

Geochemical characteristics of the Permian Changxing Formation reef dolomites, northeastern Sichuan Basin, China

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Abstract: The recent discovery of deep and ultra-deep gas reservoirs in the Permian Changxing Formation reefs, northeastern Sichuan Basin is a significant development in marine carbonate oil & gas exploration in China. Reef dolomites and their origins have been major research topics for sedimentologists and oil & gas geologists. The petrography, trace element and isotope geochemistry of the reef dolomites indicated that the dolomites are characterized by low Sr and Mn contents, relatively low Fe contents, very similar $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values and very different $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. Although the calculated results of the fluid mixing suggested that a mixture with 85%-95% meteoric water and 5%-15% seawater seemed to be the dolomitizing fluids of the reef dolomites, the low Mn contents, relatively low Fe contents, high $\delta^{13}\text{C}$ values and high homogenization temperatures of the dolomites did not support that there were large proportions of meteoric water in the dolomitization process, and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios which were close to coeval seawater also did not support the possibility of the mixture of deep-burial circulated fluids from clastic rocks. High temperature deep-burial circulated seawater with low Mn and Fe contents, high Sr content and high $\delta^{13}\text{C}$ values from the dissolution of widely distributed Triassic evaporites during the burial diagenetic processes (including dehydration of water-bearing evaporites) could have been the dolomitizing fluids of the reef dolomites.

Key words: Permian, Changxing Formation, dolomite, reef, geochemistry, northeastern Sichuan Basin

1 Introduction

Since dolomite was first described in detail by the French naturalist Dèodat de Dolomieu in 1791 (Dolomieu, 1791), dolomites which form important hydrocarbon reservoirs and host rocks of base metal deposits have always been an important research area for geologists (Warren, 2000; Machel, 2004). Although a wealth of basic information and experience has been accumulated, and a number of different dolomitization models have been established to explain the dolomite genesis, geologists still do not fully understand the 'dolomite problem' and cannot satisfactorily answer the problems of the genesis of widely distributed dolomites. Therefore it is important to recognize and understand different types of dolomites. The application of trace elements and isotopes in carbonate diagenesis have been

developed (Brand and Veizer, 1980; 1981; Banner, 1995; Hu et al, 2011). The geochemical characteristics, for example, trace elements, C, O and Sr isotopes have been issues in the study of the dolomite genesis (Katz et al, 2006; Huang et al, 2006; 2009). Dolomites with different genesis have different geochemical characteristics due to their different Mg^{2+} sources, hydrological dynamics, depths and times of the dolomitization, i.e., different geochemical characteristics of dolomites could reflect different genesis of dolomites. Thus the geochemical characteristics of dolomites could play an important role in the research of dolomite genesis.

The discovery of giant or large-scale gas fields, e.g. the Puguang, Longgang, LuoJiazhai, Yuanba, Dukouhe and Tieshanpo gas fields, has made the northeastern Sichuan Basin one of the main centers of hydrocarbon exploration in China since the mid-1990s with considerable remaining potential. The reef dolomites of the Permian Changxing Formation, which are the current main reservoir rocks of the gas fields, have been paid close attention to by sedimentary geologists and petroleum geologists for a long period. The

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dolomites may still have about 10% porosity even when buried at over 5,000 m (Ma et al, 2007b; Chen, 2008). Such good reservoir quality of dolomites in deep burial is quite rare in global marine carbonate reservoirs, and thus the genesis research of reef dolomites in the Permian Changxing Formation, northeastern Sichuan Basin has important practical and theoretical significance. A lot of detailed work has been carried out, and different dolomitization models were established for the skeletal and crystalline dolomites which are significant reservoirs, including the mixed-water dolomitization model (Mou et al, 2005; Wang et al, 2006), seepage reflux dolomitization model (Li, 1989), burial dolomitization model (Zheng et al, 2007) and a mixture of multiple dolomitization models (Li et al, 2009). However, many problems have still not been satisfactorily resolved in these studies, especially detailed geochemical studies. We analyzed the geochemical characteristics of trace elements (Fe, Mn, Sr) and isotopes (C, O, Sr) and rock textures of the typical Changxing Formation reef dolomites in the Puguang and Maoba gas fields, and discussed the dolomitizing fluids of reef dolomites, to further understand the genetic environment and mechanism of reef dolomites in the Permian Changxing Formation, northeastern Sichuan Basin.

2 Geological setting

The northeastern Sichuan Basin is located in the northeast of Sichuan Province north of Chongqing City (Fig. 1), and is an overlap region of the south-central Daba Mountains arched tectonic belt and the northeast of eastern Sichuan high-steep

structural belt on the northeastern edge of the Upper Yangtze plate (Yue, 1998). Because this zone experienced multi-phase and multi-type intense tectonic extension, compression, thrust and uplift since the Late Paleozoic, the regional tectonic patterns of the northeastern Sichuan Basin are very complex. A large number of N-E tectonic structures formed in the late Yanshan period were subsequently transformed by N-W tectonic structures in the Himalayan period (Tang et al, 2008). In spite of the general absence of Middle-Upper Silurian, Devonian and Carboniferous sediments after the late Caledonian movement, the other sedimentary strata since the Sinian with an average thickness of more than 9,000 m are complete in the northeastern Sichuan Basin (Ma et al, 2005a). Five basic sedimentary facies of the Late Permian Changxing Formation have been defined: platform, platform marginal reef-bank, platform marginal slope, shelf and basin respectively, and the rock types mainly include limestones, dolomites, cherts and carbonaceous shales (Mou et al, 2011).

Currently, at least 56 Permian Changxingian reefs have been found in the Middle-Upper Yangtze region in Southern China (in outcrops and drill cores), and most of them are concentrated in the eastern and northeastern Sichuan Basin (He and Luo, 2010). They grew and migrated gradually from east to west (Wang et al, 2008). For a long time, these reefs have been important objects of hydrocarbon exploration in the Upper Yangtze region, and many large and medium-sized gas fields have been found in the reefs where high-quality reservoirs of platform marginal reef facies act as the main gas-producing formations. Examples include the Puguang,

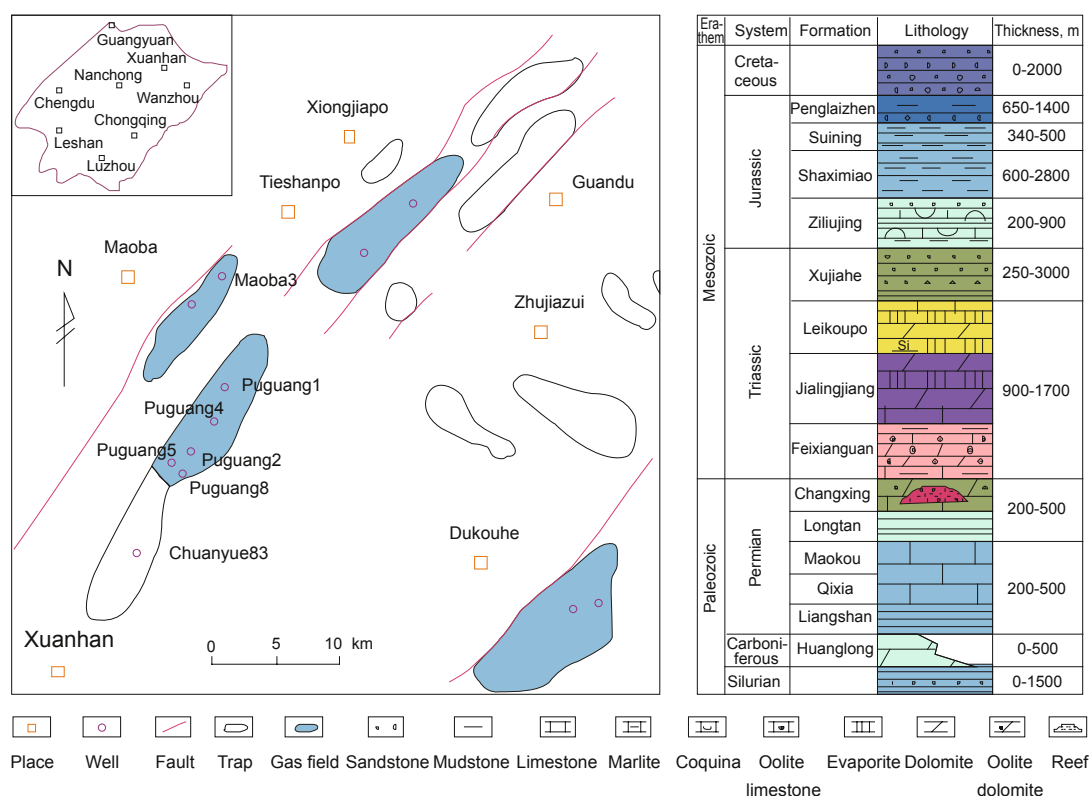


Fig. 1 Structural location and stratigraphic framework column in the northeastern Sichuan Basin (modified after Ma et al, 2007a; Ma and Cai, 2006; Qin et al, 2008)

Longgang, Yuanba, Datianchi and Jiannan gas fields. Owing to extensive dolomitization of the Permian Changxing Formation reefs in northeastern Sichuan Basin, especially in the reef core (Ma et al, 2005b; Zheng et al, 2007), these reefs are often typical research topics of the Permian Changhsingian dolomites in the Upper Yangtze region. The good petrophysical properties, considerable thickness and developed dissolution pores are advantages of the Permian Changxing Formation reef dolomite reservoirs, and the main porosity types include intercrystalline dissolution pores, intergranular dissolution pores, intragranular dissolution pores, moldic pores and solution-enlarged fractures (Ma et al, 2005b; Chen, 2008).

3 Texture types of the rocks

Because the dolomitization of the surface outcrops of the Permian Changxing Formation reefs in northeastern Sichuan Basin was very limited and mostly confined to the parts of the reef cover, the typical reef dolomites were mainly seen in drill cores, especially concentrated in some ultra-deep underground drill cores from SINOPEC blocks. Therefore our samples were chosen from the drill cores of the Puguang5 and Puguang8 wells in the Puguang gas field, and the Maoba3 well in the Maoba gas field. These drill cores are from typical reef dolomites with good reservoir quality. Detailed locations of the sampled gas wells are shown in Fig. 1. The samples include dolomites, calcitic dolomites and dolomitic limestones. Examination of standard thin sections, blue resin vacuum impregnated thin sections and scanning electron microscopy (SEM) were used to study the composition and fabric characteristics of samples. The Permian Changxing Formation reef dolomites are divided into two main end-member types on the basis of original textures and the crystal size of the dolomite. The two types are skeletal dolomites and crystalline dolomites. Good original texture is a characteristic of skeletal dolomites, and good crystalline granular texture is a characteristic of crystalline dolomites. Some samples which consist of mixture of dolomite and calcite are classified as transitional, and a few separable dolomite and calcite crystals from the reef dolomites are classified as the individual fabric types: vug dolomite and vug calcite. The main features of different texture types of reef dolomites are as follows:

1) Skeletal dolomites. These dolomites with well preserved original textures were mostly formed by dolomitization of reef limestones (the skeletons are mainly composed of reef-building sponges) (Fig. 2(a)), and some dolomites still had a small amount of binding textures (mainly blue-green algae binding texture) (Fig. 2(b)). The samples consisted mostly of microcrystalline dolomite, and some were very fine dolomite crystals. Early dolomite often forms anhedral crystals, but late dolomite deposited along the edge of vugs often forms euhedral crystals (Fig. 2(c)). The intragranular pores of dolomitic sponges are filled with sparry calcite (Fig. 2(a)), which might be related to the late sparry calcite cements. Generally speaking, the porosity of skeletal dolomites with rare visible pores is poor and intercrystalline

porosity is almost absent in the SEM micrographs (Fig. 2(c)). These dolomites are characterized by the existence of the most original textures of the carbonate grains and reef-building organisms after the dolomitization, reflecting the selective dolomitization of the early burial dolomitizing fluids.

2) Crystalline dolomites. These dolomites with poor original textures and little residual textures have mainly crystalline granular textures, but some samples with transitional texture types retain visible bioclastic textures, such as bivalves, fusulinaceas, and echinoderms (Fig. 2(d)) or reef-building skeletal textures, such as sponge (Fig. 2(e)). Crystalline dolomites are mainly comprised of very fine and fine crystalline dolomite, and some crystalline dolomites are transitional texture types with relatively fine microcrystalline or relatively coarse medium-crystalline dolomite. The crystalline dolomites could be further divided into very fine crystalline dolomites, fine crystalline dolomites and a small amount of medium crystalline dolomites. The very fine crystalline dolomites are mainly composed of very fine crystalline subhedral-anhedral dolomite (0.02-0.05 mm). The fine crystalline dolomites are mainly composed of fine crystalline subhedral dolomite (0.1-0.2 mm). The pores of crystalline dolomites included intercrystalline pores (Fig. 2(d), (e)) and vugs (Fig. 2(d), (f)), and some of them were locally filled with oil droplet-like and film-like bitumen (Fig. 2(f), (g)). These dolomites are characterized by the disappearance of the most original textures of the carbonate grains and reef-building organisms, reflecting the superimposed influence of the late inherited burial dolomitizing fluids which further affected the early formed skeletal dolomites by the strong dolomitization and recrystallization, and then the larger dolomite crystals were formed.

3) Vug dolomite and vug calcite. Some crystalline dolomites consisting of very fine and fine crystalline dolomite show a few dolomite and calcite crystals with large volumes and quantities filling dissolved pores, vugs or fractures (Fig. 2(g)), so these carbonate cements were classified as individual fabric types: vug dolomite and vug calcite (Fig. 2(f)). The vug dolomite and vug calcite are characterized by the occurrence in the early pores of dolomite reservoirs, indicating that their most important fluid source is the late burial dolomitizing fluids and calcite precipitation fluids.

4) Transitional rock type. Although the content of the residual calcite or late calcite cements is more than 50%, these rocks still contain 10%-50% of dolomite, so the dolomite could not be ignored when the samples were classified. The transitional rock types belong to the rock types with component transition, and are classified as dolomitic limestones. The dolomitization of samples which mainly consisted of residual calcite and retained complete original textures is relatively weak, and dolomite is sporadic or locally distributed (Fig. 2(h)). The other samples which mainly consist of late calcite cements are almost crystalline granular textures and are similar to crystalline dolomites. Some parts of samples retain relatively complete original textures (Fig. 2(i)). However, the content of the late calcite cements filling the pores of dolomites is more than 50%.

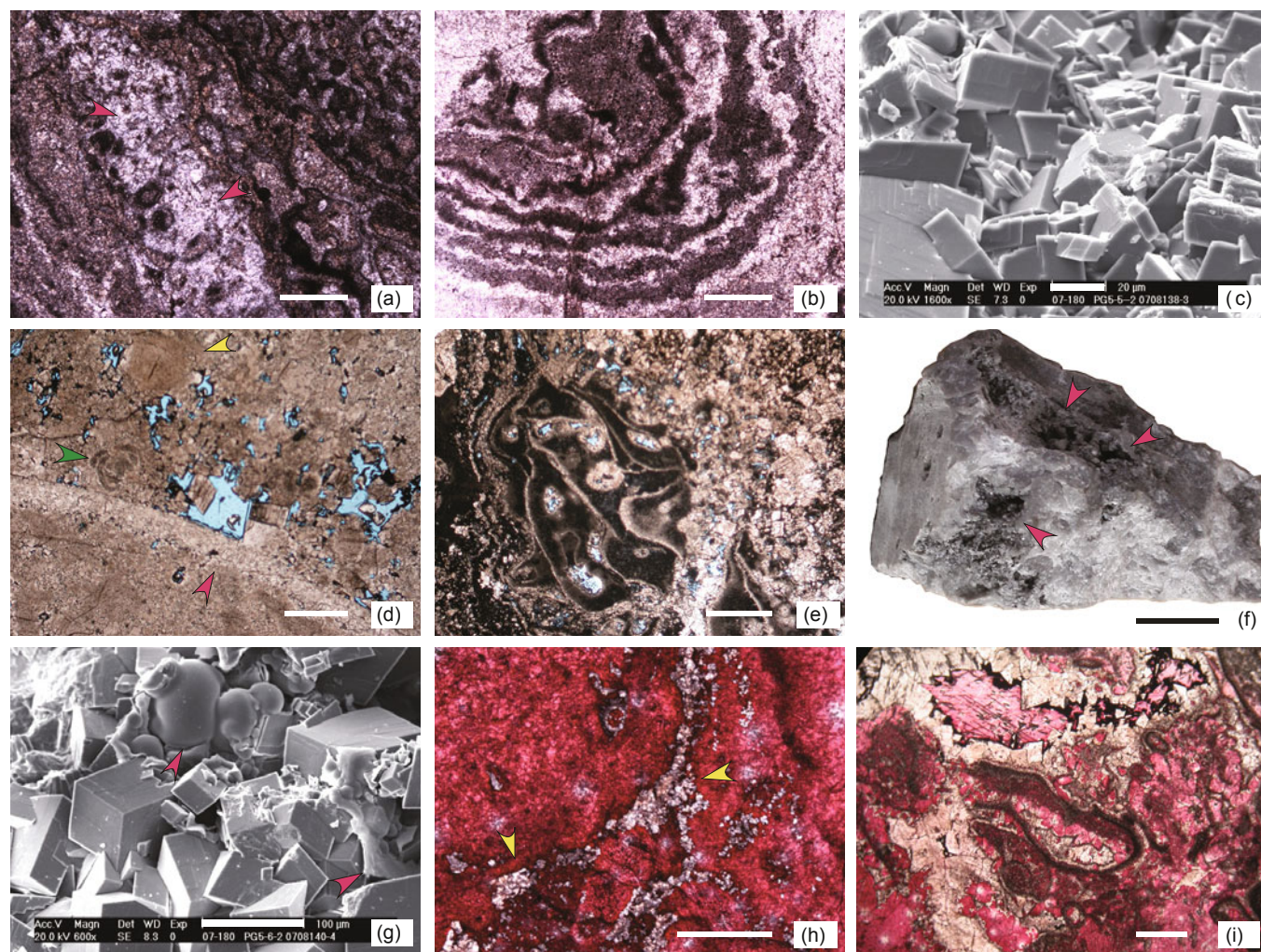


Fig. 2 Photomicrographs of main texture types of the Permian Changxing Formation reef dolomites

(a) Skeletal textures (consisted of reef-building sponges), sparry calcite filled the intragranular pores within sponges (milk-white part, such as red arrows). Puguang5 well, 5294.5 m, plane-polarized light, scale=0.5 mm. (b) Algae-binding textures. Puguang5 well, 5294.5 m, plane-polarized light, scale=0.5 mm. (c) Late dolomite cements with relatively good euhedral crystal shape filled the pores. Puguang5 well, 5157.7 m, SEM micrograph, scale=20 μm . (d) The sample mainly consists of very fine dolomite. The residual textures of the bioclasts, e.g. bivalves (red arrow), fusulinacea (green arrow), echinoderms (yellow arrow), had been replaced by dolomite. Intergranular pores and oversized dissolution pores have been relatively well developed. Puguang5 well, 5146.7 m, blue resin vacuum impregnated thin section, plane-polarized light, scale=0.5 mm. (e) The sample mainly consists of fine crystalline dolomite. The residual textures of the sponge skeleton are significant. Intergranular pores and dissolution pores have been relatively well developed. Puguang8 well, 5640 m, blue resin vacuum impregnated thin section, plane-polarized light, scale=0.5 mm. (f) Oversized dissolution pores (vugs) developed, relatively coarse euhedral dolomite crystals on the edge of dissolution pores (e.g. red arrows). Bitumen filled the intergranular pores between the dolomite crystals. Puguang8 well, 5504.5 m, scale=1 cm. (g) Intergranular pores between euhedral dolomite crystals developed in dissolved pores. Some pores were filled with oil droplet-like and film-like bitumen (red arrows). Puguang5 well, 5164.27 m, SEM micrograph, scale=100 μm . (h) A small amount of microcrystalline dolomite (yellow arrows) locally concentrated in the reef limestones. Puguang5 well, 5288.25 m, staining with alizarin red-S, plane-polarized light, scale=0.5 mm. (i) A large amount of sparry calcite (red parts) filled the pores between dolomite crystals. Puguang5 well, 5285.2 m, staining with alizarin red-S, plane-polarized light, scale=0.5 mm.

4 Geochemical characteristics

4.1 Characteristics of Fe, Mn and Sr trace elements

The average Sr contents of different types of Permian Changxing Formation reef dolomites are similar, and they are all less than 200 ppm. The average Sr contents of skeletal dolomites and crystalline dolomites are very close to 140 ppm (Table 1, Fig. 3(a)). The average Sr content of vug calcite can reach 608 ppm and the maximum Sr content of vug calcite is as high as 1,407 ppm (Table 1, Fig. 3(a)). However, the

Sr contents are not related to the different texture types of dolomites. The Sr content is related to the crystal chemistry of dolomite, and the Sr distribution coefficient in dolomite is significantly less than (theoretically only half) the Sr distribution coefficient in calcite (Vahrenkamp and Swart, 1990), so there are not very high Sr contents in dolomites (Azmy et al, 2001; Huang et al, 2006; 2009; Hartig et al, 2011). Only one crystalline dolomites sample had an exceptionally high Sr content (1,768 ppm), which might be related to the late calcite cements which filled the pores of dolomites (Table 1).

Table 1 Geochemical data of the Permian Changxing Formation reef dolomites

Well	Depth m	Rock type	Dolomite* %	MgO %	Mn ppm	Sr ppm	Fe ppm	$\delta^{13}\text{C}$ (PDB) ‰	$\delta^{18}\text{O}$ (PDB) ‰	$^{87}\text{Sr}/^{86}\text{Sr}$
Puguang5	5292.38	Skeletal dolomites	97.4	21.1	30	121	100	3.26	-5.03	0.708208
Puguang8	5640	Skeletal dolomites	95.2	20.4	40	140	200	4.23	-4.2	
Puguang8	5568.7	Skeletal dolomites	96.1	21.2	91	165	414	2.38	-5.31	
Puguang8	5656.36	Skeletal dolomites	95.6	20.5	91	131	314	2.48	-4.99	
Maoba3	4331.29	Crystalline dolomites	89.5	19.3	114	217	2614	1.81	-3.77	
Maoba3	4339.25	Crystalline dolomites	83.9	19	85	1768	442	0.83	-5.25	
Maoba3	4348.96	Crystalline dolomites	89.6	20.5	88	143	1105	2.83	-5.41	
Maoba3	4355.01	Crystalline dolomites	99.2	21.3	50	223	200	3.37	-4.58	0.707534
Maoba3	4365.62	Crystalline dolomites	87.4	20.3	73	205	544	2.49	-5.75	
Maoba3	4382.52	Crystalline dolomites	87.8	17.6	50	248	100	2.8	-4.4	0.707591
Maoba3	4393.29	Crystalline dolomites	93.1	20.7	64	139	450	3.2	-5.1	
Maoba3	4406.66	Crystalline dolomites	87.5	19.9	80	168	394	2.48	-5.28	
Maoba3	4425.7	Crystalline dolomites	94.4	20.5	50	363	100	2.69	-6.11	
Puguang5	5166.3	Crystalline dolomites	85.8	18.6	30	97	500	1.54	-6.49	
Puguang8	5528.6	Crystalline dolomites	96.8	21	60	142	300	2.37	-5.2	
Puguang8	5555.2	Crystalline dolomites	95	19.5	60	153	600	1.02	-3.55	
Puguang8	5586.1	Crystalline dolomites	97.2	21	40	103	100	1.84	-4.9	
Puguang8	5618.34	Crystalline dolomites	96.6	21	40	154	100	3.54	-4.54	
Puguang5	5146.7	Crystalline dolomites	93	20.9	66	75	537	2.69	-4.79	0.707696
Puguang5	5164.27	Crystalline dolomites	93.5	20.7	75	80	635	2.8	-5.24	0.707705
Puguang8	5508	Crystalline dolomites	94.2	20.1	266	124	1552	1.84	-5.19	0.707747
Puguang5	5157.7	Crystalline dolomites	90.4	20.3	76	100	559	2.52	-5.06	
Puguang5	5158.8	Crystalline dolomites	92.9	20.8	67	84	488	2.26	-5.31	
Puguang5	5169.8	Crystalline dolomites	94.1	21.3	71	70	293	2.52	-5.06	
Puguang8	5510	Crystalline dolomites	93.6	20.1	261	110	1333	2.14	-3.83	
Puguang8	5512.1	Crystalline dolomites	91.2	19.3	152	127	2936	2.1	-5.38	
Puguang8	5525.7	Crystalline dolomites	95.8	20.8	125	134	1221	2.17	-5.18	
Puguang8	5531.5	Crystalline dolomites	93.2	19.8	197	133	2073	2.41	-4.53	
Puguang8	5538.5	Crystalline dolomites	87.5	18	260	165	3823	2.21	-3.63	
Puguang8	5545.3	Crystalline dolomites	94	19.4	129	117	3898	2.14	-5.39	
Puguang8	5563	Crystalline dolomites	98.3	21.5	85	73	303	1.7	-5.16	
Puguang8	5572.3	Crystalline dolomites	96.4	20.8	90	128	393	2.5	-4.12	
Puguang8	5577.8	Crystalline dolomites	96	20.7	98	124	1178	2.42	-5.62	
Puguang8	5604.5	Crystalline dolomites	96.6	20.6	99	176	912	3.02	-5.32	
Puguang8	5634.5	Crystalline dolomites	95.9	20.8	69	112	418	2.76	-5.14	
Puguang5	5164.27	Vug dolomite	89.3	19.3	40	142	200	2.87	-4.8	0.708278
Puguang5	5292.38	Vug dolomite	92	18.8	40	301	1300	2.63	-6.05	0.707710
Puguang5	5351	Vug dolomite	90	18.3	30	197	50	2.86	-7.75	
Puguang8	5508	Vug dolomite	95.2	20.4	20	174	100	1.91	-4.76	0.707591

(To be continued)

(Continued)

Well	Depth m	Rock type	Dolomite* %	MgO %	Mn ppm	Sr ppm	Fe ppm	$\delta^{13}\text{C}$ (PDB) ‰	$\delta^{18}\text{O}$ (PDB) ‰	$^{87}\text{Sr}/^{86}\text{Sr}$
Puguang8	5568.7	Vug dolomite	95.8	20.8	40	123	100	2.81	-4.76	
Puguang8	5572.3	Vug dolomite	95.7	20.8	50	168	100	3.06	-3.67	
Puguang8	5577.8	Vug dolomite	95.8	20.9	40	165	100	2.95	-5.23	
Puguang8	5604.5	Vug dolomite	96.1	20.7	50	244	100	3.7	-3.62	
Maoba3	4415.54	Vug calcite	3.5	0.8	5	1407	100	-9.36	-5.67	0.707468
Puguang5	5166.3	Vug calcite	1.3	0.3	5	219	50	-0.64	-8.84	0.707746
Puguang5	5288.25	Vug calcite	1	0.2	5	199	200	3.47	-6.6	0.707591
Maoba3	4440.06	Transitional rock type	38.9	9.3	53	390	592	2.53	-5.7	
Puguang5	5288.25	Transitional rock type	11.9	2.8	45	160	184	3.26	-6.38	0.707293
Puguang5	5285.2	Transitional rock type	20.2	4.8	27	158	171	2.99	-6.42	
Puguang5	5290	Transitional rock type	39.5	9.2	64	160	591	3.23	-6.17	
Puguang5	5351	Transitional rock type	25.2	6	52	167	188	3.31	-6.32	

Notes: The Mg, Mn, Sr, Fe elemental analyses were measured on a Varian Vista MPX inductively coupled plasma-atomic emission spectrometer in the Wuxi Institute of Petroleum Geology, SINOPEC, and a Hitachi Z8000 atomic absorption spectrometer in the Testing Center of Huayang, Geological & Mineralogical Bureau of Sichuan respectively, and the relative errors are less than $\pm 15\%$. C and O isotope analyses were measured on a Finnigan MAT 253 mass spectrometer in the Wuxi Institute of Petroleum Geology, SINOPEC, the relative errors are less than $\pm 0.2\%$; Sr isotope analyses were measured on a Finnigan MAT 262 mass spectrometer in the Institute of Geology and Geophysics, Chinese Academy of Sciences, the mean analytical results of NBS987 is 0.710226 ± 0.000012 ($n=8$), blank values of all procedures are approximately $2 \times 10^{10} \text{g}$ - $5 \times 10^{10} \text{g}$.

*The contents of dolomite were calculated from MgO contents on the basis of the composition of ideal dolomite.

The average Mn contents of the Permian Changxing Formation reef dolomites are all less than 100 ppm (Table 1, Fig. 3(b)), and their average Fe contents are less than 1,000 ppm (Table 1, Fig. 3(c)). The average Mn and Fe contents of vug calcite are as low as 5 ppm and 117 ppm respectively (Table 1, Fig. 3(b), (c)). The average and maximum values of Mn and Fe contents of crystalline dolomites are the highest in all the texture types of samples, 99 ppm, 971 ppm and

266 ppm, 3,900 ppm respectively (Table 1). The Mn and Fe contents of different texture types of the reef dolomites are significantly lower than many other reported dolomites in the world, for example, Hartig et al (2011) reported that the Mn and Fe contents of Permian coarse cloud dolomite in Bravo Dome CO_2 gas fields, U.S.A. are very high, and their average contents can reach up to 5,200 ppm and 41,400 ppm, respectively.

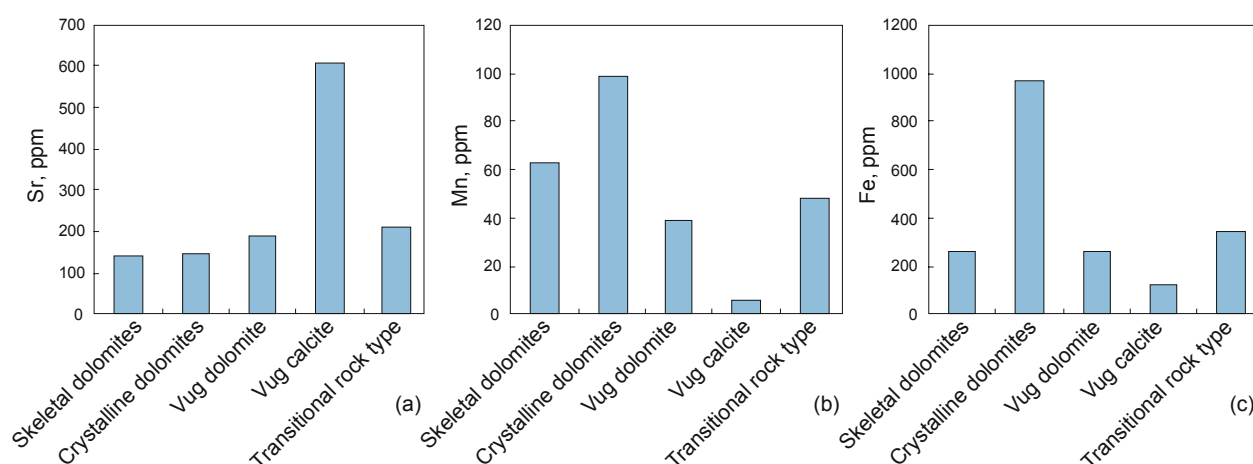


Fig. 3 The histograms of the average values of Sr, Mn and Fe contents from the Permian Changxing Formation reef dolomites

4.2 Characteristics of C and O isotopes

The average $\delta^{13}\text{C}$ values of the reef dolomites are very similar, and mainly range from 2.5‰ to 3‰ (Table 1, Fig. 4(a)). The average $\delta^{13}\text{C}$ value of vug calcite (-2.2‰) is

negative, because it is affected by one sample with very negative $\delta^{13}\text{C}$ value (-9.4‰), and the other two $\delta^{13}\text{C}$ values are -0.6‰ and 3.5‰ (Table 1). The $\delta^{13}\text{C}$ values of skeletal dolomites with original textures are consistent with those of

the transitional rock types, with the average values of 3.1‰. Although the average $\delta^{13}\text{C}$ value of crystalline dolomites is the most negative in the reef dolomites (including transitional rock types), only 2.4‰, the range of $\delta^{13}\text{C}$ value of crystalline dolomites is still similar to the other $\delta^{13}\text{C}$ values of dolomites. Only a few $\delta^{13}\text{C}$ values are less than 2‰, and these values might be affected by the late sparry calcite with strongly negative $\delta^{13}\text{C}$ values (Table 1). The range of $\delta^{13}\text{C}$ values of the reef dolomites is still consistent with that of the Permian Changhsingian seawater (1.5‰-3.5‰) (Korte et al, 2004; Korte and Kozur, 2010).

The average $\delta^{18}\text{O}$ values of the Permian Changxing Formation reef dolomites are also very similar, -4.9‰ (skeletal dolomites), -5.0‰ (crystalline dolomites) and -5.1‰ (vug dolomite) respectively, and these $\delta^{18}\text{O}$ values all concentrate

around the value of -5‰ (Table 1, Fig. 4(b)). Therefore, the three texture types of dolomites may have the same or similar formation environments and dolomitizing fluids. The average $\delta^{18}\text{O}$ values of vug calcite and transitional rock types are negative: -7.04‰ and -6.2‰ respectively (Table 1), and these values are commonly 2‰ less than the reef dolomites. Because of the different oxygen isotope fractionation between dolomite or calcite and precipitation fluids, there is a different $\delta^{18}\text{O}$ value of the coexisting calcite and dolomite from the same precipitation fluid with the oxygen isotope exchange equilibrium, i.e. ‘ Δ problems’ (Land, 1980). The different $\delta^{18}\text{O}$ value of about 2‰ does not simply equal to the different $\delta^{18}\text{O}$ value of their corresponding precipitation fluids. The $\delta^{18}\text{O}$ values of calcite and dolomite of the reef dolomites might be still related to the similar diagenetic fluids.

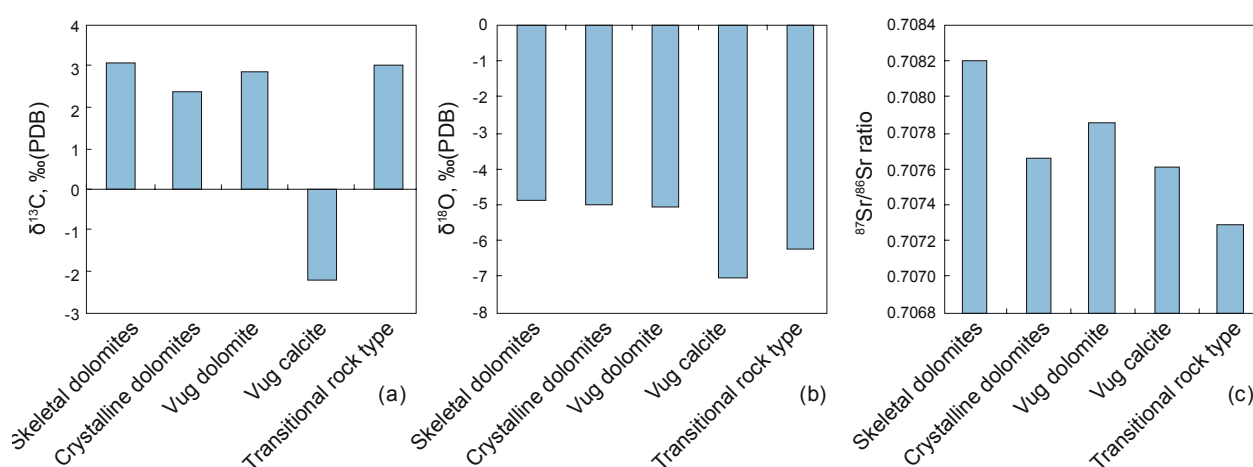


Fig. 4 The histograms of the average values of C, O and Sr isotopes from the Permian Changxing Formation reef dolomites

4.3 Characteristics of Sr isotopic ratio

The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the reef dolomites are significantly different, 0.70821 (skeletal dolomites), 0.70765 (crystalline dolomites) and 0.70786 (vug dolomite) respectively (Table 1, Fig. 4(c)), i.e. the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of skeletal dolomites is the highest, and the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of vug dolomite is moderate, and the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of crystalline dolomites is the lowest (Fig. 4(c)). These average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are higher than that of the vug calcite (Table 1). The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of dolomites and vug calcite are significantly higher than the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Permian Changhsingian seawater (0.7070-0.7073) (Veizer et al, 1999; Korte et al, 2003; 2006). Only the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the transitional rock type (0.70729) is consistent with that of the Permian Changhsingian seawater (Fig. 5). Therefore the dolomites and vug calcite are mixed with high concentrations of radiogenic strontium from external diagenetic fluids which were not coeval marine-derived fluids, and the transitional rock type samples largely retained the Sr isotope composition of the coeval seawater.

5 Source of the dolomitizing fluids

The source of dolomitizing fluids is the starting point of research into the dolomitization mechanism and dolomite

genesis. When we know the source of dolomitizing fluids (it is largely the source of magnesium), we can use the known source of dolomitizing fluids to further discuss the dolomitization mechanism and dolomite genesis. Currently, an important reason for the existing controversy is the lack of adequate knowledge and understanding of the dolomitizing fluids of the reef dolomites, and we do not really know whether the dolomitizing fluids were seawater, meteoric water or a mixture. If the dolomitizing fluids could be determined, many problems about the dolomite genesis and dolomitization mechanism of the reef dolomites would be solved, including the formation environment and genetic mechanism.

The Mn and Fe contents of the Permian Changxing Formation reef dolomites (except the vug calcite) are higher than the average Mn and Fe contents of the overlying Triassic Feixianguan crystalline dolomites, 19.6 ppm and 353.3 ppm respectively (Huang et al, 2006; 2009). A positive correlation between Mn contents and Fe contents indicated that Mn and Fe elements had a similar source (Fig. 6(a)). Moreover, the differences of the textural characteristics and the average values of Mn and Fe contents among different types of the reef dolomites reflect the differences in the timing of their dolomitization. The skeletal dolomites were probably related to the early dolomitizing fluids, and the crystalline dolomites might show the superimposed influence of the late inherited

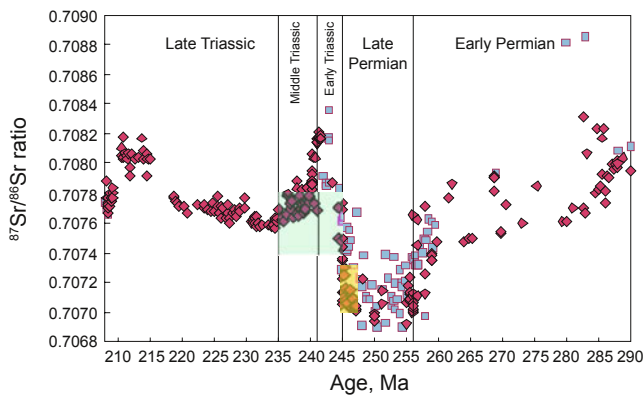


Fig. 5 Sr isotope composition of the Permian-Triassic seawater. Sr isotope data were obtained from Veizer et al (1999), diamond data points were from their Our_Data and square data points were from their Lit_Data, respectively. Geological age of data points and age boundaries were based on Harland et al (1990). The yellow shaded region is the range of the Sr isotope composition of the Permian Changhsingian seawater, and the green shaded region is the main range of Sr isotope composition of the Permian Changxing Formation reef dolomites.

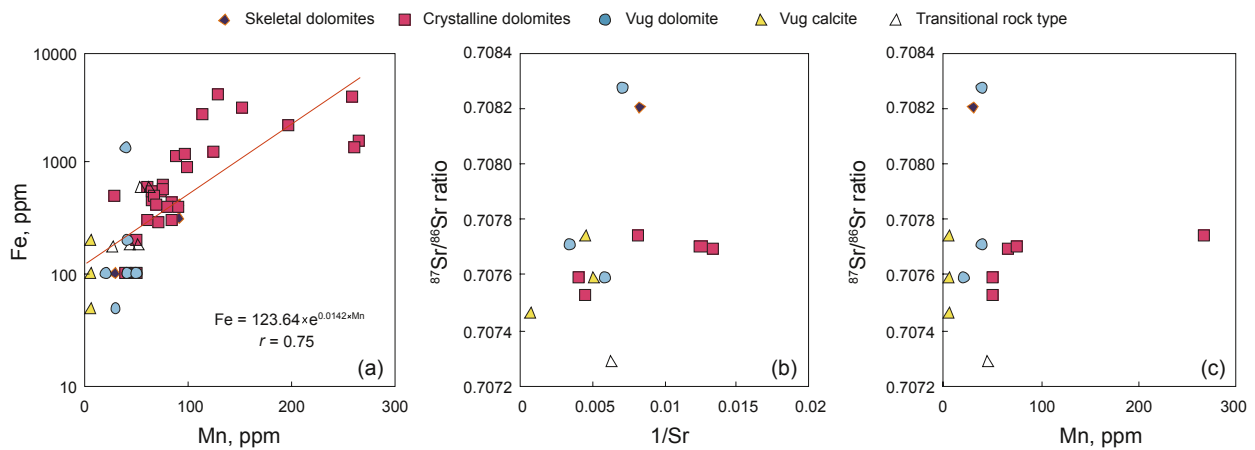


Fig. 6 Crossplots of Mn contents versus Fe contents (a); The reciprocal of Sr contents versus $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (b); Mn contents versus $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (c) of the Permian Changxing Formation reef dolomites

These dolomitizing fluids containing more radiogenic Sr might be mixtures of coeval seawater or marine-derived fluids and a little radiogenic Sr from continental crust, or the marine-derived fluids with higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from adjacent marine strata (or adjacent time interval seawater). What were these dolomitizing fluids after all? In other words, were the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Permian Changxing Formation reef dolomites similar to those of seawater, meteoric water or a mixture of seawater and meteoric water? If mixtures were the source of dolomitizing fluids, what were the proportions of the seawater and meteoric water? In order to quantitatively answer above questions, we used the formula from Stein et al (2000) to quantitatively describe the possible compositions of the Sr isotope when seawater was mixed with meteoric water:

$$\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_L = \frac{^{87}\text{Sr}_{\text{ssw}} \times (1 - f_m) + ^{87}\text{Sr}_m \times f_m}{^{86}\text{Sr}_{\text{ssw}} \times (1 - f_m) + ^{86}\text{Sr}_m \times f_m}$$

or

$$\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_L = \frac{\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{ssw}} \times \text{Sr}_{\text{ssw}} \times (1 - f_m) + \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_m \times \text{Sr}_m \times f_m}{\text{Sr}_{\text{ssw}} \times (1 - f_m) + \text{Sr}_m \times f_m}$$

or

$$f_m = \frac{\left[\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{ssw}} - \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_L\right] \times \text{Sr}_{\text{ssw}}}{\left[\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{ssw}} - \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_L\right] \times \text{Sr}_{\text{ssw}} + \left[\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_L - \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_m\right] \times \text{Sr}_m}$$

where the quantity of the respective materials is given in moles, f denoted the mole fraction, the subscripts L, ssw and m denoted the mixed fluids, the Sr contribution into the mixed fluids from seawater and from meteoric water, respectively.

Based on the above Sr isotope mixing equations, the average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Permian Changhsingian seawater

was 0.70715, i.e. $\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{ssw}} = 0.70715$. The Sr content of the

Permian Changhsingian seawater was 0.097 mmole/L (the average Sr content of modern seawater; Stein et al, 2000). The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the modern river water (0.7119; Palmer and Edmond, 1989) was substituted for the Sr isotope

composition of meteoric water, i.e. $\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{m}} = 0.7119$. The Sr

content of meteoric water was 0.001 mmole/L (the average Sr

content of modern river water; Livingstone, 1963). $\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{L}}$

are corresponding to the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the reef dolomites. As a result, the proportions of seawater and meteoric water in the mixtures were finally known, and then we can know whether or not the dolomitizing fluids were affected by mixtures.

The calculated results from the equations above are shown in Fig. 7. A very large proportion of meteoric water was contained in the dolomitizing fluids of the reef dolomites, mainly from 85% to 95%, and even the proportions of meteoric water in two samples were from 96% to 97%. In other words, the dolomitizing fluids were mixtures of 85%-95% meteoric water and 5%-15% seawater (Fig. 7). According to the typical Dorag dolomitization model which was proposed by Badiozamani (1973), the dolomitization could occur in mixtures of 70%-95% meteoric water and 5%-30% seawater, so the dolomitizing fluids of the Permian Changxing Formation reef dolomites might be mixtures of meteoric water and seawater (Fig. 7). Even if we took the maximum $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Permian Changhsingian seawater (0.7073) as the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of initial seawater

in the calculated equations, i.e. $\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}}\right)_{\text{ssw}} = 0.7073$, the

calculated proportions of meteoric water in the dolomitizing fluids were still mainly concentrated from 85% to 95% (Fig. 7), so the calculated results seemed to support that dolomitizing fluids of the reef dolomites might be related to near-surface mixtures.

If the dolomitizing fluids were very largely composed of meteoric water as derived in the above calculation, the meteoric water played an important role in the dolomitization process. However, such a large proportion of meteoric water (85%-95%; Fig. 7) resulted in a considerable increase of Mn, Fe, ^{12}C and ^{16}O . In fact, the hypothesis of mixtures is very different from the geochemical characteristics of the Permian Changxing Formation reef dolomites, because of the low Mn contents, relatively low Fe contents and high $\delta^{13}\text{C}$ values in the most reef dolomites, and very high Sr contents of some sparry calcite cements formed in the late diagenetic fluids (Table 1). Moreover, the homogenization temperatures of the saline fluid inclusions from the reef dolomites are more than 100 °C, and the main temperature ranges of the saline fluid inclusions from the dolomite and calcite are in good agreement, mainly concentrated between 120 °C and 160 °C

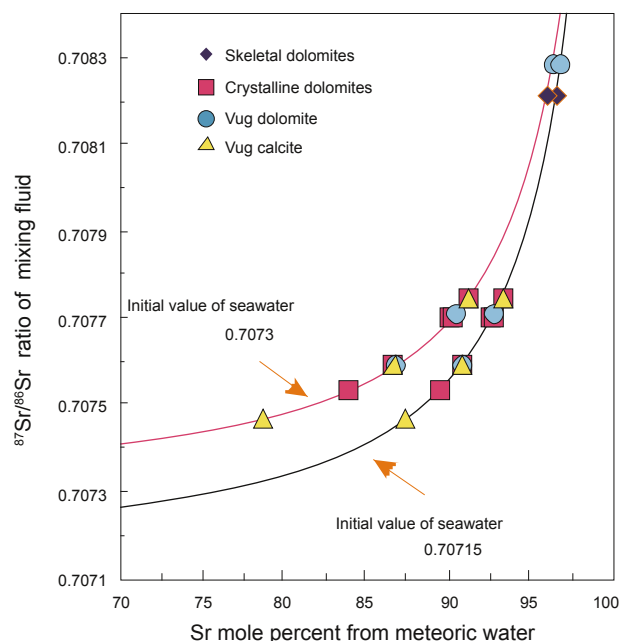


Fig. 7 The calculated proportions of different Sr sources in dolomitizing fluids which are mixtures of Permian Changhsingian seawater and meteoric water (derived from the equations above). The value of 0.70715 as an average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Permian Changhsingian seawater, and the value of 0.7073 as a maximum $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the Permian Changhsingian seawater were used

(Fig. 8), indicating the precipitation of dolomite and calcite is related to the diagenesis fluids under high temperature conditions. The fluids with such a high temperature can not be the near-surface meteoric water or mixtures. For these reasons, there were not large proportions of meteoric water in the dolomitization process, i.e. meteoric water was not a main source of the dolomitizing fluids. In addition, the mixture of migrating fluids and pore fluids from the deeply buried clastic rocks within the basin can also lead to more radiogenic Sr in the dolomitizing fluids. The mixtures would inevitably result in high Mn and Fe contents from clastic rocks. For example, the Mn and Fe contents of the latest dolomite cements from deep-burial diagenetic fluids that had circulated through clastic rocks in the Mesoproterozoic Vazante Formation, São Francisco Basin, Brazil are very high, with maximum Mn and Fe contents of 2,090 ppm and 22,010 ppm, and average Mn and Fe contents of 697 ppm and 9,920 ppm respectively (Azmy et al, 2001). The average Mn and Fe contents of the reef dolomites are low and relatively low, respectively (Table 1). If there were the mixture of throughflows or pore fluids enriched in radiogenic Sr from the deeply buried clastic rocks within the basin, the dolomitizing fluids after mixing should have much higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios than that of coeval seawater (Hu et al, 2009). However, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the reef dolomites were still close to those of coeval seawater (Fig. 5), so the possibility that the dolomitizing fluids of the reef dolomites were mixed with throughflows or pore fluids from the deeply buried clastic rocks within the basin is not supported.

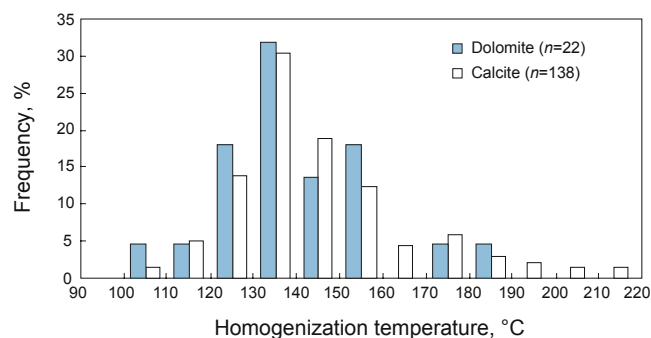


Fig. 8 The histogram of the homogenization temperatures of the saline fluid inclusions from the Permian Changxing Formation reef dolomites

Because the dolomitizing fluids of the reef dolomites were not related to the near-surface meteoric water and throughflows or pore fluids from the deeply buried clastic rocks within the basin, the dolomitizing fluids could be high temperature deep-burial circulated seawater with low Mn and Fe contents, high Sr contents and high $\delta^{13}\text{C}$ values. If this is true, were the dolomitizing fluids close to the coeval seawater or marine formation fluids? Because the ages of the reefs are Late Permian, the deep-burial diagenetic fluids from the underlying Permian strata and overlying Triassic strata were most likely to be the dolomitizing fluids. If the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the reef dolomites were taken as the tracer of dolomitizing fluids, the time intervals with the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio more than the maximum value of the Permian Changhsingian seawater (0.7073) included the Early Permian and the Triassic based on the Sr isotope curve of the Permian-Triassic seawater (Veizer et al, 1999), i.e. only the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Early Permian and Triassic seawater or marine-derived fluids were consistent with the main range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Permian Changxing Formation reef dolomites (0.7075-0.7078) (Fig. 5). However, the lower part of the Lower Permian strata is almost completely eroded, and the Lower Permian Liangshan Formation, Upper Triassic Xujiahe Formation and other overlying strata are all clastic rocks within the basin. Only the Lower Permian Qixia and Maokou formations and Lower-Middle Triassic strata are mainly marine carbonate rocks and evaporites (Geological & Mineral Bureau of Sichuan Province, 1997), so the middle-late Early Permian and Early-Middle Triassic seawater or their deeply buried marine-derived fluids could be the dolomitizing fluids of the reef dolomites. Large-scale potassium-rich and boron-rich subsurface brines have been found in the Lower Triassic Jialingjiang Formation and Middle Triassic Leikoupo Formation within the basin, with the characteristics of leaching and mixing and significant marine-derived source (Lin et al, 2004), and the upper part of the underlying Lower Triassic Feixianguan Formation with widely distributed thick evaporites (mainly consisting of gypsums) also provided favorable conditions for a large amount of brine being enriched in the neighboring strata (Chen, 2005; Zheng et al, 2011). Therefore, the high temperature deep-burial circulated seawater with low Mn and Fe contents, high Sr contents and high $\delta^{13}\text{C}$ values from the dissolution of widely distributed

Triassic evaporites during burial diagenetic processes (including dehydration of water-bearing evaporites) could be the dolomitizing fluids of the reef dolomites. Nevertheless, many questions remain to be satisfactorily answered, such as what were the specific causes of the deep-burial marine-derived brines related to the evaporites, how did the marine-derived brines distribute and migrate in the deeply buried strata, and how did the marine-derived brines participate in the dolomitization of the Permian Changxing Formation reef limestones.

6 Conclusions

1) The average Sr contents of the reef dolomites are similar and relatively low, but the Sr contents of vug calcite are very high. The average Mn and Fe contents of the reef dolomites are also low and relatively low, respectively.

2) The $\delta^{13}\text{C}$ values of the reef dolomites are very similar, mainly ranging from 2.5‰ to 3‰, and the $\delta^{13}\text{C}$ values are consistent with the range of the Permian Changhsingian seawater, but the $\delta^{13}\text{C}$ values of vug calcite can be negative. The average $\delta^{18}\text{O}$ values of the reef dolomites are also very similar, and the $\delta^{18}\text{O}$ values all concentrate around the value of -5‰.

3) The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the reef dolomites are significantly different, and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are significantly higher than the range of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the Permian Changhsingian seawater. The reef dolomites are mixed with high concentrations of radiogenic strontium from external diagenetic fluids.

4) Although the calculated results of the fluid mixing model indicate that the dolomitizing fluids of the reef dolomites could be 85%-95% meteoric water and 5%-15% seawater, the low Mn contents, relatively low Fe contents, high $\delta^{13}\text{C}$ values and high homogenization temperatures of the dolomites did not support that there were large proportions of meteoric water in the dolomitization process, and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios which were close to those of coeval seawater also did not support the possibility of the mixture of deep-burial circulated fluids from clastic rocks.

5) High temperature deep-burial circulated seawater with low Mn and Fe contents, high Sr contents and high $\delta^{13}\text{C}$ values from the dissolution of widely distributed Triassic evaporites during burial diagenetic processes (including dehydration of water-bearing evaporites) could have been the dolomitizing fluids of the reef dolomites.

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