Genetic Relationship between Natural Gas Dispersal Zone and Uranium Accumulation in the Northern Ordos Basin, China

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Abstract: The Ordos Basin is well-known for the coexistence of oil, natural gas, coal and uranium. However, there has been little research to discuss the genetic relationship between them. In this paper, a case study of the Zaohuohao area in Dongsheng, Inner Mongolia, China, is conducted to investigate the genetic relationship between the natural gas and the uranium accumulation. Fluid inclusion data from the uranium-bearing sandstone samples indicate that the fluid inclusions formed in a gas-water transition zone. Using the homogeneous temperatures of aqueous inclusions coeval with hydrocarbon-bearing inclusions, combined with the buried history and paleo-temperature data, the gas-water transition zone reached the area at about 110 Ma. On the basis of this, the contents of Uranium (U) and Total Organic Carbon (TOC) of the samples were analyzed, and there was no obvious relation between them. With regard to the available data from both publications and this study, it is found that the U mineralization has a spatiotemporal accordance with the gas-water dispersal zone. Thus, it is believed that the natural gas in the gas-water zone is an effective reducer to the U-bearing ground water abundant in oxygen, which is the main factor to U accumulation. This result can be used as the reference to the U mines predicting and prospecting.

Key words: natural gas dispersal zone, fluid inclusions, uranium mine, Ordos Basin

1 Introduction

Mesozoic-Cenozoic uranium deposits of basins in China can be subdivided into the sandstone-type uranium deposit, coal-type uranium deposit and mudstone-type uranium deposit based on U-bearing source rock (Jin and Huang, 1991; Zhang and Cai, 2002). The sandstone-type deposit is the main type of uranium deposit prospected since it has great reserves with easy exploitation (Xiao et al., 2004; Cai et al., 2005). A general view on the formation of the sandstone type U deposit is that the accumulation of U element is closely related to organic substance (Read, 1998; Landaus, 1999; Shtein and Khim, 2000; Min et al., 2000a).

The Ordos Basin is one of typical basins where oil, natural gas, coal and uranium exist together. Plenty of work has been done to study the conditions and mechanism of U mineraliztion in the basin. At present, there are some different viewpoints on it for instance, the plant oddment or oil reduces interlayer oxidated zone to result in the U accumulation (Wu et al., 2003; Zuo, 2005); exuded gas and oil from a trap structure reduces the U-bearing water containing rich oxygen to lend to the uranium deposit (Sun et al., 2004; Gong and Li, 2005); or U-bearing water driven by natural gas encounters the gas-water chemical barrier formed by surface water when a deep basin gas trap forms to cause U mineralization (Zhang, 2004). Although oil and gas reduction for U mineralization was taken into account in some papers, its dominant effect of natural gas on U precipitation is often neglected. There is also lack of research on the particular phenomenon that the U mineralization distributes in a gas-water transition zone.

The gas-water transition zone currently means a water-soluble gas transition zone between a gas zone and a water zone under the background of a hydrodynamic trap gas pool. The distribution of three zones is usually determined by drilling wells and well loggings (Liu et al., 1998; Li et al., 1999). Due to the special geological and geochemical conditions of the gas-water zone, different types of fluid inclusions coexist, usually including hydrocarbon-bearing inclusions, aqueous inclusions and gaseous hydrocarbon inclusion (Mi et al., 2004; Xiao et al., 2005). The gas-water transition zone and its migration were investigated

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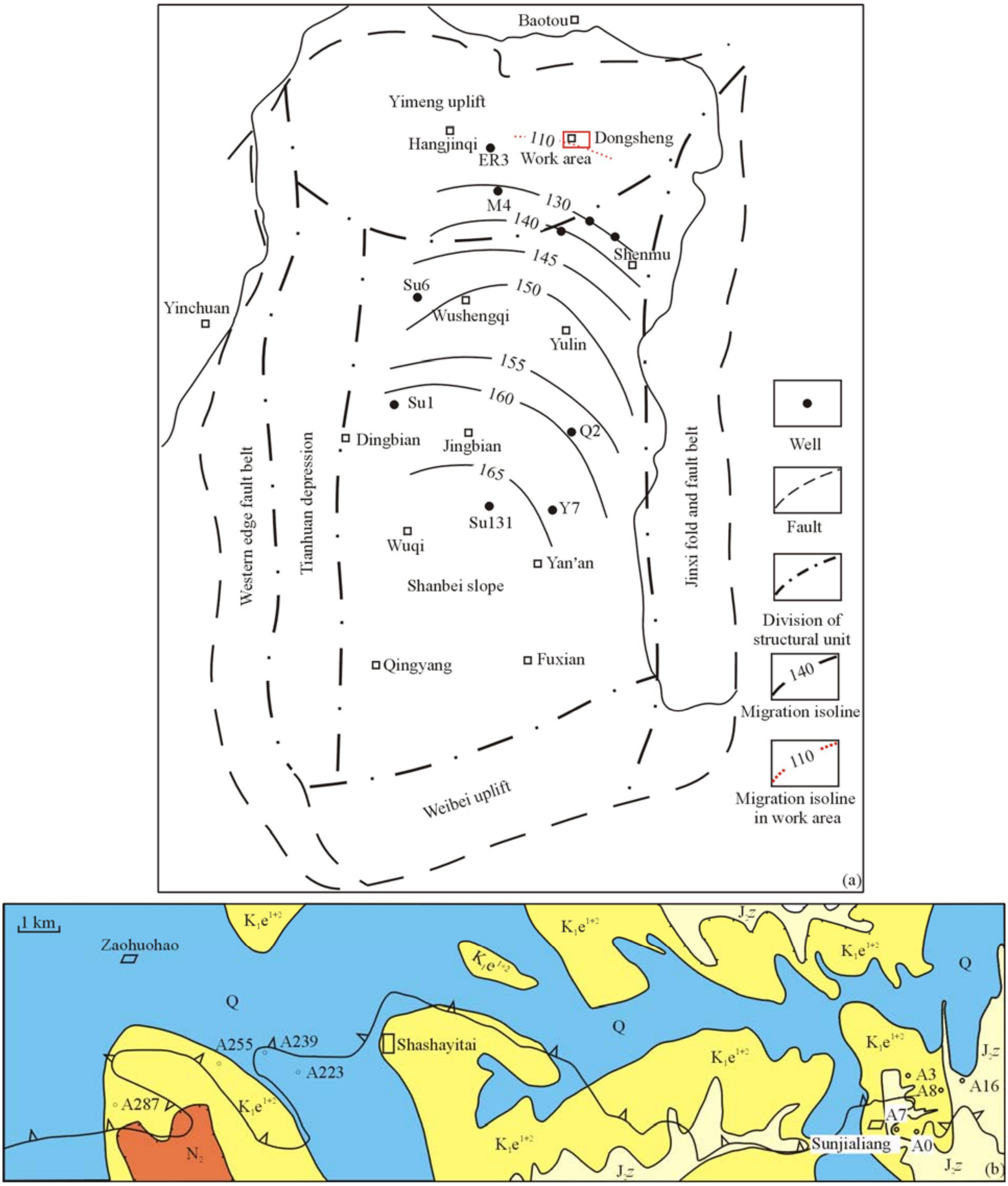

Fig. 1. Tectonic framework in the Ordos Basin and the well distribution in the work area.
(a) Tectonic framework and gas-water interface migration (Mi et al., 2004); (b) Well distribution in the work area.
Q – Quaternary; N_2 – Neogene; K_1e^{1+2} – First and second rock member of the early Cretaceous; $J_{2}z$ – Middle Jurrasic Zhiluo Formation.

in the northeastern area of the Ordos Basin by some experts (Fu et al., 2003; Mi et al., 2004). They believed that the gas-water transition zone began to develop in 165 Ma when the gas pool first formed at the center of the basin (Fig. 1a), then it migrated northward to reach the belt along well Yi12 in the north of the Yimeng uplift and well Ha2 in the Dongsheng area during the early Cretaceous (Li, 2002; Mi et al., 2004). Taking the Middle Jurassic Zhiluo Formation in the Dongsheng area as the object of study, we investigated the characteristic of the gas-water transition zone, analyzed the U distribution in the gas-water transition zone, and discussed the mechanism of U accumulation in the zone.

Table 1 Geological background of the samples and analysis results of their contents of TOC, U and Th

Well	Sample	Lithology	Depth (m)	Stratum	TOC (%)	U (ppm)	Th (ppm)
A0	4	sandstone	120.6	J_2z	0.18	20.8	8.412
	8	sandstone	110.02	J_2z	2.22	41.6	7.053
	10	sandstone	103.3	J_2z	0.2	364.9	5.943
A3	10	sandstone	147	J_2z	0.09	1.75	4.152
	11	sandstone	152.3	J_2z	0.07	2.05	4.302
	13	sandstone	156.7	J_2z	0.08	9.417	4.376
A7	10	sandstone	118.2	J_2z	0.09	0.755	2.999
A8	2	sandstone	140.6	J_2z	6.78	498.6	6.426
	10	sandstone	153.2	J_2z	0.08	5.676	2.43
A16	4	sandstone	147.55–153	J_2z	0.09	51.65	3.57
	7	sandstone	129.2–135.65	J_2z	0.1	1.43	8.508
	9	sandstone	116.4–122.9	J_2z	0.07	2.5	11.22
A223	1	coal	130	J_2z	53.2[(a)]	0.767	6.804
	3	sandstone	118.6–123	J_2z	0.07	2.194	8.883
	8	sandstone	134.9–141.3	J_2z	0.09	1.271	10.48
A239	6	sandstone	132.5–139.7	J_2z	0.1	72.25	5.795
	712	mudstone	139.7–141.1	J_2z	6.79	18.14	22.33
A255	5	mudstone	174.4–176	J_2z	0.09	2.572	11.61
	9	sandstone	187–193.45	J_2z	1.1	113.8	24.65
A287	A	sandstone	290–297.3	J_2z	0.07	1.717	7.585
	B	sandstone	282–286.25	J_2z	0.12	9.164	5.475
	C	Sandstone with coal	264.25–270.48	J_2z	22.34	10.79	2.329

Note: TOC, U and Th measured by the instruments Rock-Eval 6 standard and PE Elan 6000 ICP-MS respectively in 2005. (a) TOC of the sample was analyzed with coal procedure, and the others were analyzed with rock procedure. J_2z – Middle Jurassic Zhiluo Formation.

2 Geological Settings

The Ordos Basin is situated in the northern China (Wang et al., 2004). It is a lapped craton basin including stages of Paleozoic platform and Mesozoic-Cenozoic inland depression. The basin basement is composed of Archean and Paleoproterozoic metamorphic rock, and the sedimentary strata includes carbonates of Paleozoic, coal measures of upper Paleozoic, and river and lacustrine sedimentary cap of Mesozoic-Cenozoic inland depression. The Silurian and Devonian strata are absent (Min et al., 1998). Based on the tectonic form and evolution, the Ordos Basin can be divided into 6 units (Fig. 1a): the Yimeng uplift, Yishan slope, Weibei uplift, Tianhuan depression, Jinxi fold and fault belt, and Western edge fault belt (Tang et al., 2000).

The upper Paleozoic gas-bearing area has the characteristics of a deep-basin gas (Li, 1999; Min et al., 2000b, 2000c; Sun et al., 2001). The source rock of the natural gas is a set of Carboniferous-Permian coaly stratum with abundant organic substance and high intensity of gas generation, which provided the material basis for the gas pool. Owing to the Yanshanian Movement during the Jurassic and early Cretaceous, the source rock was matured to reach the main stage of gas generation (Li, 2002; Wang et al, 2003; Zhao et al., 2005), which led to the natural gas dispersal zone to be migrated from south to north or northeast of the basin.

The sandstone-type U deposit is discovered in the Dongsheng area. The U-bearing minerals occur mainly in coffinite, and some in pitchblende and brannerite. The ore body occurs with an irregular tabular shape in the sandstone from the lower member of the Zhiluo Formation (Zhang, 2004; Liu et al., 2006). The principal part of distributary channel in the lower member of the Zhiluo Formation has a good relationship with the U mineralization (Jiao et al., 2005; Zhao and Ou, 2006). The studied area is called as Zaohuohao in the Dongsheng area, northern Ordos Basin (Fig. 1b).

3 Samples and Experimental Methods

There are 22 core samples of the Zhiluo Formation collected from 9 wells in the Zaohuohao area (Fig. 1b), including mudstone, coal and sandstone. All the samples are accompanied with some associated mineral, such as pyrite. The geological background of the samples is shown in Table 1.

Fluid inclusions observation and measurement: The sample sections were cut into a thickness of 0.07–0.14 mm, with both sides being polished. The Leica DM RX microscope was used. Fluorescence observations of fluid inclusions were mainly conducted using a 100W Hg light, a H3 420–490-nm excitation filter, a PKP510 nm dichromatic reflector and a 515-nm protective filter. The homogeneous temperature was measured by a USGS FLUID INI air current cooling-heating stage. The heating rate was controlled at 1–5°C/min, with an error of ±1°C. RENISHAN(R) RM-2000 model Raman spectrophotometer was used for the compositional analysis of fluid inclusions, and the operating conditions are as follows: Ar^+ laser generator with laser wavelength

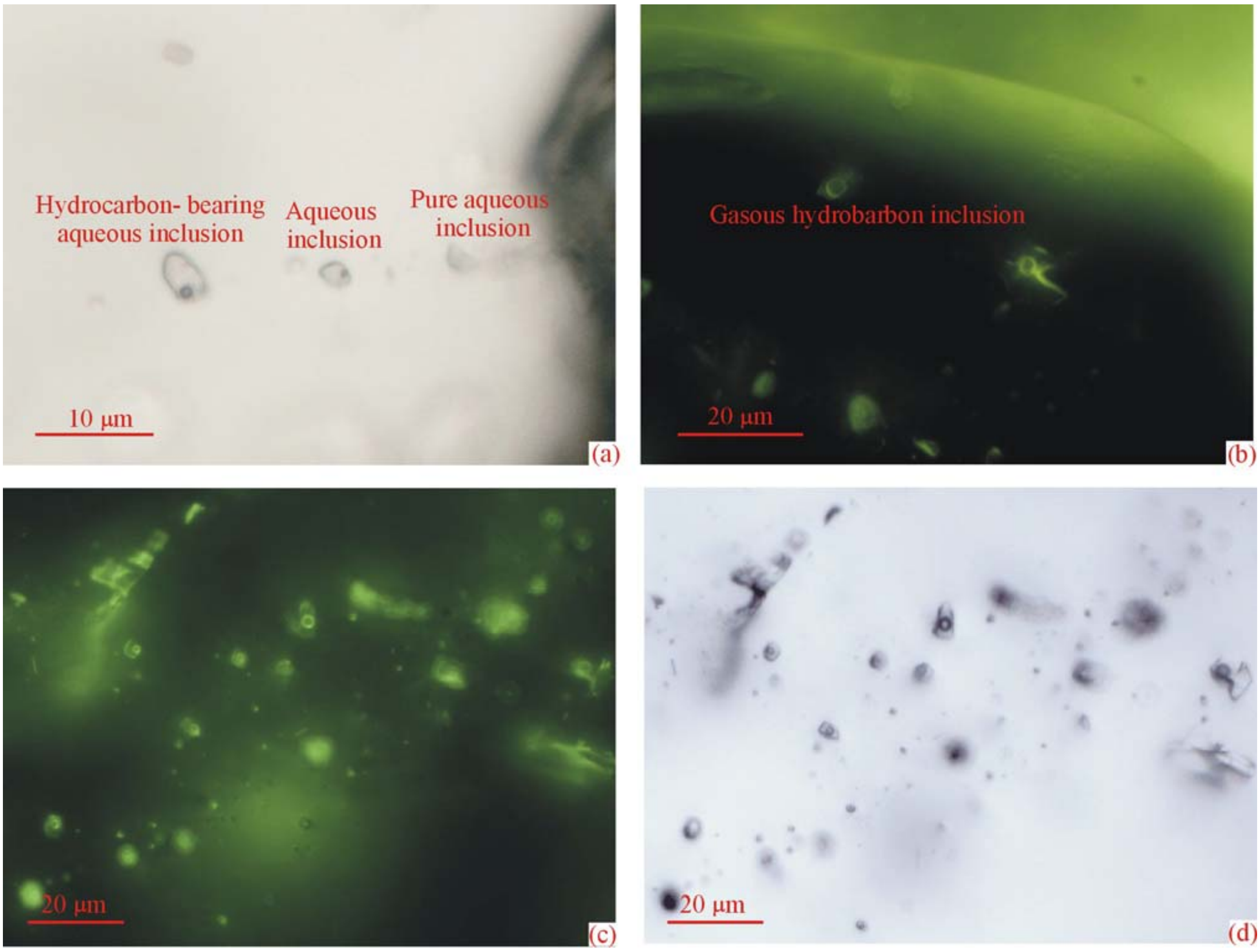

Fig. 2. Different types of fluid inclusions in the work area in the Ordos Basin.
(a) Microcopy of different kinds of fluid inclusions in A287–C, ×400;
(b) Microcopy of gaseous hydrocarbon inclusions with yellow- kelly fluorescence in A3–15, ×400;
(c) and (d) The same view field of hydrocarbon-bearing aqueous inclusions under fluorescence and transmit light in A16–4, ×400).

of 514 nm and power of 25.2 mW; grating slit of 20 μm and scanning time of 10 s.

The TOC content of the rock samples is determined using a Rock-Eval 6 standard instrument made by VINCI Ltd. Co., France on the basis of a rock or coal procedure.

The analysis of U and Th is after the following steps: (1) Powder the samples and weigh 0.5 g to put into a Pt crucible, and add 2 mL nitric acid and 5 mL hydrofluoric acid into the crucible. Shake it up, and then put the crucible on the electrothermal board to evaporate the solvent up to dryness; (2) Add 1 mL perchloric acid to the crucible, and repeat the above process to evaporate the solvent; (3) Add another 4 mL nitric acid to the crucible, and dissolve the sample; (4) Transfer the solution to a 200-mL volumetric flask for constant volume, and use the instrument PE Elan 6000 ICP-MS to measure the contents of U and Th. The instrument parameters of the mass spectrograph PE Elan 6000 are: the power of 1000 W, cooling-gas flow (Ar) of 15 L/min, assistant gas flow (Ar) of 1.2 L/min, and atomized gas flow (Ar) of 0.8 L/min.

4 Results and Discussions

4.1 Characteristic of the gas-water transition zone

(1) Fluid inclusions in the gas-water transition zone

As discussed above, the Zhiluo Formation in the studied area consists mainly of sandstones with a few thin layers of mudstone. Most of the sandstone is not tightly cemented. There are abundant fluid inclusions mainly occurring in micro-fissures of the sandstone granule and only a little in overgrowth. The fluid inclusions can be divided into aqueous inclusions, hydrocarbon inclusions, hydrocarbon-bearing inclusions and inorganic gas inclusions according to the component characteristics (Fig. 2). The aqueous inclusions have two kinds: pure liquid phase and vapor-liquid phase inclusions. Most of the aqueous inclusions are two-phase inclusions with vapor/ liquid ratios of 6%–10%, and they are non-fluorescent.

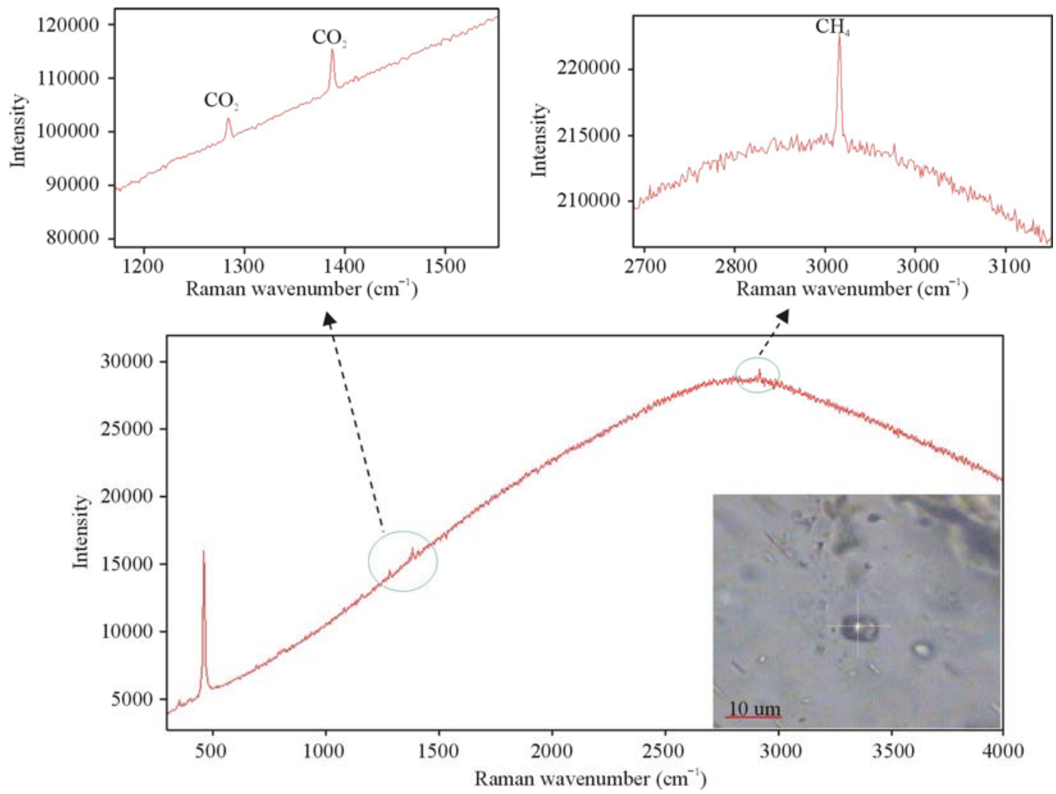

Fig. 3. Laser Raman spectra showing CH_4 and CO_2 in gaseous inclusions in A3–15.

Table 2 Interpretation of gas and water layers by well logging data in different structural units, Ordos Basin

Structural unit	Well number	Thickness of sandstone (m)	Thickness of Gas-bearing layer (m)	Thickness of Gas-water layer and water layer (m)
Yimeng uplift	18	527.4	323.1 (61%)	204.3 (39%)
Shanbei slop	327	4918.3	4761.6 (97%)	156.7 (3%)

Note: the data was modified from Min and Fu (2002).

The aqueous inclusions of the samples are small-sized, and commonly coeval with hydrocarbon-bearing inclusions (Fig. 2a). Hydrocarbon inclusions are mostly gaseous hydrocarbon inclusions and no petroleum inclusions are found, gaseous hydrocarbon inclusions are two phases with vapor/liquid ratios of 60%. It is black under transmitting light model. The ektexine of the vapor bubble also shows weak yellow-kelly fluorescence (Fig. 2b). The hydrocarbon-bearing inclusions are usually hydrocarbon-bearing aqueous inclusions with vapor-liquid two phases, and some liquid hydrocarbon clinging to ektexine of the vapor bubble causes a weak yellow to kelly fluorescence to form an aureola though both vapor and liquid phases show non-fluorescence. Since this kind of inclusions captured immiscible fluid in an inhomogeneous phase, most of them are immiscible with a very high homogeneous temperature in the area (Fig. 2c, d). The inorganic gas inclusion indicates that the inclusions mainly contain inorganic gas, such as CO_2, N_2 and vapor, and they also mainly include two kinds: one phase with pure gas and two phases with the vapor/ liquid ratios of more than 50%. The inorganic gas inclusion has non-fluorescence or very weak fluorescence.

According to the compositional analysis by using the Laser Raman Instrument (Fig. 3), the main component of gaseous part in the hydrocarbon-bearing aqueous inclusions and gaseous hydrocarbon inclusions is methane (CH_4) and CO_2, and the methane often has a high content. There are some inorganic gas inclusions only containing abundant CO_2, and they can be actually defined as CO_2 inclusion. In addition, inorganic gaseous inclusions with some nitrogen (N_2) are also found in the sample A287-C. Thus, based on the analysis result, the main gaseous components in the fluid captured by fluid inclusions in the studied area are CH_4, CO_2 and N_2.

The upper Paleozoic natural gas area in the Ordos Basin is characterized by a deep-basin gas trap. According to the well logging data (Table 2), the Shanbei slope is a gas-bearing area in reservoirs with only 3% of water layer and gas-water layer, but the Yimeng uplift is a gas-water zone, with gas-water layer and water layer of 39%, indicating the existence of gas-water transition zone (Liu et al., 1998; Li,

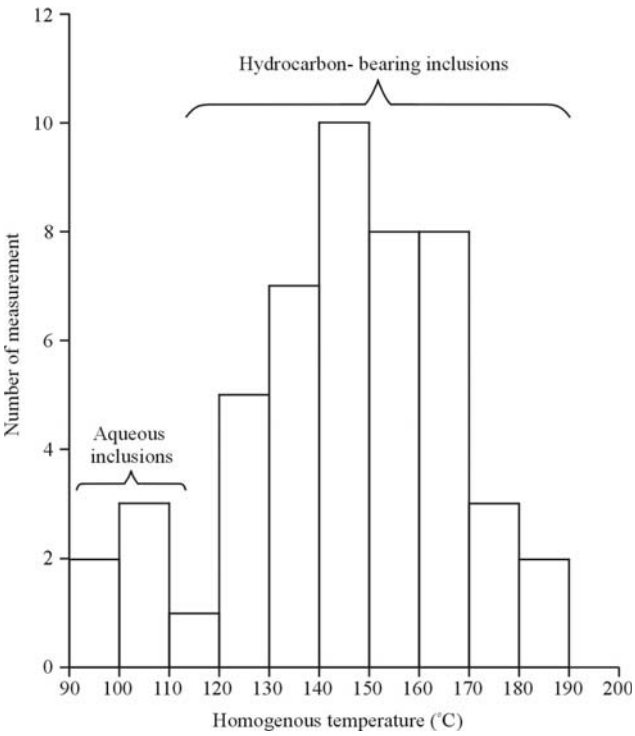

Fig. 4. Homogeneous temperature histogram of hydrocarbon-bearing inclusions and its coeval aqueous inclusions.

et al., 1999). Sandstones from this area contain plenty of gaseous hydrocarbon inclusions and aqueous inclusions, hydrocarbon-bearing aqueous inclusions and gaseous hydrocarbon inclusions coexist, with a similar characteristic as the fluid inclusions formed in the gas-water transition zone described by Mi et al. (2004) and Xiao et al. (2005), which indicates that this area is in a gas-water transition zone when the fluid inclusions form.

(2) Migration history of natural gas dispersal zone

The time of fluid inclusions growing in the gas-water transition (or gas water interface) zone will record the different position of the gas-water interface in geological history (Mi et al., 2004). The homogeneous temperatures of the fluid inclusions, combined with their buried history and paleotemperature, can be applied to probing into the migration time of the fluids (Karlsen et al., 1993; Xiao et al., 2002; Liu et al., 2003). By using this method, the geological time of gas-water transition zone reaching the studied area can be ascertained. The homogeneous temperatures of the aqueous inclusions coeval with gas hydrocarbon inclusions are in the range of 90°C–110°C, with an average of 101°C (Fig. 4). Figure 5 shows the formation time of the aqueous inclusions is about 110 Ma. The time accords with the time when the gas-water interface arrived here (Mi et al., 2004), and also matches the main period of gas generation of Carboniferous-Permian source rock.

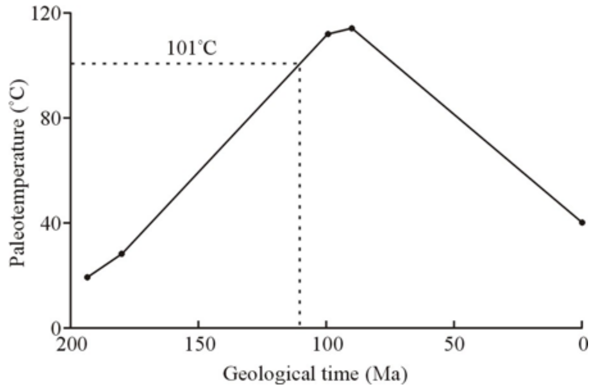

Fig. 5. Plot showing the determination of growing time of aqueous inclusions (Thickness of eroded strata) (modified from Ren et al., 1994).
Paleo-temperature gradient from the Jurassic to early Cretaceous is 4 °C/100 m, and the other geological history is about 3°C/100 m (Ren, 1996; Liu et al., 1997; Zhao and Behr, 1996; Xiao et al., 2002).

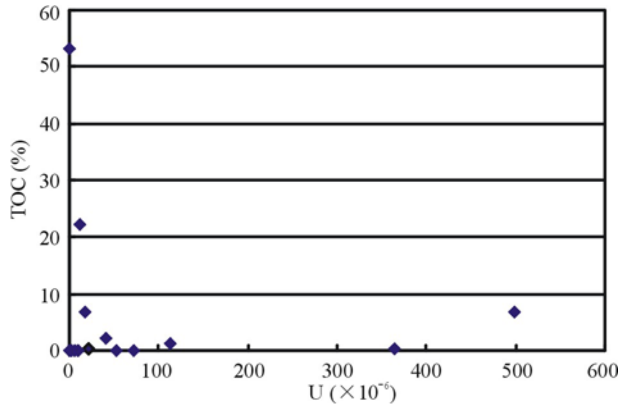

Fig. 6. Relationship between U and TOC.

4.2 Characteristic of U accumulation

(1) Relation between U and TOC

Organic substance as a reducer to the U accumulation is very common (Dong, 1996; Yang and Xia, 2004). However, there is no correlation between TOC content and U content for the studied samples in the area (Table 1, Fig. 6). Some samples have high uranium contents as well as high TOC contents. For example, the highest uranium content with a value of 498.6 ppm is present in the sample A8-2 with a TOC content of 6.78%. Some samples have a TOC contents over 20%, whereas their uranium contents are lower than 11 ppm; and some are reverse. For examples, the sample A287-C has a TOC content of 22.2%, whereas its uranium content is lower than 1 ppm; the coal sample A223-1 has a U content of lower than 1 ppm, whereas its TOC content reaches 53.2%. These results may indicate that the organic substance is not the dominant factor for uranium accumulation.

(2) Relation between contents of U and Th

U and Th are both lithophile elements. Th^{4+} and U^{4+} are difficult to be separated owing to their close relation and

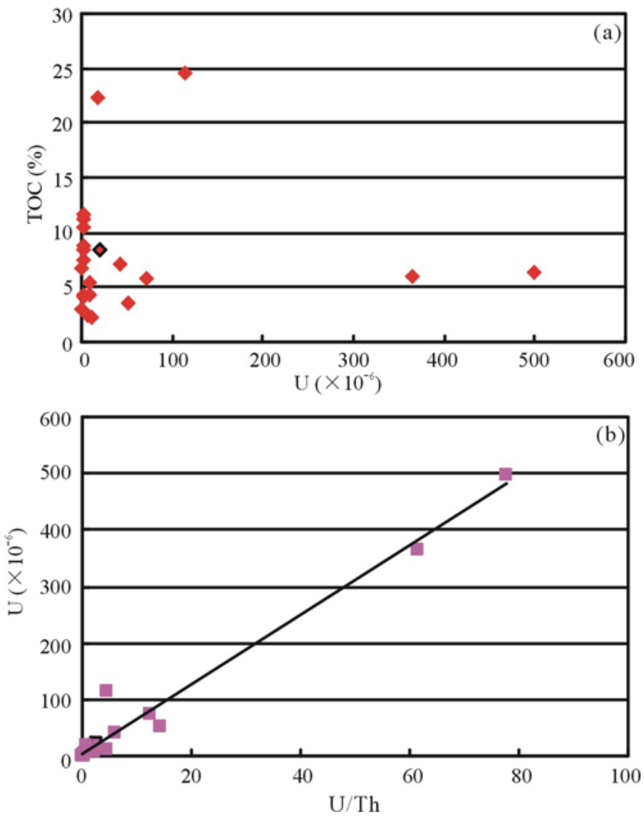

Fig. 7. Relationship between U and Th.
(a) Relations between U and Th; (b) Relations between U and U/Th.

isomorphic replacement. So, geologists usually use the U/Th ratio to investigate the origin of U (Huang and Tang, 2002). Generally speaking, a rock formed in the same term, or different rocks with the same paragenetic association or the same petrogenetic evolution sequence in stratum will have a similar value of U/Th (Xu and Yan, 1995). U/Th ratios of most studied samples are less than 1, but several over 1. In the sample A8-2, its U/Th ratio reaches 77. The contents of U and Th show no correlation for the studied samples (Fig. 7a), which indicates that the U would not be a single source but multiple sources. It should be noted that the U/Th ratios increases with the increase of the U content, showing a linear correlation (Fig. 7b). This implies that the Th content in diverse sources is similar.

4.3 Formation mechanism of U mineralization and its relation with the natural gas dispersal zone

The depositing process of U from a hydrothermal liquid usually is that dissociated UO_2^{2+} occurs when a uranyl complex compound is disintegrated; and then the UO_2^{2+} will be reduced into UO_2 (Pitchblende or Uraninite). Although there is abundant TOC content in the stratum in the Zhiluo Formation, it is not a dominant factor to U mineralization, just as pointed out before. A large amount of pyrite is discovered in the U-bearing stratum. On the basis of this, pyrite is regarded as the reducer to the U mineralization (Wu et al., 2003). However, the mineralizing experiment made by Zhao and Shen (1986) proved that reducing ability of pyrite is weak, and is controlled tightly by physical and chemical conditions. Pyrite has to be dissolved before it acts as reducer and it can reduce UO_2^{2+} to UO_2 only in acid condition. Since the physical and chemical condition of the underground water vary widely in the Dongsheng area of the Ordos Basin, the pH value in underground water increases from weak acidity to alkalinity with the burial depth of a borehole. When the depth is over 150 m, the pH value of the underground water will exceed 9 (Zhang, 2004). Thus, the pyrite has little reduction to the U mineralization.

The main metallogenic epoch of the sandstone-type U mineralization is 90–120 Ma in the Zhiluo Formation in the Dongsheng area (Xia et al., 2003; Zhang, 2004). The time of gas-water transition zone reaching this area is about 110 Ma, and has a perfect match with the U metallogenic stage. According to the discussion above, this area where U accumulated is just in the gas-water transition zone, and a large amount of reducing gases such as CH_4 is also discovered. Wang and Du (1995) proved that, under the physical and chemical conditions of U mineralization, CH_4 and H_2 can make U mineralize completely and efficiently to form a rich ore body. Wu et al. (2006) has also indicated that the U mineralization was related to the reducing gas from the natural gas in the Dongsheng area. That is to say that the U mineralization has an original relationship with the natural gas dispersal zone.

In summary, the paper proposes the process and mechanism of the U mineralization in this area. The upper Paleozoic source rock reached its peak stage of gas generation during late Jurassic and the early Cretaceous, and the generated natural gas drove underground water to migrate from the south to the north or northeast. When the gas-water interface migrated into the Dongsheng area of the basin and reached the oxidized belt with U and oxygen, the natural gas dissolved in the water would reduce UO_2^{2+}, and UO_2 would precipitate in the gas-water transition zone. Since this area uplifted and gas generation decreased during the late Cretaceous, the gas-water transition zone stopped migrating northward, which resulted in that the area became a balanced belt of natural gas supplying and dissipating. Besides CH_4, some reducing gases such as H_2S in the natural gas can also lead to U mineralization. The reaction equations are as follow:

$$4UO_2^{2+}+CH_4+3H_2O\rightarrow 4UO_2+HCO_3^{-}+9H^{+}$$

$$4UO_2^{2+}+H_2S+10OH^{-}\rightarrow 4UO_2+SO_4^{2-}+6H_2O$$

Consequently, U ore deposit in the area accumulated under a special geological and geochemical background, and natural gas was the efficient reducer to U mineralization.

5 Conclusions

(1) Secondary fluid inclusions in the sandstone-type U mineralization showed similar characteristics of fluid inclusions formed in the gas-water transition zone in the Zaohuohao area. The homogeneous temperatures of aqueous inclusions are in the range of 90°C–110°C, and their formation time was about 110 Ma, having a perfect match with the time when the gas-water transition zone migrated into the area.

(2) According to the analysis result of TOC, U and Th, the abundant organic substance in the stratum would not be the dominant factor to U mineralization. U and Th show the characteristic of multiple sources in the area and the Th content is almost the same in different sources.

(3) Pyrite cannot be the significant reducer to the U accumulation. Natural gas dispersal from the gas-water transition zone acted as a main reducer to reduce U in the underground water to form U mineralization.

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