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RESEARCH ARTICLE

Characteristics of rare earth elements of lacustrine exhalative rock in the Xiagou Formation of Lower Cretaceous in Qingxi sag, Jiuxi basin

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Abstract The exhalative rock occurring in the Xiagou Formation of Lower Cretaceous in Qingxi sag, Jiuxi basin is a sort of a rare lacustrine white smoke type, rich in ferrodolomites and albites. This paper introduces the geological background, mineral association, and lithology of the exhalative rock, and discusses its REE geochemical characteristics and connection with hydrothermal environment. It is shown that the exhalative rock has basal characteristics of hydrothermal depositional formation of LREE > HREE, with positive δCe and negative δEu, which is different from the characteristics of marine exhalative rock. Since the REE pattern and exceptional distribution of δEu and δCe are highly similar to the characteristics of alkalescent tholeiitic basalt in the same layer, the exhalative rock of Xiagou Formation is considered to be closely related to the origin of basalt in the same horizon. The fact that the amount of REE of exhalative rock decreases outwards indicates that exhalative rock in the Xiagou Formation may be connected with lacustrine hydrothermal convection circulation.

Keywords Jiuxi basin, Lower Cretaceous, exhalative rock, rare earth element, distribution pattern, Ce anomaly, Eu anomaly

1 Introduction

Exhalative rock or hot water mineralization is becoming one of the main focuses of research in the field of geoscience. However, the major achievement has been concentrated on exhalative rock formed in the oceanic basin, and little has been done with the exhalative rock formed in the lacustrine basin (Li and Feng, 2003; Ye et al., 2002; Liu et al., 2001; Wang et al., 1998; Jean-Jacques, 1994; Xiao et al., 1994; Brown, 1993; Li, 1993). Generally, exhalative rock has been classified into black smoke type and white smoke type on the basis of the mineral assemblages. The former is mainly composed of silicate, aluminosilicate, and carbonate minerals, with gypsum and barite as secondary minerals, and with oxides, hydroxides and sulfates as the minor minerals. The latter has the same major mineral assemblage as the former, but with the occurrence of abundant sulfates, such as copper, magnetic pyrite, galena and zinc blende.

2 Geological backgrounds

Located at the intersection of the northern Qilian tectonic belt and the Altun tectonic belt, the Qingxi sag is composed of three subsidiary tectonic units, i.e. Hongnan sag, Qingnan sag and Qingxi low swell. The white-smoke-type exhalative rock that occurred in the Xiagou Formation of Lower Cretaceous mainly distributes in the southern deep depression of the Qingnan sag, with the rest occurring in the north of the Qingnan sag, the east of Hongnan sag and the south of the Qinxi low swell (Fig. 1).

The macroscopic appearance of exhalative rock in the area is characteristic of muddy dolomite or dolomitic mudstone and it has been considered, in the past, as sediment tuff resulted from the sedimentation and diagenesis of the volcanic ash. However, synthetic research on the geological background, mineral compositions, lithological textures and the geochemical characteristics of trace elements and rare earth elements revealed that it is white-smoke-type exhalative rock of lacustrine facies, with a mineral assemblage of barite-albite-ferrodolomite. On the vertical profile, the exhalative rocks are intercalated with normal sedimentary shale, together with some thin to mid bedded siltstone and fine-grained

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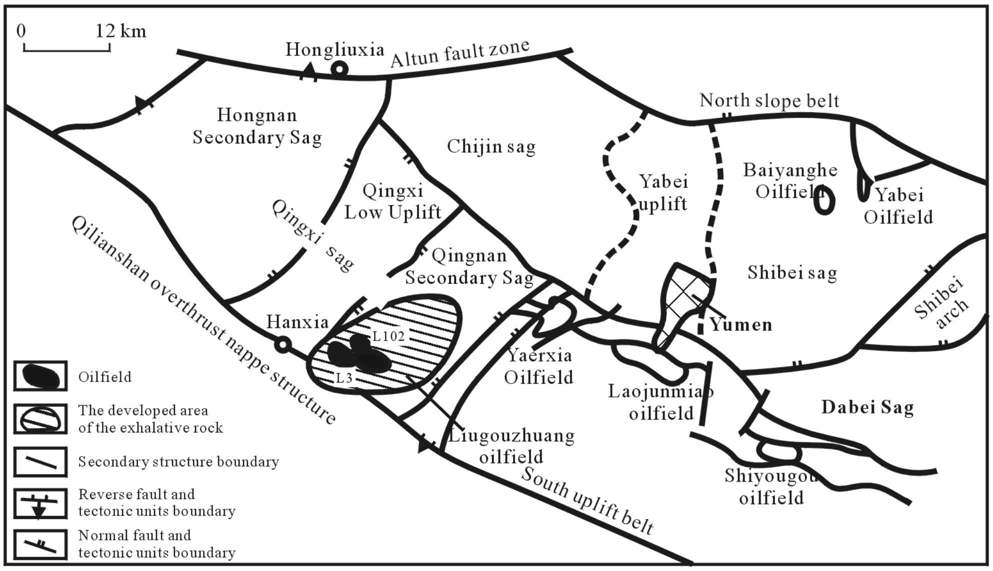

Fig. 1 Simplified map showing the location and distribution of exhalative rocks of the Xiagou Formation

sandstone. Its total thickness ranges from 800–1 000 m and it belongs to deep lacustrine sedimentation. Five exhalation-sedimentation cycles can be classified in terms of stratigraphic cyclicity. The cumulate thickness of exhalative rock for each cycle is varied, ranging from 10–250 m, with a general thickness of 50–150 m. Its distribution scope and thickness increase with the increment of the area of lacustrine basin and the depth of the water. Of the five cycles, the MSC3 and MSC4 possess the largest thickness and distribution scope, characterized by a framework of alternation of high-value sedimentary area (>100 m) with low-value sedimentary area (<50 m), which strikes in the northeast-southwest direction (Fig. 2). The exhalative rock is of micritic-aphanitic texture and laminated structure with the ferrodolomite (20%–66%) and albite (12%–75%) as its major hot water mineral components, and with barite (5%–20%), analcite (2%–18%), quartz (3%–14%), dickite (2%–15%) and pyrite (0.3%–6.4%) as its secondary mineral components. Clastic zinc blende, copper and galena occur in the rock occasionally. Microprobe analysis indicates that the elements of the mineral components are basically in accordance with those of chemical calculation. Ferrodolomite- and albite-rich white-smoke-type exhalative rock of lacustrine facies has been rarely found and studied. Therefore, its research is of great significance.

3 Characteristics of paragenetic association and fabrics for hot water minerals in the exhalative rock

3.1 Characteristics of paragenetic association of hot water minerals

Six paragenetic associations of hot water minerals in the exhalative rocks of the Xiagou Formation are recognized. They are (1) albite-ferrodolomite or ferrodolomite-albite association; (2) quartz-albite-ferrodolomite association; (3) quartz-barite-ferrodolomite association; (4) quartz-analcite-albite-ferrodolomite association; (5) dickite-ferrodolomite association and (6) ferrodolomite association. Pyrite and abundant organic materials occur in all of the paragenetic associations. Minor sulfates of Zn, Cu and Pb only appear in the (2) and (4) associations and the normal sedimentary components of quartz silt and mud mainly occur in the (1) and (6) associations.

3.2 Fabric characteristics of exhalative rock

The dominant fabrics for the exhalative rocks in the Xiagou Formation are characterized by the micritic-aphanitic textures and laminated structures. Typical hot water clastic texture, hot water breccia texture, banded structure, network vein structure, vortex structure and deformation bedding only partially exist in the rocks. These different textures and structures reflect various genetic significances.

In fact, a laminated structure results from the alternated intercalation of different minerals in the (1) and (5) associations. The laminated layers of albite, barite, analcite, and dickite appear mainly in the depth of the sag, reflecting a sedimentary environment of stable accumulation of hot brine.

Hot water intraclastic textures can be further divided into two types, the calcarenitic texture and calciruditic texture, respectively. The former is made up of aggregates of albites and barites with occasional appearance of minor zinc blende, copper, and galena. Calciruditic texture chiefly develops in the laminated exhalative rocks composed of albite and ferrodolomite. The size of breccias is varied, which are infilled and cemented by albite, barite, analcite, and dickite. Hot water intraclastic texture has been considered to be related to the

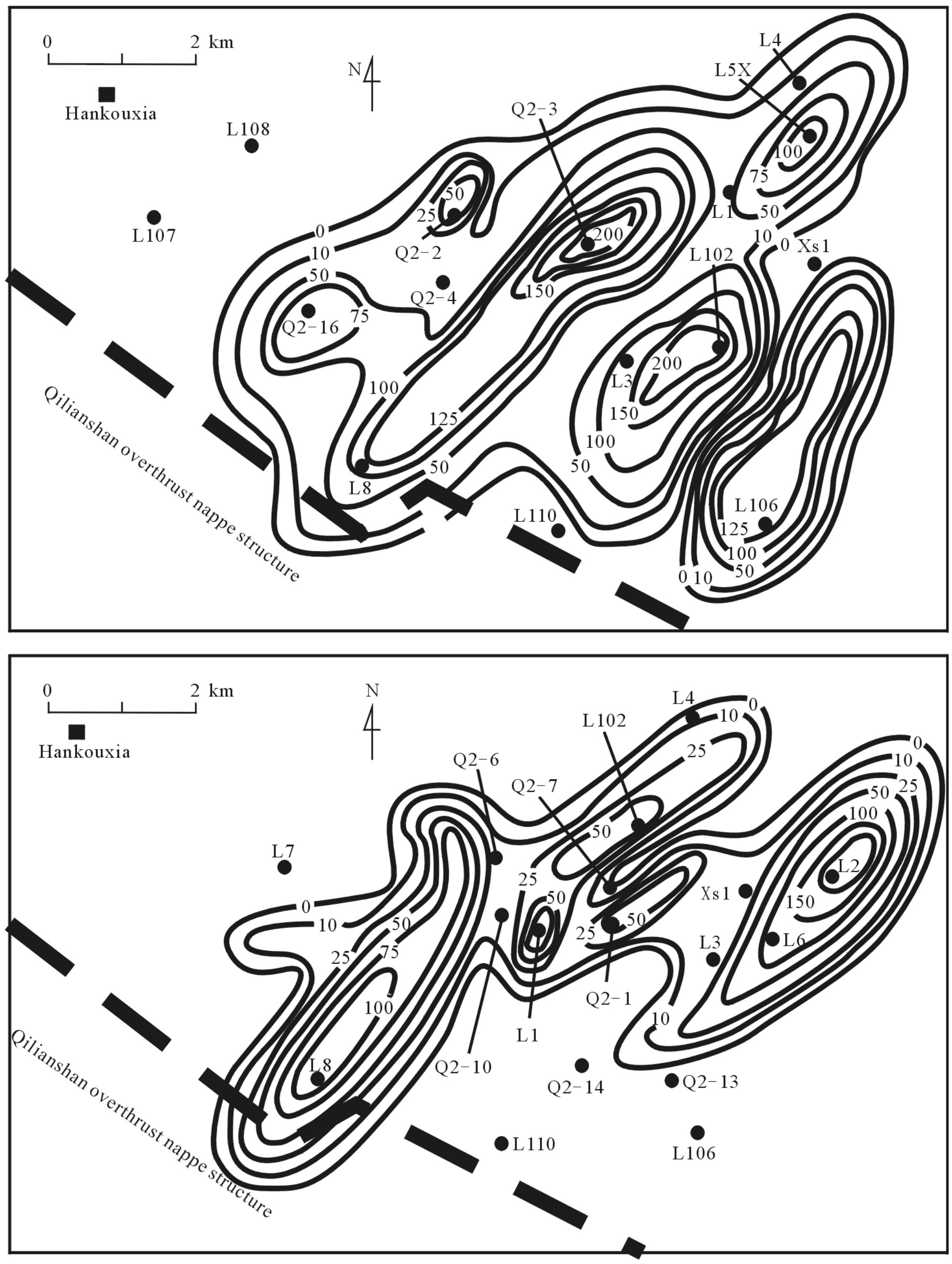

Fig. 2 Simplified map showing the sampling location and the variation of exhalative rock thickness for the MSC3 and MSC4 sequences

boil and burst of high-temperature and high-pressure hot water at the exhalative mouth. The breccias accumulated around the exhalative mouth are cemented quickly by hot water minerals, and therefore, they are usually called water-bursting breccias, which can be used to determine the position of exhalative mouth. Sandy clasts are taken by exhalative flow to the outboard of the exhalative mouth and appear as floating state in the lamellae.

Network vein structure commonly occurs in the exhalative rock and results from the filling in fissures by hot water minerals of albite, analcite, and barite. It is considered as an indicator to determine the migration passage of hydrothermal solution.

Vortex structure is uncommon in the exhalative rock and usually has a diameter of a few millimeters. Its outer circle occurs as concentric lamina composed of intraclasts of albite, barite, and ferrodolomite, and its centric pipe is filled by crystallized albite and barite, which represents the micro-exhalative mouth in the exhalative belt.

Contemporaneous deformation bedding is characterized by the irregular soft deformation, crenulation and micro-shearing. The collapse due to the steep chimney-shaped topography during the rapid accumulation of hot water minerals is considered to be responsible for the formation of this kind of special structure. Accordingly, this structure can be used to determine the location of exhalative mouth.

3.3 Classification of genetic types for exhalative rocks

The exhalative rocks in the Xiagou Formation can be classified into four genetic types on the basis of their paragenetic mineral association and fabric characters. The first type is vein filled exhalative rock formed in the exhalative mouth, with a paragenetic association of aluminosilicates and sulfates. The second one is the water-bursting exhalative rock formed at the exhalative mouth, with a paragenetic association of carbonates, aluminosilicates, and sulfates. The basin-deposited exhalative rock belongs to the third type, which is formed in the brine pool near the exhalative mouth, with a paragenetic association of carbonates, aluminosilicates, and sulfates. The last one is the regional distribution exhalative rock of carbonate mineral association which widespread in the sedimentary basin.

4 Geochemical characteristics of rare earth elements

Drill cores are sampled for rare earth elements analysis and rock from the fresh outcrop on the ground are taken as the correlative specimens. The microprobe analysis results for REE are listed in Table 1.

4.1 Σ REE and distribution pattern of REE

The variation scope of Σ REE of the exhalative rock is very large, ranging from 6.55×10^{-6} to 213.87×10^{-6}, with an average value of 121.11×10^{-6}, which is lower than the average value of basalt in the same horizon but higher than that of black shale in the same horizon. In general, the character of Σ REE of the exhalative rock is apparently different from that of normal sedimentary rocks.

Two types of exhalative rocks, the ferrodolomite-dominate exhalative rock and albite-dominated exhalative rock, are divided according to their major mineral components. Although, both of them belong to chemical sedimentary rocks, their REE compatibility and Σ REE are different. In fact, the variation scope of Σ REE of ferrodolomite-dominated exhalative rock is small, ranging from 137.88×10^{-6} to 213.87×10^{-6}, with an average value of 165.732×10^{-6}, which is almost the same as the Σ REE average value of basalt but much higher than that of the black shale in the same horizon. Since the radium of Ca (Fe) ions in the trigonal system of ferrodolomite is close to that of REE ions, it is easy for the rare earth elements to enter into the crystal lattice. That is why the ferrodolomite-dominated exhalative rock possesses relatively stronger attractive capability on REE. On the contrary, the variation scope of Σ REE of albite-dominated exhalative rock is large, ranging from 6.55×10^{-6} to 127.88×10^{-6}, with an average value of 82.588×10^{-6}, which is slightly higher than the Σ REE average value of shale in the Xiagou Formation. The radius of Na ion in the triclinic system of albite and the radius of REE ions is apparently different. Therefore, the amount of Ca ions, which can be replaced by REE, is small and limited. That is why the albite-dominated exhalative rock possesses relatively weaker attractive capability for REE. It is noted that, for the two types of exhalative rocks mentioned above, the barite-bearing ones are of relatively high REE abundance. This is because the radius of Ba ion in the barite is close to the REE ions, and therefore, can be easily replaced by REE. This is also one of the major reasons for the enrichment of REE in the exhalative rocks of the Xiagou Formation.

The REE analysis indicates that both the exhalative rocks in the Xiagou Formation and the basalt in the same horizon enrich in gas-liquid elements and show close relation in their material compositions. Their chondrite normalized REE distribution pattern is characteristic of dipping to the right slightly and of the Σ LREE > Σ HREE (Fig. 3), which is similar to the REE distribution pattern of basalt in the same horizon, to that of the hot water sedimentary rock of laminated albite in the Broken Hill Pb-Zn-Ag ore field of Australia, and to that of hot water sedimentary rock of layered albite in the west Guangdong Au-Cu-Pb-Zn-Ag ore field in China.

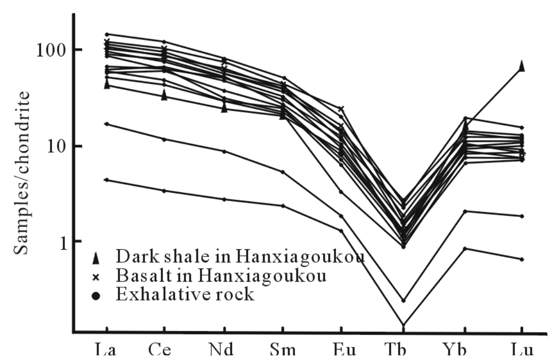

Fig. 3 Distribution pattern of REE for exhalative rock, shale and basalt in the Xiagou Formation

The similarity of REE distribution patterns of the exhalative rock and basalt in the same horizon indicates that it is highly probable that they have the same material sources. Although the main features of REE distribution pattern for the exhalative rock and the black shale in the same horizon are almost identical, and the black shale is particularly rich in heavy rare earth elements such as Yb and Lu, a typical phenomenon usually occurring in the hot water sediments on the bottom of modern ocean (Liang et al., 1997). It is indicated that the lacustrine sedimentary environment is strongly affected by the activity of hot water exhalation, so that the black shale has a similar REE geochemical character as the exhalative rock.

4.2 Ce and Eu anomalies

Ce and Eu, two variable valence rare earth elements, are very sensitive to the external environment. The change of environment leads to the transition of valence state and the

Table 1 Analytical data of REE compositions (μg/g) of exhalative rock from the Xiagou Formation (10^{-6})

Serial number for Well, sample and sampling depth (m)	Lithology	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	ΣREE	δCe	δEu	Genetic types
Long 3 Well,87/4545.00	Laminated ferrodolomite	27.22	54.57	28.92	5.91	0.86	0.72	2.05	0.33	120.58	0.993	0.749	Regional diffusing type
Long 5 Well,22/4029.97	Laminated albite-ferrodolomite	45.73	100.8	49.89	9.98	1.42	1.88	3.66	0.47	213.87	1.082	0.721	Basin deposition type
Long 103 Well,/4589.60	Laminated barite-albite-ferrodolomite	44.79	72.81	44.35	10.28	1.97	1.62	3.43	0.42	179.67	0.825	0.978	Basin deposition type
Liu 6 Well,22/4381.20	Brecciated barite-ferrodolomite-albite	21.35	52.58	22.60	5.12	0.53	0.79	2.04	0.29	105.30	1.220	0.529	Water-bursting breccia type
Long 3 Well,90/4559.00	Breccias of albite-ferrodolomite rock in sandstone	31.52	68.51	33.8	7.43	0.94	1.28	2.79	0.35	146.62	1.073	0.643	Water-bursting breccia type
Long 6 Well,/4058.60	Laminated albite	1.36	2.70	1.66	0.45	0.10	0.09	0.17	0.02	6.55	1.433	1.122	Basin deposition type
Long 3 Well,38A/4432.60	Albite laminae in deformed laminated ferrodolomite-albite	19.78	41.08	19.21	4.73	0.48	0.68	1.39	0.23	87.58	1.061	0.520	Basin deposition type
Long 3 Well,38B/4432.60	Ferrodolomite laminae in deformed laminated ferrodolomite-albite	42.75	93.48	43.02	9.69	1.11	2.11	3.15	0.45	197.76	1.104	0.576	Basin deposition type
Long 6 Well,A/4059.80	Striped ferrodolomite-albite	5.99	10.32	5.60	1.12	0.14	0.18	0.44	0.06	23.76	0.884	0.637	Basin deposition type
Long 6 Well,B/4059.80	Striped barite-albite	29.18	60.62	30.92	6.23	0.7	0.92	1.45	0.23	130.25	1.030	0.575	Basin deposition type
Long 3 Well,86/4544.00	Laminated albite-ferrodolomite	27.18	62.74	35.06	7.42	0.89	1.6	2.59	0.40	137.88	1.063	0.604	Basin deposition type
Long 6 Well, 63/4060.60	Laminated albite	18.06	52.46	18.72	4.60	0.25	0.63	1.72	0.27	96.71	1.452	0.279	Basin deposition type
Long 3 Well,85/4542.00	Laminated ferrodolomite-albite	21.89	60.86	34.52	5.82	0.82	1.17	2.49	0.31	127.88	1.176	0.711	Basin deposition type
Correlative samples: Xiagou Formation pofile in Southern Gansu	Epidote-zeolite	17.34	36.74	18.61	4.43	0.60	0.84	2.26	0.39	81.21	1.046	0.686	Basin deposition type
Correlative samples: Xiagou Formation in Hanxiagoukou	Black shale	13.63	26.78	15.48	4.09	0.68	0.80	3.46	2.61	67.53	0.950	0.841	Normal deposition
	Alkaline tholeiitic basalt	34.82	67.54	37.63	7.68	1.14	1.11	1.78	0.24	151.94	0.955	0.760	In the Xiagou Formation, interbedded partly with exhalative rock
		36.72	81.73	45.28	8.26	1.74	1.18	2.10	0.27	177.28	1.043	1.079	
Chondrite (Henderson, 1984)		0.31	0.808	0.6	0.195	0.0735	0.77	0.209	0.032 2	2.998			

separation of Ce and Eu with adjacent REE, forming Ce and Eu anomalies. The exhalative rock in the Xiagou Formation is characterized by positive Ce and negative Eu, compared with the character of negative Ce and positive Eu for marine exhalative rock (Li and Feng, 2003; Morad and Felitsyn, 2001; Ding and Liu, 2000; Wang et al., 1998; Liang et al., 1997; Xiao et al., 1994; Jean-Jacques, 1994; Brown, 1993; Li, 1993; Murray et al., 1990).

4.2.1 Ce anomaly

Under the conditions of alkalescence and oxidation in the modern seawater, Ce^{3+} is oxidized into relatively indissolvable Ce^{4+} and deposited with Fe-Mn nodules in seawater, leading to the depletion of Ce^{3+} in seawater and sediments and vice versa. Therefore, Ce anomaly can be used as an indicator for the oxidation-reduction conditions in sedimentary environment. The variation scope of δCe in exhalative rocks in the Xiagou Formation ranges from 0.825–1.452. Statistics of 13 specimens reveal that ten of them are characteristics of $\delta\text{Ce} > 1.0$ and three of them of close to $\delta\text{Ce} < 1.0$, with an average value of 1.107. It shows a weak to moderate positive Ce anomaly and indicates the enrichment of Ce^{4+} in the water and sediments at the bottom of lake, which can be used as the indicator for hot water sedimentary environment at the bottom of lake, with strong reduction and alkalescence conditions. Research by Morad and Felitsyn (2001) also showed that Ce anomaly could reflect the physical-chemical conditions of formation environment when $\text{La/Sm} > 0.35$ and La/Sm and δCe are unrelated (Luo et al., 2001). The La/Sm value from the exhalative rocks in the Xiagou Formation is 3.022–5.348, much higher than 0.35, and the correlation coefficient (R^2) of La/Sm and δCe anomalies is only 0.224 2 (Fig. 4). Therefore, the Ce positive anomaly reflects a condition of reduction, alkalescence and anoxic environment when the hot water sedimentation occurred at the bottom of the lake. It is a favorable condition and environment for the formation of black shale and quality hydrocarbon rocks. It should be pointed out that the terrestrial hot water sedimentary rocks of silicate in the Lincang germanium deposited in West Yunnan shows Ce anomaly ($\delta\text{Ce} = 0.997-1.117$), but the anomaly is considered to be related to the tectonic activity of continental margin (Qi et al., 2003).

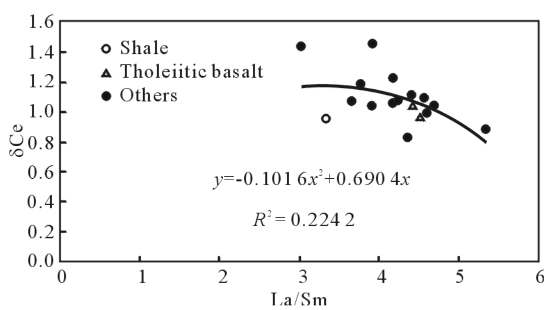

Fig. 4 Diagram showing the relation between Ce and La/Sm

4.2.2 Eu anomaly

The occurrence of Eu anomaly in water is usually under a condition of reduction and alkalescence. Many factors affect the Eu anomaly, which is very sensitive to every factor. For instance, when fluids is in a condition of reduction, the change of valence state and ion radius leads to the separation of Eu from its adjacent REE, causes the accumulation of Eu^{2+} and eventually results in the occurrence of Eu positive anomaly. However, the easiness of migration of Eu from the sediments can also cause depletion of Eu^{3+} and formation of Eu negative anomaly. Another factor affecting the Eu anomaly is temperature. In fact, the oxidation-reduction potential of Eu^{3+}/Eu^{2+} is strongly controlled by temperature. When temperature increases, the oxidation-reduction potential of Eu^{3+}/Eu^{2+} tends to shift to the direction of reduction intensification. If the temperature is high enough, a real situation at the exhalation mouth of hot water in modern oceanic ridge, Eu occurs as Eu^{2+} and forms Eu positive anomaly in water, even under a moderate reduction condition. The variation of Eu anomaly is also affected by the amount of external liquid addition. It is shown that the increase in the ratio of external liquid addition results in the decrease of δEu along with δCe. The variation scope of δEu in the exhalative rocks of the Xiagou Formation ranges from 0.279–1.122, with an average value of 0.665. Of thirteen samples, only one sample shows $\delta\text{Eu} > 1.0$, and the rest < 1.0, indicating moderate-strong Eu anomaly. The Eu anomaly characters reveal that the deposition of exhalative rocks takes place at a under relatively low-temperature condition and can be utilized as a direct indicator for hot brine sedimentary environment under strong reduction condition. The Si/Al (An%) $-\Delta(\theta)1$ diagram analysis conducted by Luo shows that the albite belongs to low-moderate temperature hydrothermal plagioclase with the An value of 2–10 (Zheng et al., 2003), and the balance temperature of O isotope calculated by the Epstein formula ranges from 66°C–94°C. These parameters are in accordance with the Eu anomaly characteristics of the exhalative rocks in the Xiagou Formation.

If variation of Eu anomaly is affected by the amount of external liquid addition, the increase in the ratio of external liquid addition results in the decrease of δEu along with δCe (Ding and Liu, 2000). However, such a case did not occur in the exhalative rocks of the Xiagou Formation, indicating that the variation of Eu anomaly is unrelated to the amount of lacustrine water addition.

To demonstrate the changing characters of flowing solution environment during the deposition processes of exhalative rocks of the Xiagou Formation, the Eu/Sm-La/Ce diagram is used to simulate the mixing processes of hot solution and lacustrine water for the Qingxi sag (Fig. 5), in reference to the Eu/Sm-La/Ce diagram analysis conducted by Ding and Liu (2000). In the (2000) diagram, the change of Ce anomaly of sea water end member is obvious and has a negative correlation with the oxidation degree of terrestrial materials and seawater, moreover, the Eu/Sm changes greatly

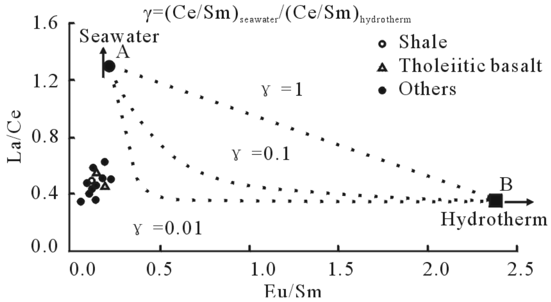

Fig. 5 Diagram showing the relation between La/Ce and Eu/Sm (modified from Ding and Liu, 2000)

for the hydrothermal solution end member. Therefore, as for the seawater end member, if the mixing ratio of seawater increases and the Ce negative anomaly is conspicuous, then the value of La/Ce of the seawater tends to become large, indicating that the influence of seawater is apparent. If a Eu positive anomaly occurs in the simulated flowing solution, then the value of Eu/Sm of the seawater tends to become large, indicating that the influence of seawater is small.

The above Eu/Sm-La/Ce diagram analysis is also used in the research on simulating the mixing processes of hydrothermal solution and lacustrine water. In the Eu/Sm-La/Ce diagram, all the samples, including exhalative rocks, basalt and shale, concentrate on a small region and lie below the $\gamma = 0.1$ curve, indicating that there is no differentiation mechanism for Ce or Eu anomaly in the hydrothermal solution and lacustrine water during the depositional processes of exhalative rocks. The character suggests that there exists a mechanism which prevents the lacustrine water from entering the hot brine pool, or that the mixing ratio of hydrothermal solution and lacustrine water is very small. The most reasonable explanation is that there are some lowland areas with a close sedimentary environment at the bottom of lake in the south of the Qingxi sag, where the great depth and pressure at the hot brine pools make the exhalative activity of the hot water relatively stable, restrain the boiling of hydrothermal solution and limit the amount of mixing hydrothermal solution and lacustrine water entering the hot brine pool, and eventually create a stable and quiet environment for the formation of hot brine pool for deep hot water sedimentation.

5 Discussion and conclusions

In summary, the exhalative rock in the Xiagou Formation of Qingxi sag, Jiuxi basin is a sort of lacustrine “white smoke type” exhalative rock with complex compositions and fabrics. The geochemical characteristics of REE indicate that the exhalative rock has a close relation with the tholeiitic basalt in the same layer. The positive Ce anomaly and negative Eu anomalies suggest that the hot water deposition occurred in the deep hot brine pool environment under reduction and alkalescence condition. The simulation by Eu/Sm-La/Ce diagram analysis further reveals that deep water, high pressure and close environment restrain the lacustrine water from entering the hot brine pool during the hydrothermal exhalative process and depositional process. Therefore, the exhalative rock in the Xiagou Formation resulted from the sedimentation of hydrothermal solution at the deep bottom of lake, and the Ce and Eu anomalies show different physical and chemical conditions from those of exhalative rocks formed in the oceanic basin. Unfortunately, little has been done on the research on the exhalative rocks formed in the lacustrine condition and little has been known about their REE geochemical characteristics. Whether the positive Ce anomaly and negative Eu anomaly represent the geochemical characteristics of fault lacustrine basin remains uncertain. Much should be done to prove the basic REE geochemical characteristics of exhalative rocks with lacustrine origin.

It should be pointed out that there is still no reasonable explanation for the coexistence of hot water minerals under different physical and chemical conditions in the complex mineral assemblage of exhalative rocks, for example, the ferrodolomite formed in alkaline condition is interbedded with laminated dickite precipitated in acidic condition, and the analcite formed in alkali condition is interbedded with the laminated ferrodolomite and albite. At least, it indicates that there exists intensified and frequent variation processes under different physical and chemical conditions during the hydrothermal exhalative activity, a phenomenon that has not been reported in previous exhalative rock research and an unsolved problem that needs to be further studied.

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