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Original Paper

The occurrence, origin, and enrichment of helium in the Wufeng-Longmaxi shale gas in the Sichuan Basin, China

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A B S T R A C T

Helium is a valuable natural resource used widely in high-tech industries because of its unique physical and chemical properties. The study of helium in shale gas is still in its infancy, and the content, genesis, and enrichment patterns of helium in shale gas are not yet clear. In this paper, the concentrations and isotopic characteristics of helium were investigated in the Wufeng-Longmaxi shale gas in the Sichuan Basin and the periphery areas. The analytical results show that the concentrations of helium in the southern Sichuan shale gas fall in the range of 0.018–0.051 vol% with an average of 0.029 vol%. The helium abundance in Weiyuan shale gas are relatively low compared to those in conventional natural gas pools from the same area (generally greater than 0.20 vol%), reflecting the significance of long distance migration to the enrichment of helium in gas pools. The relatively low ratios of ^3He and ^4He in shale gas indicate that most of the helium are crustal derived helium. Further quantitative estimate based on helium, neon, and argon isotopic ratios suggest almost 100% crustal helium source. The helium residing in shale reservoirs can be deconvoluted into the indigenous helium generated in-situ by shale and exogenous helium generated from external helium source rocks and charged through faults and/or fractures networks. According to preliminary calculations, external helium source is required to meet the threshold of an economic helium-rich field of helium concentration of 0.1 vol% except for particular areas with extraordinarily high uranium and thorium concentration. Based on detailed study on typical helium-rich shale gas reservoirs, major advantageous features for helium's enrichment in shale gas include: (1) high-quality helium source rocks, (2) effective migration paths, and (3) diminished dilution effects of shale gas. Shale gas plays with underlying ancient cratonic basement, well developed source-connecting faults, and moderate pressure coefficient are potential targets for helium exploration.

1. Introduction

As to its unique physical properties such as the extremely low boiling point (4.2 K), chemical inertness, small molecular size of 0.26 nm, helium is largely used by the cryogenic, semiconductor, nuclear industry and some other high-technique sectors (Ballentine and Burnard, 2002; Ozima and Podosek, 2001). According to the newly released data from USGS (Geological Survey, 2022), the resources of helium in China are 1.1×10^9 m^3, accounting for only 2% of the total helium resources over the world. Meanwhile, China's demand for helium continues to increase at an annual rate of greater than 10% (Siddhantakar et al., 2023). The surge in demand of helium encourages active research of helium in domestic natural gas plays in China.

Helium, as with all noble gases, is mainly sourced from the atmosphere, the crust, and the mantle. The atmospheric helium is dominantly released from volcanic eruptions and rocks weathering, and can enter petroleum systems by dissolving in circular groundwater. Mantle-derived helium originating from mantle-

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derived volatiles and magma degassing can be introduced into the crust through faults in tectonically active areas (Oxburgh et al., 1986). The crustal helium are usually released by radioactive decay of U (uranium) and Th (thorium) elements in rocks (Ballentine and Burnard, 2002). It is noteworthy that helium associated with economic reserves is always dominated by crustal helium in source (Ballentine and Sherwood Lollar, 2002).

Helium contains two stable isotopes, ^{3}He and ^{4}He. The former ^{3}He is usually associated with tectonic and volcanic activities along with magma degassing (Oxburgh et al., 1986). ^{4}He is a product of the alpha decay of ^{235}U, ^{238}U, and ^{232}Th in minerals e.g., in granite, volcanic rock, black shale and etc. (Ballentine and Burnard, 2002). After generation, helium can dissolve in and migrate along basinal fluids once releases from its source rocks. During migration process, helium can be extracted and introduced into gas phase, forming helium-bearing natural gas reservoirs in favorable traps (Brown, 2010).

Because of their chemical inertness and trace natural abundance, helium along with other noble gases and the isotopic characteristics have been successfully applied to understand the complex physical and chemical processes in different geological settings (Ballentine and Burnard, 2002). Until recent years, helium is regarded as a type of valuable resource associated with natural gas in China, and large amounts of studies have been conducted on the origin, genesis, migration, and enrichment patterns of helium in natural gas fields. Similar to hydrocarbon accumulations, economic helium accumulation is a premier results of static geological elements (source rocks, reservoirs, and seal rocks), and dynamic coupling of generation, migration, and accumulation (Brown, 2010). However, most of these studies have been confined in conventional petroleum systems so far. Our understanding of helium in unconventional natural gas systems representing a rather different subsurface environment still remain limited.

The recent decade has witnessed the rapidly growth of shale gas industry in the Sichuan Basin in China (Li et al., 2021). By the end of 2023, the cumulative proven geological reserves of shale gas in the Wufeng-Longmaxi Formation are 2.73 × 10^{12} m^3, ranking it the largest shale gas producing basin in China (Yang et al., 2021). It is timely to conduct a systematic research on helium as with other noble gases in this huge shale gas basin. The aims of this study include: (1) reveal the spatial distribution characteristics of helium in the Sichuan Wufeng-Longmaxi shale gas; (2) address the geochemical characteristics of helium in shale gas; (3) identify the key controlling factors and reveal the enrichment pattern of helium in shale gas in the Sichuan Basin.

2. Geological background

The Sichuan Basin is located on the northwestern margin of the Yangtze Platform in the southwestern region of China, bounded between the Qiyue Mountains, Longmen Mountains, Dalian Mountain-Loushan, and Micao Mountain-Daba Mountains, with a total area of about 180,000 square kilometers. This basin has experienced a complex tectonic evolution process from a craton basin to a foreland basin from the Paleozoic to the Mesozoic-Cenozoic, and is a long-developed Paleozoic-Mesozoic-Cenozoic marine and terrestrial complex superimposed basin within the Tethyan tectonic domain (Liu et al., 2018).

Six sets of source rocks of marine, marine-terrestrial transitional, and terrestrial facies are extensively developed in the Sichuan Basin and its surrounding areas. Among them, the marine Upper Ordovician Wufeng-the Lower Silurian Longmaxi Formation shale is the most favorable shale gas exploration and development series in the region (Ma et al., 2020; Nie et al., 2023). Previous studies have shown that the thickness of high-quality shale in the Wufeng-Longmaxi Group in the Southern Sichuan area is between 40 and 60 m, and the TOC content is between 2.8 and 6.0 wt% (Zou et al., 2016).

Shale gas was first discovered in the Sichuan Basin in 1965, and the industrial breakthrough was achieved in 2009. By the end of 2023, eight large shale gas fields and one small shale gas field were developed in the Wufeng-Longmaxi Formation in the Sichuan Basin and its surrounding areas. The proven geological reserves of shale gas were 2.73 × 10^{12} m^3, and the shale gas production capacity was established at 450 × 10^8 m^3/year, with an annual shale gas production of 250 × 10^8 m^3 (Yang et al., 2021).

3. Samples and experimental methods

3.1. Samples

In this study, 33 shale samples were collected from the Wufeng-Longmaxi Formation in Dazu (DZ) and Luzhou (LZ) areas to measure the U and Th contents (trace element analysis). Forty two shale gas samples were collected from the Weiyuan (WY), Changning (CN), DZ, and LZ areas in the southern Sichuan Basin. The gas samples were collected using high-pressure steel cylinder containers with double valves. The containers were flushed by shale gas five times to avoid atmospheric contamination during gas collection. Pressures in the cylinders ranged from 5 to 12 MPa. Moreover, additional data of shale gas samples from the Sichuan Basin were collected from previous studies (Cao et al., 2018; Chen et al., 2023; Dai et al., 2016; Li et al., 2021; Li et al., 2021, Luo et al., 2019; Qin et al., 2022) to investigate the contents and isotopic characteristics of helium in the Sichuan shale gas.

3.2. Analytical methods

The geochemical composition and noble gas isotope (He, Ne, and Ar) analysis of all gas samples in this study were completed at the Key Laboratory of Petroleum Resources Research, Institute of Geology and Geophysics, Chinese Academy of Sciences (Lanzhou, China).

3.2.1. Gas composition analysis

The chemical compositions of gases (hydrocarbon gases, N_2 and CO_2) were analyzed by an Agilent 6890N gas chromatograph (GC) equipped with a flame ionization detector and a thermal conductivity detector. Individual hydrocarbon gas components were separated using a capillary column (PLOT Al_2O_3 50 m × 0.53 mm). The GC oven temperature was initially set to 30 °C for 10 min, then increased to 180 °C at 10 °C/min and held at this temperature for 20–30 min. The standard deviation of each component in this analysis method were less than ±0.5%.

3.2.2. Composition and isotopic analysis of helium, neon and argon

The abundances of helium, Neon (Ne) and Argon (Ar) were determined by a Pfeiffer OmniStar 220 quadrupole mass spectrometry (QMS) using atmosphere and standard gases for calibration. QMS provide a fast, reliable and precise approach for measuring the contents of non-polar and noble gases with detection limits below 1 ppm.

A Noblesse SFT noble gas mass spectrometer was used to analyze the He, Ne, and Ar isotopic measurements. A defined amount of sample gas was transferred into a preparation line that separated and purified the noble gases and was connected to the Noblesse SFT apparatus. The standard sample used for detection was air from the top of Gaolan Mountain in Lanzhou City. Analytical procedure were described in details in Cao et al. (2018).

The standard deviations of He, Ne, and Ar isotopic ratios were less than ±3%.

3.2.3. Trace elements analysis

The shale samples were ground to 200 mesh, and then heated in a muffle furnace to 105 °C for 12 h to remove the organic matter and the crystalline water. The pulverized samples were then dissolved in pure nitric acid (HNO_3) and hydrogen fluoride acid (HF), and finally subjected to the analysis of an inductively-coupled plasma mass spectrometry (ICP-MS, Nu Instrument, Nu Attom). The relative standard deviations were typically lower than ±5%.

3.2.4. Calculation the amount of helium generated in U and Th-bearing rocks

The annual generation amount of radiogenic ^4He in U and Th-bearing rocks since deposition, denoted as F_{He} with a unit of $cm^3/(g \cdot a)$, can be roughly estimated by the following equation (Brown, 2010):

$$F_{He} = 1.22 \times 10^{-13} \times [U] + 2.92 \times 10^{-14} \times [Th] \quad (1)$$

where [U] and [Th] are the concentrations of U and Th in shale with a unit of μg/g.

Therefore, the total generation amount of radiogenic ^4He, denoted as D_{He} with a unit of cm^3/g, can be calculated as:

$$D_{He} = F_{He} \times t \quad (2)$$

where t is the duration of time (with a unit of year) since helium generation from the source rocks.

In this study, average U and Th contents were used in the calculation to provide a rough idea on the contribution of helium generated by the U and Th in shale. It was assumed that the radiogenic reactions started since the strata was deposited, and 100% of helium generated can be release from its source rock and migrated into the gas play.

3.2.5. Estimation of the generation amount of shale gas

The amount of gas generation of shale per unit mass can be calculated by Eq. (3):

$$D_g = TOC \times P_{gmax} \times T_{rg} \quad (3)$$

where: D_g—Gas generation intensity, m^3/t; TOC—Total organic carbon content, %; P_{gmax}— Maximum gas generation potential, m^3/t; T_{rg}—Gas generation conversion rate, obtained from the hydrocarbon source rock gas generation curve, %.

4. Results and discussion

Economically, a helium-rich gas field is grouped as one which has a helium volume percent of 0.3% (3000×10^{-6} cm^3 STP/cm^3 or ppmv) or greater. However, geochemically, a helium-rich field can be classified as helium concentration above 0.1 vol% (1000 ppmv), since helium occurring in most fields as trace amounts (Danabalan, 2017). Technically, the concentration of helium in natural gas should reach 0.05 vol% (500 ppmv) to be economical for helium extraction, constrained by current level of helium extraction technology (Xu et al., 1989). Through the LNG-BOG and natural gas separation method Qatar achieved commercial extraction of helium with concentration as low as 0.04 vol% (Daly, 2005). In this study, a helium-rich gas field is defined as one having a helium content of 0.1 vol% or greater, and a helium-bearing gas field is one containing a helium content of 0.05 vol% to 0.1 vol%.

4.1. The concentrations of helium in shale gas in the Sichuan Basin

4.1.1. The spatial distribution of helium

The concentrations of helium collected in Southern Sichuan Basin fall in the range of 0.018−0.051 vol% (180–510 ppmv), with an average of 0.029 vol% (290 ppmv), which is comparable with previously reported results. For instance, shale gas collected at WY area was found to contain 228 to 1286 ppmv of helium (Cao et al., 2018; Dai et al., 2016; Li et al., 2021), and shale gas in CN, Weirong (WR), FL-JSB all share similar helium concentrations range from ~200 to 600 ppmv (Cao et al., 2018; Chen et al., 2023; Dai et al., 2016; Li et al., 2021).

Relatively higher helium contents were reported in the shale gas collected from the Sichuan Basin's eastern periphery areas (Fig. 1). Shale gas from Danzhai in Guizhou contains 2200 ppmv helium (Dan et al., 2023). In addition, the Shuijingtuo shale gas in Yichang area can reach up to 3000 ppmv, making it the helium-richest shale gas reported in China (Luo et al., 2019). These data suggest that helium contents in the Sichuan shale gas vary spatially, being generally higher in the periphery areas than in the basin areas. This is probably because of the more intense tectonic activities of the periphery areas not only facilitating the release of helium from the source rocks but also resulting favorable helium migration paths and diminished dilution of shale gas. This will be discussed into details in Section 4.4.

The Cambrian Qiongzhusi and Shuijingtuo shale gas seems to contain relatively higher helium concentrations than the Wufeng-Longmaxi shale (Table 1). For instance, the Cambrian Qiongzhusi shale in WY area is 1400 ppmv (Qin et al., 2022), higher than the helium concentration in the Wufeng-Longmaxi shale in the same area. This is probably because the total generation amount of helium is positively correlated with the age of helium source rock. Also, older shale is generally located closer to the ancient basement rocks which are usually great helium source rocks.

4.1.2. Compared with adjacent conventional gas pools

Many conventional gas fields are rich in helium. For instance, the world famous Hugoton-Panhandle gas field holds as high as 2930–10470 ppmv helium (Brown, 2019). Helium concentrations in the Cambrian, conventional natural gas fields in the WY area at the Sichuan Basin fall in the range of 1200–3420 ppmv. Helium in the Tarim Basin fall in the range of 3000–3700 ppmv (Tao et al., 2019), and in the Dongsheng tight gas field falls in the range of 450–4870 ppmv (He et al., 2022).

Fig. 2(a) compares the helium concentrations in conventional natural gas and shale gas. It is clearly shown that the helium concentrations in shale gas are significantly lower than those in conventional natural gas fields. This discrepancy can be seen more clearly when compared the helium concentrations in the conventional reservoirs and shale from the same area (Fig. 2(b)). In WY area at the Sichuan Basin, for example, the helium concentrations in the conventional gas field are generally above 2000 ppmv, whereas only around 200–1200 ppmv in the shale gas. Similarly, the study of helium concentration in Haynesville shale versus the overlying Cotton Valley sandstone at the east Texas area (Byrne et al., 2020) confirms the helium concentration is much higher in the sandstone than in shale.

The higher concentration of helium in conventional gas than in the shale gas is probably related to the longer distance of secondary migration of natural gas during accumulation. Secondary migration can occur for up to hundreds of kilometers laterally and vertically through high permeable migration conduits (Demaison and Huizinga, 1991; Zhu et al., 2013). The migrating hydrocarbons can undergo phase partitioning with surrounding groundwater. Because the noble gases are less soluble in water than in

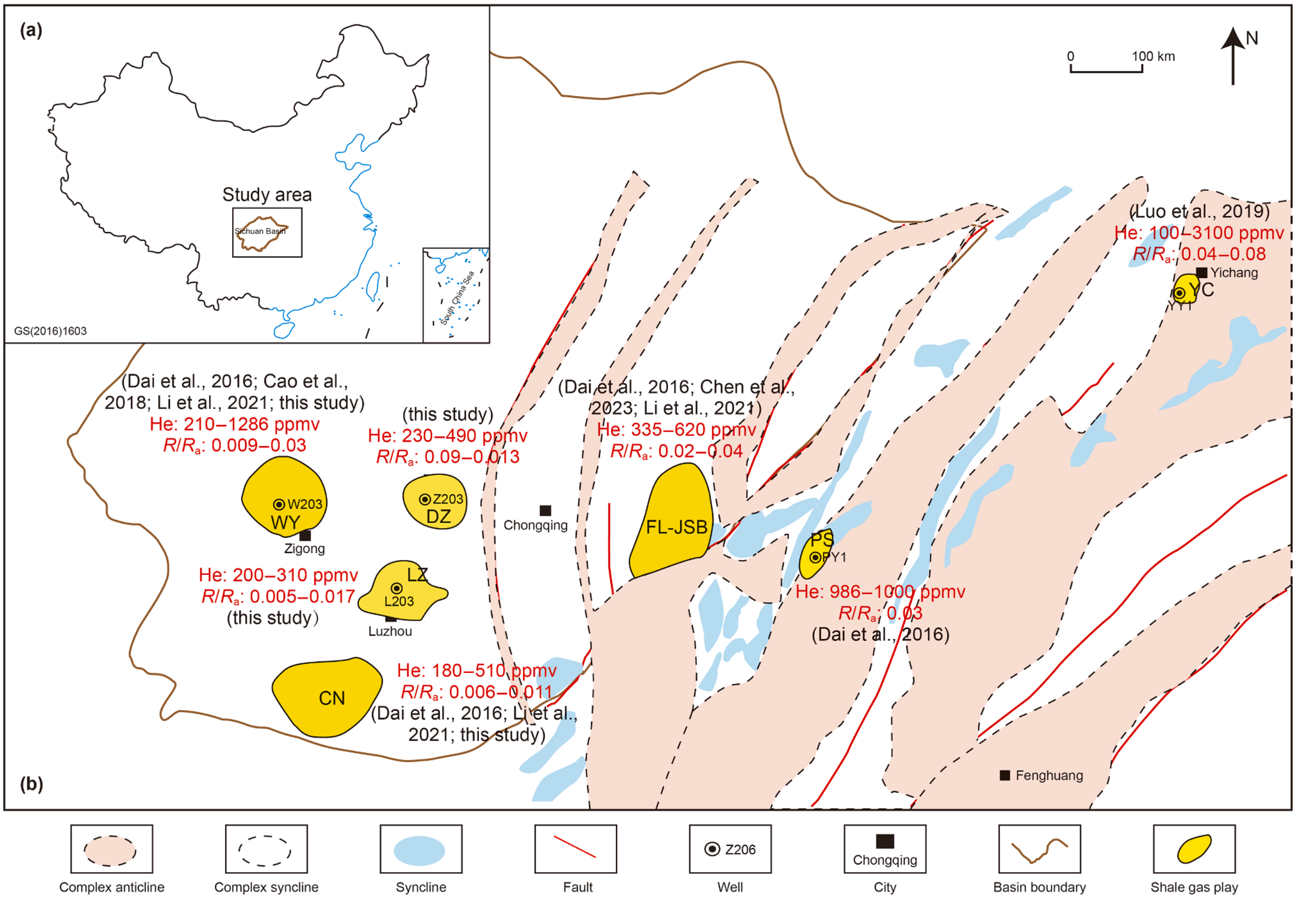


Fig. 1. The distribution of helium in Sichuan typical shale gas fields. The background is modified from Wang et al. (2022). WY: Weiyuan, CN: Changning, DZ: Dazu, LZ: Luzhou, FL-JSB: Fuling-Jiaoshiba, PS: Pengshui, YC: Yichang. The helium content data were obtained from present study, Chen et al. (2023), Dai et al. (2016); Luo et al. (2019), respectively.

hydrocarbons, as indicated in the well-defined Henry's law, the long-distance secondary migration can strip much of the noble gases off the groundwater to hydrocarbon (Ballentine et al., 1996; Byrne et al., 2017). In contrast, in systems where migration occurs over short distances, such as shale gas, it is very likely that hydrocarbons may encounter limited volumes of water with which to interact and partition noble gases. From this perspective, the larger scale and longer distance of petroleum's secondary migration, the higher concentration of helium in the gas fields. Previous studies of conventional systems (Ballentine et al., 1996) have shown contain concentrations of radiogenic noble gases far higher than that by local production, requiring external input. However, there has been no thorough investigation in the effects of secondary migration distance upon noble gas composition to date. Such study could prove an invaluable tool in assessing migration distances and pathways.

4.2. The genetic origin of helium

Helium isotopic characteristics can be used to distinguish the origin of helium.

Because of the differentiation of the crustal layers and the degassing of the mantle, the $^3He/^4He$ ratio in the atmosphere, crust and mantle vary distinctively, with typical values of 2×10^{-8}, 1.1×10^{-5}, 1.4×10^{-6}, respectively (Oxburgh et al., 1986). The $^3He/^4He$ ratio in the atmosphere is considered to be a constant and noted as R_a, which is commonly regarded as a proxy to scale helium isotopes of other sources. For instance, the typical crustal radiogenic helium is 0.01–0.05 R_a (Ballentine and Burnard, 2002), while the (MORB) and (OIB) standing for the upper and lower mantle end members are around 8 and 50 R_a, respectively (Graham, 2002).

The $^3He/^4He$ of the Sichuan shale gas ranges from 0.01 to 0.02 R_a, indicating a typical crustal helium source (Fig. 3(a)). The isotopic ratios of helium in shale gas are generally low. For instance, the $^3He/^4He$ ratios of helium in Marcellus shale gas (Hunt et al., 2012) and Barnett shale gas (Wen et al., 2017) are below 0.08 R_a, indicating typical crustal helium in these shale gas. In contrast, the $^3He/^4He$ ratios of helium in Antrim shale (Wen et al., 2015), Eagle Ford (Byrne et al., 2018) and Haynesville shale (Byrne et al., 2020) are slightly higher, suggesting relatively higher contribution of mantle-derived helium.

Similar to $^3He/^4He$, the $^4He/^{20}Ne$ ratios in the atmosphere, crust, and mantle are significantly different. The end-member values of $^4He/^{20}Ne$ of the atmosphere, crust, and mantle are 0.274, 10000, and 10000 (Castro et al., 2009; Oxburgh et al., 1986; Ozima and Podosek, 2001), respectively. Based on the three-component mixing model of the atmosphere, crust, and mantle, the contribution rates of crustal and mantle helium in shale gas can be quantitatively estimated. In this study, the $^4He/^{20}Ne$ ratios in the shale gas from the South Sichuan are 5924–38133, comparable with the results of previous studies on the Wufeng-Longmaxi Formation shale gas, ranging from 3943 to 44505 (Cao, 2017) (Fig. 3(a)). As shown in Fig. 3(a), the contribution of mantle-derived helium in the studied shale gas is generally less than 5%. The mantle helium over 0.5% contributions are only observed in

Table 1
The concentrations and isotopic characteristics of helium in shale gas plays in the Sichuan Basin and its periphery areas.

Area	Block	Formation	Depth, m	He, ppmv	R/R_a	$^{40}Ar/^{36}Ar$	$^4He/^{20}Ne$	Ref.
Southeast Sichuan	Weiyuan	Wufeng-Longmaxi	1520–1523	451.4–1286 (869)	0.03			(Dai et al., 2016)
			3882–4017	228–1286 (658)	0.019–0.025 (0.022)	340–7364 (3075)		Cao et al. (2018)
				239–250 (245)	0.019–0.029 (0.024)	570–2877 (1430)		Li et al. (2021)
				210–330 (260)	0.0095–0.013 (0.012)	464–713 (530)	5924–11752 (9113)	This study
		Qiongzhusi		1400	0.02			Qin et al. (2022)
	Changning	Wufeng-Longmaxi		386.1–445.9 (419.6)	0.008–0.011 (0.01)			Cao et al. (2018)
				154–507 (206)	0.0093–0.011 (0.01)	455–1071 (738)	3229–26825 (11320)	Liu et al. (2021)
			2002–2745	187–353 (262.75)	0.01–0.03 (0.0175)			Dai et al. (2016)
				199–414 (275.9)	0.0067–0.031 (0.021)	432–713 (573)	577–2081 (1027)	Li et al. (2021)
				180–510 (340)	0.0062–0.0011 (0.087)	23346–32686 (28596)		This study
	Weirong			201–214 (208)	0.018–0.043 (0.027)	540–843 (762)		Li et al. (2021)
	Luzhou			200–310 (260)	0.0051–0.017 (0.01)	499–662 (573)	13742–38133 (29929)	This study
	Dazu			230–490 (350)	0.086–0.013 (0.01)	448–684 (547)	10284–17357 (12438)	This study
	Fuling-Jiaoshiba		2408–2416	335–419 (377)	0.02–0.04 (0.35)			Dai et al. (2016)
				357–445 (389.4)	0.005–0.032 (0.018)	809.5–1134 (967)		Li et al. (2021)
				340–620 (450)	0.004–0.03			Chen et al. (2023)
East periphery	Pengshui		2313–2341	986–1000 (993)	0.03			Dai et al. (2016)
	Danzhai	Niutitang	950–1050	2200				Dan et al. (2023)
	Yichang	Shuijingtuo	1894.1–2113.5	100–3100 (1600)	0.04–0.08 (0.0667)			Luo et al. (2019)

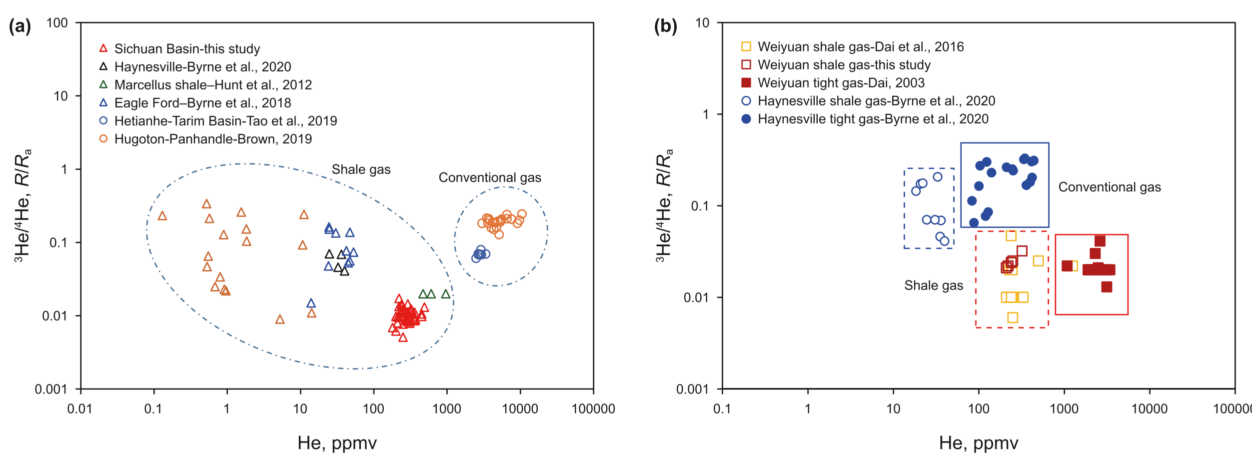


Fig. 2. The comparison in helium concentration and isotopic characteristics between conventional gas fields and shale gas globally (**a**) and from Weiyuan and Haynesville (**b**). Note that the concentration of helium in conventional gas fields are relatively higher than that in the shale gas from the same areas.

certain samples in the Antrim shale (Wen et al., 2015), Eagle Ford (Byrne et al., 2018) and Haynesville shale (Byrne et al., 2020) in the US.

$^{40}Ar/^{36}Ar$ ratio can be regarded as a mixing system between the modern atmosphere $^{40}Ar/^{36}Ar$ ratio of 298.6 (Lee et al., 2006) and the addition of radiogenic ^{40}Ar, produced mainly from crustal ^{40}K decay. Elevated $^{40}Ar/^{36}Ar$ ratios can be caused either by radiogenic production of ^{40}Ar within the crust, or mixing with mantle fluids which have high levels of radiogenic ^{40}Ar (Byrne et al., 2017). The $^{40}Ar/^{36}Ar$ ratios in Sichuan shale gas ranges from 431.8 to 713.3, which is higher than the atmospheric $^{40}Ar/^{36}Ar$ characteristic value of 295.5. It is close to previous reported results of CN (455–1360, Liu et al., 2021), WR (540–842, Li et al., 2021) and JSB shale gas (809–1134, Li et al., 2021) in the Sichuan Basin and Eagle Ford shale in the US (342–561) (Byrne et al., 2018), and much lower than that in the samples from the Songliao Basin (504–2044) (Feng et al., 2001) (Fig. 3(b)).

4.3. *The source rocks of helium*

4.3.1. *In-situ and external helium in shale gas*

Above discussion indicates that the helium in the Sichuan shale gas originates mainly from crustal U and Th radiogenesis. Based on the physical position of helium source relative to shale, the crustal helium in shale gas can be further divided into helium produced by in-situ shale radioactive decay (referred to as in-situ helium), and helium generated by rocks residing outside the shale (referred to as external helium). The external helium can enter shale via pathways such as fractures or pores through underground fluids. Therefore, total helium in the system can be calculated using the following formula:

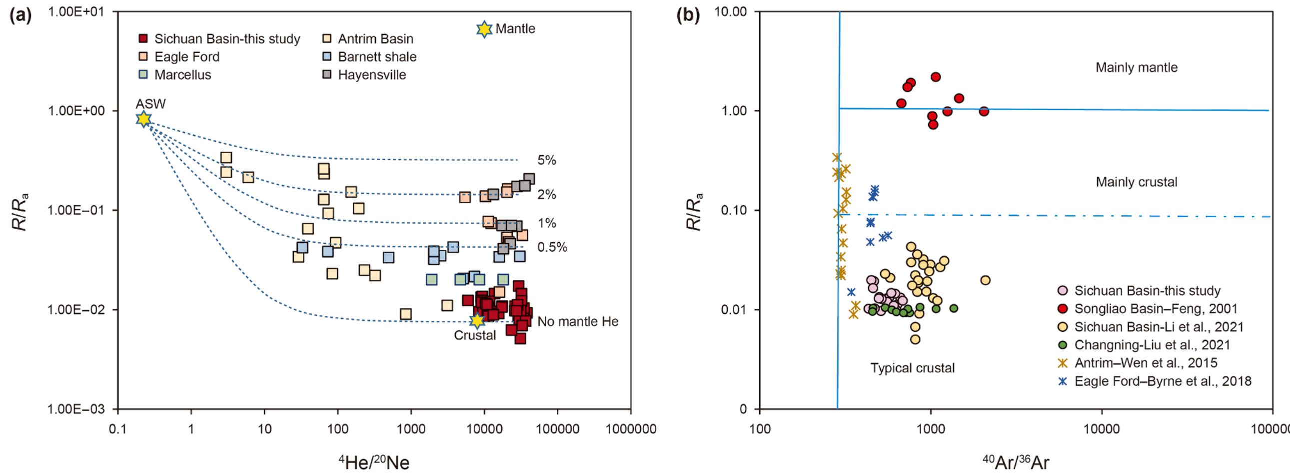


Fig. 3. Plots showing the scattering points of R/R_a vs $^4He/^{20}Ne$ (**a**) and R/R_a vs $^{40}Ar/^{36}Ar$ (**b**) in the Wufeng-Longmaxi shale gas. The results of Sichuan shale are from the present study, while the data of Antrim shale, Eagle Ford shale, Barnett shale, Marcellus shale and Haynesville shale were from Wen et al. (2015), Byrne et al. (2018), Wen et al. (2017), Hunt et al. (2012), Byrne et al. (2020), respectively.

$$[He]_{total} = [He]_{in\text{-}situ} + [He]_{external} \tag{5}$$

To reveal the enrichment patterns of helium in shale gas, it is important to evaluate the relative contributions of in-situ and external helium first, since different source of helium enriches in dramatically different ways. For in-situ helium, the most significant controlling factor is the concentration of U and Th, the volume, and the depositional age of shales. While for external helium, factors such as in the vicinity to strong external helium source and effective migration pathways are more important.

A straightforward way to solve this problem is to calculate whether the in-situ helium alone is sufficient to form an economic helium-rich shale gas play (i.e. the contents of helium greater than 0.1 vol%). Given 99% of the helium in shale gas is composed of ^4He, the ^4He amounts calculated from Eqs. (1) and (2) could be regarded as the total helium contents in the shale gas. Trace element analysis results suggest that the average U and Th concentrations of the Wufeng-Longmaxi shale are 11.8 and 17.4 μg/g in LZ area, 15.9 and 15.3 μg/g in DZ, respectively. The U and Th contents in the Wufeng-Longmaxi shale in other blocks were cited from previous studies (Table 2).

The calculation indicated that total helium generated from unit weight of Wufeng-Longmaxi Shale is around (8.46–11.9) × 10^{-4} m^3/t. The average gas-in-place contents of Wufeng-Longmaxi shale are around 1.9–7.2 m^3/t (Table 2, Yang et al., 2019; Zhang et al., 2019; Zou et al., 2016). Thus, the final helium concentration in Wufeng-Longmaxi shale gas is 0.0124 vol%–0.0593 vol% (124–593 ppmv), which is much lower than the 1000 ppmv threshold of economic helium-rich gas field. Therefore, except for particular areas with extremely high U and Th concentration (e.g. greater than 50 μg/g) and low gas contents (e.g. less than 2 m^3/t), external helium is essential for the formation of helium-rich shale gas fields.

4.3.2. External helium source in sichuan shale gas

Ancient basement rocks can usually serve as high quality helium source rocks. The Hugoton-Panhandle gas field in the United States and the WY gas field in Sichuan have both confirmed that the ancient granitic basement rocks are potential helium source (Brown, 2019; Qin et al., 2022). A number of helium-rich fields are also related to metamorphic basement, such as the Qingyang field in the Ordos Basin (Fan et al., 2023; Wei et al., 2023).

Based on gravity and magnetic anomalies results, Zhou (2016) suggested that the basement rocks of the Sichuan Basin are mainly composed of acidic rocks (mainly granite), metamorphic rocks, intermediate rocks, and basic rocks (Fig. 4). The contents of U and Th vary in different types of basement rocks. According to Meng et al. (2021)'s measurements, average U content is decreasing in the order of acidic rocks, intermediate rocks, basic rocks, ultrabasic rocks and metamorphic rocks, while average Th content is decreasing in the order of metamorphic rocks, acidic rocks, basic rocks, intermediate rocks, and ultrabasic rocks (Fig. 5, Meng et al., 2021). The helium generation rates (F_{He}) can be roughly estimated

Table 2
A list of parameters used in the calculation of amount of helium generated in the Wufeng- Longmaxi shale in typical shale gas plays in the Sichuan Basin.

Blocks	Depth, m	Gas-in-place content, m^3/t	U, ug/g	Th, ug/g	Flux of ^4He, × 10^{-12} $cm^3 \cdot g^{-1} \cdot yr^{-1}$	Time of deposition, Ma	Total ^4He, × 10^{-4} m^3/t	Calculated He concentration, ppmv	Ref.
Weiyuan	2000–3500	1.9–6.0 (4.2)	2.68–60.1 (15.7)	1.02–23.6 (11.2)	2.22	440	9.75	232	Gao et al. (2019); Zou et al. (2016)
Changing	2300–3200	2.4–5.5 (3.4)	4.34–56.9 (16.5)	8.62–46.3 (19.3)	2.55	440	11.2	329	Zou et al. (2016)
Luzhou	3500–4500	5.0–7.5 (6.8)	1.69–23.0 (11.8)	5.57–36.5 (17.4)	1.92	440	8.46	124	Yang et al. (2019)
Dazu	3500–4500	2.7–7.0 (4.1)	4.23–76.6 (15.9)	4.40–32.8 (15.3)	2.36	440	10.4	253	Zhang et al. (2019)
Fuling	2100–3500	4.7–7.2 (5.5)	4.7–54.2 (13.52)	8.73–25.1 (17.76)	2.14	440	9.42	171	Gan et al. (2018); Zou et al. (2016)
Pengshui	2000–3500	1.2–4.3 (2.0)	12.7–45.4 (19.25)	8.5–16.6 (12.94)	2.69	440	11.9	593	Wang et al.(2022); He et al. (2017)

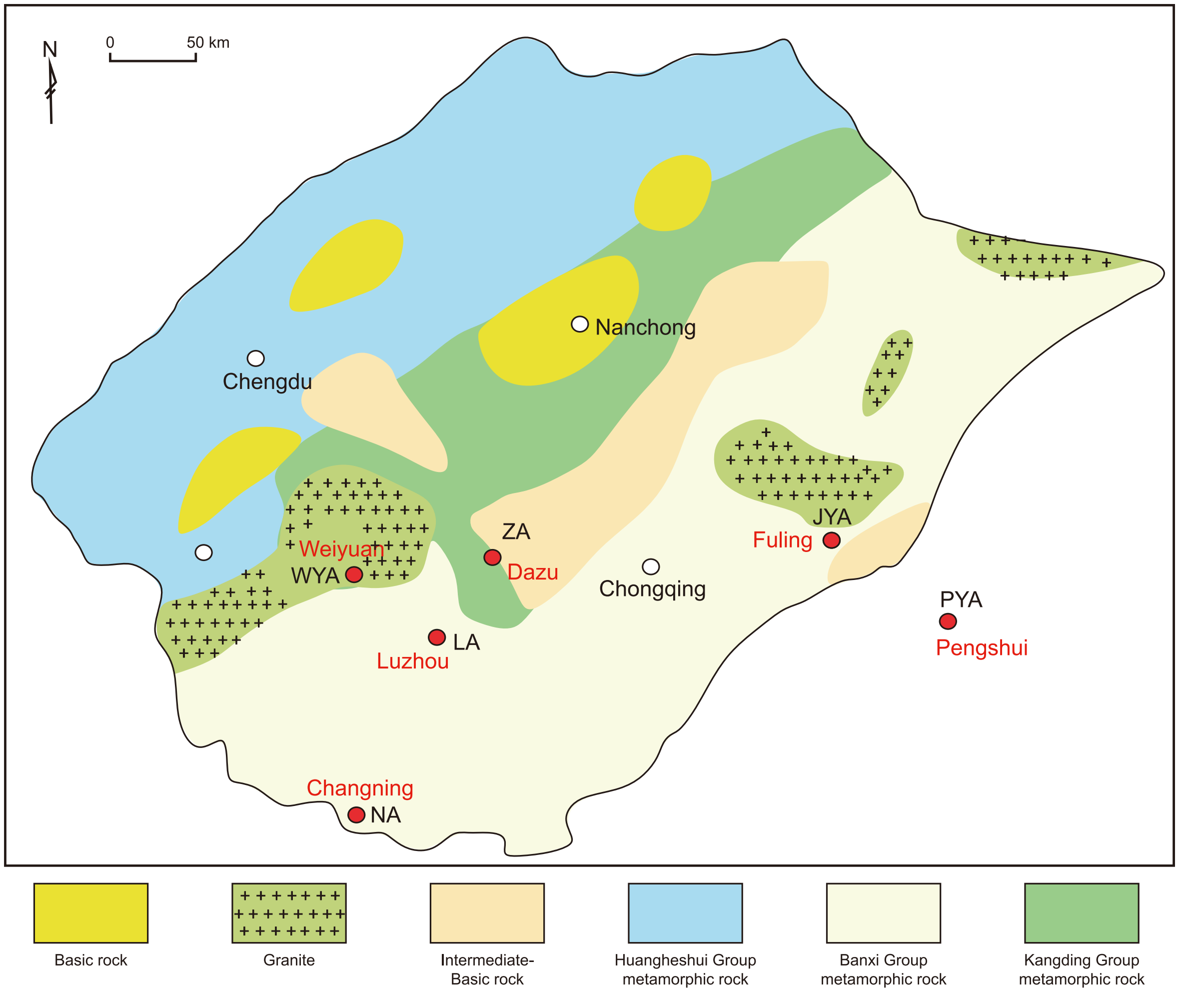


Fig. 4. Distribution of basement rock types in the Sichuan Basin (modified from Zhou, 2016).

by Eq. (1) using the U and Th contents of each type of rocks. It turns out that acidic rocks, intermediate rock, and metamorphic rocks are good helium source rocks (Fig. 5).

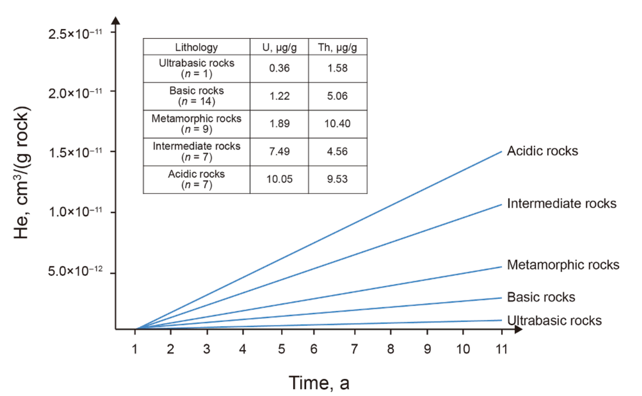
4.4. The controlling factors on the enrichment of helium in sichuan shale gas

Based on detailed investigation of typical helium-rich shale gas wells, three major factors are proposed to be vital for the enrichment of external helium: (1) sufficient helium source, (2) effective migration pathways, and (3) diminished dilution effect of shale gas resulting in relatively high helium concentrations.

4.4.1. Sufficient helium source

High quality helium source rock and sufficient helium replenishment are indispensable to form a helium rich gas field. The basement rocks of the LZ, DZ, CN, FL, and PS blocks are mainly metamorphic rocks, and of WY and a few blocks in northeastern Sichuan are granite (Fig. 4). The helium generation potential of granites are almost two times higher than the metamorphic rocks (Fig. 5). The strong helium generation capacity of the WY block basement enable sufficient helium source to the WY conventional gas filed, which is the first helium-extraction gas field in China with helium concentration up to 0.2–0.3 vol% (2000–3000 ppmv).

The relatively high helium concentration in W201-H1 well is probably also attributed to the strong helium influx from the



Lithology	U, μg/g	Th, μg/g
Ultrabasic rocks (n = 1)	0.36	1.58
Basic rocks (n = 14)	1.22	5.06
Metamorphic rocks (n = 9)	1.89	10.40
Intermediate rocks (n = 7)	7.49	4.56
Acidic rocks (n = 7)	10.05	9.53

Fig. 5. Helium generation rates of different types of rocks in the southeastern upper Yangtze region. The inset shows the uranium and thorium contents of ultra-basic rocks, basic rocks, metamorphic rock, intermediate rock and acidic rocks (modified from Meng et al., 2021).

underlying ancient granite. W201-H1 shale gas contains more than double of the helium content as in the neighboring shale gas wells. Two lines of evidence suggest the abnormally high helium contents is at least partially attributed to external helium charging. Firstly, the relatively high $^{40}Ar/^{36}Ar$ ratios in the W201-H1 well (Fig. 6(a)) indicate that the helium was generated from rocks rich

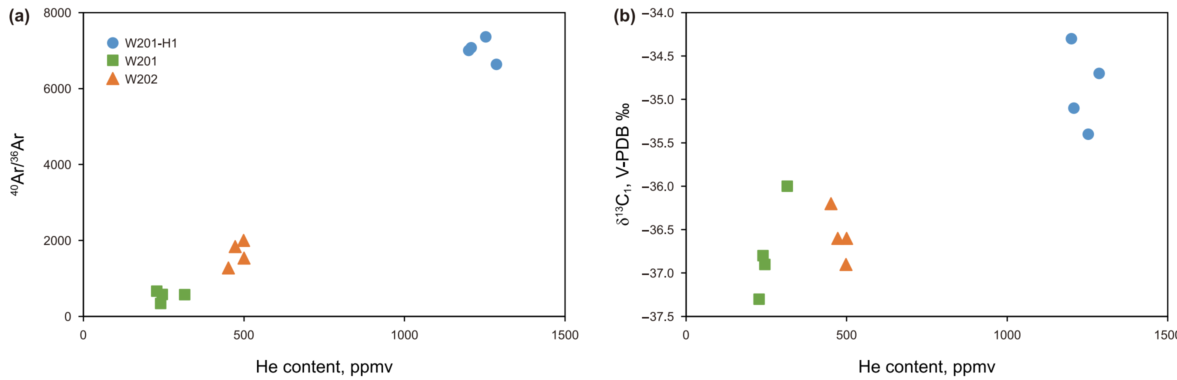


Fig. 6. Relationships between helium and $^{40}Ar/^{36}Ar$ ratios (**a**) and between helium and $\delta^{13}C_1$ (**b**) in the shale gas and conventional gas in Weiyuan in the Sichuan Basin (Data from Cao et al., 2018).

in potassium (K) from which the ^{40}Ar was decayed. The high K content (average 5.23%) in the WY granite (Liu et al., 2022) indirectly supports the point that the granite is the source of helium. Secondly, the $\delta^{13}C_1$ values of W201-H1 shale gas are much heavier than the neighboring wells (Fig. 6(b)), indicating helium partially comes from dissolved gas in water, similar to those in the WY conventional gas field (Qin et al., 2016). Helium generated over millions of years in the granite was dissolved in the pore water, and then, induced by the Himalayan tectonic uplift, transported to the reservoirs along with the underground fluids (formation water, CO_2, or supercritical fluids), and finally released into the gas field because of the decrease in pressure and temperature.

Another example demonstrating the significance of external helium is the YY-1 well. Luo et al. (2019) sampled the Cambrian Shuijingtuo shale gas at YY-1 well four times in a year, and helium content between was measured to be 0.010–0.31 vol% (100–3100 ppmv), with an average of 0.16 vol% (1600 ppmv), making the YY-1 the shale gas well with the highest helium concentration. YY-1 area is directly developed on top of the Huangling granite uplift (Luo et al., 2019). The old Huangling granite is featuring of relatively high U and Th contents. After millions to hundreds of millions of years of helium generation, considerable amount of helium were produced in the granite. Tectonic movements initiated or reactivated deep faults, connecting the underlying granite, facilitating the release of generated helium, bridging helium to the overlying shale reservoirs. Therefore, the ancient and large-scale Huangling granite body provides an exceptionally abundant external source of helium in the Yichang area.

4.4.2. *Effective migration system*

An effective conductive system connecting source and reservoir is the bridge for exogenous helium to enter natural gas reservoirs, and is essential for the contribution of external helium to the system. An effective conductive system includes two major components: migration pathways and migration carriers.

4.4.2.1. Migration pathways. Large faults bridging the U/Th-rich basement and shale reservoirs can serve as favorable conduits of the helium to shale. Overall, the faults are more developed in the peripheral areas than in the Sichuan Basin due to more intense tectonic activities. The peripheral areas of the Sichuan Basin are mainly located in the compartmentalized deformation zone and the trough transition zone, and there are many faults with large scale and great extension lengths. In contrast, the intensity of tectonic modification in the areas in south and east Sichuan is relatively weak.

Faults in southeastern Sichuan Basin can be further classified into four levels based on their development magnitude (Cai et al., 2024, Table 3). Among them, I- and II-order faults are more effective in terms of connecting basement helium source rocks. In LZ, WY and FL-JSB areas, the uplift of the strata is short in duration and small in magnitude, and the faults are mainly of the II- and III-order (Cai et al., 2024), while the CN and DZ areas has been subject to relatively strong modification, with I- and III- order faults developing at the junctions of tectonic units. The better developed I- and II-order faults in the DZ area probably lead to higher helium concentrations in the DZ area than in the LZ area.

In addition to major faults, the shale reservoir has developed various types of natural fractures under the conditions of multi-stage superimposed tectonic compression. Natural fractures, as important storage spaces and seepage pathways for gases, provide an additional migration pathway for the enrichment of helium.

4.4.2.2. Migration carriers. Because of the low intensity and quantity of gas generation, the migration of helium generally needs to be carried with underground fluids. Basinal fluids can capture free helium along the migration paths and transport it all the way to the gas reservoir. Almost all the helium in the underground fluids can be released into the gas reservoir and accumulate in the gas reservoir because helium has very low Henry's coefficient and thus ready to be displaced by other gases in the gas reservoir (such as CH_4, N_2, CO_2, etc.) (Brown, 2010).

N_2 is an effective carrier for helium and is usually directly related to its enrichment (Cheng et al., 2023). Due to similar Henry's constants of helium and N_2, they tend to enrich simultaneously in a gas field. That's why the concentrations of helium are

Table 3
Faults of different orders and their characteristics (modified from Cai et al., 2024).

Order	Characteristics
I	regional major faults with a typical displacement of 300 m
II	faults penetrating the reservoir and cap and base rocks with a typical displacement of 100–300 m
III	faults penetrating the reservoir but not cap and base rocks with a typical displacement of <100 m
IV	faults not penetrate the reservoir

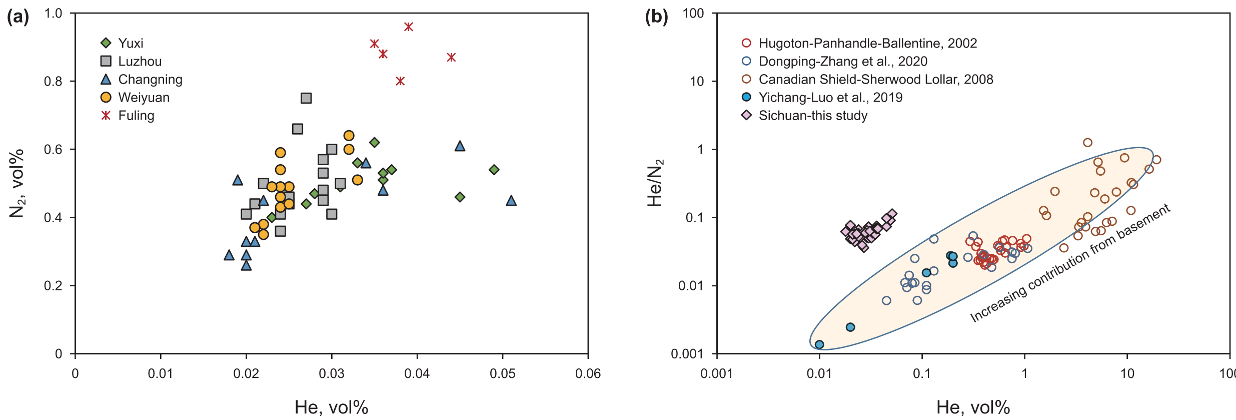


Fig. 7. The relationships between helium and N_2 in the Sichuan Shale gas (**a**) and helium and He/N_2 in typical shale gas (**b**). The data of shale gas in the Sichuan shale are from the present studies.

commonly found to be positively correlated with that of N_2 in many conventional natural gas. For example, a significant positive relationship between helium and N_2 concentration was observed in the Hugoton-Panhandle gas field in the US (Ballentine and Sherwood Lollar, 2002).

In this study, the correlation between helium and N_2 concentrations were plotted to investigate the migration mechanism of helium in shale gas. The correlation between helium and N_2 was found to be weak for shale gas in the Sichuan Basin (Fig. 7(a)). This is probably because the source and migration mechanism of helium and N_2 are different in shale gas from those in conventional gas fields. By plotting helium concentration versus He/N_2 ratio (Fig. 7(b)), gas fields showing positive helium and N_2 contents relationship fall in a linear line, with the Wufeng-Longmaxi shale gas occurring as an outlier. The $\delta^{15}N$ isotope data (−10.5‰ and −0.8‰) confirms that the N_2 in shale gas in south Sichuan was mainly generated in-situ and derived from the thermal ammonification of organic matter. The weak correlation between helium and N_2 indirectly suggest that they don't share common source rocks nor migration pathways.

4.4.3. Moderate tectonic activities resulting diminished dilution effects of shale gas

4.4.3.1. Dilution effects of shale gas. A distinction between unconventional petroleum system and conventional system regarding on helium enrichment is that large amounts of natural gas generated from source rocks are retained in-situ which can heavily dilute the concentration of helium, preventing the formation of a helium-rich gas field.

A simple calculation here can demonstrate the dilution effects of shale gas on helium enrichment. Assuming that the TOC of the Longmaxi Formation shale is 1 wt%, the maximum gas generation potential is 500 m^3/t (Ma et al., 2020), and the gas generation conversion rate is taken as 80%, then the gas generation intensity per unit mass of this set of shale is approximately 4.0 m^3/t according to Eq. (3). The helium generation intensity of shale per unit mass is calculated by Eqs. (1)–(3), and is approximately 0.001 m^3/t. The generation intensity of natural gas is about 4000 times as high as that of helium.

The dilution effects can be clearly demonstrated by the relationship between pressure coefficient and helium contents. Pressure coefficient is a good proxy for shale gas content, with higher pressure coefficient indicating better preservation and higher gas content (Nie et al., 2019; Yang et al., 2021). The isochore map of pressure coefficient for the Wufeng-Longmaxi Formation in the Sichuan area was drawn based on drilling data (Fig. 8). As can be seen, overpressure is widely developed in the Sichuan Basin, with the highest pressure coefficient reaching up to 2.25 in the southeastern Sichuan, and gradually decreases towards the southern and eastern basin margin, and the western Wufeng-Longmaxi shale line directions. The pressure coefficient in the basin's the peripheral areas is generally less than 1.0, indicating overall moderate preservation conditions and relatively low shale gas content.

Helium concentration in the areas with high and ultrahigh pressure coefficients are usually lower than that in the areas with lower pressure coefficients (Fig. 8). Specifically, helium contents in the LZ area with high pressure coefficient (>2) is generally lower than that in the DZ area with lower pressure coefficient (~1.5). These two blocks share similar basement rock lithology and regional tectonic background. Helium contents in the PS area (pressure coefficient ~1) and W201-H1 well (pressure coefficient ~0.9) almost double that in the southeastern Basin with pressure coefficient >1.2.

The higher pressure coefficient, the higher shale gas contents and the greater dilution effect, leading to the lower helium contents. Areas with great shale gas yields usually contain low helium concentration and is not advantageous for helium exploration. Therefore, moderate tectonic activities adjusting shale gas and helium relative contents, is beneficial for the enrichment of helium in unconventional system.

4.4.3.2. Tectonic activities adjusting the relative contents of shale gas and helium. The shale gas content in the Sichuan Basin is largely affected tectonics activities, and is closely related with the degree of uplift and the timing uplift (Fig. 9(a)). Tectonic uplift and erosion lead to a reduction in the thickness of the rock layers above the shale gas section, or even expose the shale to the surface. Under the influence of tectonic stress and pore fluid pressure, closed fractures reopen, causing the shale gas to seep and dissipate.

The intensity and scale of tectonic activities in the Sichuan Basin vary spatially. In general, the southeastern Sichuan region, located in the tectonically stable area west of the basin boundary fault (Qi Yue Mountain Fault), generally has a higher shale gas content. In contrast, the eastern periphery areas experiences relatively more intense tectonic activity, has a relatively lower shale gas content.

The shale gas content and helium concentration are also largely affected by the timing of uplift (Fig. 9). The earlier uplift leads to earlier end of shale gas generation and destruction of preservation,

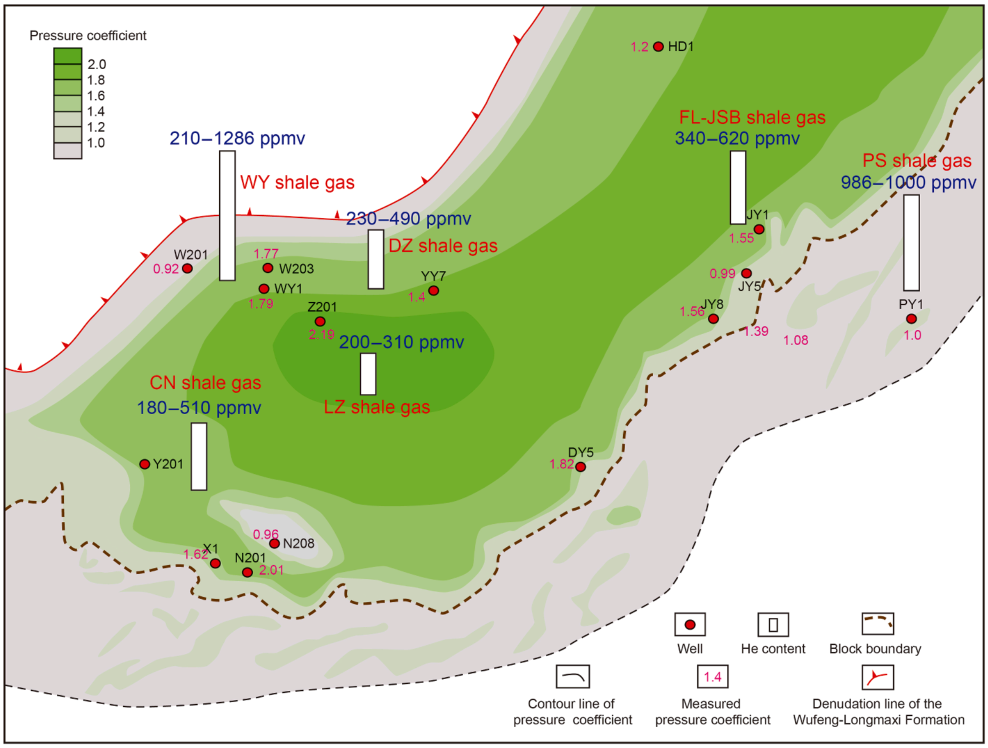


Fig. 8. The relationship between He and pressure coefficient (modified from Wei, 2015)

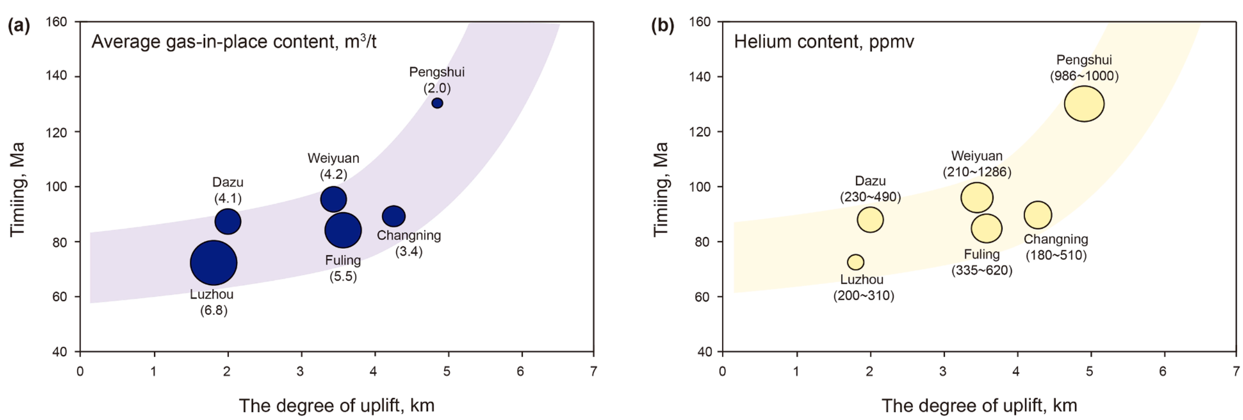


Fig. 9. The relationships between the timing and degree of uplift with shale gas contents (**a**) and helium concentrations (**b**).

resulting in a stronger reduction in the gas content of the shale. Affected by the Yanshan-Himalayan orogeny, the tectonic uplift started earlier the western Hunan-Hubei area (about 165 Ma ago) than the basin area (about 80 Ma ago). The FL-JSB and PS blocks have experienced earlier uplift and a long period of shale gas loss. In contrast, the LZ, DZ, WY, and CN areas have experienced later uplift, and the overpressure state resulting a high shale gas content and meantime leads to the low helium content. It is noteworthy that moderate tectonic activities causing shale gas (and helium) loss and connecting external helium source and shale reservoirs is significant for the enrichment of helium in shale gas. However, too strong tectonic activities severely damage preservation is not favorable for helium enrichment since helium can escape with shale gas.

4.4.4. Sealing and preservation

The effects of preservation on the enrichment of helium in gas fields are still controversial. The smaller size of helium compared with CH_4 enables the greater diffusionality of helium. Therefore, a lot of researchers argued that the conservation of helium does require more effective seals than CH_4. In contrast, another opinion insists that the enrichment of helium doesn't require better seals since the general trace helium content in the gas field results in low partial pressure and slow diffusion.

To clarify this issue, it is necessary to first understand the migration mechanism of helium in different physical property reservoirs (especially low-permeability reservoirs). Through He-CH_4 binary gas mixture seepage experiments, Sun et al. (2023) found that the CH_4/He volume ratio dropped from an initial value of 7:1 to about 1:1 after seepage through a shale plug (Fig. 10(a)). In comparison, the CH_4/He volume ratio remained similar in tight

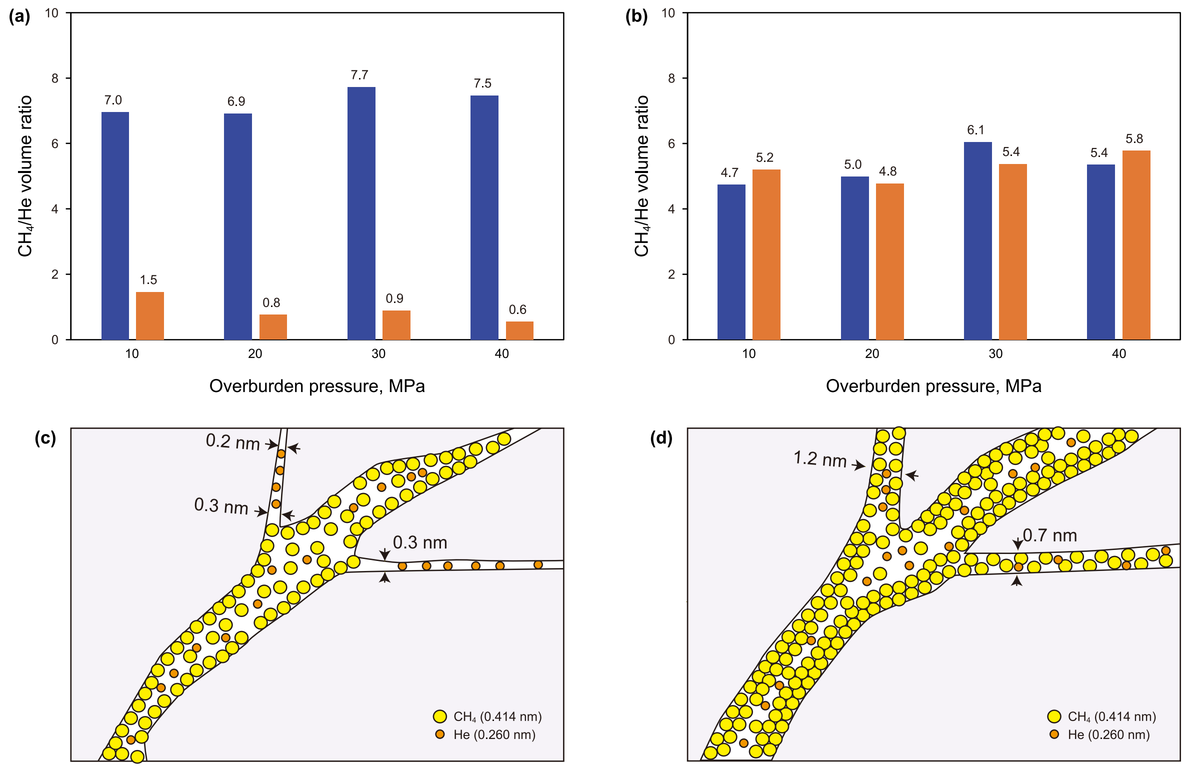


Fig. 10. Comparison of CH_4/He content ratio before and after shale plug permeation (**a**) and tight sandstone plug permeation (**b**). Schematic drawings showing fractionation of CH_4 and helium in shale (**c**) but not in tight sandstone (**d**). Note that helium molecule could transport through a pore with diameter less than 0.4 nm where CH_4 might be trapped.

sandstones with higher permeability and larger pores (Fig. 10(b)). In other words, the abundant of micro- and nano-pores in the low permeable shale carried out physical fractionation to separate helium from CH_4. Helium has a migration advantage due to its extremely high Knudsen number in the migration through micro- and nano-channels. Thus, the micro- and nano-pores in the shale can seal methane, but cannot effectively seal helium (Fig. 10(c) and (d)). Helium has higher requirements for the effectiveness of the trap, seal, and preservation conditions than natural gas reservoirs.

A helium-rich shale gas play definitely needs good sealing and preservation otherwise helium will dissipate rapidly. However, when considering the effects of preservation for helium enrichment in shale gas, it is wise to evaluate the dilution effects of shale gas meanwhile. Better preservation leading to greater helium amounts can also results in higher shale gas content which is not good for helium enrichment.

4.5. The enrichment of helium in sichuan shale gas

4.5.1. Dynamic coupling of controlling factors

The enrichment of helium shale gas is usually attributed to favorable dynamic coupling of aforementioned controlling factors. Take the Y1 well in the Tiangongtang structure as an example. Our results showed that Y1 shale gas contains the highest helium content of 0.051 vol% among our CN samples. The Tiangongtang structure has experienced multiple phases of subsidence, deep burial, and uplift modification (Zhang et al., 2022). Liquid hydrocarbons were generated in the Wufeng -Longmaxi Formation since the end of the Silurian, which were thermally cracked in to gases since the Early Jurassic because of increasing burial depth and temperature. During the Early Cretaceous to the Late Cretaceous, increasing amount of gas hydrocarbon was generated because of thermal cracking of kerogen, forming the initial overpressure shale gas play. The strata uplifted rapidly since the Cenozoic, reactivating the main fault of the Tiangongtang structure, Fault No. 1, and leading to the loss of formally generated shale gas. The leakage of shale gas lead to the gradual adjustment of the overpressure shale gas play to a moderate-pressure shale gas play (Fig. 11).

Based on the burial history and hydrocarbon generation history studies, a semi-quantitative evolution chart of shale gas content over time can be made (Fig. 11). The shale gas content reached its peak at the maximum burial depth and then gradually dissipated due to uplift-induced fracturing. Unlike the generation of shale gas, which need to meet a certain threshold temperature for the source rock to generate hydrocarbons, sedimentary rocks can generate helium radioactively since the time of deposition, and this is positively correlated with the depositional age. Until the early Paleogene strata were uplifted, the completely closed system was broken, and helium, with a smaller molecular diameter and higher diffusion rate, dissipated from the cracks at a greater rate. However, at the same time, the reactivated Fault No. 1 connected the base helium source, and a large amount of exogenous helium generated from the base, using the fault as a conduit, continuously moved upward along the direction of the overlying strata, driven by buoyancy, and converged at the higher positions. The continuously inflow of exogenous helium, on top of compensating for the dissipated endogenous helium, resulted in an increase rather than a decrease in the total absolute helium content (Fig. 11). In summary, the amount of shale gas decreases, and the total amount of helium increases, ultimately leading to an increase in the relative content of helium in the shale gas, forming a helium-rich shale gas reservoir.

4.5.2. Tectonic movement is a primary driver

Above discussion has mentioned from time to time that the effects of tectonic movement on helium migration and shale gas

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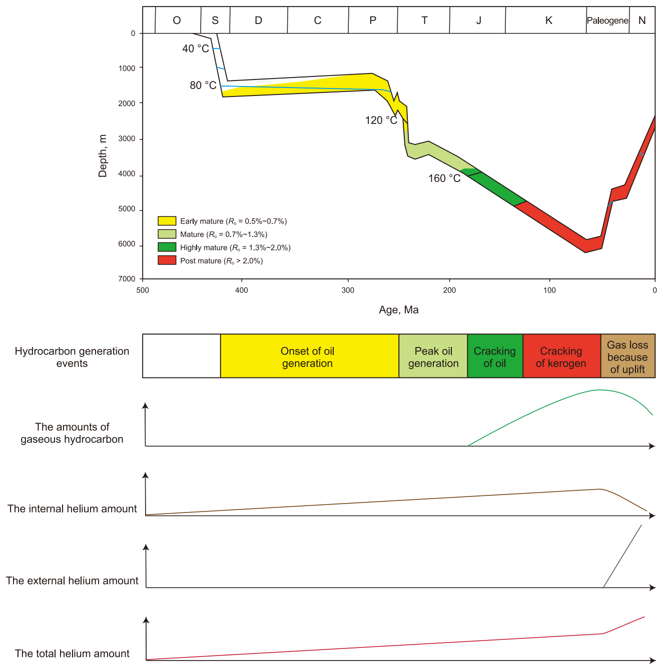

Fig. 11. Evolution of shale gas and helium content with burial history in Changning area.

content. Here we want to stress that the tectonic movement can influence each controlling factor of helium enrichment at varying degrees, and is a major driver initiating the conducive dynamic coupling of these controlling factors.

Unlike conventional natural gas reservoirs, shale gas reservoir systems are generally more self-contained, with generated hydrocarbons largely accumulating in-situ in the shale system. Meanwhile, external helium sources cannot easily replenish the shale systems. Therefore, a prerequisite for helium enrichment in shale gas is that the sealed, self-contained system has once been moderately damaged, leading to improved system openness and enhanced external helium charging.

The weakening of the sealing is often caused by moderate tectonic activities, forming effective migration system bridging external sources and shale reservoir. Faults are the result of stress release from tectonic movements, and they are usually accompanied by the development of fractures systems. The development of faults and fractures creating high permeable migration pathways, not only allowing helium migration upwards to shale, but also promoting shale gas to migrate towards the faults through seepage and diminish the dilution effect of shale gas. Therefore, moderate development of faults and fractures is advantageous for helium enrichment. However, excessively intense tectonic activities might cause severe preservation destruction, preventing helium enrichment.

Spatially, faults and fractures are more developed in the peripheral areas than in the tectonically stable Sichuan Basin due to more intense tectonic activities. The peripheral areas of the Sichuan Basin are mainly located in the compartmentalized deformation zone, and there are abundant faults with large scale and great extension lengths. The intensity of tectonic modification in the areas in South and East Sichuan is whereas relatively weak. Specifically, in LZ area, the main depth of burial is greater than 3500 m, and the uplift of the strata is short in duration and small in magnitude, which leads to a smaller scale of fault development in this area (Cai et al., 2024). Therefore, the preservation conditions are overall excellent, resulting relatively high shale gas content and low helium enrichment.

In addition, tectonic compression activities can also facilitate the release of helium that is generated and stored in the mineral lattice (Roques et al., 2020). The helium contents were 240–500 ppmv in the shale gas of Wei 201 and Wei 202 wells that were not horizontal fractured, while climbed to 973–1375 ppmv in the shale gas of Wei 201-H1 and Wei 201-H3 wells which were subject to horizontal fracturing. The large difference in helium content probably suggest that hydraulic fracturing (can be treated an artificial tectonic activity) significantly promotes the release of helium from rocks (Lin et al., 2024). The high helium content (1.877%) found in the Precambrian granite fracture gas in the late 1980s (Xu et al., 1989), also indicate that fractures are conducive to the large-scale release of helium and argon from granite.

A typical enrichment pattern of helium rich shale gas play can be illustrated as in Fig. 12.

4.5.3. Balances important for helium enrichment in shale gas

Helium enrichment requires a balance between system openness and preservation. Pressure coefficient is a straightforward proxy to measure the preservation conditions and openness of

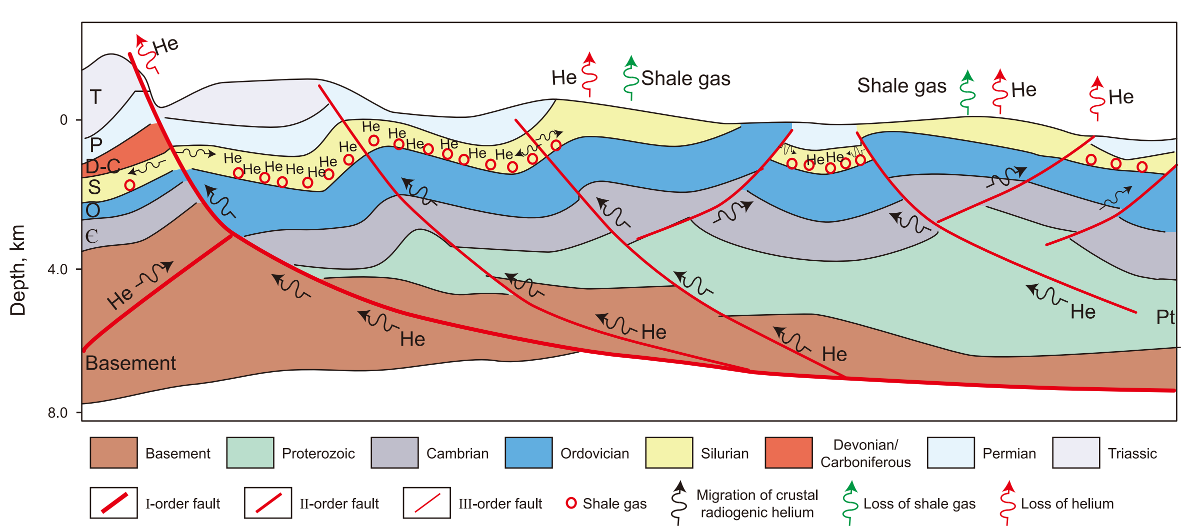

Fig. 12. A schematic diagram of the enrichment pattern of helium in Sichuan shale gas.

shale gas. Either underpressure or overpressure is not conducive to helium enrichment, with the former results in too much helium loss while the latter leading to overly strong dilution effect and too little external helium supplementation. Therefore, shale gas wells with a moderate pressure coefficient might be more favorable for helium enrichment.

In addition, a balance between the generation of shale gas and helium is also crucial for helium enrichment. Source rocks begin to generate a large amount of hydrocarbons when they reach a certain depth and temperature. If the strata continue to be buried without tectonic uplift or other disruptive effects, generated hydrocarbons will accumulate in large quantities in place. The generation of helium, on the other hand, is a continuous, linear, and stable decay process of radioactive elements, lack of peak generation rate. Its production rate depends only on the abundance of U and Th elements. Therefore, taking advantage of the contrast in the generation mechanisms of shale gas and helium, periods before large-scale generation of shale gas or after its significant loss are favorable timings for helium enrichment.

5. Conclusions

In this study, the concentrations and isotopic characteristics of helium were investigated in the Wufeng-Longmaxi shale gas in the Sichuan Basin and the periphery areas. The following conclusions can be reached:

(1) Based on the analysis data of this study, the concentrations of helium in the shale gas collected from Weiyuan, Changning, Luzhou and Dazu areas fall in the range of 0.018–0.051 vol% (180–510 ppmv). Generally, helium is more enriched in the eastern periphery areas than in the eastern and southern Sichuan Basin.
(2) The relatively low ratios of ^{3}He and ^{4}He in shale gas indicate that most of the helium is crustal derived helium. Further quantitative estimate based on helium, neon, and argon isotopic ratios suggest almost 100% crustal helium source.
(3) External helium source is required to meet the threshold of an economic helium-rich field, except for specific areas with extraordinarily high U and Th concentration and low shale gas content.
(4) Major controlling factors of a helium-rich field include sufficient helium source, effective migration pathways, and tectonic activities adjusting the relative contents of shale gas and helium. In general, shale gas plays with underlain ancient cratonic basement, well developed source-connecting faults, and moderate pressure coefficient are potential targets for helium exploration.

Although the overall content of helium in shale gas is not high, the reserves of helium are considerable because of the large reserves of shale gas. With the development and innovation in helium extraction technology, shale gas with sub-economic level of helium is expected to become a promising field in the exploration and development of helium resources.

CRediT authorship contribution statement

Yan-Yan Chen: Writing – review & editing, Validation, Investigation. **Shi-Zhen Tao:** Resources, Investigation, Data curation. **Wei Wu:** Writing – original draft, Data curation. **Xiang-Bai Liu:** Writing – review & editing, Investigation. **Cheng-Peng Song:** Writing – review & editing, Validation. **Zuo-Dong Liu:** Writing – review & editing, Writing – original draft, Formal analysis, Data curation, Conceptualization. **Qing-Yao Liu:** Writing – review & editing, Investigation. **Lin Wei:** Zhengjun He, Writing – review & editing, Validation, Zhaoming Wang, Writing – review & editing, Investigation. **Jian-Rong Gao:** Writing – review & editing, Investigation. **Yue Chen:** Writing – review & editing, Investigation.

Declaration of competing interest

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

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