Noble gas constraints on hydrocarbon accumulation and groundwater flow in the central area of Western Sichuan Basin

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Abstract The noble gas concentrations and isotope ratios of seven natural gas samples from the central area of the Western Sichuan Basin were measured. The samples all have $^{40}Ar/^{36}Ar$ ratios greater than the atmospheric values, and the $^{3}He/^{4}He$ ratios (R/R_a) are entirely consistent with the crustal radiogenic He values. The vertical variation of the calculated $CH_4/^{36}Ar$ ratios with depth clearly indicates that the CH_4 and ^{36}Ar are intimately associated, indicating a common reservoir intermediate to the sampled reservoirs, where they are well mixed and stored together prior to entrapment into gas reservoirs. Meanwhile, the calculated $CH_4/^{36}Ar$ ratios range between 8×10^6 and 64×10^6 very much greater than the $CH_4/^{36}Ar$ values for pure water and 5 mol/L NaCl brine at low temperature and hydrostatic conditions, reflecting the presence of "excess" thermogenic CH_4 over that supplied by a CH_4-saturated groundwater at low temperature, and the excess CH_4 saturation and dissolution to be at depth greater than the sampled reservoirs. This conclusion is consistent with the $\delta^{13}C(CH_4)$ and $\delta^{13}C(C_2H_6)$ values. In addition, the $^{4}He/^{36}Ar$ ratio is correlated with depth, showing that the crustal radiogenic ^{4}He are well mixed with the atomosphere-derived ^{36}Ar before introducing into gas reservoirs. The $^{4}He/^{36}Ar$ ratio vertical variation with depth can be attributed to the preferential transport of ^{4}He relative to ^{36}Ar in fluxes from lower aquifers through water-filled pores into the upper one. The increasing $^{4}He/^{40}Ar^{*}$ ratios with decreasing depth, from 1.3 times to 29 times greater than the crustal production ratio, are also assumed to be the results of preferential transport ^{4}He relative to $^{40}Ar^{*}$ from the production site into groundwater.

Keywords: noble gas isotopes, groundwater flow, hydrocarbon migration, Western Sichuan Basin.

1 Introduction

In recent years noble gases have been extensively used as natural tracers to study sources, water-rock interactions, migration, and mixing of basin fluids. The noble gas species in basin fluids can originate from the atmosphere-equilibrated groundwater, and any flux into basin traps. Therefore, they are greatly suitable for a complementary tool to study the role of groundwater on transporting hydrocarbon[1,2]. However, so far these properties of noble gas have been little exploited in the central area of Western Sichuan Basin, and the fluid-related phenomena of gas accumulation are still poorly understood. The purpose of the present paper is to present helium and argon isotope abundance in hydrocarbon-rich reservoirs of the area, and discusses the degree of interaction between hydrocarbon and groundwater. Such studies would impose further constraints in the understanding of the phenomena of hydrocarbon accumulation in the area.

2 Regional geology

The Western Sichuan Basin is located along the western margin of the Sichuan Basin, and is a NE-SW elongate foreland basin superimposed up on the Paleozoic basement during the Meso-Cenozoic tectonic episodes. The central area of the foreland basin is bounded on the west by the Longmenshan Thrust-Nape Belt and on the east by the Longquanshan Fracture Zone, covering approximately 9000 km^2. The area began to subside during the late episode of Indosinian Orogeny, and accompanied to accumulate 3000 — 4000 m thick of Upper Triassic marine-continental organic-rich, classic and pelitic sediments with coal beds, as well as continued to be filled with interbeded fluvio-lacustrine red sandstone and mudstone up to 2500 m thick in the Lower Jurassic through Lower Cretaceous time. These strata and the basement were subjected to intense disturbance during the Yanshan and Himalayashan Movements, resulting in regional erosion surfaces and well-developed fissures swarms, and were overly by the younger Quaternary sediments[3].

3 Hydrologic setting

The marine-continent alternating and fluvio-lacustrine sedimentions produced the multilayered aquifer system. The Upper Triassic and Jurassic formation waters of the system are Na-Cl-Ca types, and the salinity derives from dissolution of evaporites in the Upper Triassic and Lower Jurassic strata. The Upper Triassic brine has a high and variable salinity, ranging from 46 g/L up to 180 g/L. Generally speaking, the aquifers are recharged by meteoric water at their outcrops in the west and southwest and by Triassic leaching water through vertical faulting, and the flow occurs toward the east and northeast. Because of the presence of the brine leakage from the bottom to top, the recharged freshwater (< 0.4 g/L) soon becomes saturated with salt. The hydrodynamic component flowing with gas migration seems to have affected the distribution of gas pools in the area. The main gas fields in the area are found in the catchment regimes at Xiaoquan, Xinchang and Hexinchang[4].

4 Hydrocarbon characteristics

Seven samples were collected from the Xiaoquan, Xinchang and Hexinchang gas fields, taking at depths from 650 m to 4 566 m. A detailed study of the total gas composition and carbon isotopic ratios of all samples have been given elsewhere[5]. Briefly, the samples are dominated by hydrocarbons with content of >99% by volume and wetness >0.5%, their methane $\delta^{13}C(CH_4)$ and ethane $\delta^{13}C(C_2H_6)$ values are > −37‰ and > −27‰, respectively, the data indicate that the hydrocarbons in these gas fields are thermogenic; in addition, their isotopic and compositional variations with depths clearly show that the Xiaoquan and Hexinchang hydrocarbons are in situ accumulation in the Upper Triassic strata, while the Xinchang Jurassic hydrocarbons are derived from the deeply buried and mature source rocks of Upper Triassic age and have experienced a long distance migration.

5 Noble gas data

The compositional and isotopic data of noble gases are summarized in Table 1. The samples all have $^{40}Ar/^{36}Ar$ ratios greater than the atmospheric values (295.5). This indicates the ^{40}Ar component of the samples to be a simple mixture of atmosphere-derived ^{40}Ar and a resolvable radiogenic component $^{40}Ar^{*}$ (in subsequent discussions of argon, isotope an * will denote radiogenic origin), and reveals the atmosphere-derived ^{40}Ar reaching 40% to 80% of the total ^{40}Ar in the samples. The ^{36}Ar in the samples can be considered to be entirely atmosphere-derived and its calculated relative abundance is also reported in Table 1. In addition, the measured $^{3}He/^{4}He$ ratios (R) for the samples are given as the percent deviation from the atmospheric $^{3}He/^{4}He$ ratio (Ra = 1.40×10^{-6}), and shown R/Ra values range from 0.014 to 0.05, as well as reflected R/Ra to be entirely consistent with the crustal radiogenic production ratios of R/Ra (0.01—0.10)[6]. The helium within the samples can therefore be considered to be entirely crust radiogenic in origin.

The calculated $CH_4/^{36}Ar$ and $^{4}He/^{36}Ar$ ratios for thermogenic CH_4 and radiogenic ^{4}He to atmosphere-derived ^{36}Ar, and $^{4}He/^{40}Ar^{*}$ ratio for radiogenic ^{4}He and $^{40}Ar^{*}$ in the samples have been estimated and are also given in Table 1. These calculated ratios of each sample are shown as functions of depth (Fig. 1(a), (b), (c)).

Table 1 Noble gas concentrations and isotope ratios in the central area of the Western Sichuan Basin

Well	Age^a)	Depth/m	$^{4}He/10^{-6}$	$^{40}Ar/10^{-6}$	$^{36}Ar/10^{-6}$	$^{40}Ar/^{36}Ar$	R/R_a	$CH_4/^{36}Ar$ ($\times10^{6}$)	$^{4}He/^{36}Ar$	$^{4}He/^{40}Ar^{*}$
Xinchang										
CX136	J(U)	650	171.8	5.8	0.0156	372	0.02	63.40	11013	143.9
CX134	J(U)	1710	269.6	26.9	0.0364	739	0.01	26.80	7407	16.70
CX129	J(M)	2356	77.2	12.2	0.0224	545	0.01	43.83	3446	13.83
CX152	J(M)	2730	101.2	21.6	0.0465	465	0.02	21.06	2176	12.84
CX135	J(L)	2752	214.8	42.9	0.0635	676	0.02	15.15	3383	8.887
Xiaoquan										
CX96	T(U)	2628	161.4	49.8	0.1112	448	0.05	8.75	1452	9.485
Hexinchang										
CH127	T(U)	4566	82.1	30.9	0.0628	492	0.01	15.70	1307	6.656

a) J = Jurassic; T= Triassic; U = Upper; M = Middle; L = Lower.

6 Discussion

6.1 CH_4 and atmospheric noble gases in groundwater

Based on the He, Ar isotopic and compositional analyses in the previous section, the atmosphere–derived noble gases are clearly known to exist in each natural gas sample. The atmospheric noble gases are believed to have originated from groundwater that equilibrated previously with atmosphere [1,2,7]. Furthermore, the vertical variation of all $CH_4/^{36}Ar$ ratios but one (CH127) with depth observed in Figure 1(a), clearly indicates that the CH_4 generated from sedimentary organic material by thermocatalytic processes and the ^{36}Ar derived from atmosphere are intimately associated. In the fluids the coherence of the differently source components requires a common reservoir intermediate to the sampled reservoirs, where they can be well mixed[8], and, therefore, suggests that these different source components are stored together in groundwater prior to entrapment into gas reservoirs.

In addition, the calculated $CH_4/^{36}Ar$ ratios have range between 8×10^6 and 64×10^6 (Fig. 1(a)), very much greater than that 1.9×10^6 for pure water and 3.4×10^6 for a 5 mol/L NaCl brine at low temperature and hydrostatic conditions[9]. This reflects the presence of "excess" thermogenic CH_4 over that supplied by a CH_4-saturated groundwater. Furthermore, the "excess" reflects CH_4 saturation and dissolution to be at depth greater than the sampled reservoirs due to the CH_4 solubility depending upon the subsurface pressure, temperature and salinity[9]. This conclusion is consistent with the explanation for the $\delta^{13}C(CH_4)$ and $\delta^{13}C(C_2H_6)$ values of the samples. The $\delta^{13}C$ values suggest that the shallower Xinchang CH_4 of Jurassic age are derived from the deeply buried and mature source rocks of Upper Triassic age, while the deeper Xiaoquan and Hexinchang CH_4 in Upper Triassic strata are produced by the deeper a bit over source rocks and in situ accumulation[5].

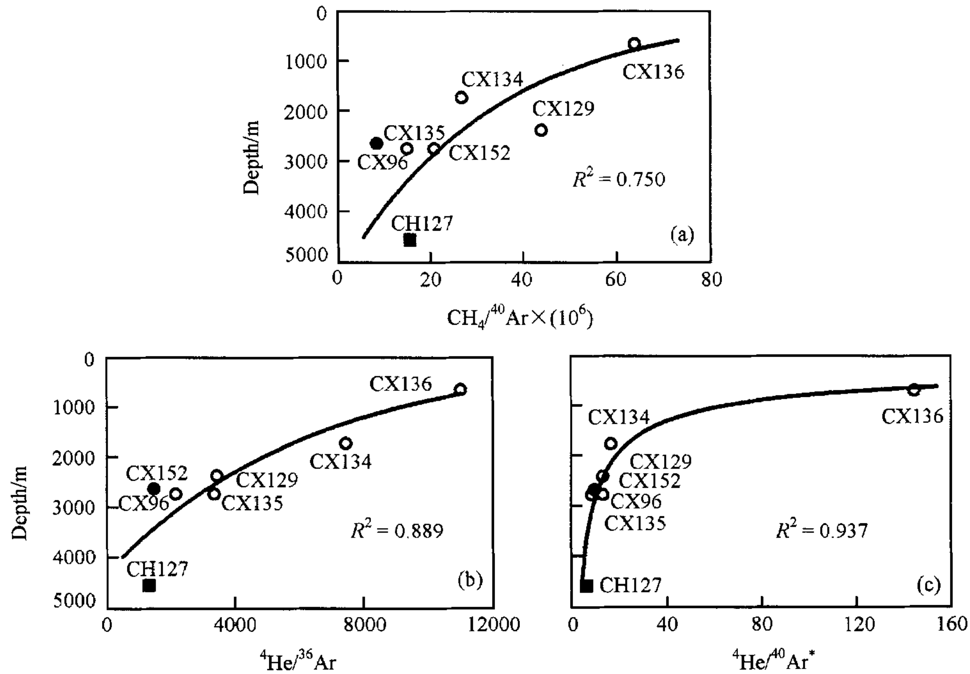

Fig. 1. The variation of $CH_4/^{36}Ar$ and $^4He/^{40}Ar^*$ ratios shown as functions of depth of hydrocarbon-rich natural gases in the Xiaoquan, Xinchang and Hexinchang gas fields; R^2 correlation coefficient; ○ Xinchang; ● Xiaoquan; ■ Hexinchang.

6.2 Vertical transport processes of radiogenic He and Ar

The observed correlation between $^4He/^{36}Ar$ ratio and depth in Figure 1(b), clearly shows that the crustal radiogenic ^4He is well mixed with the atomosphere-derived ^{36}Ar before introducing into gas reservoirs, and shows that they are transported vertically and their ratio progressively become greater with the depth decreasing. The vertical variation of the $^4He/^{36}Ar$ ratio with depth can be attributed to the preferential transport of ^4He relative to ^{36}Ar in fluxes from lower aquifers through water-filled pores into the upper one.

Figure 1(c) also show the correlation between the $^4He/^{40}Ar^*$ ratio and depth. Because the $CH_4/^{36}Ar$ and $^4He/^{36}Ar$ ratios have the correlation with depth (Fig. 1(a), (b)), the $^4He/^{40}Ar^*$ ratio also has a correlation with the $CH_4/^{36}Ar$ and $^4He/^{36}Ar$ ratios. So the correlation among the $^4He/^{40}Ar^*$, $CH_4/^{36}Ar$ and $^4He/^{36}Ar$ ratios strongly suggests a together vertical transport of radiogenic ^4He, $^{40}Ar^*$ and thermogenic CH_4 as well as atmosphere-derived ^{36}Ar within the multilayered aquifer system.

Furthermore, the radiogenic $^4He/^{40}Ar^*$ values of the samples are compared with the $^4He/^{40}Ar^*$ production ratio of 4.92 for an average crustal composition[10], which are 1.3 times to 29 times greater than the crustal production ratio. And the greatest fractionation value of the ^4He to $^{40}Ar^*$ components occurs in the shallowest gas. The characteristic of $^4He/^{40}Ar^*$ ratios indicates that the ^4He preferentially transports relative to $^{40}Ar^*$ from their production site into groundwater, along with the transfer process is controlled by depth and temperature. Moreover, $^{40}Ar^*$ having a much higher closure temperature than ^4He in radioactive minerals, only a small portion of $^{40}Ar^*$ transports into the groundwater, and its solubility in water gradually becomes larger with decreasing temperature. So the $^4He/^{40}Ar^*$ ratio in fluxes and gas phase progressively become higher with decreasing depth. As a result, the radiogenic $^4He/^{40}Ar^*$ ratio of the shallowest Xinchang gas (depth at 650 m) become the highest one.

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