

Salt-Gathering and Potassium Formation of Potassium-Rich Brine during the Triassic in the Sichuan Basin, China

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Abstract: Potassium-rich brine in the Sichuan Basin has been much studied in recent years, but few studies have focused on the distribution and migration of salt basin and the differences of potassium formation mechanisms. This work examined the salt-gathering and potassium formation of potassium-rich brine during the Triassic in the Sichuan Basin using lithofacies palaeogeographic depiction and geochemical analyses. (1) The favorable sedimentary facies controlling the formation of potassium-rich brine during the Triassic in the Sichuan Basin are evaporation platform and restricted platform, whereas the salt basin is one of the main factors controlling the poly-salt center. (2) The distribution and migration of this salt basin were affected by certain factors. The salt basin of the Jialingjiang Formation was mainly distributed in the east and central Sichuan Basin, whereas that of the Leikoupo Formation was mainly distributed in the central and west Sichuan Basin. The sedimentary centers have gradually moved westward and become smaller. (3) Three main formation mechanisms were identified for the potassium-rich brine during the Triassic in the Sichuan Basin, i.e., evaporation and concentration of seawater, surface fresh water leaching, and deep water-rock reaction. Fresh water leaching was characterized by low anomaly $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values. Water-rock reaction was mainly related to temperature, and high temperature environment (caused by burial depth, overthrust and deep hydrothermal fluids) was beneficial to water-rock reaction. The characteristics of water-rock reaction do not correspond to the increase ratio of $\text{K}\cdot 10^3/\text{Cl}$ and $\text{Br}\cdot 10^3/\text{Cl}$ in brine, and the Rb^+ content of the brine was high. (4) The formation mechanisms of potassium-rich brine differed between different areas of the Sichuan Basin. In east Sichuan, the evaporation and concentration of seawater, together with meteoric fresh water leaching, was the main formation factor, whereas the evaporation and concentration of seawater and water-rock reaction predominated in west Sichuan. This study of the sedimentary environment and formation mechanisms is of significance to the exploration and exploitation of potassium-rich brine in the Sichuan Basin.

Key words: potassium-rich brine, sedimentary environment, formation mechanism, evolution model, Sichuan Basin

1 Introduction

China's potash dependence on foreign countries reaches up to 70%, and the shortage of potassium resources has seriously restricted the development of agriculture (Zheng Mianping et al., 2006, 2016). The Sichuan Basin is not only an important petroliferous basin (Ma Yongsheng et

al., 2007, 2017; Guo Tonglou, 2011; Liu Shugen et al., 2011; Li Yanjun et al., 2013; Nie Haikuan et al., 2015; Zhao Wenzhi et al., 2015; Ran Bo et al., 2016; Yu Yu et al., 2016; Guo Xusheng et al., 2017; Xie Zengye et al., 2017; Zhang Yuanyin et al., 2018) but also an important base of potassium resources (Chen Yuchuan et al., 2007; Zheng Mianping et al., 2010). Therefore, it is particularly important to strengthen the study and exploration of

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potassium resources in this basin.

Scholars have carried out numerous studies on the paleoenvironment of potash mineralization. European and American scholars (Condie, 2015; Haq et al., 2005; Hay et al., 2006) believed that giant marine salt basins developed in cratonic basins. Based on the analysis of sedimentary structure and paleogeographic characteristics, Wang Dongsheng (1985) considered that the Sichuan Basin is a shallow-water carbonate evaporation platform during the periods T_{1j}^2 , T_{1j}^4 , T_{1j}^5 , and T_2^1 . Lin Yaoting et al. (1997) focused on the paleoenvironment in relation to the formation of potassium-rich brine in the western Sichuan Basin, and considered that potassium-rich brine was produced in the Chengdu salt basin which is one of the best salt-bearing basins for potash formation and prospecting. Qi Wen et al. (2010) pointed out that potash was mainly formed in the marine platform basin lacking compensation, and that the individual was formed in the craton margin graben/rift basin of marine and continental alternative deposition. Hu Mingyi et al. (2010), Xu Guosheng et al. (2012a), and Zheng Mianping et al. (2012) further divided the typical carbonate platform facies of the Sichuan Basin of the Leikoupo and Jialingjiang Formations into open platform, restricted platform, and evaporate platform. Xu Guosheng et al. (2012b) considered that the salt basins during the T_{1j}^4 and T_{1j}^5 periods were mainly developed in northeast Sichuan and central Sichuan. Through the study on distribution of the evaporate in the salt-forming period of the Sichuan Basin, Zhou Jiayun et al. (2015) believed that salt basins were distributed across a wide area in T_{1j}^4 , T_{1j}^5 , and T_2^3 . Salt basins of T_{1j}^4 and T_{1j}^5 are mainly distributed in western Sichuan, central Sichuan, and eastern Sichuan. The salt basins of T_2^3 are mainly distributed in southwest Sichuan and central Sichuan. Chen Anqing et al. (2015) focused on the salt accumulation environment of the Triassic in northeast Sichuan and concluded that Bazhong–Xuanhan–Quxian was the development zone of a gypsum salt basin during the Jialingjiang Formation; the basin moved westward during the Leikoupo Formation, distributed in the Bazhong–Yilong–Nanchong area. Research on the palaeoenvironment formation of potassium-rich brine in the Jialingjiang and Leikoupo Formations of the Sichuan Basin has been more thorough and uniform, and it is believed that the evaporate and restricted platforms in the arid/hot period are the main phases. However, the distribution scope and scale of the salt basin, as well as its migration behavior, are still poorly understood owing to the limitations of well data. Most existing well data are from oil and gas wells. The lithofacies palaeogeography characterizations of potash suggest a problem of "insufficient salt taste". For this

purpose, we collected more than 100 well date including oil and gas drilling, old potash wells and newly drilled potash wells over the previous two years. Based on the obtained well data, the aims of this paper were to (i) re-characterize the paleoenvironment of potassium-rich brine, (ii) discuss the sedimentary environment of potassium-rich brine formation in the Sichuan Basin (west Sichuan and east Sichuan), and (iii) determine the distribution and migration behavior of salt basins. Our goal is to provide geological guidance for the selection of areas favorable to the production of potassium-rich brine.

Based on many previous studies, it is generally believed that there are three mechanisms of potassium formation in the Sichuan Basin: (i) evaporation and concentration, (ii) atmospheric leaching and (iii) dissolution under high temperature and high pressure. Yuan Jianqi et al. (1985) believed that some saline deposition resulted from evaporation and concentration. Zhou Xun et al. (1997) suggested that the brines of the Leikoupo and Jialingjiang Formations originated from sea water and were the residual bitter brines remaining after the precipitation of evaporates in marine sedimentary environments. Song Hebin (1997), Zhang Shubin et al. (2003), and Zhang Chengjiang et al. (2012) also demonstrated that the formation of potassium-rich brine was mainly caused by the evaporation and concentration of sea water. Zhao Yanjun et al. (2015) proposed that the formation of potassium-rich brine is the result of high-intensity evaporation and concentration of saline lake brine. Comparing brine from the Chengdu Basin with that obtained abroad, Lin Yaoting et al. (2002) considered that potassium-rich brine has the characteristics of leached and filtered solid potassium salts. Zheng Mianping et al. (2006, 2010) and Xu Guosheng et al. (2012a) proposed that in the Late Triassic of the Sichuan Basin, some salt layers were transformed into brine by the leaching effect of precipitation. Li Yawen et al. (1998) clarified that the original solid potassium salt was destroyed or even completely dissolved, and that the potassium salt was transferred to the liquid phase. Zhou Xun et al. (2015a) believed that there was a high content of K^+ in some brines of the Triassic formation in the east of Sichuan, not only because of the evaporation and concentration of seawater but also due to later metamorphism and the effect of leaching of the potassium salt formations. Although there are many studies on the mechanism of potassium formation, there are no detailed studies on the source of the original material. Moreover, data on the genetic mechanisms of the geochemistry, the relationship between the geochemistry and tectonic movements, and the differences between the mechanisms of potassium formation in the Sichuan Basin are insufficient. Based on

geochemical data from drilling wells in western Sichuan and eastern Sichuan, this paper (i) studies the mechanisms of potassium accumulation in the Sichuan Basin, (ii) explores the differences between these various mechanisms, and (iii) attempts to establish a genetic model for potassium-rich brine to provide some guidance for its exploration and exploitation.

2 Geological Setting

The Sichuan Basin is located in the northwest of the upper Yangtze platform in China (Zi Jinping et al., 2017), with an area of about 1.8×10^5 km². It is a composite basin formed and developed on the Yangtze Plate and craton platform (Fig. 1a) (Zhang Yueqiao et al., 2011). In the Early Triassic and Middle Triassic, against a background of transgression and regression in the upper Yangtze platform (Liu Ying et al., 2017), the Sichuan Basin was in a carbonate-evaporate platform environment. At the end of the Middle Triassic, influenced by the Indo–China movement, the Ghangnania in the eastern part of the basin rose sharply and the seawater retreated (Lin Yaoting et al., 2002). With the uplift of Luzhou–Kaijiang (Li Zhongquan et al., 2011), the basin shifted from “west high, east low” to “east high, west low”, and developed a NE trend structural framework with uplift and depression. As a result, the denudation area expanded. During the Late

Triassic and Mesozoic, the Yanshanian movement caused the basin to again subside. Due to the strong orogenic movement, some areas were uplifted, resulting in an unconformable contact with the overlying strata. The subsequent Himalayan movement caused strong folding in the basin, forming a series of ejective folds in the eastern part of the basin. The salt-exposed surface was in anticline and suffered denudation and leaching. However, in the syncline and vast areas of the western part of the basin, salt remained deeply buried.

During the Triassic period, global sea level was low (Vail et al., 1977; Haq et al., 1987) and the environment was hot and dry (Woods, 2005; Retallack, 2013; Frakes et al., 1992). Therefore, the Sichuan Basin was exposed to a hot and arid climate in the Triassic (Wang Mingquan et al., 2015). During this period, six salt-forming periods have been identified for the Sichuan Basin (T_{1j}^2 , T_{1j}^4 , T_{1j}^5 , T_2l^1 , T_2l^3 , and T_2l^4) (Fig. 1b) (Lin et al., 2002), together with three potassium formation periods (T_{1j}^4 , T_{1j}^5 – T_2l^1 , and T_2l^4), which are related to sea level fluctuation (Zhou et al., 2015). The Jialingjiang Formation (T_{1j}) is mainly composed of carbonate rocks and evaporate. The lithology of T_{1j}^2 is mainly limestone, dolomite and anhydrite, and halilith. The lithology of T_{1j}^4 is mainly anhydrite, dolomite and halilith, and some regions of polyhalite and langbeinite. The upper lithology of T_{1j}^5 is mainly anhydrite, and dolomite and halilith, together with

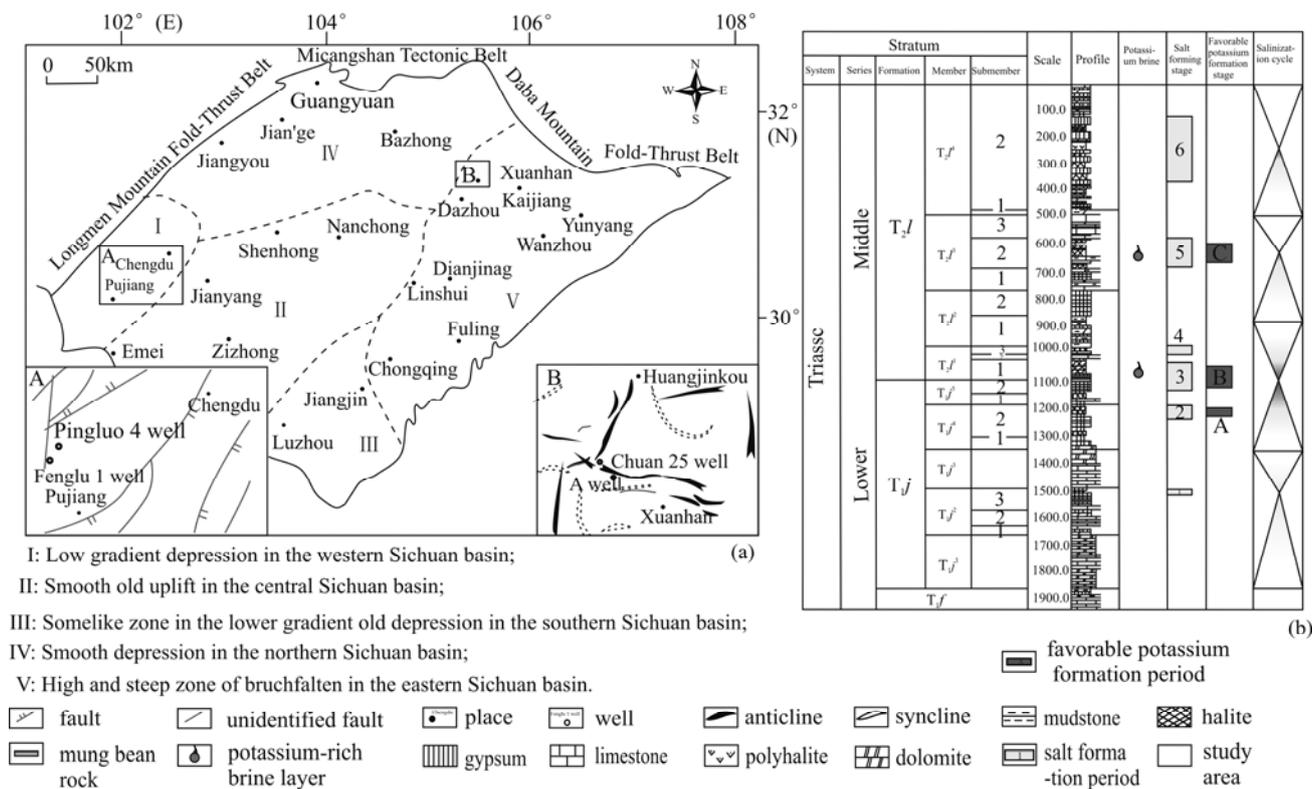


Fig. 1. Tectonic geological map of the Sichuan Basin (a) and the synthesis histogram of T_{1j} and T_2l (b) (modified from Gong Daxing et al. (2014) and Zhou Jiayun et al. (2015)).

polyhalite and langbeinite in certain areas; the lower lithology is mainly grain limestone and dolomite. The Leikoupo Formation (T_1l) is mainly composed of carbonate rocks, gypsum salt, and salt-rock. The lithology of T_2l^1 is mainly dolomite with anhydrite, although some regions contain halite, polyhalite, and langbeinite. The lithology of T_2l^3 is limestone, dolomite, anhydrite, and halilith. The lithology of T_2l^4 is mainly dolomite, interbedding of limestone and anhydrite, halilith, and polyhalite.

3 Sedimentary Paleoenvironment and Evolution Model

3.1 Sedimentary facies type

According to the analysis of regional geotectonics and the lithofacies palaeogeography background, the Sichuan Basin is mainly a sedimentary system with both evaporation and restricted platforms. During each sedimentary cycle, the Sichuan Basin experienced the

sedimentation of transgression and high system tracts (Fig. 1b). The sediments in the different system tracts are very different; using the system tract as a research unit, three sedimentary facies and many kinds of subfacies and microfacies are identified in the Jialingjiang and Leikoupo Formations of the Sichuan Basin.

3.1.1 Open platform

The seawater circulation of the open platform is good, with a depth of several meters to tens of meters; moreover, its salinity is normal to slightly high. The platform is biologically rich, containing gastropoda, lamellibranchia, echinoderms, foraminifera, and all kinds of algae. The open platform is divided into the tidal flat and the open intra-platform bank, mainly developed limestone, dolomitic limestone, micrite, bioclast limestone, powder crystal limestone, and granular micrite (Figs. 2a and 2b).

3.1.2 Evaporation platform

The evaporation platform was developed at relatively

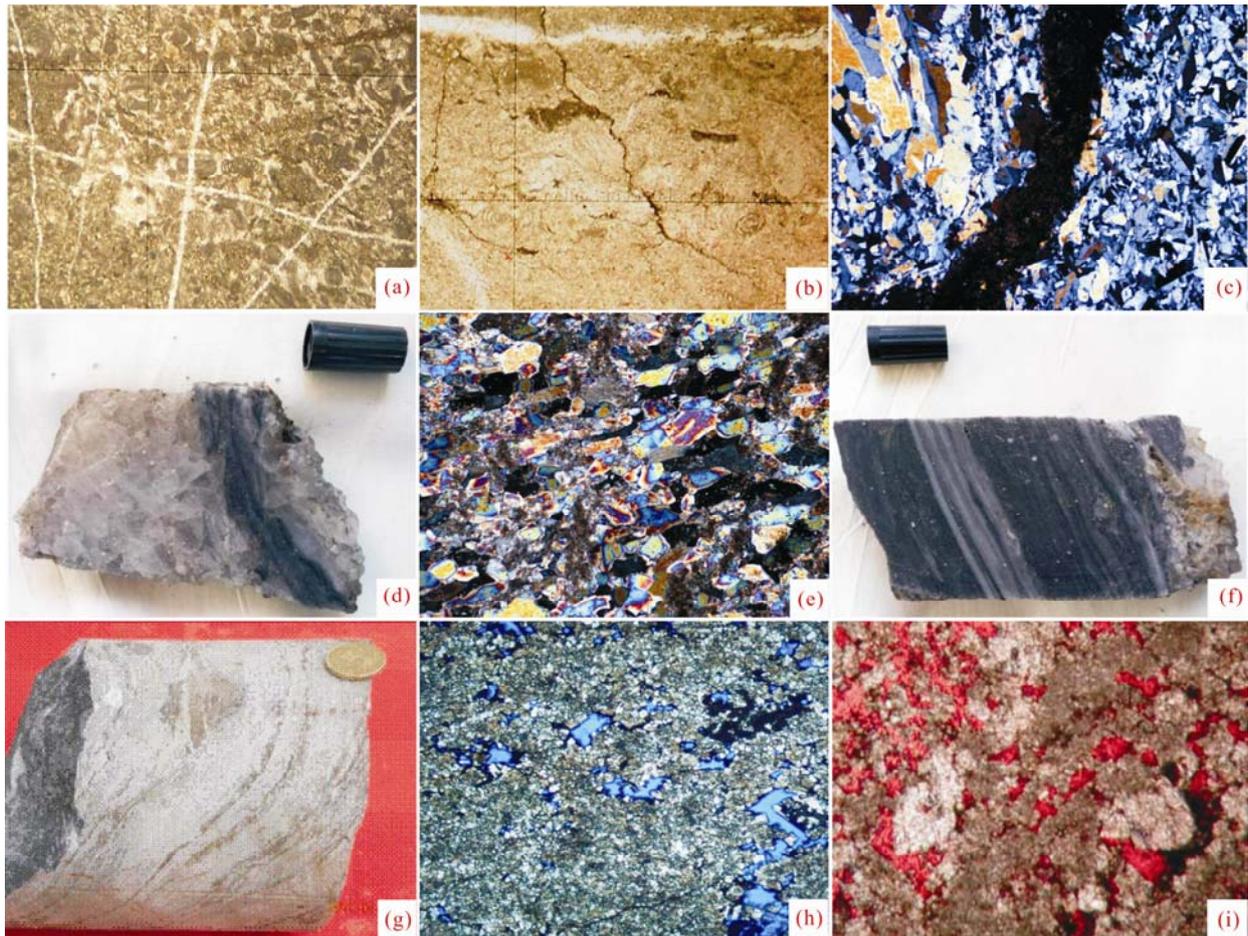


Fig. 2. Rock types in different sedimentary facies during Jialingjiang Formation and Leikoupo Formation in Sichuan Basin.

(a), Mud-sparry bioclastic limestone, A well, 3041.3m, single polarization, $d=4.4\text{mm}$; (b), dolomite-bioclastic micrite, A well, 3408.6 m, single polarization, $d=3.2\text{ mm}$; (c), gypsum rock, Pingluo 4 well, 4806.7 m, orthogonal $4^{\circ}10'$; (d), halite with thin polyhalite, A well, 3447.42 m, drill core; (e), laminar micrite gypsum dolomite, Fenglu 1 well, 4859.32m, orthogonal $10^{\circ}10'$; (f), anhydrite with striped polyhalite, A well, 3550.21m, drill core; (g), gypsum dolomite, Fenglu 1 well, 4835.2m, drill core; (h), powder crystal dolomite, Pingluo 4 well, 4856.7m, casting thin section, single polarization; (i), micro-powder crystal dolomite, Fenglu 1 well, 4459.55, $10^{\circ}10'$.

low sea level. Compared with the restricted platform period, the connectivity between the platform and open sea is more obstructed. Evaporation is more intense owing to the hot and dry climate (Hu et al., 2010). The evaporation platform is divided into gypsum dolomitite flat, dolomitite gypsum flat, salt basin, gypsum basin, and evaporation lagoon, with dominated gypsum, halite, and dolomite gypsum (Figs. 2c, d, e, f).

3.1.3 Restricted platform

The slope of the restricted platform is relatively low. The platform is located across a wide area (range of tidal zones) between the high tide line and the low tide line. Under the influence of the platform depression marginal shoal, the "platform-basin" phase, and paleo underwater uplift, circulation of the water is poor. The restricted platform can be divided into limestone dolomite flat, dolomite flat, mud dolomite flat, platform margin, restricted intra-platform shoal, and semi restricted-restricted lagoon. It has mainly developed dolomite, muddy dolomite, calcite dolomite, gypsum dolomite, etc. (Figs. 2g, h, i).

3.2 Characteristics of the sedimentary paleo-environment

Chen Anqing et al. (2015) studied the lithofacies palaeogeography in eastern Sichuan based on changes of the sea level cycle. Hu Mingyi et al. (2010) conducted a detailed study of the paleoenvironment of the Jialingjiang Formation by compiling the sequence-facies palaeogeography, but they did not study the Leikoupo Formation. Gong Daxing et al. (2015) studied the lithofacies palaeogeography of the Early and Middle Triassic in the Sichuan Basin, but only included the four main salt layers T_1^{4-2} , $T_1^{5-2}-T_2^{1-1}$, T_2^{3-2} , and T_2^{4-2} . Therefore, the lithofacies palaeogeography of the Jialingjiang Formation and Leikoupo Formation over the entire basin is not complete, and the distribution of the salt basins has not been fully reported. Zhong Yijiang et al. (2012) divided the Jialingjiang and Leikoupo Formations into four three-level sequences (SQ1–SQ4). The present paper uses these sequences as a reference for the description of the lithofacies palaeogeography.

Based on the basic principles of sedimentology and the compilation of a lithofacies palaeogeography map, we collected more than 100 well date including salt-bearing oil and gas drilling, old potash wells and newly drilled potash wells (especially in east Sichuan). The lithologic thicknesses of representative facies of salt rock, gypsum rock, polyhalite, limestone, dolomite, etc. in the Jialingjiang and Leikoupo Formations in each borehole were determined. In combination with the salt well cross

section, point-linking was performed in the geographic base map such that areas with fewer well data were connected by interpolation. In the process, the distribution ranges of salt subfacies and microphases in the salt forming region were emphasized. Then, based on a comprehensive analysis, the distribution range of the gypsum salt basin was redrawn, and the sedimentary facies and denudation area were delineated. Finally, we drew the lithofacies palaeogeography map for the six periods (the six important reservoirs) of T_{1j} (Fig. 3) and T_{2j} (Fig. 4) and analyzed the paleoenvironmental characteristics and evolution of the gypsum salt basin in the different periods.

3.2.1 Jialingjiang Formation (T_{1j})

The sedimentary paleoenvironment of T_{1j} in the Sichuan Basin is mainly the restricted evaporation flat, and the main sediments are marine carbonate rock and platform evaporate. During the three processes of transgression and regression, the sedimentary characteristics of T_{1j} show the process of shoal→restricted→salinization. The gypsum basin and salt basin are mainly developed in T_{1j}^2 , T_{1j}^4 , and T_{1j}^5 , laying an important material foundation for the formation of potassium-rich brine.

T_{1j}^2 At the end of the first-sequence late high-water level, the northern part of the basin mainly developed as a gypsum basin (Fig. 3a), the western part was mixed tidal flat and mud dolomite flat, and the central basin was mainly dolomitite gypsum flat. During this period, there was no favorable enrichment environment for potash.

T_{1j}^4 At the late high-water level of the second sequence, evaporation increased sharply. The basin was an evaporation platform or evaporative tidal platform. The salt basin was more developed and scattered, being mainly distributed in northeast Bazhong, Xuanhan, Guang'an, Liangping, Quxian, and southeast Chongqing (Fig. 3b). The gypsum basin mainly developed in Wangcang, Nanchong, Dalian, Linshui, and Changshou. The western and southern regions of the Sichuan basin mainly developed dolomitite gypsum flat.

T_{1j}^5 This period was equivalent to the third sequence. As a result of uplift and erosion, evaporation was further increased and the whole basin existed in a salinized and arid climate (Williams, et al., 2007, Zhou et al., 2015). The salt basin was mainly distributed in Wangcang–Bazhong of northern Sichuan, Daxian–Xuanhan of northeast Sichuan, and Nanchong–Zizhong of central Sichuan (Fig. 3c). The gypsum basin was mainly developed in southeastern Chongqing. The K^+ concentration in the Chuan 25 well was 25.96 g/L and in the A well was 31.96 g/L. These two wells are located in

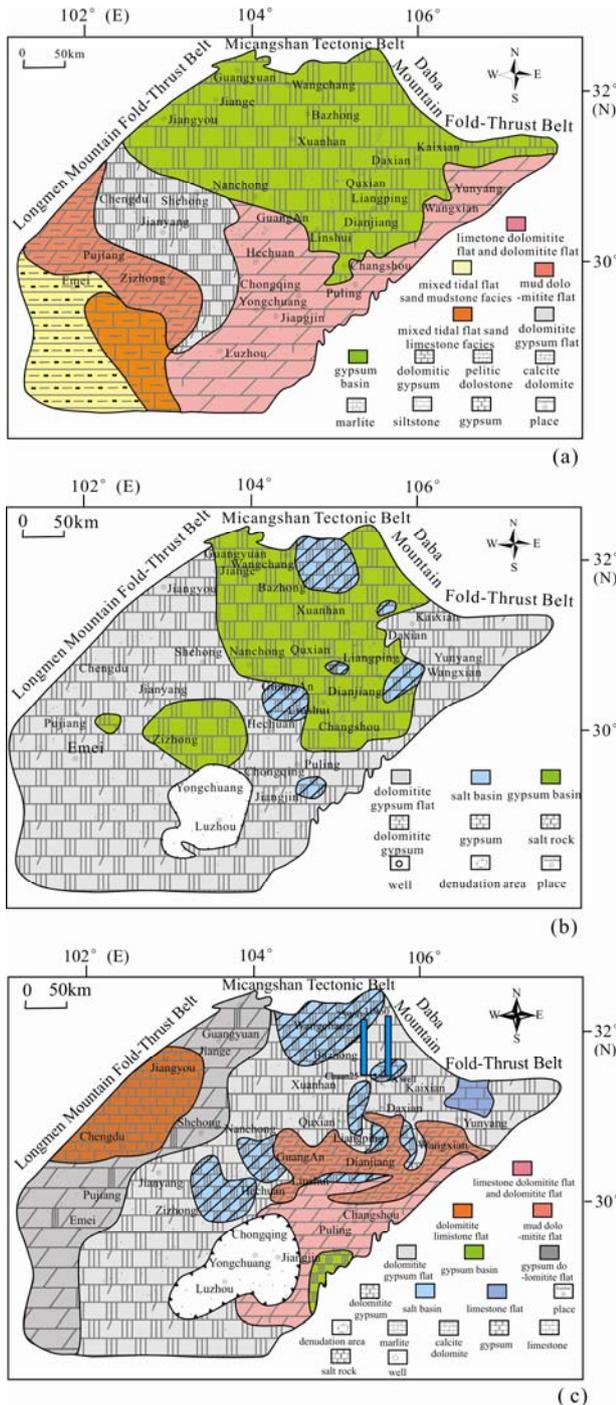


Fig. 3. Lithofacies palaeogeography map of Triassic Jialingjiang Formation in Sichuan Basin. (a), Lithofacies palaeogeography map of T_{1j}^2 in Sichuan Basin; (b), lithofacies palaeogeography map of T_{1j}^4 in Sichuan Basin; (c), Lithofacies palaeogeography map of T_{1j}^5 in Sichuan Basin.

the salt basin, indicating that the salt basin is the dominant factor in the formation of potassium-rich brine.

3.2.2 Leikoupo formation (T_2l)

Compared with the Jialingjiang Formation, the paleoenvironment of T_2l in the Sichuan Basin changed markedly, and the evolution of sedimentary facies

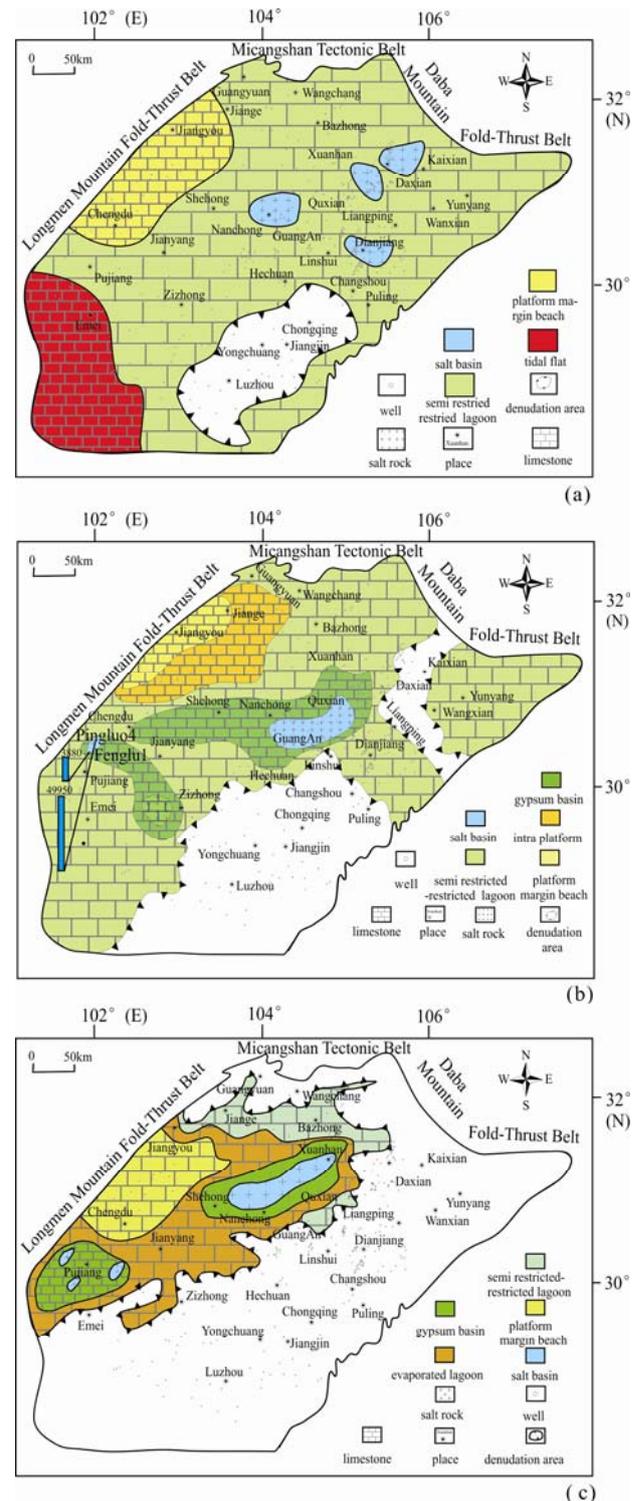


Fig. 4. Lithofacies palaeogeography map of Triassic Lei Leikoupo formation in Sichuan Basin (a), Lithofacies palaeogeography map of T_2l^1 in Sichuan Basin; (b), Lithofacies palaeogeography map of T_2l^3 in Sichuan Basin; (c), Lithofacies palaeogeography map of T_2l^4 in Sichuan Basin.

occurred more frequently. From T_{1j} to T_{2l} , the sedimentary centers of the gypsum basin and salt basin in the Sichuan Basin have gradually moved westward from the east. The salt basin mainly developed in T_{2l}^1 , T_{2l}^3 , and

T_2^4 .

T_2^1 The Sichuan Basin mainly developed tidal flats and restricted lagoons, and the development area of the salt basin was small (Fig. 4a). It was mainly distributed in Daxian–Xuanhan, Dianjiang, and Nanchong, and the lithology was mainly halite. The remaining areas of the basin were semi-restricted–restricted lagoon deposits, owing to the Luzhou paleo uplift and Indosinian movement, which caused the loss of the T_2^1 formation in Chongqing–Yongchuan–Luzhou.

T_2^3 Salt basins in the T_2^3 basin were mainly distributed in the Nanchong–Quxian–Guang'an (Fig. 4b) in northwestern Pujiang and western Weiyuan. The gypsum basin was mainly distributed in the Chengdu–Shehong–Nanchong–Quxian area and the Pujiang–Weiyuan area. The Pingluo 4 well is located in the salt basin and had a K^+ concentration of 49.950 g/L. Fenglu 1 well is located at the salt basin edge and had a K^+ concentration of 3.880 g/L. Although both wells are located in the salt basin, the concentration of K^+ is very different. This indicates that the salt basin is not a predominant factor in potassium enrichment; rather, other reasons affect the concentration of K^+ . Given that the scope of the denudation was enlarged, the Xuanhan–Liangping and Hechuan–Chongqing–Jiangjin–Luzhou areas lacked the strata of the formation.

T_2^4 The Sichuan Basin mainly developed a platform margin, an evaporated lagoon, and a semi-restricted–restricted lagoon. The salt basin was mainly distributed in northeastern Shehong and Pujiang (Fig. 4c). The gypsum basin was mainly distributed in Shehong–Nanchong and Pujiang. The denudation area of the basin was further extended, and the strata were absent in the northeast, south, and east of the basin.

3.3 Evolution model

Researchers have previously reported numerous achievements relating to a salt formation model. Borthert and Muir (1964) proposed the “multi sub-basins” sedimentary model for marine evaporate deposits. Schmalz (1970) proposed two distribution models for evaporates: the “Bull’s eye” model and the “Tear drop” model. Hsu (1972) proposed a “desiccated deep basin” model to account for the origin of saline giants. Yuan et al. (1983) studied the Qaidam Basin Saline Lake using the “high mountain–deep basin” salt formation model. Liu Chenglin et al. (2008, 2013) found that the structural inversion of the basin plays an important role in the formation of salt and potash; they proposed an “inverse lake-chain” model. Zhang Yongsheng et al. (2013) presented a salt forming model of the salt basin in northern Shanxi as the “multiple pan” model. Lin Yaoting

et al. (2003) proposed the “multi-channel evaporation and salt formation model for large shallow water basin”, emphasizing the influence of multi-stage structure on evaporation and deposition in the Sichuan Basin. Based on the sedimentary characteristics of the Middle Cambrian gypsum basin in the Sichuan Basin, two genetic models of gypsum salt rock have also been established: an evaporation mechanism on the supratidal and a restricted saline lake mechanism on the sandbank (Lin Liangbiao et al., 2014). Chen Anqing et al. (2015) established a “sea water concentrated and salinized centripetally” model for northeast Sichuan, emphasizing the importance of the sedimentary environment and the distribution characteristics of the evaporate.

According to the paleotectonic, paleoclimate, and paleogeographic features of the study area, two sedimentary evolution models for T_1j and T_2l in the Sichuan Basin have been proposed. Liu Chenglin et al. (2015) proposed that the formation of salt deposits was closely related to three factors: provenance, tectonics, and climate. The arid and hot climate is beneficial to the evaporation and concentration of seawater in the salt basin. Through the determination of carbon and oxygen isotopes at the Chengdu Geological and Mineral Research Institute, the average paleotemperature of the Early and Middle Triassic of the basin is thought to have been 34.6–36.9°C (Lin Yaoting, 1994), which had a positive effect on the deposition of gypsum and halite. From T_1j (Fig. 5) to T_2l (Fig. 6), influenced by Luzhou paleo uplift tectonics, the Sichuan Basin changed from “west high, east low” to “east high, west low”, which led to the migration of the gypsum and salt basins to the west, showing the characteristics of a “multi-salt basin center”. Affected by sedimentation, seawater evaporated, resulting in concentration and providing a material basis for the accumulation of potash. Therefore, the salt-forming model in the Sichuan Basin is a “secondary deep depression” model controlled by the dual factors of tectonics and sedimentation.

4 Experimental Sample Preparation and Test Methods

The geochemical samples collected for this study were mainly drilling brine, cores, and cuttings from both A well in east Sichuan and Fenglu 1 well in west Sichuan (Fig. 1a). The samples of potassium-rich brine were bottled immediately and sealed with paraffin to preserve the sample. The collected core and cutting samples were ground to 200 mesh. X-ray fluorescence (XRF) was used for the analysis of SiO_2 , CaO , Al_2O_3 , Na_2O , K_2O , MgO , TiO_2 , and MnO_2 . Loss on ignition was also measured.

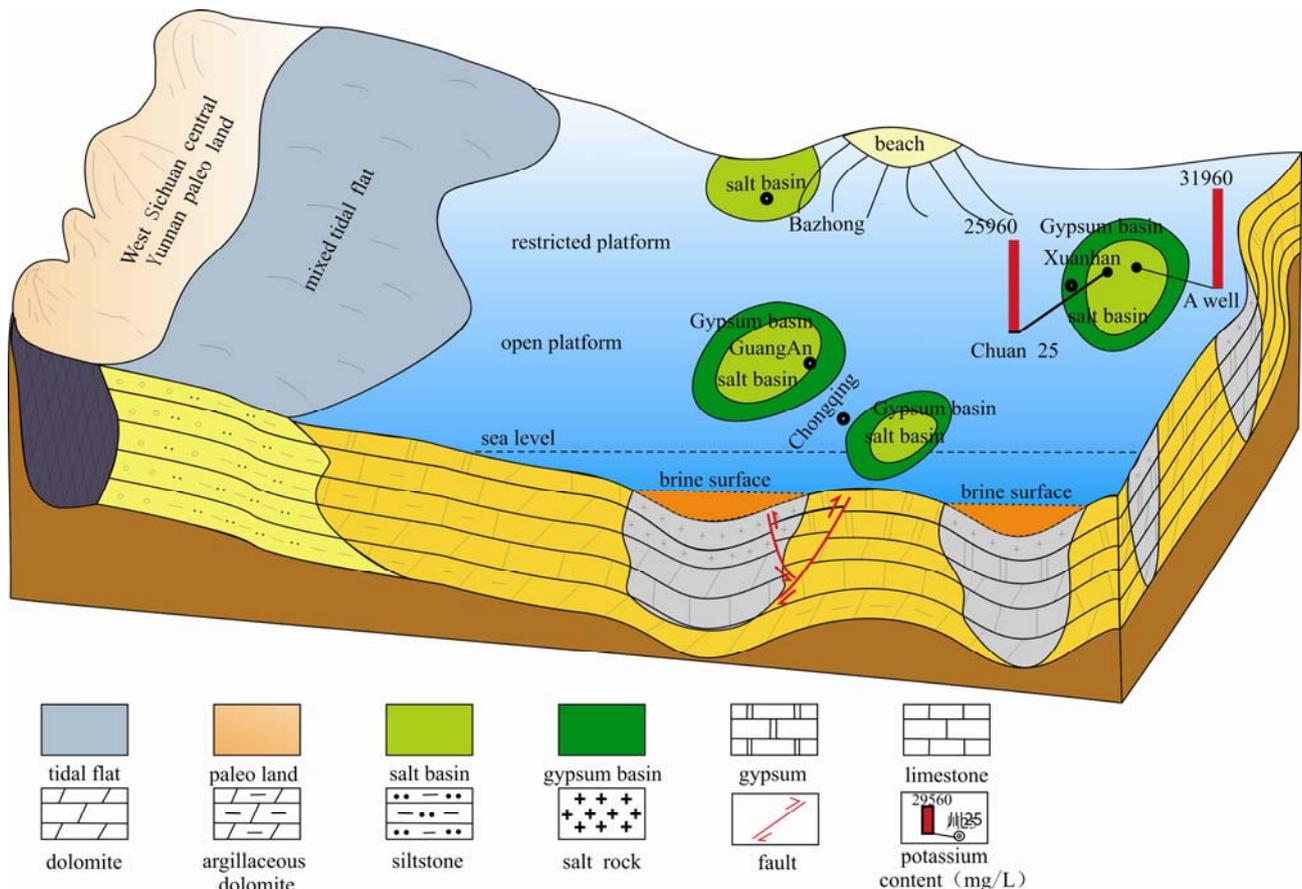


Fig. 5. Sedimentary model diagram of gypsum salt basin in of T_{1j} in Sichuan Basin.

Inductively coupled plasma–mass spectrometry was used for trace element analysis.

Carbon and oxygen isotopes were analyzed using a MAT-253 instrument. This was done by initially taking a small amount of carbonate rock sample into a quartz tube filled with helium and then adding high purity phosphoric acid. After reaction for about 1.5 h, the carbon and oxygen isotopes of the CO₂ generated were measured using a gas isotope mass spectrometer.

Strontium isotope analysis was conducted by placing about 0.3 g of ground sample into a polytetrafluoroethylene cup, adding 5 mL of 1.25 mol/L HCl, and allowing the mixture to dissolve overnight. The supernatant after centrifugation was separated and purified using a strong acid cation exchange column to obtain the enriched Sr. An isotope mass spectrometer was used for the determination of Sr isotopes and the results for ⁸⁷Sr/⁸⁶Sr were used in this study.

5 Discussions on the Genesis Model of Potassium-Rich Brine

The genesis of potassium-rich brine in the Sichuan Basin is not simply a result of sea water concentration and

evaporation (Lin Yaoting et al., 2004). It also has multi-stage and multi-genesis characteristics owing to the influence of stratigraphy, lithology, sedimentary facies, structure, climate, and deep metamorphism (Lin Yaoting et al., 2002). The formation mechanism of potassium-rich brine in the Sichuan Basin can be divided into three processes: evaporation and concentration of seawater, surface fresh water leaching, and deep water–rock reaction. The origin of the brine in different areas may differ slightly; however, in principle, it has undergone the process of evolution from the surface to underground (i.e., from an open to a closed system).

5.1 Material source

The Sichuan Basin of the Early Triassic period was a vast marine environment. Under the influence of tectonic and sedimentation, the stage evaporation and concentration of Sichuan Basin providing an important material basis for the enrichment of potassium. During the Late Permian–Early Triassic, the Sichuan and its adjacent areas were in an extensional setting. This was accompanied by many volcanic movements (Yin Hongfu et al., 1989; Mundil et al., 2004), with volcanic eruptions providing large amounts of potassium-containing

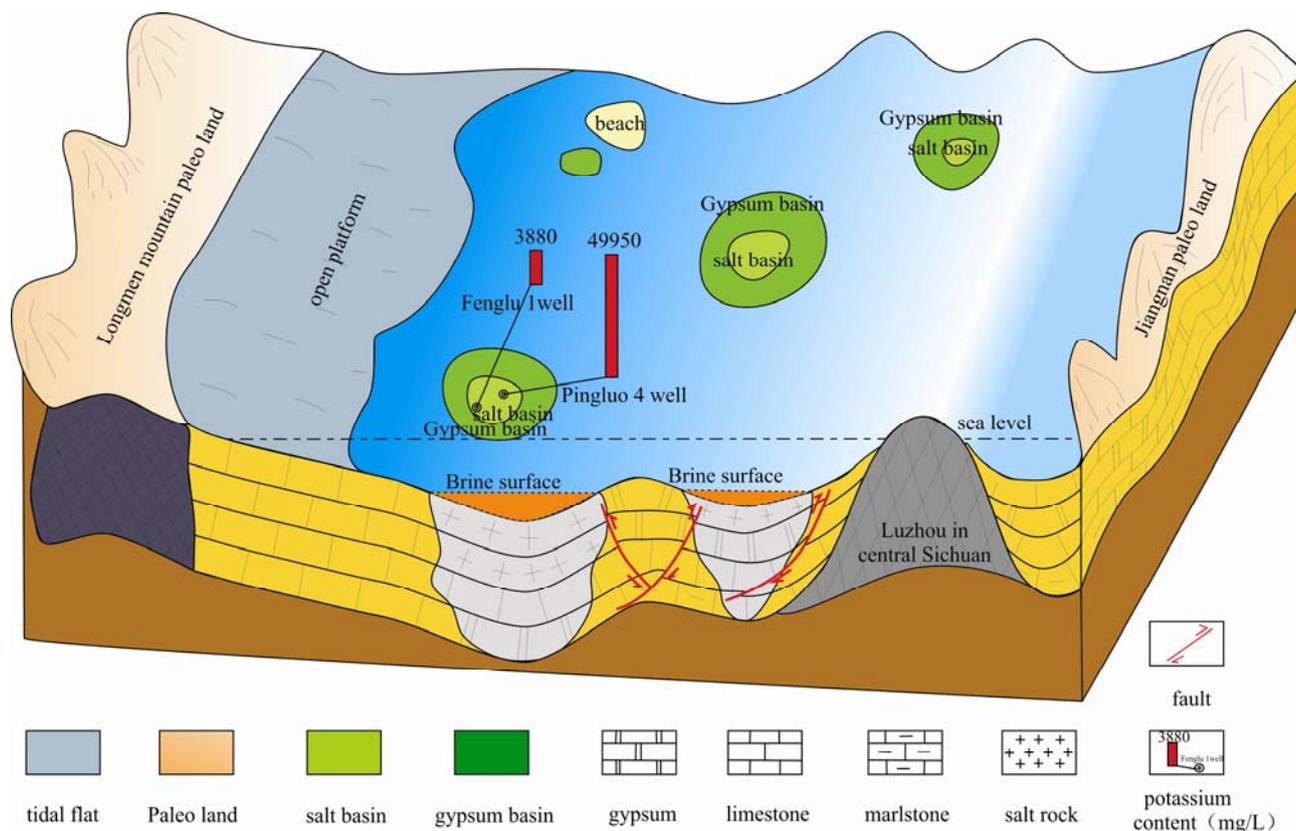


Fig. 6. Sedimentary model diagram of gypsum salt basin in of T_2l in Sichuan Basin.

substances (Zhang Chengjiang et al., 2012). At the same time as development of the boundary layer of the Jialingjiang and Leikoupo Formations (Huang Keke et al., 2016), “mung bean rock” was widely developed in the basin (Liu Chenglin et al., 2016) with a cumulative thickness is more than 1 m (Zhu Zhongfa et al., 1986). The prominent feature of “mung bean rock” is that its potassium content is generally high, with an average value of about 5.38% (Zhu Lijun et al., 1995). In addition, the polyhalite in the Jialingjiang and Leikoupo Formations in the Sichuan Basin is a ubiquitous insoluble potassium mineral with a theoretical potassium content (K_2SO_4) of up to 28% (Anlian Ying et al., 2010). Therefore, the sources of potassium-rich brine are diverse.

5.2 Potassium-rich brine formation mechanisms

5.2.1 Evaporation and concentration of seawater

At the beginning of the Permian period, the Yangzi platform completely sank (Li Zhongquan, 2011). The transgression of the Sichuan Basin reached its peak in the middle of the Early Triassic. Influenced by the Indosinian movement, the Luzhou-Kaijiang paleo uplift increased significantly, causing the Yangtze platform to uplift and the sea gradually withdraw from the Yangtze platform (Chen Hongde et al., 2011). As a result, the Sichuan Basin evolved from the ocean to an isolated basin. From T_1^1 to

T_2^4 , seawater evaporation and concentration took place (Fig. 7), with the accumulative thickness of marine evaporate deposits gradually increasing. The occurrence of potassium minerals is one of the most important indicators for evaluating potassium forming conditions in the basin (Liu Chenglin et al., 2010). From the Jialingjiang Formation to the Leikoupo Formation in the six salt formation periods, the thickness of the gypsum rock was 600–800m, of which the salt rock was tens of meters to more than 100 meters thick (Chen Jizhou, 1990, Li Yawen et al., 1998; Lin Yaoting and Chen Shaolan, 2008). As a result, a variety of elements (K, Li, I, Br) are more abundant than originally was the case (Song Hebin, 1997). Therefore, the evaporation and concentration of seawater is an important reason for the increase in K^+ content in

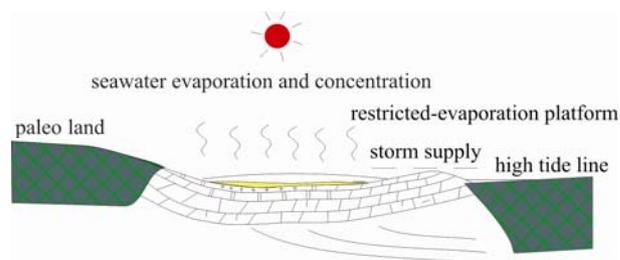


Fig. 7. Model of evaporation and concentration of seawater.

potassium-rich brine (Xu Zhengqi et al., 2017).

5.2.2 Surface fresh water leaching

Before the Middle Triassic, the topography of the basin was generally “west high, east low”. With the influence of the Indosinian movement, the crust of the eastern Sichuan uplifted, and the basin topography evolved to “west low, east high”, which led the strata of the eastern Sichuan to outcrop at the surface. These strata were weathered and eroded, providing suitable conditions for the infiltration, leaching, and erosion of meteoric water (Fig. 8).

For carbonate rocks, a $\delta^{18}\text{O}$ value of lower than -10‰ is an indication that the oxygen isotopes have significantly changed from their original composition (Qian Yixiong et al., 2005). The normal range of $\delta^{13}\text{C}$ values of marine

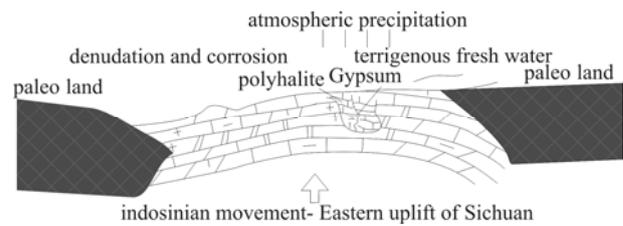


Fig. 8. Model of surface fresh water leaching.

carbonate rocks is -5‰ to 5‰ (Veizer and Demovic, 1974). The C and O isotopic analyses of cores in Well A indicate that the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ contents show a low anomaly in the middle–lower part of T_2^2 and the upper part of T_2^1 (Fig. 9; Table 1). This indicates that after the Indosinian movement was uplifted, freshwater or surface runoff was mixed during marine carbonate deposition in

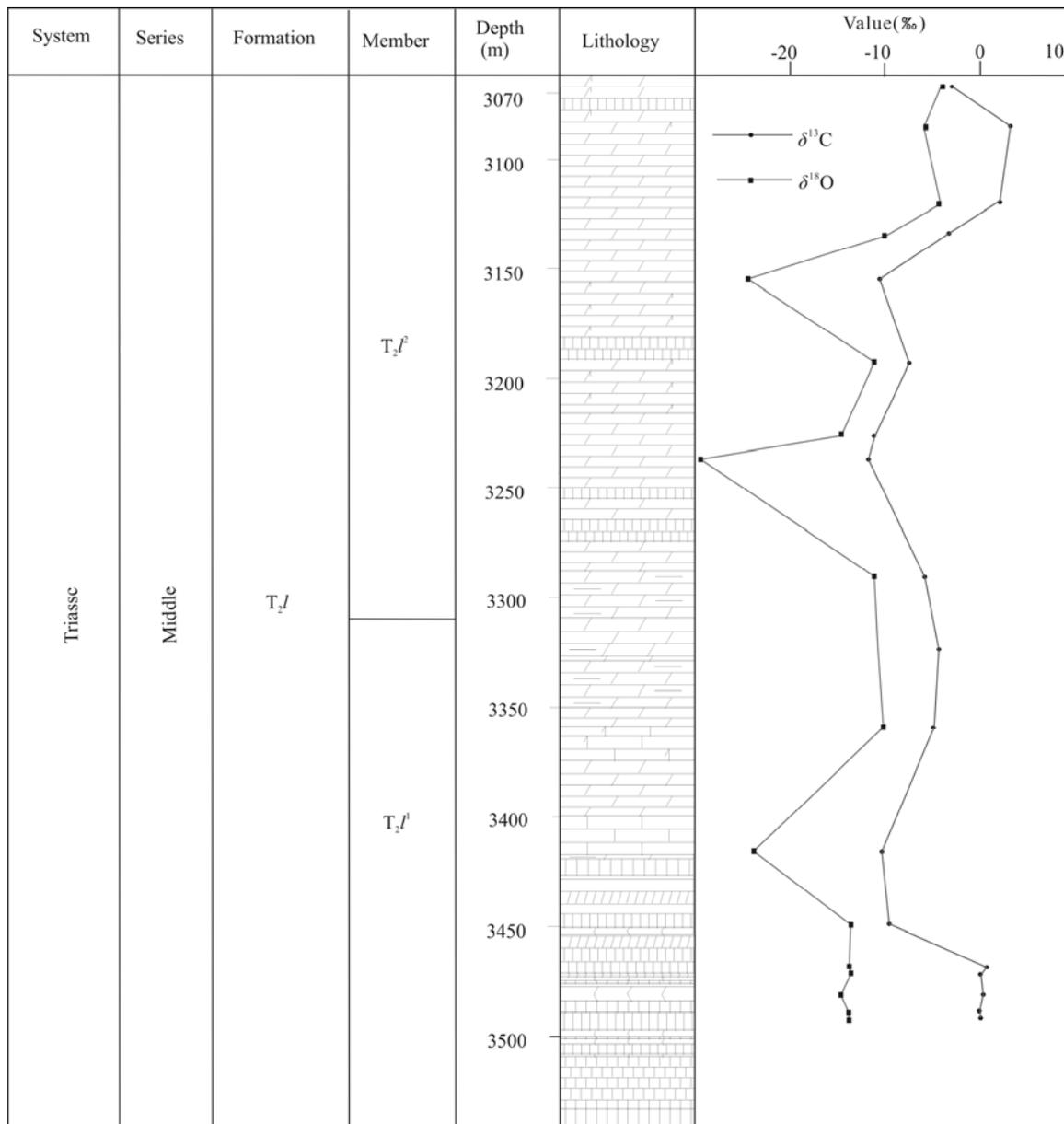


Fig. 9. Chart of C and O isotopic compositions of A well cores and cuttings in Northeast Sichuan.

Table 1 Test results of carbon and oxygen isotopes and trace element of A well

Num	Depth (m)	Lithology	$\delta^{13}\text{C}_{\text{VPDB}}$ (%)	$\delta^{18}\text{O}_{\text{VPDB}}$ (%)	Element content/Carbonate average					
					Co	Cr	Mo	Sr	Zr	Nb
3	3060–3066	Dolomitic limestones	-0.472752	-3.950648	4.8	3.818182	3.625	5.278689	1.105263	7.666667
6	3082–3088	Calcite dolomite interbedded with gypsum	3.310850	-6.5043524	4.0	4.272727	8.075	4.967213	1.052632	8.333333
10	3120–3124	Dolomite	1.995128	-4.8669440	6.5	4.363636	5.450	2.352459	1.842105	12.66667
12	3128–3138	Dolomite	-4.097116	-9.8130596	10.7	19.00000	30.625	4.016393	2.526316	16.00000
14	3150–3156	Dolomite interbedded with gypsum	-11.524858	-25.135495	5.8	6.363636	13.625	3.213115	1.684211	9.666667
17	3178–3188	Calcite dolomite interbedded with gypsum	-7.3513285	-12.722694	4.3	11.09091	8.400	3.270492	1.263158	7.666667
21	3220–3230	Dolomite interbedded with gypsum	-12.91400	-15.463000	5.5	10.45455	4.400	2.245902	2.000000	12.00001
22	3230–3240	Dolomite interbedded with gypsum	-13.514523	-27.017734	6.9	7.363636	4.275	1.629508	2.526316	15.00000
27	3280–3290	Argillaceous dolomite	-6.3837822	-11.801858	5.4	7.545455	11.300	2.655738	1.842105	11.00000
31	3325	Dolomite	-4.612700	-11.17994	8.0	3.000000	3.125	0.570492	3.473684	21.33333
34	3357.5	Dolomite	-5.1232302	-10.169934	1.3	0.272727	0.550	0.381967	0.157894737	0.333333
36	3416	Dolomite	-10.56300	-23.899000	1.1	0.181818	0.400	0.295902	0.105263	0.333333
40	3449	Halite	-9.79400	-13.453000	1.3	0.363636	1.000	3.737705	0.315789	1.666667
44	3468	Anhydrite	1.925918	-4.308304	1.8	1.818182	3.025	5.737705	0.210526	2.333333
46	3471	Anhydrite	-0.7672897	-4.524684	1.0	0.181818	2.500	2.885246	0.157895	0.666667
47	3477	Halite	0.977531	-4.923591	1.0	0.272727	2.325	3.688525	0.157895	0.666667
52	3488	Anhydrite interbedded with halite	-0.2033446	-4.2609796	2.1	0.727273	0.675	0.486885	0.684211	4.00000
53	3490	Anhydrite interbedded with halite	-0.0895778	-4.2605608	1.8	0.909091	1.525	3.04918	1.105263	4.00000

the east of Sichuan. Fresh water is relatively rich in ^{12}C and ^{16}O owing to the injection of fresh water, which causes hydrogen and oxygen isotope fractionation; the values of delta ^{18}O and delta ^{13}C are thus lower under the influence of isotopic fractionation (Wang Kun et al., 2011). In the west of Sichuan, because of its low-lying condition, the influence on fresh water leaching was relatively small compared with that in the East.

5.2.3 Deep water-rock reaction

5.2.3.1 Burial stage

Temperature has an important effect on water-rock reaction (Zhang Ronghua et al., 2016). During the deposition of marine sediments, water-rock reaction was continually underway, although the reaction is initially weak. As the deposition thickness increases, temperature also increases and water-rock reaction become more and more intense. After drilling, the blowout temperature of well Pingluo 4 was 118°C and the pressure was 89 MPa. The potential temperature in the fault at a depth of 4643 m was about 318°C (data from the former Southwest Petroleum Bureau, second geological team). Moreover, the energy released by tectonic movement also increases the strength of water-rock reaction and speeds up the reaction (Fig. 10). An Lianying et al. (2004) conducted dissolution experiments on polyhalite using aqueous solvents at three different temperatures (25°C , 50°C , and 75°C). Their experimental results showed that the equilibrium

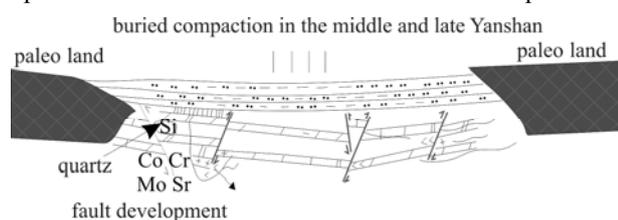


Fig. 10. Model of water-rock reaction.

concentrations of K^+ were 19.13 mg/mL, 21.30 mg/mL, and 36.78 mg/mL, respectively, and the dissolution equilibrium concentration of K^+ increased with the increase in temperature. During the middle-late diagenetic stage, when the temperature increased to a certain extent, some of polyhalite and potassium-bearing minerals were destroyed and K^+ , Mg^{2+} , and SO_4^{2-} were released (Zhang Chengjiang et al., 2012). The K^+ component in the solid potassium salt was transferred into the liquid phase (Lin Yaoting and Cao Shanxing, 2001), leading to an increase in the concentration of K^+ . There was a small amount of debris of the dissolved polyhalite in the brine storage section of the Chengdu salt basin in western Sichuan (Lin Yaoting et al., 2002), and the dissolution is obvious. According to data from more than a hundred wells, there are almost no potassium-rich brines in the layers where the polyhalite is preserved, and there is almost no storage of the polyhalite in the layers with potassium-rich brine (Huang Jianguo, 1998). Much polyhalite (Figs. 2d, f) is found in the east Sichuan Basin (Zhou Jiayun et al., 2015). Therefore, the water-rock reaction in west Sichuan are stronger than those in east Sichuan; this is an important reason for the high concentration of potassium ions in the western Sichuan brine.

Liu Wei (2012) leached “mung bean rock” at different temperatures (25°C , 90°C , and 180°C) and different salinities (0%, 1.7%, and 3.4%) (Fig. 11). The results showed that potassium in the “mung bean rock” could be leached. Moreover, with the increase in the concentration and temperature of the leachate, the potassium content of the leachate showed an increasing trend. During the diagenetic process, especially after the tectonic movement, the increase of temperature promoted the release of potassium in the “mung bean rock”, causing a portion of the potassium to be transferred into the liquid phase

Table 2 Ion ratio of normal sea water and deep brine (modified from Lin Yaoting et al., 1997; Zhou Xun et al., 2015b)

Sample name	K·10 ³ /Cl	Br·10 ³ /Cl
Normal water*	20.62	3.46
Plaster	18.91	3.50
Halite	47.90	3.99
Epsomite	85.70	15.65
Sylvine	100.78	22.94
Carnallite	3.36	26.31
Bischofite	2.18	34.68
Western Sichuan		
Pingluo 4 well brine	253.57	12.06
East Sichuan		
Chuan 25 well brine	128.51	8.29
A well brine	151.93	6.38

Note: * according to Hanor (1988) seawater chemical composition data calculation.

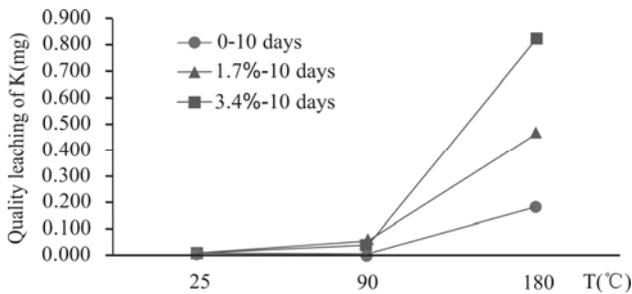


Fig. 11. Effect of different temperature and salinity on K⁺ leaching of “mung bean rock” (modified from Liu Wei, 2012).

(Huang Jianguo, 1998).

(1) Incoordination with increasing ratio of K·10³/Cl and Br·10³/Cl in brine

During the process of seawater evaporation and concentration, the K·10³/Cl ratio first increases with the change in concentration and then decreases rapidly after the deposition of sylvine; whereas the Br·10³/Cl ratio increases gradually with the change in concentration. The K·10³/Cl ratio of the Pingxiao 4 well in the western Sichuan was 253.57 and the Br·10³/Cl ratio was 12.06. The K·10³/Cl ratio of the Chuan 25 well in eastern Sichuan was 128.51 and the Br·10³/Cl ratio was 8.29. The K·10³/Cl ratio of the A well in eastern Sichuan was 151.93 and the Br·10³/Cl ratio was 6.38. Compared with the K·10³/Cl and Br·10³/Cl values (Table 2) at the sedimentary stages of normal seawater, it is obvious that Br·10³/Cl was still within the range of evaporation and concentration of seawater; however, the K·10³/Cl ratio was not in this range (Lin Yaoting et al., 1997), indicating that seawater evaporation and concentration is not the only factor causing increases in the concentration of potassium ions (Zhou Xun et al., 2015b; Cao Qin et al., 2015). During the later burial stage, the salt layer was dissolved by raw brine or crystal water, especially under high temperature and high pressure, which accelerated the dissolution of salts. This caused K⁺ to enter the brine and increase the concentration of K⁺. K·10³/Cl and Br·10³/Cl values of brine in western Sichuan, as observed for the Pingluo 4

well, were higher than those in eastern Sichuan, indicating that water–rock reaction in western Sichuan were stronger than those in eastern Sichuan.

(2) High content of Rb⁺ in brine

The contents of Rb⁺ in brine of the Ping 4 well were as high as 37.5 mg/L and in the brine of the Chuan 25 well were as high as 32.2 mg/L; both values are higher than that for the sedimentary stages of seawater. Rb⁺ and K⁺ have similar ionic radii (respectively, 1.49 Å and 1.3 Å) and are similar in elemental geochemical characteristics. Rb⁺ does not form an independent mineral in nature; it replaces K⁺ via isomorphism and enters the lattice of potash minerals. Therefore, the high content of Rb⁺ in brine is synchronous with the high anomaly of K⁺, which is a result of the leaching of solid potassium salt (Lin Yaoting et al., 1997, 2004). According to the Rb⁺ content in the Chuan 25 well in eastern Sichuan and the Pingluo 4 well in western Sichuan, the water–rock reaction in western Sichuan are clearly stronger than those in eastern Sichuan.

5.2.3.2 Hydrothermal activity promotes water–rock reaction

In this study, it was found that the distribution area of potassium-rich brine has a certain relationship with deep hydrothermal activity; we believe that the high temperature environment caused by the hydrothermal activity promotes water–rock reaction.

(1) Strontium isotope reduction

Based on measurements of the strontium isotopes of the carbonate rock, it was found that the Sr isotopic composition in the middle and late periods of T₂^l of the Fenglu 1 well was low overall, and that the ⁸⁷Sr/⁸⁶Sr ratio decreased (Fig. 12). In the western Sichuan area, the exploitable high-K brine resources were concentrated on the fault zone inside the anticline (Zhou Xun et al., 2015a). Faults and fractures developed abnormally in the brine migration zone, which was mainly related to the overthrusting effect of the Longmen Mountains. The deep hydrothermal fluid carried low strontium isotopes along the fracture. At the same time, it increased the underground temperature and accelerated the water–rock reaction. The dissolution of potassium minerals in the formation resulted in a significant increase in the potassium content of the brine.

(2) High silicon content

The content of silica in both limestone and dolomite was very low in T₂^l. Through analysis of the major elements in core samples of T₂^l in the Fenglu 1 well (Table 3), it is evident that K is positively related with Si, Al, Fe, Mn, Ti, and P. Moreover, the siliceous content of some sections of T₂^l in the Pingluoba structure was higher.

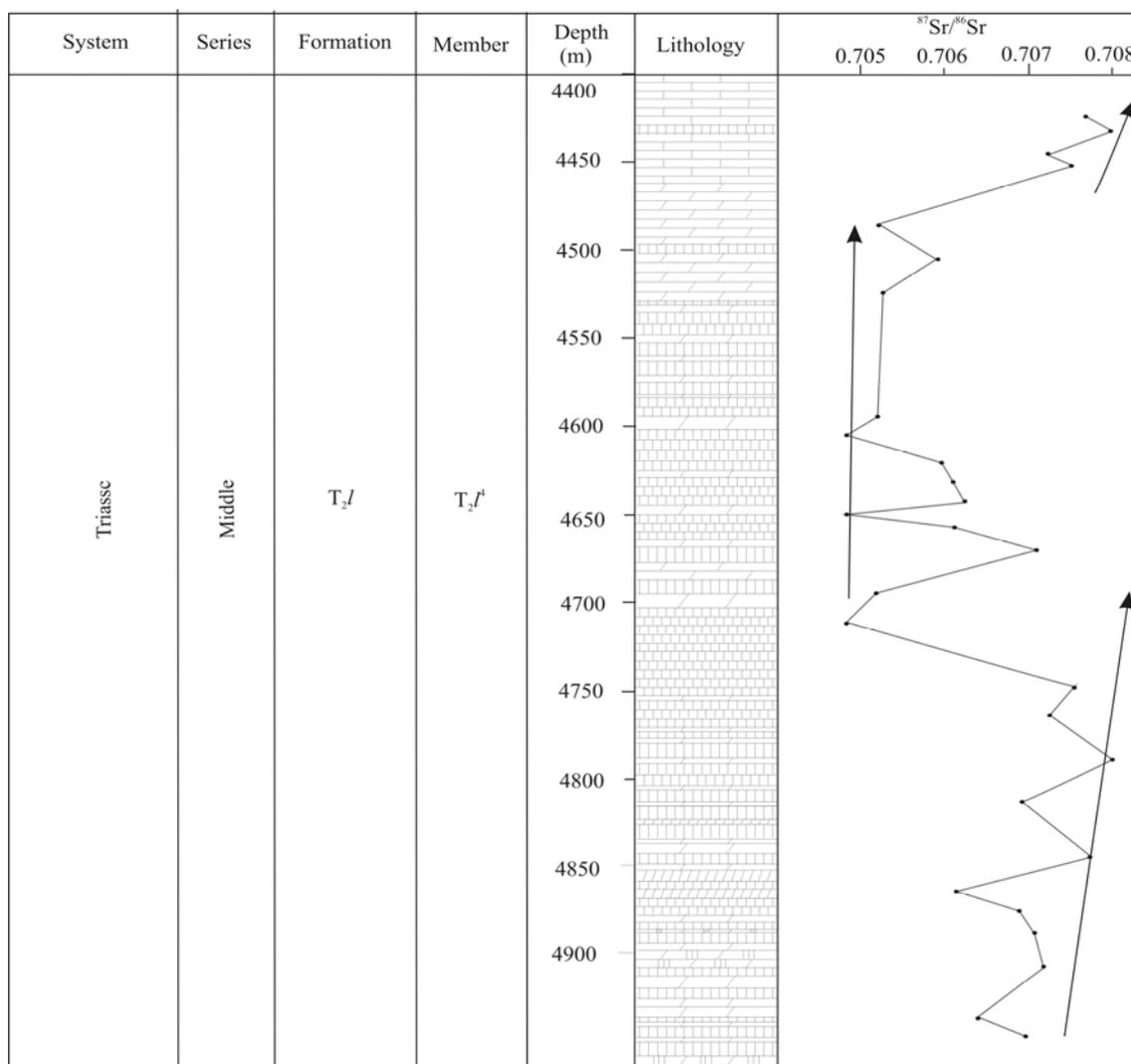


Fig. 12. The characteristic curve of Sr isotope in the T_2l of Fenglu 1 well.

The content of potassium in high silica samples was also higher, with a correlation coefficient of 0.88. X-ray diffraction analysis showed that the mineral composition of the Leikoupo Formation was mainly dolomite, calcite, gypsum, rock salt, etc. The local area contains quartz. The increase in Si content generally reflects the existence of hydrothermal activity, indicating that the formation of potassium-rich brine has a certain relationship with the activity of deep hydrothermal fluid.

(3) High contents of Co, Cr, Mo, Sr, Nb, and Zr

The analysis of trace elements in core samples from the A well in northeast Sichuan (Table 4) shows that the contents of Co, Cr, Mo, Sr, Nb, and Zr were high; higher than the average in ordinary carbonate rocks and higher than the average of the crust. These elements are generally associated with deep high-temperature fluid, which carries a large (Fig. 13) number of elements related to Co, Cr, Mo, Sr, Nb, and Zr; moreover, the high temperature environment also strengthens the water-rock reaction.

6 Conclusions

(1) Based on a detailed description of lithofacies palaeogeography, it is considered that the favorable sedimentary facies controlling Triassic potassium-rich brine formation in the Sichuan Basin are the evaporation platform and restricted platform. The salt basin is one of the main factors controlling the poly-salt center, which provides an important material foundation for the formation of potassium-rich brine.

(2) Based on the palaeotectonics, paleoclimate, and palaeogeographical features of the Sichuan Basin, the sedimentary evolution patterns of the T_1j and T_2l periods are summarized. The salt-forming model in the Sichuan Basin is "secondary deep depression" controlled by the dual factors of tectonics and sedimentation. It is considered that with the change of geological structure, the salt basin sedimentary center gradually migrated to the west and became increasingly smaller. Effect of

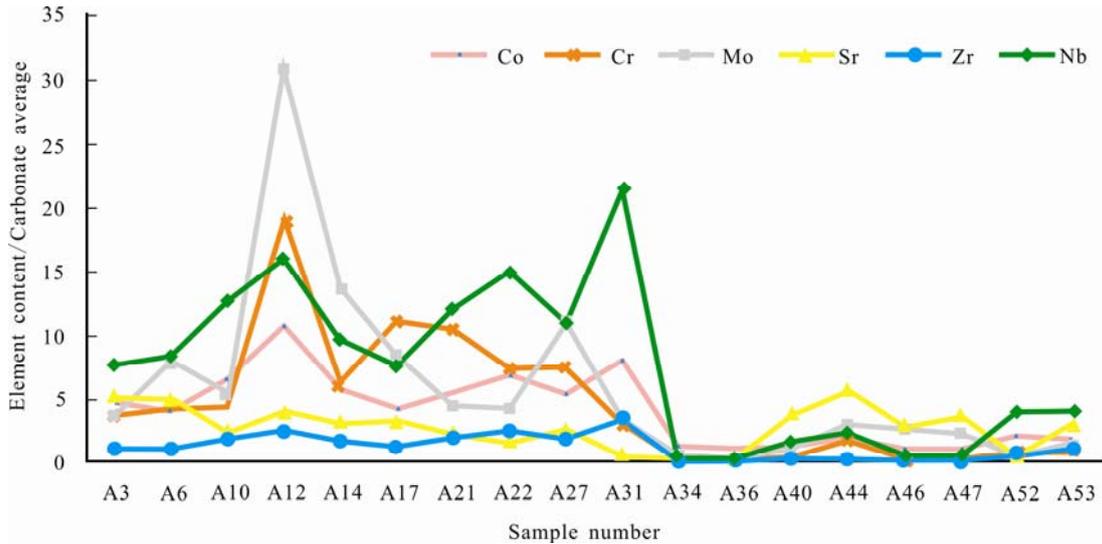


Fig. 13. Ratio of Co, Cr, Mo, Sr, Zr and Nb contents in core samples of A well.

Table 3 Correlation of major elements in cuttings of Leikoupo Formation from the Fenglu 1 well

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO	Na ₂ O	K ₂ O	MnO	TiO ₂	P ₂ O ₅	Ignition loss	FeO
SiO ₂	1											
Al ₂ O ₃	0.95	1										
Fe ₂ O ₃	0.94	0.95	1									
MgO	-0.18	-0.11	-0.01	1								
CaO	-0.13	0.13	0.03	0.08	1							
Na ₂ O	0.06	0.00	-0.02	-0.17	-0.39	1						
K ₂ O	0.88	0.96	0.85	-0.09	0.11	0.06	1					
MnO	0.78	0.86	0.88	0.02	0.16	-0.10	0.79	1				
TiO ₂	0.97	0.94	0.96	-0.04	-0.11	0.00	0.89	0.82	1			
P ₂ O ₅	0.93	0.85	0.86	-0.15	-0.25	0.10	0.81	0.65	0.89	1		
Ignition loss	-0.11	0.11	0.09	0.62	0.75	-0.21	0.11	0.16	-0.04	-0.16	1	
FeO	0.924	0.90	0.97	0.05	-0.04	-0.05	0.78	0.81	0.94	0.83	0.056	1

Table 4 Test results of strontium isotopes in Fenglu 1 well

Num	Depth(m)	Lithology	⁸⁷ Sr/ ⁸⁶ Sr	Num	Depth(m)	Lithology	⁸⁷ Sr/ ⁸⁶ Sr
FL-54	4420-4430	Limestone interbedded with mudstone	0.707692	FL-78	4663-4672	Gypsum interbedded with dolomite	0.707108
FL-55	4430-4440	Limestone interbedded with dolomite	0.708011	FL-81	4691-4700	Gypsum interbedded with dolomite	0.705228
FL-56	4440-4448	Limestone interbedded with dolomite	0.707212	FL-83	4711-4720	Gypsum interbedded with dolomite	0.704769
FL-57	4450-4460	Dolomite interbedded with gypsum	0.707533	FL-86	4740-4753	Gypsum interbedded with dolomite	0.707521
FL-58	4480-4490	Dolomite interbedded with gypsum	0.705184	FL-88	4763-4772	Gypsum interbedded with dolomite	0.707225
FL-60	4500-4510	Dolomite interbedded with gypsum	0.705943	FL-90	4783-4792	Gypsum interbedded with dolomite	0.708011
FL-62	4520-4530	Dolomite interbedded with gypsum	0.705253	FL-93	4813-4822	Gypsum interbedded with dolomite	0.706858
FL-64	4580-4592	Gypsum interbedded with dolomite	0.705030	FL-96	4843-4852	Gypsum interbedded with dolomite	0.707748
FL-66	4601-4608	Gypsum interbedded with dolomite	0.704758	FL-98	4865-4869	Corroded gypsum	0.706168
FL-70	4616-4625	Gypsum interbedded with dolomite	0.705910	FL-100	4873.5-4878.5	Gypsum interbedded with dolomite	0.706996
FL-72	4630-4638	Gypsum interbedded with dolomite	0.706105	FL-103	4888.5-4893.5	Gypsum interbedded with dolomite	0.707079
FL-73	4639-4645	Gypsum interbedded with dolomite	0.706238	FL-107	4908.5-4913.5	Gypsum interbedded with dolomite	0.707226
FL-76	4648-4651	Gypsum interbedded with dolomite	0.704861	FL-112	4933.5-4938.5	Gypsum interbedded with dolomite	0.706353
FL-77	4652-4662	Gypsum interbedded with dolomite	0.706191	FL-114	4943.5-4948.5	Dolomite interbedded with gypsum	0.706975

sedimentation makes sea water evaporation and concentration towards central and saline deposit is distributed in concentric circle.

(3) Three main genetic mechanisms exist for the formation of potassium rich brine during the Triassic in the Sichuan Basin: evaporation and concentration of seawater, surface fresh water leaching, and deep water-rock reaction. Evaporation and concentration are widely distributed in the whole region under the influence of climate and tectonics, and provide material sources for potassium-rich brine. Meteoric fresh water leaching is

characterized by low anomaly $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values owing to the fresh water that enters at the surface of the salt-bearing system. The water-rock reaction is closely related to the high-temperature environment. The burial depth, tectonic movement, and the high-temperature environment caused by the deep hydrothermal fluid contribute to the water-rock reaction. The reduction in Sr isotopes, the high content of silica, and the high content of heavy metals prove the existence of a deep hydrothermal solution. When the temperature reached a certain level, the solid potassium salt was dissolved and filtered. Therefore, the

characteristics of water–rock reaction are not corresponding to the increase ratio of $K \cdot 10^3/Cl$ and $Br \cdot 10^3/Cl$ in brine and the content of Rb^+ in the brine is high.

(4) Due to tectonics, lithology, climate, and other reasons, there are differences in the genesis of potassium-rich brine in the Sichuan Basin. In east Sichuan, evaporation and concentration of seawater and meteoric fresh water leaching are the main factors, whereas evaporation and concentration of seawater and water–rock reaction predominate in west Sichuan. Various factors control the potential of the Sichuan Basin in terms of potassium formation and these can be used as a basis for the exploration of potassium resources.

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