usagaran Belt, Mozambique Belt and the Tanzanian Craton (this calculation is confined to the Gregory, eastern, rift arm 42 500 km\(^2\); and lastly, on a local scale, (5) the Rukwa Basin asymmetrical half-graben (Roberts et al. 2012a, b) (basement and sediment fill) within the Ubendian Belt (12 800 km\(^2\)) (Fig. 2).

For each region we convert its surface area into a volume by considering helium generation to a depth of 10 km. The crustal thickness in these regions varies from 21 to 27 km (Getachew et al. 2011) but radioelement (U + Th) concentrations (2.8 ppm U and 10.7 ppm Th, respectively, values used in the calculations that follow) tend to be higher in shallower, less mafic regions of the crustal section above c. 10 km (Rudnick and Fountain 1995). A 10 km depth therefore provides a minimum estimate for the helium generation potential of any individual region. It is further assumed that this crust has an average crustal density of 2.7 cm\(^3\) g\(^-1\) and an average porosity of 0.64% (Chaki et al. 2008). The thickness of the sediment-fill in the Rukwa Basin has a maximum depth of about 11 km (Roberts et al. 2012a, b), and the sandstone reservoir (Red Sandstone Group) porosity of up to 28% has been measured (Roberts et al. 2012a, b) (Table 5). The Red Sandstone Group occurs below the Lake Beds, which in turn has the potential to act as a regional seal.

In addition, an age is needed before which the helium content of the regional rock volume is assumed to be negligible, and from which subsequent helium generation will have accumulated and/or been released. For the Tanzanian Craton, the ‘resetting age’ is assumed to be the last phase of metamorphism at 2.4 Ga (Pinna et al. 1994; Weeraratne et al. 2003). For the NTD, this is assumed to be when it was accreted onto the Tanzanian Craton at 2.0 Ga (Ebbing et al. 1997; Dawson 2008). The resetting age for the Ubendian Belt, the southern Usagaran Belt and the Rukwa Basin (basement) is likely to be when they were each reworked during the Pan African Orogeny at 570 Ma (Reddy et al. 2004; Boniface and Schenk 2012; Boniface et al. 2012). The deposition of the Karoo Supergroup sediments within the Rukwa Basin occurred at 260 Ma and forms the accumulation age for the deeper basin infill (Wescott et al. 1991; Delvaux et al. 1998; Baiyeguhi et al. 2014). The volume of helium produced since the nominal resetting age for each of these regions is shown in Table 5 and ranges from a combined sediment and basement helium generation volume in the Rukwa Basin of 0.16 × 10\(^{12}\) m\(^3\) \(^4\)He (STP) (5.65 × 10\(^3\) billion cubic ft (BCF)) to 170 × 10\(^{12}\) m\(^3\) \(^4\)He (STP) (6 × 10\(^6\) BCF) within the Tanzanian Craton. For context, the global annual industrial use of helium is c. 160 × 10\(^6\) m\(^3\) \(^4\)He (STP) (5.65 BCF). The combined efficiency of helium release and gas-phase trapping will determine the recoverable helium volume in any one area.

In the absence of any thermal perturbation there is evidence that rocks forming the crystalline basement dominantly lose helium slowly by diffusion (Torger sen 2010; Neretnieks 2013) and, as a result, retain much of their generated helium, on timescales of up to billions of years (Lippmann-Pipke et al. 2011; Holland et al. 2013; Warr et al. 2018). Helium release from each of the different regions in Tanzania requires identification of a mechanism. The development of the EARS is an obvious source of geologically recent and regional thermal input, with surface exhibition of enhanced heat flow shown by Cenozoic volcanic activity and extant geothermal springs. Over 15 geothermal areas have been identified in Tanzania, and summaries of their water and gas chemistry have been published (James 1967a, b; Walker 1969). Five of these occur on the Archean Tanzanian Craton and its Precambrian surrounds, the remaining geothermal centres are found near Quaternary volcanism both in the Tanzanian divergence zone and within the western branch of the EARS. We cannot at this stage identify whether the region has released helium during earlier thermal events.

In Figure 2 we superimpose the major volcanic provinces, geothermal springs and regions over a base map of Tanzania that shows significant sedimentary overburdens, potentially capable of forming stratigraphic or structural traps. This provides a first ‘play fairway’ for Tanzania.
Table 5. Radiogenic 4He volumes generated to 10 km depth since the last tectonic event within different parts of the Tanzanian system |

| Region and age of last thermo-tectonic event prior to current Rifting | Source area (km²) | Radiogenic 4He volume (m³ STP) | References |
|---|---|---|---|
| Tanzania Craton (2.4 Ga) | 350,000 | 1.7 × 10¹³ | Pinna (2003) |
| Ubendian Belt (570 Ma) | 75,000 | 7.4 × 10¹² | Boniface et al. (2012) |
| Southern Usagaran Belt (570 Ma) | 75,000 | 5.7 × 10¹² | Reddy et al. (2004) |
| North Tanzanian Divergence (2.0 Ga) | 50,000 | 1.6 × 10¹³ | Engega et al. (1997), Boniface et al. (2012) |
| Rukwa Basin (basement) (570 Ma) | 50,000 | 1.3 × 10¹³ | Wescott et al. (1991), Delvaux et al. (1998), Babu et al. (1999) |
| Rukwa Basin (sediments) (260 Ma) | 12,800 | 3.3 × 10⁶ | |

Refining the play fairway

It is clear that in any release of helium from the basement formations proximal to volcanic centres risks being diluted by CO₂ associated with volcanic outgassing. This risk is clearly demonstrated at Yellowstone in the USA where a significant flux of ⁴He is liberated from the crust by the extant volcanism, but the helium concentration within the dominant CO₂ gas is far too low to be commercially viable (Lowenstein et al. 2014). Determining the size of a thermal aureole around a volcanic system, where helium (and likely nitrogen) is liberated from the basement rocks and yet is suitably distal as to reduce the risk of CO₂ dilution (Fig. 1), is still to be determined and beyond the scope of this paper. It is also important to consider the hydrocarbon generation potential of sedimentary systems, as this subsurface gas source may also act to dilute helium concentrations. Nevertheless, both CO₂ and hydrocarbon (CH₄) subsurface gas sources, while diluting any pristine basement helium, may provide a mechanism that either prevents helium from being dissolved entirely in the groundwater or strips helium dissolved in groundwater into the gas phase to form a subsurface gas field (e.g. Ballentine and Lollar 2002). In practice, therefore, this means that helium-prone areas will have sweet spots or Goldilocks zones in which there is neither too much nor too little of these often-associated gases.

Despite uncertainty as to the precise boundaries of the CO₂ dilution/concentration zone, the Albertine Graben north of the Kivu Volcanic Complex in East Africa can be regarded as a lower-priority helium province because of the proximity of three volcanic complexes (Toro Ankole, Virunga and Kivu), each with potential for CO₂ dilution. These are in addition to multiple hydrocarbon seeps containing CO₂ around Lake Albert, and high concentrations of both CH₄ and CO₂, as well as mantle-derived helium, reported in Lake Kivu (Schoell et al. 1988; Tedesco et al. 2010; Abeinomugisha and Kasande 2012).

The Tanganyika rift area appears to be a good prospect due to both its size and distance to current volcanic activity. Helium-rich geothermal springs to the east of the lake at Uvinza produce highly saline fluids and gases that contain 2.5% ⁴He. The ³He/⁴He ratios of the gas are 0.28Rₑ indicating a crust-dominated helium source (Pflumio et al. 1994; Kraml et al. 2016). ³He/⁴He are reported as Rₑ values, which is ³He/⁴He of the sample normalized to the air ³He/⁴He of 1.4 × 10⁻⁸ (Rₑ). At the northern end of Lake Tanganyika at Pemba and Cape Banza, fluids contain predominantly magmatic-origin CO₂ (60–90%) and CH₄ with heavier hydrocarbons (Botz and Stoffers 1993; Tiercelin et al. 1993). Lake Tanganyika is an area identified for oil exploration with viable trapping structures in the rift basin (Roberts et al. 2016). The southern regions of the Tanganyika rift are more distal to current volcanism and may not be experiencing the same thermal perturbation as the northern section. While these observations do not discount the area as a potential helium-rich province, there is an increased CO₂ risk as the Kivu Volcanic Complex is approached and CH₄ dilution risk within the basin depocentres. Regions where the balance of these additional gas sources provide the mechanism to create or maintain a free-gas phase into which the helium partitions may provide prospectivity, but with lower helium concentrations than a primary ⁴He and N₂ gas system.

The North Tanzanian Divergence Zone (NTDZ) provides significant interest for investigating helium occurrence. This, one of the youngest sections of the EARS, is an example of extension within the Tanzanian Craton coupled with significant Quaternary volcanic activity in the form of the Crater Highlands Volcanic Complex (Fig. 2). Numerous geothermal springs are reported in both the Eyasi-Wembere Rift and the Manyara Rift (separated by the Mbulu Plateau), which are variably CO₂ rich and helium poor or nitrogen rich and helium rich. The sediment cover is not well
developed but is reported to be up to 2 km thick (James 1967a, b; Macheyeck et al. 2008).

The Rukwa Basin is of similar interest; it has recent rifting and is proximal to the Rungwe Volcanic Complex from which there are reports of nitrogen- and helium-rich gas seeps (James 1967a, b). The area is also now known from interpretation of seismic data to have a substantial thickness of sedimentary rocks (up to 7 km) within which seal and candidate reservoir horizons are known to occur (Mulaya et al. 2022).

**Helium in the NTDZ and Rukwa**

Noble gas abundance and isotopes within natural gases can be used to determine the contribution from mantle and crustal sources (e.g. 3He/4He), as well as the degree of interaction with the groundwater system (e.g. 20Ne and 36Ar) (e.g. Ballentine et al. 1991; Ballentine and Burnard 2002). Four seeps from the NTDZ (Balangida, Gonga, Eyasi and Mponde) and three seeps from the Rukwa Basin (Idindiro, Rukwa and Ivuna) were sampled for noble gas composition and isotope analysis at the University of Oxford, UK (Fig. 3; Tables 6 and 7). The major gas composition of seeps in both regions was first recorded by James (1967a, b) who found that they were predominantly N2 and 4He rich. A mixed mantle–meteoric source for the origin of gases from the seeps was originally proposed. In addition, Barry et al. (2013) measured low 4He concentrations and high CO2 concentrations in sampled thermal springs from the Ngozi-Songwe hydrothermal system and the Rungwe Volcanic Province, leading to the observation of an increasing crustal signal distal to the active volcanoes in the region.

**Helium, neon and argon determinations**

In all samples 4He/20Ne values are significantly higher than that of air (0.032), and range between 330 and 8920. Air corrections to the 3He/4He ratio of samples, therefore, have no significant effect on the R4 value (ratio of isotopes relative to that in air, which is defined as R4 = 1). This indicates that there are negligible air contributions to 4He concentrations in samples and that 4He/3He ratios that deviate from purely crustal ratios (0.02R4) are due to a mantle contribution.

The He isotope value of samples varies considerably both within and between study areas. Samples from the NTDZ have consistent 3He/4He ratios that range from 0.039 to 0.053R4, whereas samples from Mbeya have a larger range of values (0.18–3.45R4) (Table 6). From the He isotope values alone it is evident that samples from the NTDZ show predominantly crustal R4 values (where 0.02R4 = crust: e.g. Ballentine and Burnard 2002), whereas the Rukwa Basin shows a transition towards a more magmatic signature (where 6.10R4 = subcontinental lithospheric mantle (SCLM): Gautheron and Moreira 2002; Day et al. 2015).

Concentrations of 3He in all gases are orders of magnitude greater than the air concentration of 5.4 ppm. Samples from the NTDZ are consistently high in 3He, with a narrow concentration range of 2.7 × 10−2–10.6 × 10−2 cm3 (STP) 3He cm−3. In contrast, the Rukwa Basin samples vary in 4He concentrations from 4.3 × 10−5 to 2.5 × 10−2 cm3 (STP) 4He cm−3. From the He isotope values alone it is evident that samples from the NTDZ show predominantly crustal R4 values (where 0.02R4 = crust: e.g. Ballentine and Burnard 2002), whereas samples from the NTDZ have a higher gas/water ratio than those in the NTDZ, and those closest to the Rungwe volcanic zone the highest gas/water ratio.

Argon isotopes provide similar information to the helium system since subsurface 40Ar* is derived from the radioactive decay of 40K, while 36Ar is dominantly from air dissolved in groundwater similar to 20Ne (Ballentine and Burnard 2002). It is therefore no surprise that 36Ar concentrations show a similar pattern to 20Ne, with only a small variance in concentrations between samples from the NTDZ and significantly lower concentrations in the Rukwa samples. The latter show a similar trend in gas/water ratios recorded in the argon as that seen in neon. The greatest risk to the discovery of a commercial helium reservoir remains whether the gas/water ratio is sufficiently high to exceed the groundwater saturation/bubble point at the depth of the structure being considered.

20Ne can be used as a proxy for the degree of groundwater contact of a gas phase (e.g. Ballentine et al. 1991; Ballentine and Sherwood Lollar 2002; Barry et al. 2016). Neon concentrations similarly show two distinct patterns. Within all the helium-rich NTDZ samples, neon shows a small range of concentrations from 1.1 × 10−2 to 1.88 × 10−3 cm3 (STP) 20Ne. In contrast, samples proximal to the Rungwe volcanic zone in the Rukwa Basin have low helium concentrations, and neon concentrations almost two orders of magnitude lower. Even the helium-rich Ivuna seep has a neon concentration intermediate between the concentrations seen in the NTDZ samples (Rukwa Basin samples range from 0.012 × 10−2 to 0.22 × 10−2 cm3 (STP) 20Ne). Gas seeps in the Rukwa Basin therefore have a higher gas/water ratio than those in the NTDZ, and those closest to the Rungwe volcanic zone the highest gas/water ratio.

Fig. 3. (a) and (b) The two regions of interest outlined in Figure 2 and the approximate location of the local major tectonic units. Volcanoes are shown by red triangles. Helium concentrations in seeps reported by James (1967a, b) (green circles) are shown as per cent of seep-gas composition. Helium concentrations and 3He/4He ratios (R4) from this work are also shown (yellow circles). Similar to Figure 1, samples with higher 3He/4He contain only trace levels of helium (~0.05%). Samples with 3He/4He < 1R4 can contain up to 10.5% helium. NMBR = Natron–Manyara–Balangida Rift.
samples have a larger range with values of 331–787, with the samples closest to the Rungwe Volcanic Complex showing a 4He/40Ar* closest to the air ratio. Qualitatively, these results are consistent with mantle-derived gases in the samples closest to the Rungwe Volcanic Complex, being mostly overprinted by an atmosphere isotopic signature. This is most likely to have been caused by even a small amount of contact between the volcanic gas phase and groundwater containing dissolved atmosphere-derived noble gases.

**Binary mixing between crustal and mantle gas end members**

It is observed from gas composition analyses that natural gases containing 4He concentrations, which exceed the economic threshold of 0.1%, also contain high concentrations of N2 relative to 4He (>10:1) (e.g. Danabalan 2017). In the Rukwa Basin study, radiogenic 3He/4He (3He/4He = 0.02–0.03) are observed as the sample localities become more distal from the volcanic system. Similar spatial patterns of 3He/4He (R3), N2 concentrations and CO2 concentrations as a function of distance from volcanic centres have been observed in other regions (e.g. Fig. 1), and indicate binary mixing between crustal and mantle end members, the latter having likely been derived from the SCLM (Weinlich et al. 1999; Barry et al. 2013; Karolyte et al. 2019).

It is assumed that 3He/4He (R3) values are primarily controlled by the extent of mixing between crust-derived (N2-rich) fluids and associated 3He. In this context the ratio of 4He/40Ar* is correlated with changes in 3He/4He. The observed trend is fully consistent with simple two-component mixing between a crustal radiogenic end member with a 4He/40Ar* between 9 and 16, and a mantle end member represented by 4He/40Ar* < 0.35 and 3He/4He = 6.1Ra (Fig. 4). While consistent with binary gas mixing between crust (here dominated by 4He and N2) and magmatic gases (dominated by CO2), we note that the crustal end member resolved in this study is not the ratio at which 4He and 40Ar* are produced by radioactivity in the crust (crustal 3He/40Ar* = 4.9: Ballentine and Burnard 2002). The data show an enrichment in radiogenic helium relative to argon in the gases associated with the crustal end member. Similar trends have also been observed in the Hugoton–Panhandle giant gas field (Ballentine and Sherwood Lollar 2002).

Table 6. Noble gas isotope ratios (±1σ errors are shown in brackets)

| Sample name | 3He/4He (R3) | 20Ne/22Ne | 21Ne/22Ne | 40Ar/36Ar | 38Ar/36Ar |
|-------------|--------------|-----------|-----------|-----------|-----------|
| Study area 1 (NDT) | | | | | |
| Balangida | 0.053 (0.001) | 9.74 (0.030) | 0.031 (0.0003) | 544 (1.2) | 0.186 (0.0005) |
| Balangida | 0.052 (0.001) | 9.73 (0.030) | 0.031 (0.0003) | 549 (1.0) | 0.186 (0.0005) |
| Gonga | 0.039 (0.001) | 9.71 (0.030) | 0.029 (0.0003) | 432 (1.1) | 0.183 (0.0006) |
| Eyasi | 0.046 (0.004) | 9.72 (0.030) | 0.029 (0.0003) | 440 (1.5) | 0.187 (0.0006) |
| Mponge | 0.040 (0.002) | 9.71 (0.030) | 0.030 (0.0004) | 410 (0.8) | 0.184 (0.0005) |
| Study area 2 (Mbeya) | | | | | |
| Idindiro | 0.69 (0.01) | 10.04 (0.033) | 0.030 (0.0003) | 303 (0.2) | 0.187 (0.0003) |
| Rukwa | 3.45 (0.005) | 10.04 (0.033) | 0.030 (0.0003) | 331 (0.9) | 0.182 (0.001) |
| Rukwa | 3.45 (0.005) | 10.04 (0.033) | 0.030 (0.0004) | 336 (0.6) | 0.184 (0.001) |
| Ivuna | 0.18 (0.01) | 9.68 (0.029) | 0.032 (0.0004) | 787 (0.8) | 0.185 (0.0003) |
| Air (Pepin and Porcelli 2002) | 1 | 9.80 (0.080) | 0.029 (0.0003) | 295.5 (0.5) | 0.188 (0.0004) |

Table 7. Noble gas and N2 concentrations (±1σ errors are shown in brackets)

| Sample name | 4He (×10−2 cm3 cm−3 STP) | 20Ne (×10−5 cm3 cm−3 STP) | 40Ar (×10−2 cm3 cm−3 STP) | N2 (cm3 cm−3 STP) |
|-------------|--------------------------|---------------------------|---------------------------|------------------|
| Study area 1 (NDTZ) | | | | |
| Balangida | 10.6 (0.42) | 1.19 (0.015) | 1.47 (0.042) | 0.90 |
| Balangida | 10.4 (0.42) | 1.17 (0.016) | 1.59 (0.021) | 0.90 |
| Gonga | 8.4 (0.35) | 1.88 (0.033) | 1.69 (0.043) | 0.95 |
| Eyasi | 4.3 (0.29) | 1.30 (0.016) | 1.21 (0.020) | 0.95 |
| Mponge | 2.7 (0.11) | 1.10 (0.019) | 1.11 (0.019) | |
| Study area 2 (Mbeya) | | | | |
| Idindiro | 0.04 (0.002) | 0.028 (0.0004) | 0.73 (0.017) | 0.96 |
| Rukwa | 0.0047 (0.0002) | 0.014 (0.0004) | 0.029 (0.0008) | 0.78 |
| Rukwa | 0.0043 (0.0002) | 0.012 (0.0003) | 0.025 (0.0006) | 0.78 |
| Ivuna | 2.5 (0.04) | 0.22 (0.0011) | 0.46 (0.002) | 0.96 |
| Air (Pepin and Porcelli 2002) | 0.000524 (0.000006) | 1.65 (0.0036) | 0.93 (0.001) | 0.96 |

The principles of helium exploration
Helium One completed two exploration wells at Rukwa in August of 2021 (Tai 1 and Tai 2). Both demonstrated the presence of thick well-developed seals and reservoir intervals in the Lake Beds with helium shows. The next phase of drilling will target the Red Sandstone reservoir interval (Helium One Global 2021).

Conclusions

We have detailed the principles of helium generation, primary release, secondary migration and considerations of the role of the groundwater system that allow for a 4He–N2 gas phase to develop, with or without volcanic CO2 or sedimentary CH4, and thus form economically viable helium exploration provinces. Geological regions that contain old (billions of years) basement rocks will have accumulated substantial 4He through the radiogenic decay of U + Th. Within regions of recent tectonism causing geothermal perturbation, stored helium will be released along with associated gases such as nitrogen. Consideration of the migration of either a buoyant 4He–N2 free-gas phase, or 4He-N2 dissolved in the groundwater system and its subsequent exsolution, have been detailed together with the need for the presence of suitable trapping structures. These components enable the formation of a ‘play fairway’ approach to developing an exploration strategy; regions that exhibit all of these characteristics have the prospect of delivering commercially viable helium resources.

We show that sections of the Tanzanian East African Rift contain most, if not all, of the prerequisites for an economic helium province. Noble gas data from gas seeps in the west and east branches of the Tanzanian section of the East African Rift System contain 4He concentrations of up to 10.5%. This, combined with the potential 4He generated by the Tanzanian Craton and surrounding mobile belts of c. 7.0 × 10**13** BCF (2 × 10**13** m**3** 4He (STP)), implies that even with inefficient release, migration and trapping, these regions could provide high-helium concentration reservoirs. In the region of the Rukwa Basin independent prospective resource estimates undertaken on behalf of the operator and their financial backers suggest that there is a P**10** (best estimate) of 138 BCF 4He (2.78 × 10**13** m**3** 4He (STP)) potentially trapped within existing trapping structures (Helium One Global 2020), which, if recoverable, would alone supply the current world consumption for 14 years.

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Data availability

All data are included in the published article.

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The principles of helium exploration

13

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