



$$K = \frac{a_{\text{Ca}^{2+}} a_{\text{Dol}}}{a_{\text{Mg}^{2+}} a_{\text{Cc}}}$$

where  $K$  is an equilibrium constant,  $a_{\text{Ca}^{2+}}$ ,  $a_{\text{Mg}^{2+}}$ ,  $a_{\text{Cc}}$  and  $a_{\text{Dol}}$  are the activity of calcium ion, magnesium ion, calcite and dolomite, respectively. The former studies basically assumed that the calcites and the dolomites are pure component minerals, and did not consider the issue of dolomites' degree of order for this formulation, and so  $a_{\text{Cc}} = a_{\text{Dol}} = 1$ . As a result, the above equilibrium constant expression can be simplified as  $K = a_{\text{Ca}^{2+}} / a_{\text{Mg}^{2+}}$ .

Due to the non-occupational assignment of positive ion for calcites, their activity can be replaced by the activity of pure component minerals, that is  $a_{\text{Cc}} = 1$ . However, the activity of dolomites is more complicated than that of calcites. If the components of dolomites have a departure with ideal compositions and the arrangement of ions are not well ordered, it was called protodolomite by Graf et al.<sup>[9]</sup>. Even if the components of dolomites during the period of diagenesis are very close to the ideal chemical compositions, the part-ordered phenomenon of ions arrangement still exists. So a variable for degree of order,  $s$ , is introduced into this paper to determine the above equilibrium constant. The ideal crystalchemical formula for dolomites is  $\text{CaMg}(\text{CO}_3)_2$ , in which calcium ion and magnesium ion are distributed in the place of M2 and M1, respectively. Based on the theoretical model of Bragg-Williams<sup>[10]</sup>,  $s = 2X_{\text{Ca}}^{\text{M2}} - 1 = 2X_{\text{Mg}}^{\text{M1}} - 1$ , where  $X_{\text{Ca}}^{\text{M2}}$  and  $X_{\text{Mg}}^{\text{M1}}$  represent the mole fraction of Ca and Mg in the place of M2 and M1, respectively. If  $s = 0$ , it represents complete disordered dolomite; if  $s = 1$ , it represents ordered dolomite. Consequently, the activity of ordered components of dolomites ( $a_{\text{oDol}}$ ) and the activity of disordered components of dolomites ( $a_{\text{dDol}}$ ) in dolomites crystals are

$$a_{\text{oDol}} = X_{\text{Ca}}^{\text{M2}} X_{\text{Mg}}^{\text{M1}} = (1 + s)^2 / 4$$

$$a_{\text{dDol}} = 4(X_{\text{Ca}}^{\text{M2}} X_{\text{Mg}}^{\text{M2}} X_{\text{Ca}}^{\text{M1}} X_{\text{Mg}}^{\text{M1}})^{0.5} = 1 - s^2$$

and then the above equilibrium constant expression can be substituted by the activity of dolomites and related calculations can be performed.

During the period of diagenesis for clastic rocks,

dolomites can be regarded as deuterite cements, which have been explicitly proven by the research findings of buried diagenesis of sandstones<sup>[11, 12]</sup>. This kind of dolomites generally contains  $\text{Fe}^{2+}$ , and its content increases with depths and finally ankerites may appear<sup>[11]</sup>. Although quite a bit of  $\text{FeCO}_3$  ( $< 0.05 - 17 \text{ mol}\%$ ) can be contained in dolomites, they only have a very small impact on the above reaction equilibrium, especially, the ratio of Ca/Mg for water solution in the equilibrium system does not change obviously<sup>[13]</sup>. Therefore, the impact of  $\text{Fe}^{2+}$  in the activity of dolomites expression can be ignored.

For the dolomites crystals with closely ideal components their degree of order ( $s$ ) depends on temperature. When dolomites are in the equilibrium system under some geological environments, their degree of order changes a little and can be regarded as a constant<sup>[6]</sup>. Thus, the equilibrium constant ( $K$ ) becomes the measurement of  $a_{\text{Ca}^{2+}} / a_{\text{Mg}^{2+}}$  in water solution. The result is that the chemical equilibrium between calcites and dolomites is controlled by the ratio of Ca/Mg for groundwater in the environment of diagenesis. If the ratio of Ca/Mg for groundwater decreases, the expression of (1) will change to the direction of dolomites' sedimentation and calcites' solution and the dolomites will gradually replace calcites in clastic rocks cements. Otherwise, if the ratio of Ca/Mg for groundwater increases, the dolomites will dissolve and be replaced by calcites. The equilibrium constant ( $K$ ) is the function of temperature and pressure, among which they have the following thermodynamics expression:  $\Delta G^0 = -RT \ln K$ , where  $R$  is the real gas constant, and  $\Delta G^0$  is the Gibbs free energy difference in the condition of standard state, which is the function of temperature and pressure. The thermodynamic parameters for calcites and dolomites originate from the minerals database of Helgeson<sup>[14]</sup> and Berman<sup>[15]</sup> and those for calcium ion and magnesium ion from the thermodynamic model for hydrated ion<sup>[16, 17]</sup> in the referential condition of  $25^\circ\text{C}$  and  $10^5 \text{ Pa}$ . By using the calculation software SUPCRT92<sup>[18]</sup>, which is for the simulation of thermodynamics phase equilibrium, a thermodynamics phase relationship diagram for the system of calcites, dolomites and water can be constructed (Fig. 1).

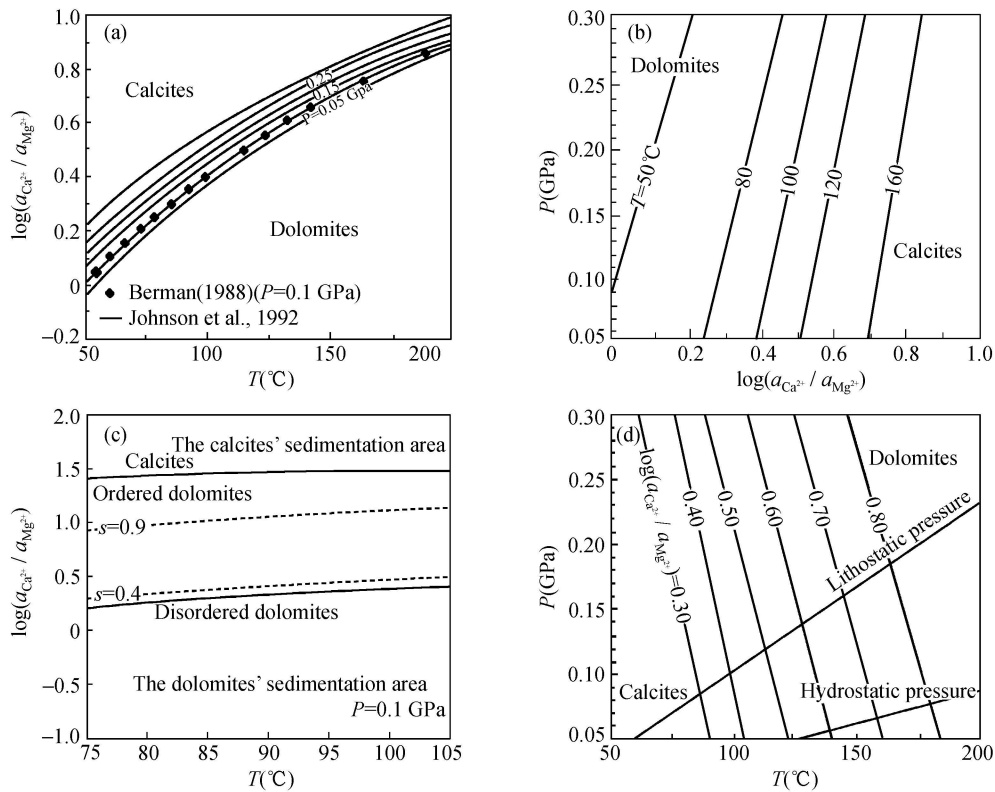


Fig. 1. Phase diagram for the system of calcite, dolomite and water. (a) The pressure contour; (b) the temperature contour; (c) the degree of order for dolomite; (d) the lithostatic pressure based on the rock density of  $2.65 \text{ g/cm}^3$ .

## 2 The interaction between pores and fluids of Kela 2 gas field in Kuche sag

The Kela 2 gas field in Kuche sag, which is in the northern part of Tarim basin, is one of the major large gas fields in China. The buried depths are generally greater than 3600 m, and the reservoirs are clastic rocks from the Palaeogene to the Cretaceous System. From 3600 m down to 4000 m, the porosity and permeability are abnormally high, where the porosity is from 10% to 20%, averaged about 12.5%, which greatly deviates from the change trend of pore volume for the normal compaction of sediments (Fig. 2). The belts with high porosity and high permeability are completely in accord with the belts with abnormally high pressure in the studied area. Taking the Kela 201 well as an example, the authors aim to discuss the interaction between carbonate cements and pore fluids and its impact on reservoir quality for clastic rocks reservoirs.

### 2.1 The relationship between carbonate cements and porosity and permeability

In the studied area, two main carbonate cements in clastic rocks reservoirs are calcites and dolomites,

and the cement content is closely related to the porosity and permeability (Fig. 2). Porosity and permeability increase by depths from 3600 m to 3800 m, corresponding to an obvious descending trend for the total content of carbonate cements. However, they decrease by depths from 3800 m to 4000 m and with an increasing trend for the total content of carbonate cements. Apparently, there is the feature of high porosity and permeability but low content of carbonate cements around 3800 m, which shows that a large amount of carbonate cements were dissolved, moved out and formed a lot of secondary pores at the depths. In fact, further analysis of the content of carbonate minerals shows that the change of total content of carbonate minerals is really caused by the change of the content of calcites in cements. The content of dolomites shows little change along the section, but that of calcites is in negative correlation with porosity and permeability (Fig. 2). The relationship between the content of calcites in cements and porosity and permeability as well shows that the dissolution and sedimentation of calcites play a critical role in forming secondary pores. Although dissolution of feldspar grains has some contributions to the formation of secondary pores, it is actually small in amount and has a

relatively uniform longitudinal distribution in the section<sup>[19]</sup>. Consequently, speculation can be made as that the change of pore fluid property during the dia-

genesis at the middle depths in the studied area results in the dissolution of calcite cements and causes the formation of intergranular dissolved secondary pores.

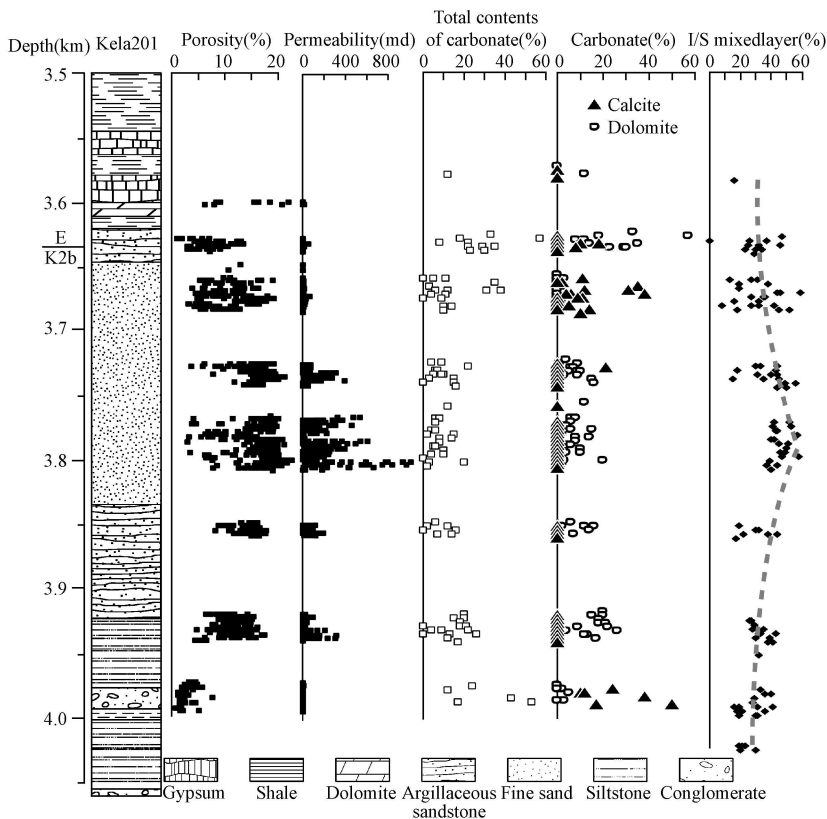


Fig. 2. Porosity, permeability, content change of carbonates minerals and I/S mixedlayer minerals of Kela 201 well in Kela 2 gas field

2.2 Pore water chemistry and interaction between water and rocks

The geothermal gradient of Kela 2 gas field is 2.1 °C/100 m. The logging temperature during drilling shows that the reservoir temperature is basically from 92 °C to 100 °C and the formation pressure

is around 0.07–0.09 GPa. The components of groundwater in the studied area were measured by spectrophotometer and the ion strength of most solution is lower than 3.0. Therefore, the activity of Ca<sup>2+</sup> and the activity of Mg<sup>2+</sup> can be approximately computed using the activity model of Debye-Huckle (Table 1).

Table 1. Formation water components (mMol/L), ion strength (I), and ionic activity of a<sub>Ca<sup>2+</sup></sub> and a<sub>Mg<sup>2+</sup></sub> of Kela 2 gas field

Horizon	Depth (m)	T (°C)	pH	mMol/L									I	a(Ca <sup>2+</sup> ) × 10 <sup>3</sup>	a(Mg <sup>2+</sup> ) × 10 <sup>3</sup>	
				CO <sub>3</sub> <sup>2-</sup>	HCO <sub>3</sub> <sup>-</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Na <sup>+</sup>	K <sup>+</sup>	Fe <sup>2+</sup>				B <sup>3+</sup>
E	3600	92.4	6.68	0.0	22.1	892.5	49.7	18.7	10.8	910.4	28.2	0.1	5.4	1.1	2.97	2.42
E	3600	92.4	6.28	0.0	21.0	817.3	30.9	19.2	7.6	798.6	29.6	0.1	6.0	1.0	3.17	1.74
E	3600	92.4	6.2	0.0	19.0	786.0	44.0	27.2	9.7	776.7	26.6	0.3	5.0	1.0	4.47	2.23
E	3600	92.4	7.57	0.0	17.1	535.5	16.2	16.0	8.1	505.5	19.3	0.0	4.0	0.6	3.01	2.03
E	3630	93.1	7.7	0.0	29.0	128.1	14.0	6.2	0.0	175.0	0.0	0.2	0.0	0.2	1.70	0.00
E	3630	93.1	7.42	0.0	30.4	814.2	2.6	3.2	4.9	812.9	12.9	1.0	2.0	0.9	0.55	1.14
K2b	3665	94.0	8.1	0.0	12.0	55.5	5.7	1.1	2.8	70.1	0.8	0.1	0.0	0.1	0.38	1.14
K2b	3665	94.0	8.07	0.0	22.6	144.1	7.9	4.8	5.9	127.7	19.9	0.6	4.0	0.2	1.30	1.93

To be continued

Continued

Horizon	Depth (m)	$T(^{\circ}\text{C})$	pH	mMol/L										I	$a(\text{Ca}^{2+}) \times 10^3$	$a(\text{Mg}^{2+}) \times 10^3$
				$\text{CO}_3^{2-}$	$\text{HCO}_3^-$	$\text{Cl}^-$	$\text{SO}_4^{2-}$	$\text{Ca}^{2+}$	$\text{Mg}^{2+}$	$\text{Na}^+$	$\text{K}^+$	$\text{Fe}^{2+}$	$\text{B}^{3+}$			
K2b	3665	94.0	8.1	0.0	23.0	187.9	6.3	5.3	5.4	197.7	2.8	0.6	0.0	0.2	1.38	1.70
K2b	3665	94.0	7.65	0.0	15.1	43.8	9.4	5.3	2.7	38.9	13.7	0.0	3.0	0.1	1.83	1.05
K2b	3665	94.0	7.48	0.0	16.6	40.7	6.8	3.2	4.3	25.9	17.9	0.0	4.0	0.1	1.12	1.71
K2b	3728	95.6	6.8	0.0	2.8	3175.8	3.0	202.2	40.1	2646.6	34.7	2.6	3.8	3.4	23.99	7.42
K2b	3728	95.6	6.1	0.0	3.0	3073.5	20.5	194.0	28.7	2620.1	34.0	2.0	4.4	3.4	23.13	5.33
K2b	3770	96.0	8.6	1.9	30.1	97.4	13.5	0.0	2.7	155.4	0.0	0.3	0.0	0.2	0.00	0.91
K2b	3770	96.0	8.08	0.0	15.8	40.5	5.2	3.2	2.7	15.2	23.7	0.0	5.4	0.1	1.13	1.08
K2b	3770	96.0	6.41	0.0	7.9	223.0	6.8	24.0	5.4	134.2	30.7	0.5	6.6	0.3	5.77	1.61
K2b	3770	96.0	7.85	0.0	23.9	51.7	0.5	0.5	7.6	0.0	35.8	0.0	9.2	0.1	0.17	2.84
K2b	3770	96.0	7.23	0.0	17.7	440.9	9.9	35.0	8.4	352.0	24.1	0.0	5.3	0.5	6.90	2.17
K2b	3770	96.0	7.9	0.0	43.5	110.5	4.2	0.5	3.2	125.8	17.3	0.0	3.9	0.2	0.15	1.09
K2b	3770	96.0	6.78	0.0	18.6	507.8	39.8	42.4	10.8	465.7	21.3	0.4	3.9	0.7	7.71	2.65
K2b	3918	98.8	10	52.8	0.0	1986.5	0.0	87.2	8.8	1468.8	196.7	0.1	78.2	2.5	11.17	1.71
K2b	3918	98.8	9.13	66.0	0.0	2044.0	1.4	87.2	4.4	2086.0	0.0	0.2	11.7	2.5	11.14	0.86
K2b	3918	98.8	8.59	26.4	0.0	2038.8	0.0	87.2	5.9	1973.0	0.0	0.2	0.0	2.3	11.40	1.16
K2b	3926	98.9	6.8	0.0	9.9	2608.9	8.6	152.4	40.8	2190.3	37.3	0.0	7.4	2.9	18.87	7.77
K2b	3936	99.1	6.63	0.0	11.9	2453.9	8.4	142.0	33.5	2062.3	42.9	0.1	8.7	2.7	17.83	6.44
K2b	3936	99.1	6.7	0.0	9.5	2255.2	6.0	128.4	32.7	1882.5	44.4	0.0	9.2	2.5	16.46	6.36
K2b	3936	99.1	5.91	0.0	4.8	2749.8	8.9	161.2	37.0	2315.4	38.0	0.6	7.1	3.0	19.73	7.00
K2b	3936	99.1	5.56	0.0	2.5	2666.7	2.1	158.0	38.1	2225.0	35.3	0.5	6.6	2.9	19.51	7.24

With the increase of depth, the groundwater in the studied area can be classified as sodium sulphate type ( $< 3600$  m), sodium bicarbonate type (3630—3795 m), and calcium chloride type ( $> 3795$  m). The pH value of groundwater is from 5.6 to 8.1, presenting weak alkali to neutrality. The pH value of sodium bicarbonate type groundwater is generally higher than 7, but the pH value of the calcium chloride type is mostly lower than 7. Degree of mineralization is usually 5—17 g/L, among which most sodium sulphate type water below rock salts and calcium chloride type water deeper than 3900 m obviously have higher degree of mineralization than that at the depth from 3750 m to 3850 m which is usually lower than 50 g/L. Groundwater with low degree of mineralization benefits to the mineral dissolution in rocks. Sedimentation and dissolution of authigenic minerals such as calcite formed during the diagenesis are closely related to the chemical property of groundwater. The horizons in which large quantities of calcites occur correspond completely to the sodium bicarbonate type water and those in which calcites are basically

dissolved out with the calcium chloride type water. The horizons with the sodium sulphate type water usually scatter with a small amount of calcites.

The components of formation water control the chemical equilibrium between pores and fluids in the process of diagenesis. Particularly, the ratio of Ca/Mg in water has a great impact on the equilibrium between calcites and dolomites. The change of the degree of order of dolomites in sedimentary rocks results in great differences for the equilibrium conditions among calcites, dolomites and fluids. Fig. 3 shows that the requirements of the solution strength of Ca/Mg for the equilibrium between calcites and ordered dolomites are much stricter than that between calcites and disordered dolomites. The groundwater ( $T < 90^{\circ}\text{C}$  and  $T > 103^{\circ}\text{C}$ ) in the high pressure belt of Kela 2 gas field in Kuche sag is mainly distributed in the equilibrium accrete area of calcites and dolomites with the degree of order from 0.4 to 0.9 plus calcites, corresponding to the widespread appearance of carbonate cements of dolomites plus calcites observed in the reservoirs at the depth shallower than 3700 m

and deeper than 3900 m, in which the porosity and permeability apparently become low (Fig. 2), where their corresponding flux of pore fluids is small, infiltration rate is low, and the pore fluids have enough residence time in rocks bringing about tightly approximate chemical reaction equilibrium between pores and fluids. Consequently, expression (1) of chemical reaction between calcites and partly ordered dolomites (the degree of order is from 0.4 to 0.9) and pore fluids basically reaches an equilibrium state. In such a low porosity and low permeability condition, the system of pores and fluids is a fairly good buffer system. That is to say, the ratio of Ca/Mg in solutions in such a condition depends mainly on the equilibrium reaction between calcites and dolomites in rocks and the degree of order of dolomites. The limited change of fluids components cannot change the ratio of Ca/Mg in solutions and the equilibrium reaction between calcites and dolomites in rocks at all. This is just significant evidence to show that people measure the equilibrium between calcites and dolomites at low temperature by means of the system of carbonate and fluids in nature<sup>[6]</sup>.

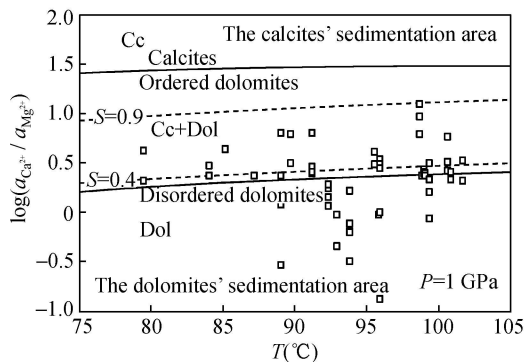


Fig. 3. Pore water and carbonate mineral phase diagram of Keh 2 gas field ( $s$  is the dolomites' degree of order).

Fig. 3 shows that the groundwater at the middle depths where the temperature is 90–103 °C of Kela 2 gas field in Kuche sag is basically aviated from the equilibrium syngenetic zoning of dolomites plus calcites but is included within the sedimentation area of dolomites. When this type of groundwater penetrates into rocks, the calcites cements in pores will be dissolved, which results in the sedimentation of dolomites. Even if the dolomites cements in pores are completely disordered (such kind of dolomites exists hardly in nature), there is still considerable amount of solution which is severely undersaturated to calcites, causing almost complete dissolution of the cal-

cites in the reservoirs at the middle depths in the studied area, and generating a great number of pores. This is fully consistent with the situation that the calcites almost disappear and the porosity and permeability are very high around 3800 m as shown in Fig. 2. On the other hand, the features of high porosity and permeability in the reservoirs at the middle depths in the studied area contribute to the high flux of groundwater and high speed of penetration. The pores in rocks are always in a condition of eluviation and erosion by groundwater and so expression (1) of chemical reaction is hard to get equilibrium among calcites, dolomites and groundwater in pores. Relatively, the calcites with a higher solubility are gradually dissolved by pore water and move out from rocks resulting in more secondary pore generated in rocks. However, the dolomites with an extremely low solubility can constantly precipitate from the pore fluids. Meanwhile, the  $Fe^{2+}$  released from the transformation of montmorillonites to illites and the decompose of chlorites combines into the crystal lattice of dolomites and forms ankerites. Obviously, these systems of pores and fluids in the belts with high porosity and permeability are not ideal buffer systems and it is impossible to achieve the equilibrium thermodynamic parameters of calcites, dolomites and water solution in a low temperature condition from these systems of water and rocks with high porosity and permeability.

### 3 Discussion and conclusions

Based on the thermodynamic database of carbonate minerals and related hydrated ion, according to the theoretical model of Bragg-Williams, this paper establishes a thermodynamic phase relationship diagram among calcites, dolomites and water solution by introducing a parameter  $s$ , degree of order, into dolomites. Study shows that the temperature has a major impact on the chemical reaction system of calcites, dolomites and water solution, but the pressure has a minor impact on it. Due to the penetration and fluidity of groundwater in clastic rocks, the temperature of these pore water changes with the environment of ground temperature when water penetrates and flows in rocks. Consequently, it will definitely have an impact on the equilibrium between calcites and dolomites in the pores. The surface water or the water close to the surface such as the depositional water in equilibrium between calcites and dolomites originally will result in unsaturated calcites with the rising temperature when it penetrates into the deep part.

Thus, calcites will be dissolved and transform into dolomites, causing the dolomitization. Similarly, the compaction water from deep thermobaric water or mudstones migrating upwards along the decreasing trend of pressure gradient will lead to the dissolution of dolomites and the precipitation of calcites, because the compact water will become cold and be unsaturated for dolomites, causing the dedolomitization (Fig. 1 (a)). Such an underground reaction has been proven by Land et al.<sup>[20]</sup> and Budai<sup>[21]</sup>.

In the reservoirs of Kela 2 gas field in Kuche sag, the equilibrium system of calcites, dolomites and pore water appears in rocks with low porosity and low permeability both at the depths of shallower than 3700 m and deeper than 3900 m, while the calcites in carbonate cements in the belts with high porosity and high permeability at the middle depths (about 3800 m) almost disappear completely (Fig. 2). Based on the above thermodynamic phase relationship diagram, speculation can be made as follows: ① The ratio of Ca/Mg for groundwater at the middle depths ( $90^{\circ}\text{C} < T < 103^{\circ}\text{C}$ ) is obviously lower than that at the low depths and at the deep depths in the studied area. Thus, such type of groundwater chemistry shows that it is impossible for abundant sedimentary water to infiltrate to the reservoirs at the middle depths and dissolve the calcites. In addition, the caprock of rock salts overlying the reservoirs with huge thickness prevents from the infiltration of sedimentary water. ② The low ratio of Ca/Mg for pore water at the middle depths can interpret the dolomitization in reservoir rocks but cannot fully explain the appearance of abundant secondary pores at the depths (Fig. 2). Only the dissolution of calcites caused by the formation water with rich  $\text{H}^+$  into reservoirs is the real factor of the appearance of the belts with high porosity and high permeability at the middle depths.

The ratio of Ca/Mg for pore fluids in the process of diagenesis depends not only on the chemical equilibrium between calcites and dolomites in pores and fluids, but also on the degree of order of dolomites. The magnitude of degree of order parameter reflects the age and the temperature of diagenesis. Generally, the higher the temperature is, the longer the time will be, and also the higher the degree of order will be. The calcites and dolomites at the lower depths (lower than 3700 m) and at the deeper depths (deeper than 3900 m) will basically keep a state of thermodynamic equilibrium phase between calcites to

dolomites and pore fluids in rocks with low porosity and low permeability. Dolomites show a partly ordered condition and its degree of order parameter is between 0.4 and 0.9. Due to the low geothermal gradient in the studied area, speculation can be made that the high degree of order is the direct response to the long time of burial diagenesis and the temperature has a minor impact on it. Similarly, high degree of order corresponds to high ratio of Ca/Mg for the system of calcites, dolomites and pore fluids in reservoirs (Fig. 3).

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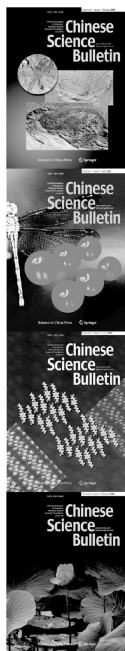
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