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Research Progress on Dynamic Transport and Accumulation Coupling Mechanism of Crust-sourced Helium Gas Across Rock Strata

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Abstract

As an important scarce strategic material, helium is an associated resource in natural gas, but its enrichment mechanism is not well studied. Based on the analysis of helium enrichment in major helium-rich gas reservoirs at home and abroad, aiming at the difficult problem of how to enrich helium in the helium-rich gas reservoirs that have attracted much attention, helium in natural gas is mainly caused by shell source radioactivity, the distribution of helium-rich gas is closely related to ancient granite or ancient basement, and the occurrence modes of helium are mainly water-soluble and free-form. A large amount of ancient formation water is one of the main controlling factors of helium enrichment in natural gas. The basement granite of the basin can be used as the effective helium source rock of the helium-rich natural gas reservoir, and the organic-rich shale can also be used as the helium source rock. The main reservoir-forming models of shell source helium are helium-magma-rich gas reservoirs. Helium-groundwater - helium-rich gas reservoir formation model; Helium gas accumulation model in geothermal field. The important mechanism of helium enrichment is that the ancient formation water dehelium enrichment, helium dissolved and preserved in water, accompanied by late tectonic movement, resulting in fracture, stratum rise, and stratum movement above water. Under the action of Henry's law, the dissolved helium in water decreases with temperature and pressure and is released into the gas reservoir, forming helium-rich natural gas reservoir. By studying Henry's law, the proportion of water soluble and free states in helium migration is analyzed. It is concluded that helium dissolved in the pore water of surrounding rock diffuses to the migration gas, and most of the helium and nitrogen in the pore water are decomposed into the migration gas, and even the gas dissolved in the shale pore water near the gas reservoir. This process concentrates the helium in the gas, and highly enriched helium is more likely to form in shallow layers; A cooler temperature gradient is more conducive to helium enrichment in the gas.

Keywords

Helium; Crust-sourced helium gas; Helium source rock; Helium-rich gas reservoirs; Water-soluble；Henry's law.

1. INTRODUCTION

Helium gas as an important rare strategic resource mainly exists in natural gas with trace components, because of its chemical inertia, low boiling point, low density and other special

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physical and chemical properties and has important industrial value, is widely used in national defense, aerospace industry, nuclear industry, deep diving (strategic nuclear submarine cruise), superconducting technology, clinical medicine, laser and industrial analysis and other high-tech fields. And with the development of science and technology, the application of helium is more and more widely. In the whole universe, helium accounts for 23% by mass, second only to hydrogen. On Earth, however, helium is so rare that the main source is not air, but natural gas. Natural gas can contain up to 7.5% helium, which is 15,000 times more than air. However, there are not many deposits of such high helium natural gas, because helium in natural gas is the product of the decay of radioactive elements such as uranium, and it is generally believed that the content of helium in natural gas can be used for industrial use when it reaches 0.1%.

According to the United States Geological Survey (USGS), the current estimated global helium resources amount to $519\times10^8m^3$. The United States is the world's richest helium resource, and although it has been exploited on a large scale for more than 60 years, the helium resource still accounts for more than 40% of the world's total. According to a 2016 survey by the U.S. Geological Survey, the United States, Qatar, Algeria and Russia together hold 88 percent of the world's helium resources. The United States $206\times10^8m^3$, Qatar $101\times10^8m^3$, Algeria $82\times10^8m^3$, Russia $68\times10^8m^3$, Canada $20\times10^8m^3$, China $11\times10^8m^3$, and other countries $31\times10^8m^3$. The main helium producing countries in the world are the United States, Qatar, Algeria, Russia and Poland. The annual production of helium in the world from 2001 to 2016 was $(1.44\sim1.75)\times10^8m^3$, with little change in general. In 2009, the helium production reached the highest value; In recent years, helium production has fallen slightly, with a significant decline in 2009. Before 2012, the United States dominated the helium industry, accounting for at least 76 percent of production. Since then, Qatar has dramatically increased its helium production from liquefied natural gas in the North Field, producing 32 percent of the world's helium in 2016. Meanwhile, the United States, which produced 55 percent of the world's helium in 2016, has been producing less and less each year.

Although China also has a certain amount of helium-rich natural gas resources, at present, only Sichuan has obtained helium utilization from the gas reserves in Gongweiyuan, which is far from meeting the domestic demand, and the resources are close to exhaustion, and it is heavily dependent on imports. The United States, whose helium production accounts for more than 80% of the world, limits the production of helium as a strategic reserve resource, and threatens the export of rare earth resources in China, and the price of liquid helium has soared all the way, seriously affecting China's normal demand for helium. Therefore, the search for helium resources is one of the most urgent tasks in the field of resource investigation in our country. Although China's helium resources show the characteristics of many points and wide areas, the overall proved degree is relatively low. Helium resources are relatively poor, the exploration progress is slow, and the exploration degree is also relatively low. Because China has never carried out a national helium resource evaluation work, the understanding of the specificity of helium reservoir formation is insufficient, the existing work lacks objectives, the prospective areas are unclear on the plane, and the current vertical production layer may not contain the best helium, so very few helium rich gas reservoirs have been discovered so far, and most of them are helium poor. There are two main types of helium sources: mantle source and shell source, and shell source helium related to reservoir formation is the most common in nature. So far, no single pure helium reservoir has been found in the world. Helium is mostly associated with natural gas (including hydrocarbon gas, carbon dioxide gas, etc.) in the same gas reservoir to form associated gas or dissolved in water to form water-soluble gas. Among them, in natural gas reservoirs, as long as the amount of helium accounts for more than $0.05\%\sim1\%$, it can achieve industrial exploitation value. After the helium is released from the mineral, it will enter the adjacent fluid system. The amount of helium released is too small to allow it to move as a single helium gas, but only with other fluids. These fluids can be oil, gas, water, or magma. At present, the helium used in the global industry is the associated helium in natural gas (mainly methane), and methane plays an important role in the enrichment of

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uranium (strong source), and the desolubilization and preservation of He. Therefore, it is of great significance to establish mantle-source and crust-source helium migration dynamics models by studying the physical and chemical properties of helium and carrier fluids.

2. STUDY ON CRUST-SOURCE HELIUM SOURCE ROCKS

At present, a large number of studies have shown that basement granite in the basin can be used as an effective helium source rock for helium-rich natural gas reservoirs, and organic-rich shale can also be used as a helium source rock (Table 1, Table 2).

Table 1. Evaluation parameters of source rock and helium source rock

Source rock evaluation parameters		Helium source rock evaluation parameters	
Types of organic matter	Maceral composition, hydrogen index, H/C-O/C, etc	**Helium source rock type**	Granite, metamorphic rock, sedimentary rock, etc
Abundance of organic matter	Total organic carbon content (TOC)	**Uranium and thorium abundances in helium source rock**	U, Th content
Organic matter maturity	Vitrinite reflectance (Ro) or source rock pyrolysis peak temperature (Tmax)	**Age of helium source rock formation**	Radioactive reaction time
Source rock volume size	Distribution characteristics of source rock	**Helium source rock volume scale**	Distribution characteristics of helium source rock

Table 2. Types and characteristics of shell source helium Potential helium source rocks

Potential helium source rock types	Times	Volume	U/ppm	Th/ppm	Raw helium intensity	Hydrocarbongeneration intensity	Porosity
Basement granite	Old	Big	7.78	32.96	Stronger	\	Low
Organic-rich shales	New	Small	16.23	11.66	Strong	Extremely strong	High

Note: The average content of uranium thorium takes Weiyuan gas field as an example

2.1. Basement granite of basin

J.f. Chen, etal. (2021) calculated the distribution of global helium resources and found that most of the helium-rich natural gas fields in the world are mainly distributed in or around the craton, and the older the Craton age, the higher the helium abundance in the helium field. The main reasons are as follows: the rocks of the Proterozoic to Archaean crust/craton are mainly composed of granites or metamorphic rocks. On the one hand, these rocks are rich in uranium and thorium elements and have strong ^{4}He generation potential; On the other hand, the basement formed early, and the ^{4}He production gradually accumulated over time, which either remained in the interior of minerals, or diffused or released into the pore fluid of rocks. Therefore, a large amount of helium may have accumulated in the ancient cratons, providing helium sources for the rich accumulation of helium in the later sedimentary basins. For example, in the Archean craton below Yellowstone National Park in the United States, helium accumulated for hundreds of millions of years and was rapidly released within 2 million years under the influence of tectonic and

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magmatic activities. In addition, basement rocks are usually large in size and can accumulate to produce large helium resources. There are huge basement granite rock under the helium-rich natural gas fields in the Panhandle-Hugoton area, Cliffside area, Riley Ridge area of America, and the Weiyuan gas fields in Sichuan and Weihe basin in northern Qinling orogenic belt of our country, which may be the main source rock for helium.

Based on the principle of mass balance, the contribution of basement granites to helium in natural gas reservoirs in sedimentary basins can also be calculated. The detailed calculation method is described in the following article. Ballentine and Lollar (2002) and Danabalan (2017) calculated the amount of helium produced in situ by reservoir rocks in the Hugoton-Panhandle basin and the contribution of helium from external sources (underlying sediments and basement granite). The results show that even if all the helium produced by the reservoir rocks is released, migrated, accumulated and stored in the gas reservoir, its helium content is still lower than the detected radioactive helium content in the gas reservoir, and if the helium produced by the underlying sediments is taken into account, it is still insufficient to form the discovered helium resource in the Hugoton-Panhandle gas reservoir. Therefore, it is inferred that a large amount of helium from the basement granites of the basin must accumulate in the gas reservoir. This indicates that the basement granites in the Hugoton-Panhandle area have an important contribution to the helium in the gas reservoir. It should be noted that the exact relative source of helium in gas reservoirs is still a difficult problem due to the high uncertainty of helium release efficiency in helium source rocks, helium loss caused by uplift and denudation, and helium recharge and escape in gas reservoirs in geological history. For the Hugoton-Panhandle area, Brown (2019) established a model for the generation and migration of helium from sediments and basement in the atmospheric field, and identified the source and migration characteristics of helium in different locations. However, due to the lack of laboratory calibration of geological parameters in the model (such as diffusion coefficient, Henry coefficient, etc.), and insufficient understanding of the thickness of the basement helium source rocks and the mechanism of helium release and migration, the model is poorly constrained by actual geological parameters. Therefore, the relative contribution of the two types of helium source rocks can only be determined qualitatively, and the contribution of different types of helium source rocks cannot be evaluated quantitatively. Cheng et al. (2021) identified the important contribution of basement rocks in the Willliston Basin of North America to helium and neon in the sedimentary basin by calculating the mass balance of helium and the characteristics of neon isotope composition, and proved the control effect of diffusion and helium flux in basement rocks on the distribution of helium in the sedimentary basin by establishing a helium transport model. Halford et al. (2022) also applied the mass balance principle to confirm the helium sources of four helium-rich natural gas reservoirs in the Colorado Plateau of the United States, and believed that the Precambrian granite basement was the main helium source, and the deep rift provided an effective channel for helium migration. In addition, helium was extracted from the basement granites of Guanzhong Basin in China by vacuum crushing, heating melting and stage heating, and it was verified that granite can be used as an effective helium source rock for the helium-rich gas reservoir in Weihe Basin. The experimental results show that the helium in the granite mainly exists in the mineral lattice, of which the helium release rate is more than 80%, and temperature is the primary factor controlling the helium release in the granite.

The above research shows that the basement granite of the basin can be used as an effective helium source rock for the helium-rich natural gas reservoir.

2.2. Organic-rich shale

According to the uranium and thorium content of various rocks, the helium-generating capacity of granite is stronger than that of sandstone and carbonate rock, but weaker than that of shale, especially organic-rich mud shale or source rock. In the Weiyuan gas field, the uranium content of the organic rich shale of Qiongzhusi Formation as a source rock ranges from

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13.7 to 47.7ppm, while that of the granite rock ranges from 2.55 to 16.94ppm, which is significantly lower than that of the shale. The relative contribution of the two to the helium in the Weiyuan gas field is still unclear. Although uranium content in the shale of Weiyuan gas field is higher, the helium generating capacity is stronger, and the hydrocarbon gas can be used as a carrier to carry helium together to migrate into the reservoir. However, along with the process of hydrocarbon generation by the thermal evolution of organic matter, shale can produce both hydrocarbons and helium, and the intensity of hydrocarbon generation is much higher than that of helium generation, which will inevitably lead to the dilution of helium concentration by the generated methane and other hydrocarbon gases, making helium concentration lower than the abundance of economic utilization. Brown (2010) calculated that the shale with the lowest hydrocarbon generating potential (S_2=2 mg HC/g rock) produced 3000 times more hydrocarbon gas than helium in 1 billion years. It can be seen that the helium production was very low compared with that of natural gas. In addition, in sediments, uranium and thorium elements are mainly enriched in clay minerals or particle envelope. Meanwhile, the diameter of mineral particles in shale is much smaller than that in granite. According to the decay reaction release mechanism of 4He, 4He produced in shale is more likely to be lost and not easily preserved, that is to say, pore fluids of rocks are more enriched in helium than solid rocks and minerals. However, the porosity of rocks also has an important influence on the enrichment rate of helium in the pore fluid. Helium is enriched slowly in the pore fluid of the high-porosity helium source rock, while it is easy to enrich in the pore fluid of the low-porosity helium source rock (Figure 1). Sedimentary rocks tend to have higher porosity than igneous and metamorphic rocks, which may be the reason why they are less likely to form high concentrations of helium. In general, whether organic-rich shales can form natural gas reservoirs under the influence of hydrocarbon capacity and high porosity remains to be further discussed.

The current exploration practice shows that most of the helium reservoirs that reach the level of industrial utilization are related to granite and ancient strata, and the gas field with organic-rich shale as the helium source rock is not easy to form a helium rich gas reservoir. For example, the organic-rich shale of the Lower Silurian Longmaxi Formation in Sichuan Basin has a high uranium content (5.99~13ppm) and is a high-quality source rock for the shale gas of the Longmaxi Formation. However, due to its strong hydrocarbon generation, the helium concentration in the gas reservoir is severely diluted, and the helium content of most gas reservoirs is lower than 0.1%. For another example, Chang 7 oil shale of Triassic Yanchang Formation in Ordos Basin, as the main source rock of Mesozoic oil and gas reservoirs, has an average content of 35.46 ppm and 13.56 ppm of uranium and thorium, and a helium-producing intensity of 9.8 L/km^3 ·a, but no helium or helium content below 0.1% has been detected in natural gas reservoirs. Therefore, in the absence of external helium sources or special enrichment mechanisms, it is difficult to form a helium rich or high helium gas reservoir when organic-rich shale is used as a helium source rock alone, which can be said to be a non-major helium source rock at this time. However, this does not mean that the contribution of helium produced from organic-rich shales to the whole gas reservoir is zero. Because the current quantitative evaluation methods of helium sources are not perfect, we can only assume that in sedimentary basins with granite basement, shell source helium in helium-rich gas reservoirs is mostly mixed and mainly derived from basement granite. As for whether organic-rich shale can be used as an effective helium source rock and the evaluation criteria of effective helium source rock, further studies are needed.

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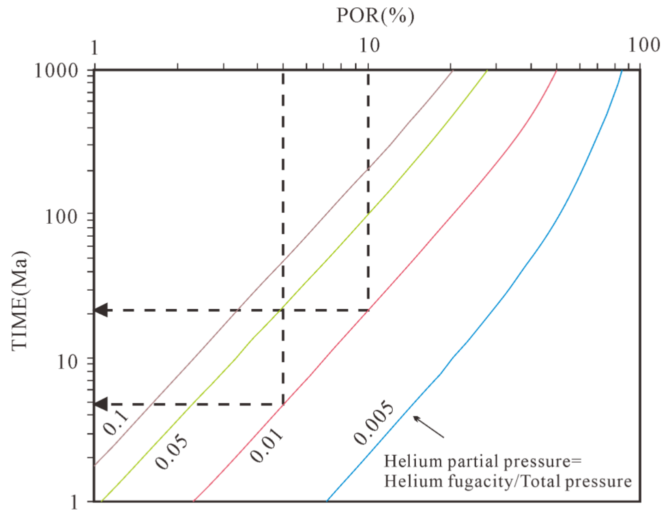

Figure 1. Effect of porosity on the partial pressure of helium in pore fluids
Note: The ordinate time means the time required to enrich the helium of the corresponding helium partial pressure in the pore fluid

3. STUDY ON THE ACCUMULATION MECHANISM OF HELIUM GAS

There are two stable isotopes of helium, ^3He and ^4He. The $^3He/^4He$ of mantle source, atmospheric source and shell source are 1.1×10^{-5}, 1.4×10^{-6}, 2×10^{-8}, respectively. It can be seen that ^4He is the main helium in nature, especially shell source helium related to helium reservoir.

The gas intensity of shell source helium is very low, and it is a typical weak source gas compared with conventional oil and gas. Conventional oil and gas geological theory believes that the most important and efficient secondary migration mode of natural gas is to enter the trap driven by buoyancy in the form of free state after discharging from the source rock. Compared with the huge groundwater circulation system, the weakly sourced helium produced by slow radioactive decay in the crust is difficult to form a free gas column like conventional natural gas and enter the trap to form a helium reservoir driven by buoyancy, so it can only use groundwater as a carrier (water-soluble phase) for secondary migration and accumulation to form a reservoir. The migration and accumulation coefficient of oil and gas with concentrated mass generation and episodic migration is generally only 1%-10% (natural gas is lower). Faced with a huge geological body and groundwater circulation system, how can the slowly generated weak source helium overcome dissolution and consumption, improve the migration and accumulation coefficient to form a helium reservoir?

Li Yuhong et al. put forward two points worth considering in the process of migration and accumulation: (1) All the helium used in the global industry is associated helium in natural gas (mainly methane), natural gas is organic gas, and helium is inorganic gas. The two sources are different, but they exist in the same reservoir space, and the formation mechanism and interaction relationship are unknown. ② It is now believed that helium has two types: free state and water soluble state, and the influence of its occurrence state and its spatial relationship on migration and accumulation needs further study.

Helium resources usually exist in two phases: water-soluble phase and free phase. However, in the current actual exploration process, helium-rich natural gas associated with methane gas is the focus of the current exploration. Therefore, the study on the phase change of helium gas is conducive to the search for helium resources in free phase.

At present, the helium used in the global industry is associated helium in natural gas (mainly methane), and methane plays an important role in the enrichment of uranium (strong source) and the dissolution and preservation of He. According to the preliminary summary of the helium enrichment model diagram (Figure 2), a small amount of helium can be precipitated from the

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surface granite mass, dissolved into the surface water, and then participated in the water circulation to the underground along with the surface water. At the same time, the underground underwater cycle can also carry U^{6+} formed by oxidation of the surface granite to the underground. When U^{6+} meets the reduction environment of organic matter enrichment such as CH_4 in the underground, it is reduced to U^{4+}, which is enriched in the formation of uranium enrichment layer (mineralization), and becomes an important source rock of helium.

Shell source helium and a small amount of mantle source helium formed by radioactive decay of uranium and thorium elements in uranium enriched layer, buried granite and sedimentary strata dissolve into helium containing groundwater, further increase the concentration of helium in groundwater, form helium rich groundwater, enter the groundwater circulation system, and participate in groundwater circulation in a water-soluble state. The groundwater circulation channel can be a fracture or a dredge-out layer (pore). No matter the groundwater goes down or up, the fracture is the most efficient channel, so it is also easy to form a uranium-rich formation near the fracture. The helium-rich fluid enters the trap through the fracture and has a high efficiency of enrichment, which can effectively reduce the consumption in diffusion and improve the transport and accumulation coefficient of the limited quantity of helium. During the upward migration of helium-rich groundwater, the solubility of helium gas decreases with the decrease of pressure. At the same time, when the helium-rich groundwater encounters the CH_4, CO_2 and other gas-bearing layers, due to the competitive dissolution of multi-component gases, helium gas is replaced from the water by easily soluble carrier gas, and a large number of de-dissolved into the carrier gas reservoir to form a helium-rich gas reservoir.

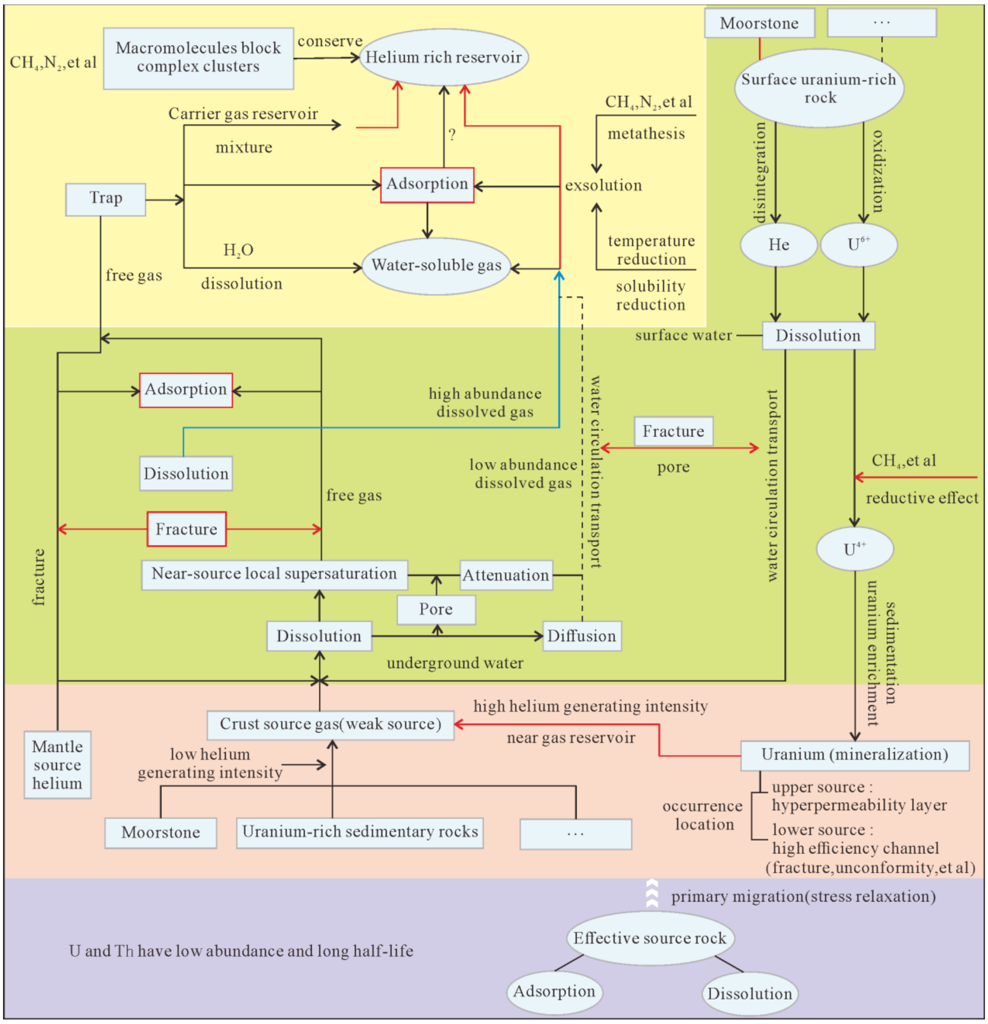

Figure 2. Conceptual model of weakly source helium rich reservoir

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3.1. Release of helium from minerals

^{4}He is produced by the radioactive decay of minerals, rocks containing uranium and thorium, so helium must first be released from the earth's crust minerals. Helium is released from Earth's crust minerals in four main ways: decay recoil release, diffusion release, rupture release and mineral transformation release.

(1) Decay recoil release. The high-energy alpha particles produced by the decay of the uranium element in the mineral will be ejected back a certain distance, called the "stopping distance", which is mainly determined by the density of the mineral. When the grain size of a mineral containing uranium is close to the "stopping distance", a considerable amount of helium is released from the mineral's grain.

(2) Diffusion release. Diffusion is the most direct way in which helium is released from minerals in the Earth's crust, often accompanied by other release mechanisms. The diffusion of helium in minerals is controlled by the size and time of mineral particles, the smaller the mineral particles, the greater the diffusion coefficient, and the shorter the time it takes for helium to be released from minerals.

(3) Rupture release. Brittle rock cracks under the pressure of the overlying load to form cracks. After mineral cracks appear, the helium gathered in the grain will be released quickly along the cracks, and the amount of helium released is proportional to the volume of the cracks, and it will be released along the cracks in the way of diffusion.

(4) Mineral transformation release. The changes of minerals in diagenesis and metamorphism will lead to the release of helium in their lattice, the most typical is the recrystallization of minerals. Although this mechanism is microscopic, it can lead to large-scale releases of helium from the Earth's crust.

3.2. Helium reservoir formation mechanism

So far, no single pure helium reservoir has been found in the world. Helium is mostly associated with natural gas (including hydrocarbon gas, carbon dioxide gas, etc.) in the same gas reservoir to form associated gas or dissolved in water to form water-soluble gas. In natural gas reservoirs, as long as the amount of helium accounts for more than 0.05 ~ 1%, it can achieve industrial exploitation value. After the helium is released from the mineral, it will enter the adjacent fluid system. The amount of helium released is too small to allow it to move as a single helium gas, but only with other fluids. These fluids can be oil, gas, water, or magma. Fractures and fissures are efficient channels for helium migration. Fractures can trap helium in the surrounding rock at any depth and can be used as a channel for helium to flow to the surface. At deep depths, faults can be inserted into magma puncturing structures and transporting helium and other escaping gases to reservoirs where they can accumulate. Mantle source or deep crust helium migrates vertically through deep large faults and fissures, and then migrates and accumulates in oil and gas traps through faults or fissures.

(1) The accumulation pattern of helium-magma-helium-rich gas reservoirs: After He, CO_2, N_2, CH_4 and other natural gas in the mantle are mixed into the magma, as the magma migrates to the shallow part of the crust, with the decrease of temperature and pressure, the magma degasses the natural gas such as He and other natural gas such as He, and the released natural gas such as He can be charged into the adjacent reservoir to directly form the gas reservoir, or it can continue to be transported up along other faults and then accumulated into the reservoir. The existence of magmatic activity is the primary condition for the formation of such accumulation.

(2) Helium-groundwater - helium-rich gas reservoir accumulation model: After helium is released from minerals, it first dissolves into the pore water and migrates with the pore water. During this period, if another gas (hydrocarbon gas, carbon dioxide gas, etc.) enters the water, helium will be pumped into the gas and will migrate with the gas. If the gas migrates into the trap,

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it will accumulate to form the gas reservoir. There are three main accumulation modes: ① Hydrocarbon gas migrates in the helium-enriched pore water, and the gas will pump the helium dissolved in the pore water. Helium migrates into the trap with the gas and accumulates into the gas reservoir; ② When the water flows through the bottom or transition zone of the gas reservoir, the helium in the pore water will be sucked into the gas reservoir and directly form a reservoir; ③ No matter the organic mantle-derived CO_2 or the CO formed by organic decarbonization, CO_2 migrates in the pore water enriched with helium, nitrogen and methane, CO_2 will suck the helium dissolved in the pore water, and CO_2 will dissolve in the water to enrich the helium in the remaining gas.

(3) Helium accumulation patterns in geothermal fields: In the study of geothermal fields at home and abroad, it is found that there are helium anomalies in soil gas and geothermal water in geothermal fields, such as the Roosevelt hot spring area in the United States and the Weihe Basin in China. There is a corresponding relationship between surface helium distribution and geothermal.

According to the study of surface soil helium in the geothermal field of Roosevelt Hot Springs, Utah, USA, the concentration of helium in soil gas is higher in the geothermal field producing hot water. The high surface helium anomaly is not necessarily related to the depth of geothermal Wells.

There is a unique consistency between heat and helium in geothermal systems, as 75% of the Earth's geothermal energy and almost all ^4He is produced by the emission of uranium, thorium, etc., and the migration of heat and helium in groundwater is also coupled. Therefore, helium can be used as a guide for geothermal exploration.

Helium can be released from cooling magma beneatha geothermal field and diffused directly through the overlying strata, or mixed with hot water, water vapor, and steam released from the cooling magma, and then moved out through permeable rocks or along fracture zones. This gas slowly seeps into ground water at the surface and enriches the soil gas with helium. This rising gas and hot water system can also transport helium originally stored in the surrounding rock to the surface, which can form helium-containing hot springs, steam vents or geysers.

3.3. Phase changes of the water-soluble and free states of helium

In some cases, the water-soluble phase migration helium will change into the free state. Some scholars have found that when the ground water containing helium is in contact with other free gas reservoirs, the helium in the water soluble phase will be "sucked" out of the aqueous solution by other free phase gases, and then migrate together to form a reservoir. This "suction" effect occurs because the partial pressure equilibrium of helium dissolution is broken. The dissolved partial pressure balance is closely related to the mole fraction of solute inside and outside the solution, which is a manifestation of the phase equilibrium between the aqueous and free phase solute molecules inside and outside the solution. In phase equilibrium, there is a corresponding phase equilibrium constant -- the ratio of the mole fraction of a component in the gas phase to the mole fraction of the component in the liquid phase when the gas-liquid phase reaches the equilibrium state under a certain temperature and pressure. The following relationship exists when the phase is in equilibrium:

$$y^*=mx$$

Where: x -- molar fraction in the liquid phase of the solute gas;

m -- phase equilibrium constant;

y* - molar fraction of the solute in the gas phase in equilibrium with the liquid composition.

When the solute gas reaches the phase equilibrium state, the solution can exist stably. When the molar fraction of the solute in the gas phase is small, the process of solute desorption will occur, and the water-soluble phase gas becomes the free phase. On the contrary, when the molar fraction of

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the liquid solute is small, the dissolution process will occur (Figure 3). Therefore, according to the principle of phase equilibrium, when other gases are in contact with helium-containing water, the mole fraction of free phase helium becomes smaller, the phase equilibrium of helium-containing aqueous solution is broken, and the dissolved helium becomes a free phase and mixed with other gases. Therefore, the phase state of helium will change during migration, and the migration characteristics of water-soluble helium also play an important role in searching for free helium.

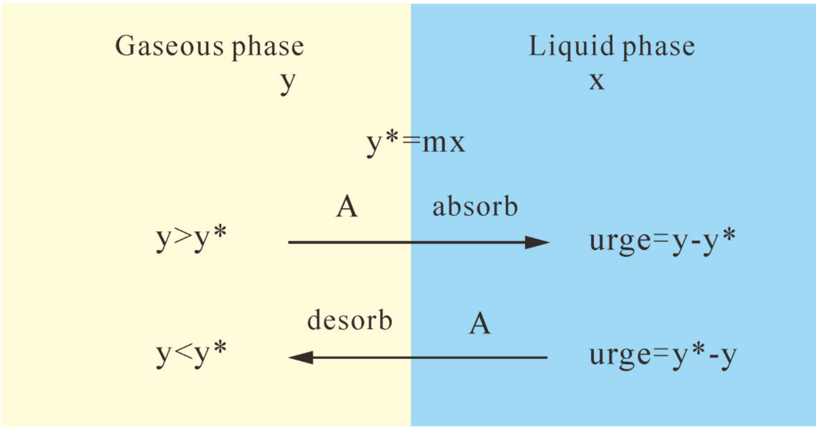

Figure 3. Schematic diagram of phase equilibrium and gas dissolution and decolution

The shell source helium generated by radioactive decay not only has a very slow generation rate, but also has a strong diffusion ability due to its small molecular mass and diameter. Therefore, helium is difficult to migrate in the crust in a single gas phase in a free state, mainly in a water-soluble state or carried by other gases. Brown studied the formation process of natural gas with high helium content in the United States and believed that a large amount of ancient formation water was one of the main controlling factors of helium enrichment in natural gas. Later, the above helium enrichment phenomenon was characterized semi-quantitatively by using the original gas-water ratio (V_g/V_w) calculated from the rare gas isotopes (^{36}Ar and ^{20}Ne) derived from air. In other words, the larger the volume of ancient formation water where natural gas meets, the higher the helium content in natural gas will be with the continuous occurrence of gas-water interaction, indicating that ancient formation water plays an important role in the process of helium enrichment and accumulation.

The helium dissolved in the pore water keeps dispersive distribution in the water; It cannot gather helium into the trap. Helium becomes concentrated by interacting with the gas phase. The amount of helium in a gas that coexists with helium dissolved in water is described by Henry's law:

$$X_{He} = \lim_{f_{He}\to 0} \frac{f_{He}}{H_{He}}$$

Where: F_{He} is the fugacity of helium; Fugacity is approximately equal to the partial pressure, which is the total pressure multiplied by the mole fraction of helium in the gas;

X_{He} is the mole fraction of He in water;

H_{He} is the Henry constant of helium in water, which is a function of temperature and water salinity.

The Henry constant is always much greater than 1 (Figure 4), so helium naturally condenses into the gas phase. The greater the Henry constant, the greater the proportion of helium in the gas phase. At diagenetic temperatures, helium is more difficult to dissolve than N_2 and methane

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(Henry's constant is larger). At metamorphic temperatures, nitrogen is less soluble in water than helium.

The lower curve is the solubility of the gas in fresh water. The top (purple) curve is the helium Henry constant in high salinity water. Salinity reduces gas solubility (increases Henry's constant). Salinity reduces the solubility of ammonia and methane, just as it affects his solubility.

The Henrys constant, combined with changes in temperature and pressure with depth, can be used to predict the ratio of helium's mole fraction in gas to its mole fraction in water as a function of depth and thermal gradient (Figure 5): Pressure has an important effect on the distribution of helium. There is more fractionation of He in the shallow gas phase than in the deep gas phase. A lower fluid pressure gradient (underpressure) favors more helium in the gas. The thermal gradient has less effect on the distribution of helium, and more helium is in the gas phase with a lower geothermal gradient.

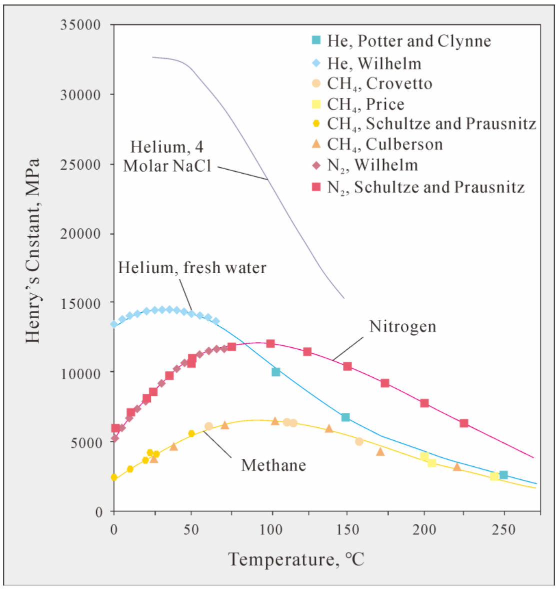

Figure 4. Henry's constant vs. temperature function

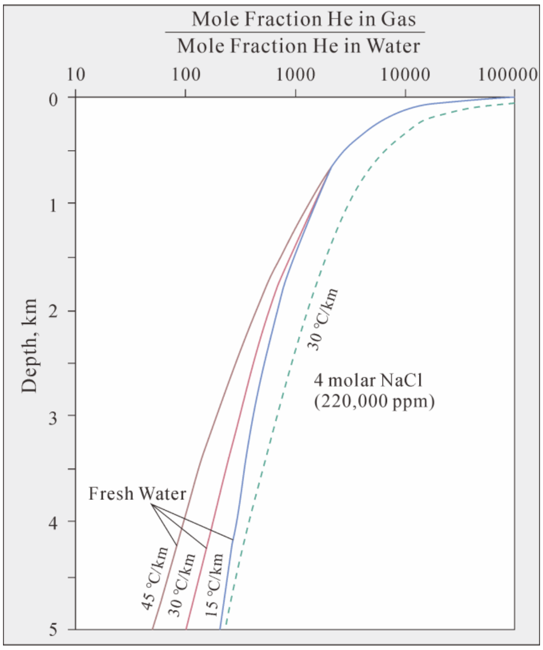

Figure 5. The ratio of the mole fraction of helium in gas to the molefraction in water as a function of depth and thermal gradient

Henry's law describes the molar concentration equilibrium between two static phases. The concentration is in equilibrium, so the actual mass of helium in each phase depends on the mass of each phase. Where the gas has less mass than water, the concentration of helium in the water gas will be higher.

Where the gas moves through water, the situation is a little more complicated. The first part of the gas that interacts with the water absorbs most of the helium. The concentration of ammonia in this gas will be the greatest because it balances out to the water with the greatest concentration of ammonia. The migrating gas removes helium from the system, so the gas that later migrates through the water-saturated reservoir will see the lower concentration of helium dissolved in the pore water. These gases will therefore pick up less gas and their concentration of helium will there by decrease. This process continues as described by the figure at right(Figure 6).

The gas extracts helium from pore water that is not in direct contact with the gas. When dissolved helium is extracted from the pore water, the helium dissolved in the pore water of the surrounding rock diffuses into the moving gas. Assuming a diffusion time scale of 1 million years, 90% of the dissolved helium in the thickness of the rock 200 to 500 meters above and below the migrating gas is extracted by the gas (Figure 7). Most of the helium and nitrogen in pore water are decomposed into migration gases, or even dissolved in the shale pore water near the gas reservoir. This process concentrates the helium in the gas and results in the helium/nitrogen correlation in most high helium gases. High helium is more likely to form in shallow layers than deeper into the Earth. A cooler temperature gradient favors the higher helium in the gas. The less gas there is relative to water, the greater the concentration of helium in the gas.

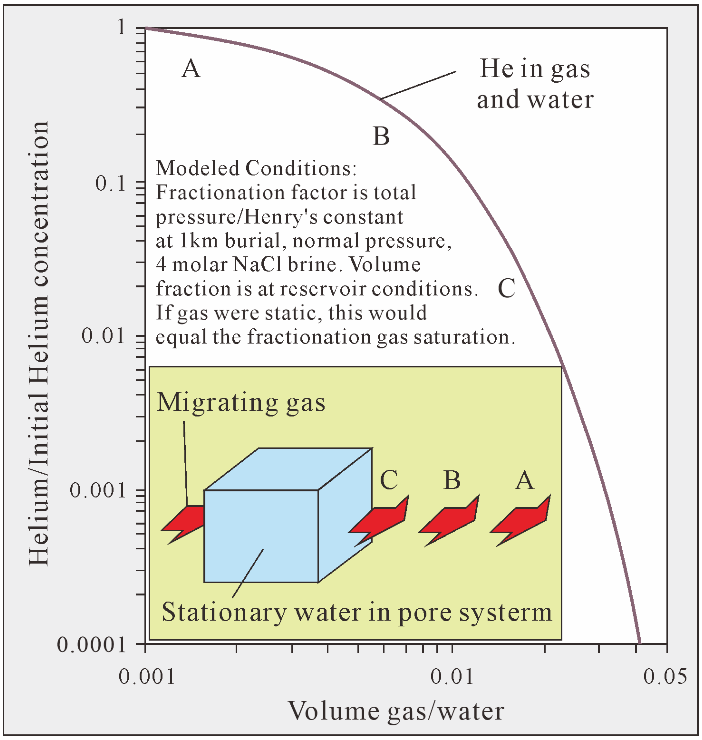

Figure 6. Helium fractionation simulation diagram

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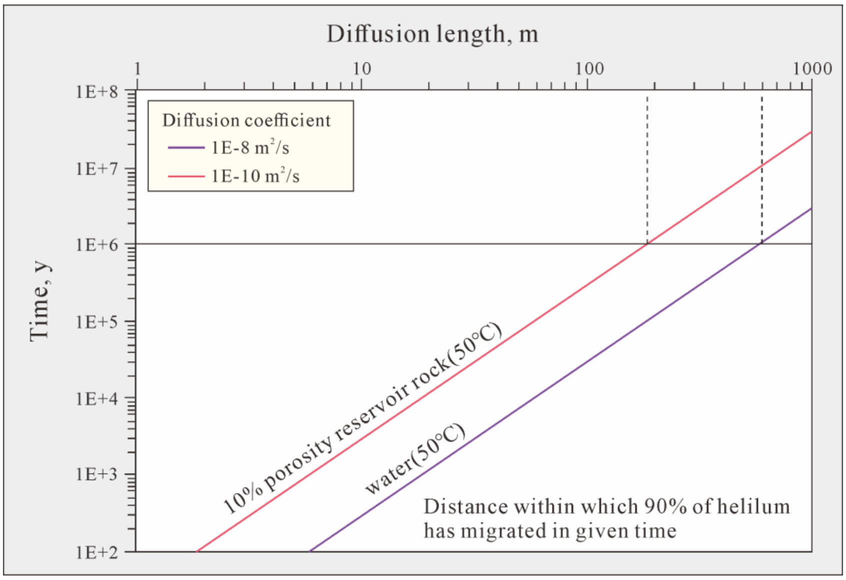

Figure 7. Gas Schematic diagram of extracting helium from pore water that is not in direct contact with the gas

4. CONCLUSION

(1)^{4}He is produced by radioactive decay of minerals, uranium and thorium contained in rocks. Helium is released from Earth's crust minerals in four main ways: decay recoil release, diffusion release, repture release and mineral transformation release. Helium is mostly associated with natural gas (including hydrocarbon gas, carbon dioxide gas, etc.) in the same gas reservoir to form associated gas or dissolved in water to form water-soluble gas. After helium is released from minerals, it will enter the adjacent fluid system. The released helium can only migrate together with other fluids. Fractures and fissures are the effective channels for helium migration. The reservoir forming modes of shell source helium mainly include helium-magma-helium-rich gas reservoir; Helium-groundwater - helium-rich gas reservoir accumulation model; Helium accumulation in geothermal field.

(2) Helium in natural gas is mainly of radioactive origin from shell sources, and the distribution of helium-rich natural gas is closely related to ancient granite or ancient basement. A small part of helium in the helium-rich reservoir comes from source rocks and reservoirs, and the occurrence modes of helium are mainly water-soluble and free-form, among which a large amount of ancient formation water is one of the main controlling factors for helium enrichment in natural gas. The important mechanism of helium enrichment is the dehelium-enrichment of old formation water. After the decay of U and Th in ancient rock strata over a long geological period, the helium produced is dissolved in water and preserved. With the later tectonic movement, faults occur, strata uplift occurs, and strata move over water. The dissolved helium in water decreases with temperature and pressure and is released into gas reservoirs under the action of Henry's Law, forming helium-rich gas reservoirs. If the natural gas migrates along the ancient reservoir, the helium content in the natural gas will gradually increase with the increase of migration path.

(3) Gas extracts helium from pore water that is not in direct contact with gas. When dissolved helium is extracted from the pore water, the helium dissolved in the pore water of the surrounding rock diffuses into the moving gas. Most of the helium and nitrogen in the pore water are decomposed into migration gas, and even dissolved in the shale pore water near the gas reservoir. This process concentrates the helium in the gas and results in the helium/nitrogen correlation in most high helium gases. High helium is more likely to form in shallow layers than

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