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Enrichment mechanism and resource potential of shale-type helium: A case study of Wufeng Formation-Longmaxi Formation in Sichuan Basin

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Abstract China's helium resource is highly dependent on overseas imports. Organic-rich and U/Th-rich shale reservoirs generally contain helium, and although the helium content is low, the total reserve is large. Therefore, the effective development and utilisation of shale-type helium resources is a realistic way to improve the security of helium resources in China. In this study, the generation mechanism, helium source and content, migration modes and pathways, controlling factors of enrichment, distribution pattern, and resource potential of the helium were analysed, using the Wufeng-Longmaxi shale in the Sichuan Basin and its periphery. Furthermore, countermeasures were proposed for shale-type helium exploration and development. The results show that the Wufeng-Longmaxi shale has a high content of U and Th and a good ability to generate helium. The helium is generated by a typical crustal source of helium and is characterised by self-generation, self-storage, and wide distribution. The helium resource potential is a product of its content and the resources of the associated natural gas. The continuous supply of helium and effective preservation are the main geological factors that control the enrichment of shale-type helium. The preliminary evaluation results show that the reserves of helium in proven shale gas reserves are 10.8×10^8 m^3 in the Sichuan Basin and its periphery, where the extra-large helium fields are likely to be discovered. Additionally, 0.0912×10^8 m^3 of helium was produced, along with the annual production of shale gas. To avoid the waste of helium and to improve the self-supply ability, it is suggested that research on the resource potential, enrichment mechanism, and distribution pattern of shale-type helium should be carried out as soon as possible, and helium extraction techniques for helium-bearing natural gas should be studied.

Keywords Shale, Helium content, Enrichment mechanism, Resource potential, Sichuan Basin

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1. Introduction

Helium, which has the lowest boiling point, is characterised by a low density, strong inertness, and excellent thermal conductivity, and plays an important role in the military, aviation, medical, semiconductor, nuclear industries, large scientific research, and other high-tech fields. Therefore, it is considered the key strategic material for future science and technology competitions (Dai et al., 2017; Huang et al., 2021). The production of helium is extremely low in China, and only 53×10^4 m^3 of helium was produced in 2020, making the market demand for helium highly dependent on imports. In 2020, the amount of imported helium was approximately 2076×10^4 m^3, and the external dependency on helium was as high as 97.5% (Zhang et al., 2022), seriously affecting national security. In the current severe international environment, the exploration of helium resources has become one of the most urgent tasks in mineral resource exploration in China and is of great practical significance.

There are three sources of helium in Earth's environment: the atmosphere, mantle, and crust (Xu et al., 1990; Mtili et al., 2021). Among these, crust-source helium is produced by the decay of radioactive elements of U and Th in crustal rocks (Tao et al., 2019; Liu et al., 2022; Peng et al., 2022). This type of helium occurs in three main forms. The first is the water-soluble gas in the geothermal field (Danabalan, 2017; Zhang et al., 2019). However, the amount of helium in the geothermal water and spring water is very low, even though the solubility of helium is relatively high. Therefore, the total reserve of water-soluble helium is small, which makes it difficult to form a large helium field (Tao et al., 2019). The second is non-hydrocarbon-associated gas in non-hydrocarbon gas reservoirs, such as nitrogen and carbon dioxide reservoirs (Xu et al., 1996). For example, in the petroliferous basins that are distributed along the Tan-Lu Fracture belt, previous researchers discovered multiple nitrogen gas and carbon dioxide reservoirs that contain helium. The helium content in these reservoirs is high and meets or exceeds the industrial utilisation standard (Xu et al., 1996). However, because the production of carbon dioxide is low and nitrogen gas has no exploitation value, the commercial development value of the associated helium is low. The third category is associated with gas in oil and gas reservoirs (Cao et al., 2018; Tao et al., 2019). This is the primary type of helium being developed and utilised worldwide. Although the associated helium reserves are relatively large, their commercial value varies significantly because of the different total natural gas volumes and helium content in different oil and gas reservoirs (Cao et al., 2018; Tao et al., 2019).

In general, it is difficult for the deep crust- and mantle-source helium to form an economical helium reservoir (Brown, 2010). Therefore, current research predominantly focuses on shallow crustal sources, such as granite, volcanic rock, and sandstone. Through a certain distance of migration, the helium produced by these ancient rocks can accumulate in the conventional reservoirs in the shallow part of the basin and eventually form a natural gas reservoir containing helium, such as the Oklahoma and Panhandle-Hugoton Gas Field in Oklahoma and Kansas, USA (Brown, 2019). In addition, shale rich in U and Th can also produce helium. For example, helium was discovered in the Antrim shale of the Michigan Basin, the New Albany shale of the Illinois Basin, hot shales of the Middle East and North Africa, and the Wufeng-Longmaxi shale of the Sichuan Basin (Brown, 2010, 2019; Schlegel et al., 2011; Wen et al., 2015; Wang et al., 2020; Chen et al., 2021; Chen et al., 2023). However, the helium content in these shales is very low; therefore, no attention has been paid to these helium resources in the past. With the worldwide exploration and development of shale gas in recent years, its associated helium has attracted the attention of scholars and industry (Chen et al., 2021). In this study, the helium that is produced by the decay of radioactive elements of U and Th in organic-rich shale, which exists in the free state in shale and its interlayers, and that occurs with shale oil and gas, is referred to as shale-type helium. The shale-type helium is the result of nearby accumulation following the generation of helium in the shale and is a typical in-situ accumulation. Similar to shale oil and gas, it exhibits the characteristics of self-generation, self-storage, and wide distribution. The enrichment degree of shale-type helium is largely controlled by the U and Th contents of the shale and the gas preservation conditions. The resource potential of shale-type helium is controlled by the content of helium and the reserves of its associated shale oil and gas.

Shale-type helium and its associated shale gas are self-generated and self-stored; therefore, shale gas dilution makes the abundance of helium very low. In such cases, shale-type helium generally cannot form into industrial accumulations. However, shale has a large distribution area, high volume, and high U and Th content. Therefore, the total amount of shale-type helium is very large, its resources are vast, and the exploration and development potential of shale-type helium has bright prospects. Currently, little attention has been paid to shale-type helium, and more research should be conducted. In this study, the enrichment characteristics of the shale-type helium of the Wufeng-Longmaxi shale in the Sichuan Basin were studied, considering the generation mechanism, source and content, migration methods and pathways, controlling factors, and distribution pattern of helium. The resource potential of shale-type helium and countermeasures for its exploration and development are then discussed to support the development and utilisation of shale-type helium.

2. Geological setting

The Sichuan Basin is a rhomboid basin in which both Palaeozoic marine facies and Meso-Cenozoic continental facies developed. It has experienced a complex history of tectonic movements, including the Caledonian (Late Sinian-Silurian), Hercynian (Devonian-Permian), Indosinian (Triassic), Yanshanian (Jurassic-Late Cretaceous), and Himalayan (Palaeocene-Quaternary) (Zhang et al., 2013). Presently, the boundaries of the Sichuan Basin include the Longmenshan thrust belt in the northwest, the Micangshan-Dabashan thrust belt in the northeast, the Qiyueshan-Daloushan fault belt in the southeast, and the thrust strike-slip belt in Southwest Sichuan (Figure 1). Owing to the different basements of the Sichuan Basin in different areas and the different effects of palaeo-structural uplift and depression on sediment deposition, different areas have different structural deformations and styles. According to the structural characteristics, the Sichuan Basin can be divided into four major regions: a gentle fold area in the central region, a high and steep fold area in the east, a nappe fold area in the west, and a low and flat fold area in the north. The Sichuan Basin and its periphery are mainly composed of the Archean, Palaeozoic, Mesozoic, and Cenozoic strata. Devonian sediments are missing, and most Carboniferous sediments were eroded owing to the Caledonian orogeny.

The thickness of the Palaeozoic to Cenozoic strata is approximately 6000–12000 m, and the thickness of the current residual Silurian strata is between 0 and 1200 m. Black shale is mainly formed in the Lower Cambrian, Upper Ordovician Lower Silurian, Lower Permian, Upper Permian, Lower Triassic, and Lower Jurassic strata. Among them, the Upper Ordovician Wufeng-Lower Silurian Longmaxi Formation is predominantly formed in the shallow shelf and deep water shelf that are anoxic, with a thickness of approximately 200–600 m (average thickness of 260 m), and the distribution area is approximately 13.7×10^4 km^2 (Dong et al., 2014). The high total organic carbon (TOC) content and radioactive elements such as U and Th lay a good foundation for the generation and storage of both shale gas and shale-type helium. In 2012, a high-yield gas production of 20.3×10^4 m^3 d^{-1} was obtained from the Jiaoye 1HF well, achieving

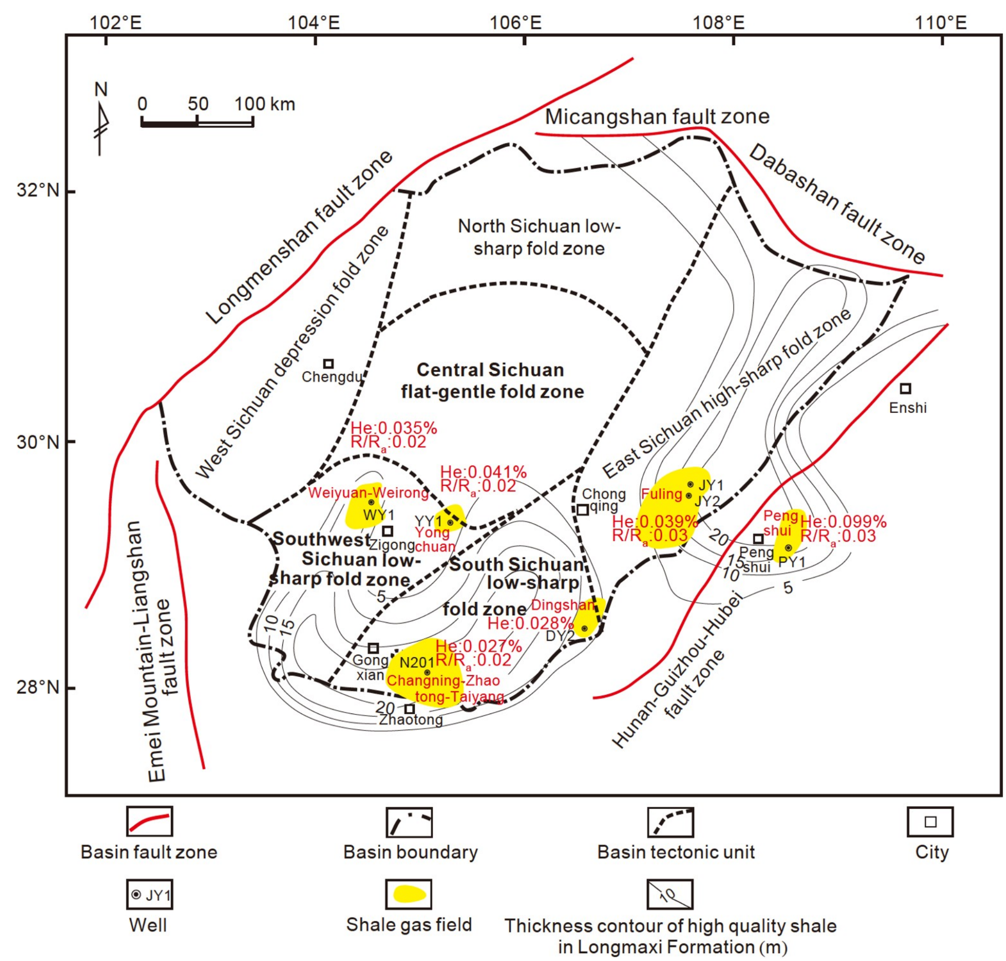

Figure 1 Structural division of Sichuan Basin and helium content of Wufeng Formation-Longmaxi Formation shale gas fields (Some data sources: Cao et al. (2018), Feng et al. (2020), Tenger et al. (2020), Liu et al. (2021), Li et al. (2021) and Qin et al. (2022)). R/R_a is the ratio of stable isotope $^3He/^4He$ in natural gas to $^3He/^4He$ in the atmosphere, and "He: 0.035%" is the helium content.

the commercial breakthrough of Wufeng-Longmaxi shale gas (Guo, 2014). In 2014, the Fuling shale gas field in the Sichuan Basin reported its proven reserves of shale gas of 1067.5×10^8 m^3 and became the first shale gas field outside North America (Jin et al., 2016). By the end of 2021, the proven reserves of shale gas of the Wufeng-Longmaxi Formation in the Sichuan Basin and its periphery exceeded 2.7×10^{12} m^3, and the annual production reached 228×10^8 m^3, making it a major shale gas-producing area in China (Sun et al., 2021; Nie et al., 2022).

3. Enrichment mechanism of shale-type helium

3.1 Generation mechanism of helium

In nature, helium exists in the mono-atomic state and has two types of stable isotopes: ^3He and ^4He. Among them, ^3He predominately originates from the deep mantle and its relative abundance is low (O'Nions and Oxburgh, 1988), while ^4He originates from the α-decay of ^{238}U, ^{235}U, and ^{232}Th in the crust rock (e.g., shale and granite). The relative abundance of ^{238}U and ^{235}U in naturally occurring U are 99.28% and 0.72%, respectively, while the relative abundance of ^{232}Th in naturally occurring Th is 99.995%. The decay equations for the three nuclides are as follows:

$$^{238}_{92}U\rightarrow^{206}_{82}Pb+8^4_2He+6^0_{-1}e,$$
$$T_{1/2}=44.68\times10^8\ yr.$$

$$^{235}_{92}U\rightarrow^{207}_{82}Pb+7^4_2He+4^0_{-1}e,$$
$$T_{1/2}=7.10\times10^8\ yr.$$

$$^{232}_{90}Th\rightarrow^{208}_{82}Pb+6^4_2He+4^0_{-1}e,$$
$$T_{1/2}=140.5\times10^8\ yr.$$

It can be seen from the above equations that the generation of crustal source ^4He is dependent on the U and Th content and their formation time. The longer the formation time, the greater the amount of helium produced. In crustal rocks, organic-rich shale contains the highest content of U and Th and produces the largest amount of helium (Meng et al., 2021). In the Ordovician Wufeng-Silurian Longmaxi shale of China and the hot shale of the Middle East and North Africa, the U and Th content is much larger. The U ranges from 15 to 50 ppm, and the Th ranges from 6 to 25 ppm. The U content in a unit mass of hot shale is much greater than that in granite, neutral rock, and other rock types, and the amount of helium produced by hot shale is also much greater than that produced by other rocks. Previous studies have found that the amount of helium produced by hot shale is approximately 9–10 times that produced by granite in 100 million years (Brown, 2010). Therefore, with regard to shale-type helium, the amount of helium is controlled by the U and Th content and is not influenced by temperature, pressure, or thermal maturity. Shale at any buried depth and maturity can produce helium, and the higher the content of U and Th in the shale, the greater the amount of helium produced.

To evaluate the amount of helium produced quantitatively, a helium volume calculation equation was generated, based on the ^4He atomic number generation equation proposed by Ballentine and Burnard (2002). This equation was used then to calculate the amount of helium produced by the Wufeng-Longmaxi shale in the Sichuan Basin.

$$V_{4_{He}}=\left\{\frac{(3.115\times10^6+1.272\times10^5)[U]+7.710\times10^5[Th]}{N_A}\right\}\times V_m\times(\rho_s\times v)\times yr, \quad (1)$$

where $V_{4_{He}}$ is the amount of helium produced by shale (m^3); [U] and [Th] are the U and Th contents in the shale (ppm), respectively; N_A is the Avogadro constant (6.02×10^{23} mol^{-1}); V_m is the molar volume of gas, (2.24×10^{-2} m^3) mol^{-1}; ρ_s is the density of rock (2.6 t m^{-3}); v is the volume of shale (m^3); yr is the time (yr). Based on the geological characteristics and radioactive log data of Wufeng-Longmaxi shale in the Sichuan Basin, the above unknown values were obtained. The average contents of U and Th in the shale are 30 and 15 ppm, respectively; the formation time of the Wufeng-Longmaxi shale is 4.4×10^8 yr; the average thickness of the shale is 260 m, and its distribution area is 13.7×10^4 km^2. By substituting these values into eq. (1), it was calculated that the accumulated amount of helium produced is approximately $1,650\times10^8$ m^3, since the formation of the Wufeng-Longmaxi shale in the Sichuan Basin.

There is a strong positive correlation between U and Th content, and the TOC content in shale. Additionally, the TOC content is an important index for screening the enrichment and high-production section of shale gas. The content ratio of U and Th can also be used to evaluate the redox environment and reservoir quality of shale (Ma et al., 2020). Therefore, there is good consistency between the amount of helium generated and that of shale gas. The shale intervals with high gas content and high helium content are primarily the intervals with a high U, Th, and TOC content (Figure 2). When the organic-rich shale is in the mature stage, it can produce both shale gas and helium, and the amount of shale gas produced is approximately 10,000 times that of helium, resulting in diluted helium in the shale reservoir. Presently, the relative abundance of helium in shale gas is approximately 0.01% (Qin et al., 2022).

3.2 Source and content of helium

As mentioned above, ^3He is formed from the mantle, whereas ^4He is predominantly from the crust. Therefore, the ratio of $^3He/^4He$ reflects the binary mixing of the mantle and

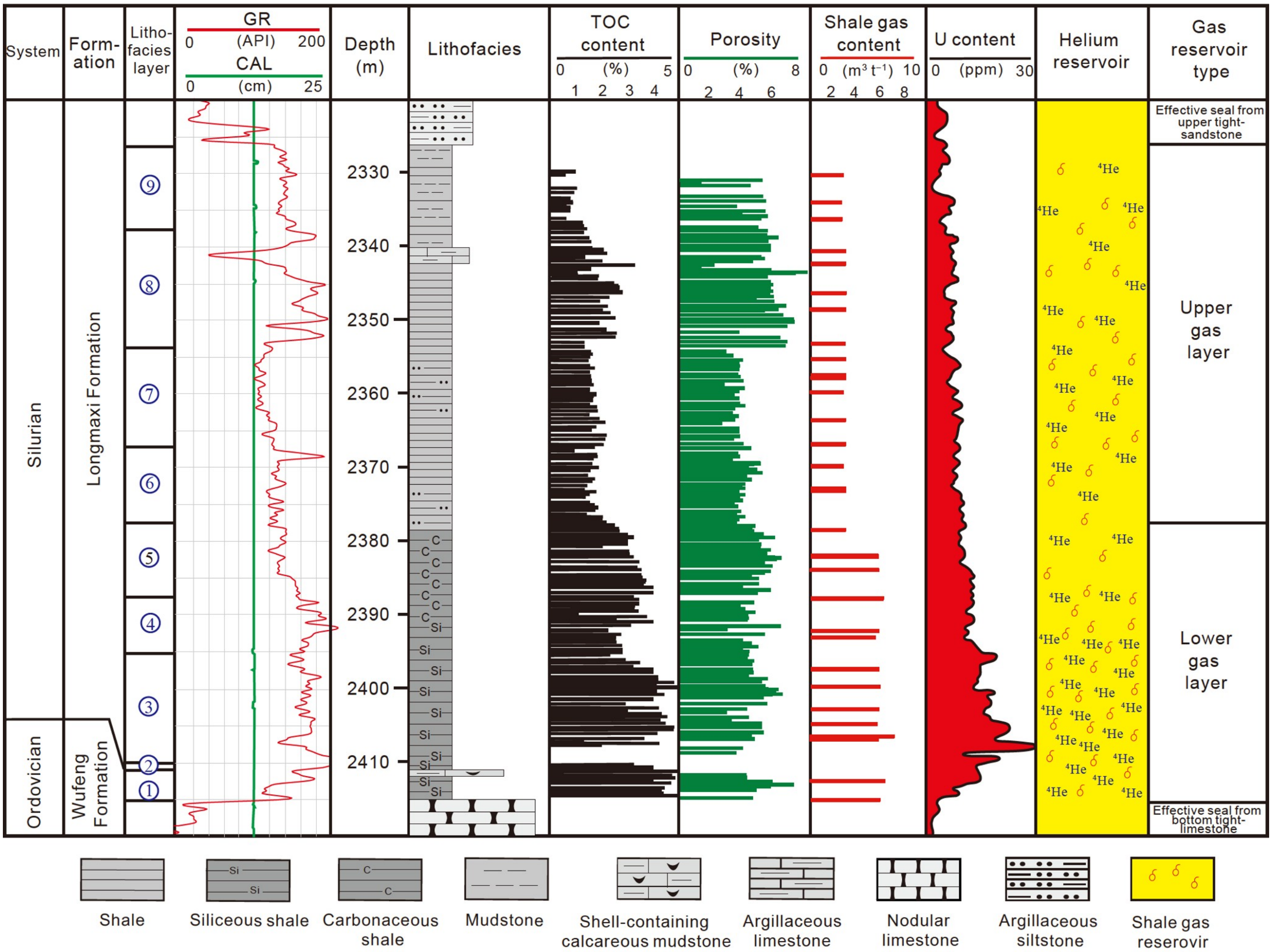

Figure 2 Comprehensive histogram and division of exploration and development of Shale Gas and Helium in Well Jiaoye 1 in the Sichuan Basin. GR is the gamma logging curve, unit: API; CAL is caliper logging, unit: cm; TOC is the total organic carbon content; ①–⑨ is the number of lithofacies sub-member.

crustal helium sources. To further understand the origin and source of helium in natural gas, we designated the ratio of $^3He/^4He$ in natural gas and the atmosphere as R and R_a, respectively. Then, $R/R_a=(^3He/^4He)_n/(^3He/^4He)_a$. In general, when $R/R_a<0.1$, all of the helium in natural gas is from a crustal source (Qin et al., 2022). Table 1 shows that the content of helium in Wufeng-Longmaxi shale gas ranges from 0.0145% to 0.1286%, with an average value of 0.0389%. The value of $^3He/^4He$ ranges from 0.76×10^{-8} to 6.35×10^{-8}, with an average value of 2.87×10^{-8}. The value of R/R_a ranges from 0.02 to 0.03 and all the values are lower than 0.1, suggesting that the helium in Wufeng-Longmaxi shale gas is a crustal source and is predominantly composed of ^4He. Therefore, this shale-type helium is produced by the α-decay of ^{238}U, ^{235}U, and ^{232}Th in shale.

In addition, by comparing the content of helium in each shale gas field in the Sichuan Basin and its periphery, it can be observed that the helium content of the shale gas fields in the basin generally ranges from 0.02% to 0.03%, and the ratio of $^3He/^4He$ ranges from 2×10^{-8} to 3×10^{-8}. These values vary little among shale gas fields, indicating that the main sources of helium in the basin are the same; that is, the shale produces helium. However, in the Pengshui area outside the basin and the Yongchuan and Dazu areas near the great faults inside the basin, the average content of helium in shale gas exceeds 0.05%, and the highest value is close to 0.10%. Additionally, the value of $^3He/^4He$ was 4.53×10^{-8}. These values are greater than those inside the basin. This may be due to complex tectonic plate movements, the development of faults, strong structural uplift, poor preservation conditions outside the basin, and the lack of replenishment of shale gas after loss. However, the U and Th in the shale continue to produce helium, weakening the dilution effect of shale gas and increasing its relative content of helium. Considering the relatively low shale gas content outside the basin, the resource potential of helium outside the basin is not as high as that inside the basin. Overall, the shale-type helium of the Wufeng-Longmaxi Formation in the Sichuan Basin and its periphery is a typical crustal helium source.

Table 1 The compositions and helium content in Wufeng-Longmaxi shale gas fields in Sichuan Basin and its periphery[a)]

Shale gas field	Gas compositions (%)								$^3He/^4He$ ($\times10^{-8}$)	R/R_a
	C_1	C_2	C_3	N_2	CO_2	H_2	H_2S	He		
Weiyuan	95.52–99.27 (97.84)	0.32–0.70 (0.53)	0.01–0.03 (0.02)	0.01–2.95 (0.58)	0.001–1.52 (0.66)	0.00–0.03 (0.01)	0.00–0.53 (0.18)	0.0172–0.1286 (0.0348)	1.20–4.49 (2.71)	0.01–0.03 (0.02)
Weirong	96.40–96.81 (96.61)	0.40–0.46 (0.43)	0.01–0.03 (0.02)	0.44–0.72 (0.60)	1.48–1.68 (1.58)	0.00–0.05 (0.012)	0.62–0.80 (0.73)	0.0201–0.0214 (0.0208)	/	/
Changning	97.69–99.28 (98.56)	0.32–0.54 (0.42)	0.00–0.10 (0.02)	0.00–0.75 (0.28)	0.00–0.91 (0.41)	0.00–0.01 (0.01)	0.45–1.00 (0.76)	0.0145–0.0507 (0.0267)	0.93–2.85 (1.58)	0.01–0.03 (0.02)
Zhaotong	98.21–99.45 (98.67)	0.47–0.62 (0.54)	0.00–0.01 (0.01)	0.03–0.63 (0.37)	0.00–0.51 (0.23)	0.00–0.02 (0.02)	0.00–0.57 (0.43)	0.0173–0.0414 (0.0266)	0.76–6.35 (2.20)	0.01–0.05 (0.02)
Fuling	97.54–98.95 (98.27)	0.43–0.74 (0.58)	0.00–0.05 (0.02)	0.08–1.36 (0.71)	0.00–1.16 (0.36)	0.00–0.01 (0.01)	0.00–0.56 (0.11)	0.0199–0.0445 (0.0372)	1.02–6.01 (3.23)	0.01–0.04 (0.02)
Fushun Yongchuan	95.32–99.59 (97.42)	0.23–0.60 (0.37)	0.00–0.01 (0.001)	0.01–0.53 (0.38)	0.06–1.60 (1.05)	/	/	0.041	2.61–3.26 (2.94)	0.02
Pengshui	98.46–98.77 (98.62)	0.55–0.71 (0.63)	0.00–0.01 (0.01)	0.05–0.15 (0.10)	0.35–0.94 (0.64)	/	/	0.0830–0.1000 (0.0993)	4.43–4.62 (4.53)	0.03
Dingshan	98.17–99.02 (98.60)	0.44–0.68	0.00–0.01	0.44	0.42–0.44	/	/	0.0272–0.0294 (0.0282)	/	/

a) The values in brackets are the average values; sources of data: Dai et al. (2014, 2016); Gao (2015); Cao et al. (2018); Feng et al. (2020); Liu et al. (2021); Li et al. (2021); Qin et al. (2022).

3.3 Helium migration modes and channels

Similar to conventional oil and gas, helium migration and accumulation is the process of migration, through channels in a certain mode and under various dynamic conditions. However, compared with natural gas generation, the amount of helium generated through slow decay is low. Therefore, it is difficult for helium to form a free gas column and the helium enters the trap under the drive of buoyancy to form a helium reservoir. Therefore, helium is usually aggregated and transported in the form of molecular diffusion and pore water (i.e., water-soluble phase), oil/gas, or non-hydrocarbon gases (such as nitrogen and carbon dioxide) through fractures and pore channels driven by pressure and concentration differences. The molecular diameter of helium is only 0.26 nm (methane is 0.42 nm) with strong diffusion and penetration ability. Therefore, the helium in shale reservoirs is likely to undergo primary and secondary migration, resulting in a loss of helium or migration to sandstone or carbonate reservoirs for enrichment. Helium in shale may escape in two different ways, including slow diffusion and large-scale release accompanied by hydrocarbon expulsion. The enrichment of helium is similar to that of shale gas, which is self-generated and self-stored in tight shale formations. There are four dynamic mechanisms of helium migration from the shale layer, among which differential pressure, concentration diffusion, and buoyancy drive mainly occur in a relatively stable structural environment, while the flow of natural gas tends to occur in areas with large hydrocarbon expulsion or developed faults.

3.4 Main controlling factors and distribution law of shale-type helium enrichment

Helium enrichment is a dynamic equilibrium between generation, accumulation, preservation, and escape. A rich helium reservoir requires a greater supply and less loss. Therefore, a rich supply of helium sources is fundamental, and preservation is key. Given a sufficient gas supply, helium enrichment is strongly controlled by the preservation conditions. The world's largest helium field, the North-South Pars gas field, is the Permian-Early Triassic Khuff Formation carbonate gas reservoir, in Lower Silurian hot shale, and the cap rock is Triassic Dashtak Formation gypsum salt. Gypsum salt provides the key conditions for the preservation and enrichment of conventional natural gas and helium. The helium content of the gas field is only 0.04%, but the proven reserve of natural gas is as high as 25.5×10^{12} m^3, with a helium reserve of 102×10^8 m^3, accounting for 25% of global helium reserves (Tao et al., 2019). In 2021, this field produced 0.51×10^8 m^3 of helium, accounting for about 31.9% of the annual helium production in the world (Data source: https://www.statista.com/statistics/925214/helium-production-worldwide-by-country/). Although the helium content is much lower than the 0.1% helium-rich grade, it still achieves great commercial development and utilisation value owing to its large reserves. The shale-type helium of the Wufeng-Longmaxi Formation in the Sichuan Basin and its periphery has geological conditions similar to those of the North-South Pars helium field. The helium content in different regions is significantly affected by Triassic salt. In the anticline area capped with extensive Triassic gypsum salt, the helium content is positively correlated with shale quality (U content). For example, horizontal wells such as the JY 6-2 HF well and JY 8-2 HF well target the second and first layers of the Wufeng-Longmaxi Formation, respectively. The U contents in these two wells are relatively high, and the helium contents are as high as 0.06% and 0.044%, respectively. In contrast, the JY 1HF well travels through the fourth to the sixth layers where the helium content is 0.037%, and the rest of the wells passing through the third layer have helium contents ranging from 0.04% to 0.05% (Figure 3). Hence, layers with the highest shale gas content, such as the fourth to sixth layers, tend to show a higher helium content in the Jiaoshiba area. The shale gas and helium contents are significantly positively correlated (Figures 2 and 4). In summary, the regional cap rock is the key to helium preservation. In addition, a large number of CH_4 or CO_2 molecules in shale gas reservoirs flowing into the cap rock block porous channels, which slows the escape of helium.

It should be noted that because shale is usually the source rock of conventional gas reservoirs, the helium generated from shale is expelled and migrates with the shale gas to conventional reservoirs to form conventional gas reservoirs. Therefore, conventional gas reservoirs contain a certain amount of helium. However, because the content and industrial or commercial value of helium is low, it cannot be extracted directly. In areas close to the basement of the basin or faults, the faults provide a good pathway for the upward migration of helium from the deep basement granite to shale reservoirs. Therefore, deep granite is another helium source for conventional gas reservoirs in addition to shale. Double helium sources provide a fundamental condition for a high helium content in conventional gas reservoirs. Under suitable geological conditions, a conventional gas reservoir with a high helium content can be formed (Wang et al., 2020), such as the Precambrian and Cambrian gas reservoirs in the Weiyuan area.

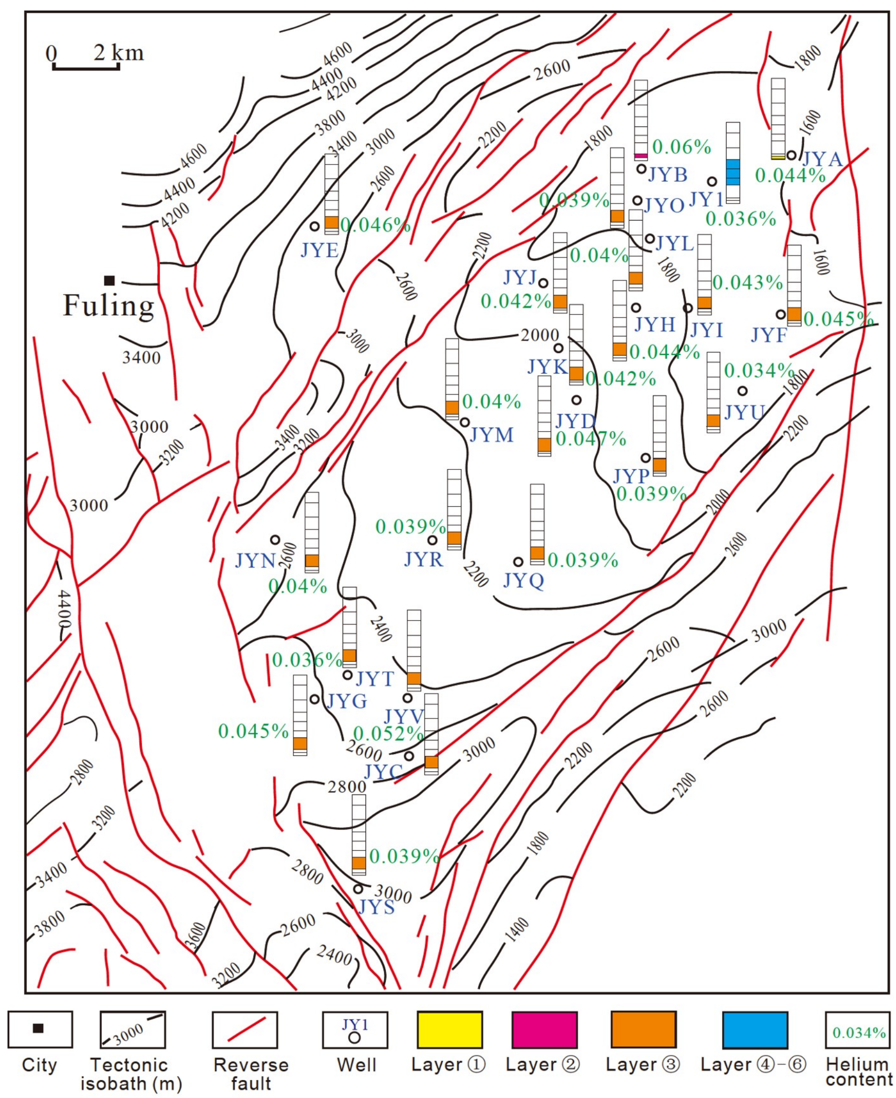

Figure 3 Distribution of helium content of the Jiaoshiba shale gas field in the Sichuan Basin. Data sources: Dai et al. (2016); Feng et al. (2020); Tenger et al. (2020); Liu et al. (2021); Li et al. (2021); Qin et al. (2022), and Chen et al. (2023).

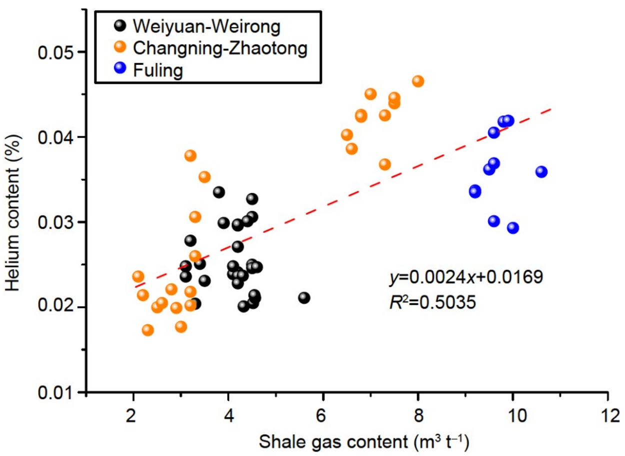

Figure 4 Relationship between helium and shale gas contents of the main shale gas fields in Sichuan Basin.

For shale-type helium, helium, and shale gas exhibit good consistency in enrichment and distribution. Shales with high U and Th contents are mainly distributed at the bottom of the Wufeng-Longmaxi shale. This is consistent with the high production and enrichment sections of shale gas (Figure 2). A previous study on the preservation conditions of shale gas in the Wufeng-Longmaxi Formation showed that the earlier the uplift and fracture closed after J_3-K_1, the more favourable it is for shale gas enrichment (Jin et al., 2018), and the simulated storage time of helium in sediments is approximately 120 Ma (Zhou and Ballentine, 2006; Schlegel et al., 2011). Therefore, if there is no fault damage, the time required for all the helium to dissipate through slow diffusion is approximately 100 million years, which coincides with the last uplift of the Wufeng-Longmaxi shale gas. Therefore, areas with fewer faults inside the Sichuan Basin generally have better helium storage conditions. For example, in the Jiaoshiba anticline and Pingqiao anticline, the content of helium in shale is relatively high, ranging from 0.041% to 0.049% with an average value of 0.045%. This may be due to the enrichment of helium caused by intra-source migration at higher structures and more favourable preservation conditions (Figure 5).

4. Shale-type helium resource potential as well as exploration and development strategies

The helium abundance in shale gas reservoirs is generally low owing to the dilution of natural gas. As a result, the scale of shale-type helium resources and reserves is primarily controlled by total natural gas resources or reserves. The shale gas geological reserves in China are approximately 21.84×10^{12} m^3 (Oil and Gas Resources Strategic Research Center of the Ministry of Land and Resources, 2016). The shale-type helium geological reserves in China will reach 87.36×10^8 m^3, under the assumption that the helium content is 0.04%. The proven shale gas reserves in the Sichuan Basin and its surroundings are 2.7×10^{12} m^3, and the helium reserves are 10.8×10^8 m^3. Helium reserves of 1×10^8 m^3 are defined as a super-large helium field based on the industrial classification standard proposed by Dai et al. (2017). The shale gas fields (or blocks) mentioned above are potential super-large helium fields in China. It is estimated that the helium in shale gas geological reserves and proven reserves can be used for approximately 500 and 50 yr, respectively, based on the helium resource consumption of 0.21×10^8 m^3 in 2021 (almost all of which was imported). Taking Fuling Shale Gas Field as an example, the proven reserves of shale gas are approximately $9{,}000\times10^8$ m^3, and the helium reserve is 3.6×10^8 m^3, indicating a super large helium field. The shale gas production

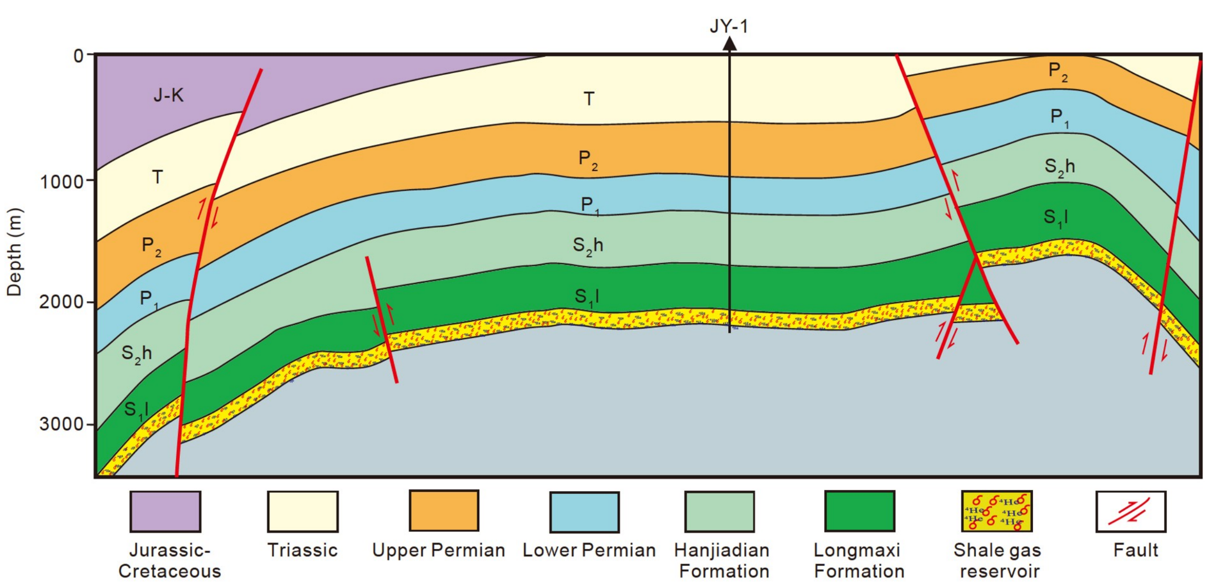

Figure 5 Accumulation model of shale gas and helium in Jiaoshiba, Sichuan Basin.

in the Sichuan Basin and its surroundings was 228×10^8 m^3 in 2021, and the annual helium production associated with shale gas development was up to 0.0912×10^8 m^3. Consequently, the economic value is about 3.6 billion RMB based on the price of 400 RMB per cubic meter.

It is generally believed that helium production has commercial value only when the relative helium content in natural gas reaches above 0.05–0.1% (Xu et al., 1996). However, this study believes that the associated natural gas reserves should also be considered. The relative total amount of helium and hydrocarbon gases is critical for determining whether helium can be commercially developed. Specifically, although the helium content is high, the limited overall natural gas reserves reduce the industrial value of helium. Helium resources cannot be developed for regions with low or no natural gas production. In contrast, although the shale-type helium content is low, the helium reserves are considerable because of the large scale of the shale gas reserves. The low helium content in shale gas has led many oil companies to ignore helium resources in the past because they did not know the helium value, or that the natural gas they mined contained high-value helium, resulting in the waste of hundreds of millions of cubic meters of helium resources annually.

Presently, China has mastered the helium extraction technology at a helium content of 0.1% to extract high-purity helium, including the small-scale helium-rich natural gas cryogenic and normal temperature methods, which has achieved industrial application in the Weiyuan Gas Field in the Sichuan Basin. The natural gas helium extraction unit was completed in Rongxian County, Zigong City, Sichuan Province in 2012, which was the only operated natural gas helium extraction unit in China. The helium content was 0.18%, with an annual production of pure helium of approximately 21×10^4 m^3, and the crude helium purity was between 90% and 95%. However, the technology for extracting helium from large helium-poor natural gas reservoirs is not yet mature. Shale-type helium is rich in reserves, whereas its grade is relatively low, bringing difficulty to direct extraction. However, to increase commercial, the LNG tail gas concentration can be used to increase the helium content from 0.04% to 50% (Daly, 2005). Globally, a large amount of helium is commercially extracted in this manner, such as in the well-known North Gas Field in Qatar. Presently, natural gas sold via LNG from Fuling Shale Gas Field is approximately 100×10^4 m^3 (accounting for ~5% of the total production). Therefore, this extraction technology can produce 14.6×10^4 m^3 helium in one year. Shale gas fields in China have centralised dehydration treatment stations, for example, Baitao in Fuling, Dongsheng in Nanchuan, and Weiyuan. These dehydration stations should be selected as pilot test areas for helium resource development and utilisation to accelerate the feasibility of building helium strategic reserve bases.

5. Conclusions

Shale-type helium of the Wufeng-Longmaxi Formation in the Sichuan Basin and its periphery is typically crustal-sourced and is characterised by self-generation, self-storage, and large-area distribution. The enrichment of shale-type helium depends on its generation intensity of helium, preservation conditions, structural characteristics, etc. The content of helium ranges from 0.02% to 0.1%, and the resource potential of helium is controlled by both the helium content and its associated natural gas resources.

The reserves of shale-type helium in the Wufeng-Longmaxi Formation in the Sichuan Basin and its periphery are relatively large, and a large amount of helium is produced every year along with shale gas. However, because of its low abundance and difficulty in extracting helium, it has not attracted the attention of oil companies. Owing to the large amount of shale-type helium and its large economic value, in-depth research on the helium extraction technology of helium-deficient and helium-containing natural gas is recommended, such as helium extraction membranes and other containment technologies. It is of great practical significance to improve the economy of helium production, ensure safe helium consumption, and promote the development of China's helium extraction industry from natural gas.

China's shale-type helium fields have significant resource potential and development value. It is recommended to conduct research on shale gas experimental analysis and testing technology, helium enrichment mechanisms, and distribution laws, and to explore and evaluate the helium resource potential in hydrocarbon and non-hydrocarbon natural gas reservoirs.

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