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Isotope geochemistry of ore fluids for the Dongsheng sandstone-type uranium deposit, China

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Abstract The Dongsheng sandstone-type uranium deposit is one of the large-sized sandstone-type uranium deposits discovered in the northern part of the Ordos Basin of China in recent years. Geochemical characteristics of the Dongsheng uranium deposit are significantly different from those of the typical interlayered oxidized sandstone-type uranium ore deposits in the region of Middle Asia. Fluid inclusion studies of the uranium deposit showed that the uranium ore-forming temperatures are within the range of 150–160℃. Their $^{3}He/^{4}He$ ratios are within the range of 0.02–1.00 R/Ra, about 5–40 times those of the crust. Their $^{40}Ar/^{36}Ar$ ratios vary from 584 to 1243, much higher than the values of atmospheric argon. The $\delta^{18}O_{H2O}$ and δD values of fluid inclusions from the uranium deposit are -3.0‰– -8.75‰ and -55.8‰– -71.3‰, respectively, reflecting the characteristics of mixed fluid of meteoric water and magmatic water. The $\delta^{18}O_{H2O}$ and δD values of kaolinite layer at the bottom of the uranium ore deposit are 6.1‰ and -77‰, respectively, showing the characteristics of magmatic water. The $\delta^{13}C_{V\text{-}PDB}$ and $\delta^{18}O_{H2O}$ values of calcite veins in uranium ores are -8.0‰ and 5.76‰, respectively, showing the characteristics of mantle source. Geochemical characteristics of fluid inclusions indicated that the ore-formation fluid for the Dongsheng uranium deposit was a mixed fluid of meteoric water and deep-source fluid from the crust. It was proposed that the Jurassic-Cretaceous U-rich metamorphic rocks and granites widespread in the northern uplift area of the Ordos Basin had been weathered and denudated and the ore-forming elements, mainly uranium, were transported by meteoric waters to the Dongsheng region, where uranium ores were formed. Tectonothermal events and magmatic activities in the Ordos Basin during the Mesozoic made fluids in the deep interior and oil/gas at shallow levels upwarp along the fault zone and activated fractures, filling into U-bearing clastic sandstones, thus providing necessary energy for the formation of uranium ores.

Key words He-Ar isotopes; fluid inclusion; uranium ore

1 Introduction

The sandstone-, stratigraphical unconformity- and breccia-type uranium deposits are the most important three types of large-sized uranium deposits. About 70% of uranium products come from these three types of uranium deposits (Dahlkamp, 1993). Owing to their easy exploitation and low cost, since the 90's of the last century, the sandstone-type uranium ore deposits have become the most important target of exploration and development throughout the world, and the output has increased drastically, with the proportion of the total yield of uranium ores being increasing steadily. It is predicted that the output of uranium ore will still increase continuously in the future time (Underhill, 2000). Located in the northern part of the Dongsheng region of the Ordos Basin of China, the Dongsheng uranium deposit is a large-sized sandstone-type uranium deposit discovered recently in our country (Fig. 1). Many scholars carried out investigations into the metallogenesis of the Dongsheng uranium deposit. It is now commonly accepted that the Dongsheng deposit is an interlayered, oxidized sandstone-type uranium deposit. It is considered from the metallogenic model that O-, U(U^{6+})-bearing fluid was transported in the permeable sandstones with mudstones as water-resisting layers in the upper and lower parts, and, when the fluid encountered with reduction obstacles, U^{6+} would be reduced and precipitated at the redox interface, forming a rolled tongue-shaped uranium orebody (Huang Shijie, 1994). However, results of the study of uranium metallogenesis showed that the ore-forming temperature of the Dongsheng uranium deposit is relatively high, higher than that of local normal burial

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thermal evolution (Xiao Xinjian et al., 2004). It is considered that there must have been involved other sources of energy in metallogenesis. Recent studies have indicated that the characteristics of the Dongsheng uranium deposit are quite different from those of the typical interlayered, oxidized sandstone-type uranium deposits in the Yili Basin of Xinjiang, China, as well as in the Middle Asia. According to Zhu Xiyang et al. (2003), the Dongsheng uranium deposit is rich in Zr, Hf and other mantle-phile elements and LREE, which were enriched obviously in the process of metallogenesis. It is thought that uranium mineralization is related to mantle fluids. As viewed from uranium mineral assemblages and fluid inclusion evidence, Xiao Xinjian et al. (2004) considered that the formation of the Dongsheng uranium deposit is associated with the epithermal solutions derived from the deep interior of the Earth.

Isotope geochemistry can play a unique role in tracing geological processes and constraining the sources of geofluids. As He and Ar isotopic compositions are significantly different for meteoric waters, mantle and crust, the ^{3}He/^{4}He ratios in the crust (^{3}He/^{4}He=0.01–0.05 R/Ra) are different from those of the mantle (^{3}He/^{4}He=6–9 R/Ra) by a factor of 1000 (Stuart et al., 1995). Although crust-derived fluids contain a minor amount of mantle-derived helium, geofluids of these two sources can also be easily distinguished from each other. Therefore, He and Ar isotope geochemistry is an effective approach to constrain the source of geofluids. And it provides a very effective tracer for the study of the source of geofluids, multi-source fluid mixing effect and metallogenesis, particularly it has found wide applications in revealing the involvement of mantle fluids in metallogenesis (Ozima and Podosek, 2002). In recent years He and Ar isotope geochemical studies have been expanded to trace the sources of ore-forming paleo-fluids which were preserved as fluid inclusions during geological history, including studies of the sources of ore-forming fluids for Pb-Zn, Au, W-Mo sulfide polymetal ore deposits. The host minerals for fluid inclusion measurement are dominated by quartz and pyrite, followed by stibnite, galena, etc. (Li Zhaoli et al., 2005; Hu Ruizhong and Bi Xianwu, 1999; Stuart et al., 1995; Burnard et al., 1999; Burnard and David, 2004; Mao Jingwen et al., 2002, 2003). According to the He-Ar and H-O isotopic compositions of secondary fluid inclusions from the uranium ore-associated sandstones, in combination with the H-O isotopic composition of kaolinite and the C-O isotopic composition of calcite from the bottom of the Dongsheng uranium deposit, the authors discussed the sources of uranium ore-forming fluids.

2 Geological background

The Dongsheng uranium deposit is located in the eastern part of the Yimeng uplift in the northern part of the Ordos Basin, China, and it is a large-sized uranium deposit discovered recently in our country. The Yimeng uplift is separated by the Hetao fault depression from the Daqingshan uplift in the north, with the North Shaanxi slope, the Tianhuan depression and the West Shanxi fold zone as the fault boundaries (Liu Shaofeng, 1998). Like the Ordos Basin, the Dongsheng uranium mining district is characterized by gently extending strata with a dipping angle of 1°–3°, present in the form of a monoclinal structure. The strata exposed in this region tend to become younger from east to west, i.e., Precambrian metamorphic rocks (the West Shanxi uplift zone), Early Paleozoic platform-facies carbonate rocks, Late Paleozoic marine/continental transitional-facies coal-bearing strata, Triassic fluoviolacustrine-facies clastic rock series, Jurassic continental-facies coal-bearing series and Cretaceous fluvial clastic series (Fig. 1). Uranium ores are hosted in the Middle Jurassic Formation (J_2z). The Zhiluo Formation is composed mainly of mottled sandstones, and overlies the Middle Jurassic Yan’an Formation (J_2y) coal-series strata. Petrographically, the Zhiluo Formation can be divided into the upper and lower segments. The upper segment is a suite of mottled (yellow, red, greyish-green) medium-fine- grained sandstone and mudstone. The lower segment can be further divided into the upper and lower sub-segments. The upper sub-segment is a suite of greyish-green mudstone, siltstone and sandstone while the lower sub-segment is a suite of grey, greyish-white medium-coarse-grained sandstones, with thin-layered coal seams appearing at the top. Uranium ores are hosted in the greyish-white coarse-grained sandstones of the lower sub-segment, the hanging-wall rocks of the uranium deposit are green mudstones and its foot-wall rocks are the white kaolinite layers at the top of the Yan’an Formation.

3 Samples and experiments

The samples used to study fluid inclusions were collected from a certain drill core in the Dongsheng uranium deposit, including the red siltstone in the upper segment of the Zhiluo Formation and the greyish-green siltstone in the upper sub-segment and the greyish-white uranium ore sandstone in the lower sub-segment of the lower segment. Uranium analyses of the samples showed that the contents of uranium tend to increase progressively (Table 1). Microscopic observations revealed that fluid inclusions in the sandstone of the Dongsheng uranium deposit are dominated by secondary fluid inclusions which filled

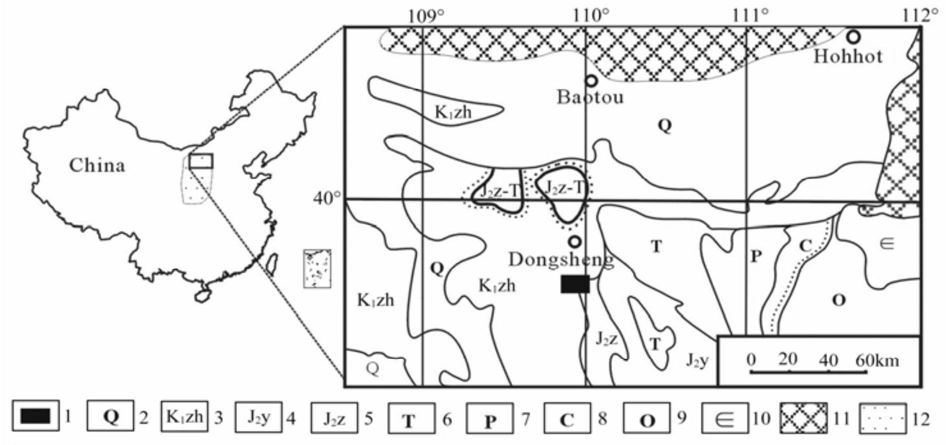

Fig. 1. Geological sketch map of the Dongsheng region, northern Ordos Basin, China. 1. Uranium deposit; 2. Quaternary; 3. Lower Cretaceous Zhidan Group; 4. Middle Jurassic Yan'an Formation; 5. Middle Jurassic Zhiluo Formation; 6. Triassic; 7. Permian; 8. Carboniferous; 9. Ordovician; 10. Cambrian; 11. Precambrian; 12. Ordos Basin.

mainly in the cracks and fissures of quartz clastic grains, mostly orientationally distributed in the form of breeds, colorless or light yellow. The fluid inclusions are commonly small in size, generally 2–8 μm, dominated by double-phase ones with vapor/liquid ratios being mostly within the range of 5%–10%. NaCl-bearing cubic daughter mineral inclusions are occasionally observed.

3.1 Measurement of the homogenization temperatures and salinities of fluid inclusions

The aim of measuring the homogenization temperatures of fluid inclusions is to determine their forming temperatures. As only the homogenization temperatures of homogeneous-phase fluid inclusions can reflect their forming temperatures, we chosed pure saline-water fluid inclusions for the measurement of homogenization temperatures and ice-point temperatures in this study. The salinities of fluid inclusions were worked out with the equation of Bodnar (1993) on the basis of their ice-point temperatures. The thermometric measurement of fluid inclusions was conducted on a THMS 600 Type Freezing Stage manufactured by the UK Linkam Scientific Instruments Company. Leica DMR microscope was used with the eyelens being 10X and the object lens being 50X. The laboratory temperature was kept at 25 ºC.

3.2 He and Ar isotopic measurement of fluid inclusions

Uranium ore samples whose fluid inclusions are most abundant and dominated by secondary fluid inclusions were selected and opened and analyzed for He and Ar isotopes by using the thermal decrepitating method at the State Key Lab of Natural Gas Geochemistry, Lanzhou Institute of Geology, Chinese Academy of Sciences. The samples were ground as fine as 60 mesh and wrapped with aluminum foil, then sealed into the sample stage for vacuum pumping. When the vacuum pressure was less than 1×10^{-5} Pa, the stage was heated with an external heater to 130ºC and baked for more than 10 hours to remove the air adsorbed on the sample surface. For the details of the concrete measuring method and procedure, refer to Ye Xianren et al. (2001). The analytical instrument is a VG5400 Type Nobal Gas Static Mass Spectrometer. Analytical condition: voltage of the ion source: 9 kV; trap current: 800 μA; working standard: air from the top of the Gaolan Mountain at Lanzhou [$^3He/^4He=(1.40\pm0.03)\times10^{-6}$].

3.3 Measurement of $\delta^{18}O_{H2O}$,δD and $\delta^{13}C$ for ore fluids

Two main methods were employed to work out the H and O isotopic composition of ore fluids. As for H-free minerals (e.g. quartz), one can directly measure the $\delta^{18}O_{H_2O}$ and δD values of inclusion fluid of mineral fluid inclusions, which are indicative of the H and O isotopic composition of ore fluids. As for H- and O-bearing minerals (e.g. kaolinite), one can directly measure the $\delta^{18}O_{H_2O}$ and δD values of the minerals, and then, on the basis of the mineral/water isotopic equilibrium fractionation equation, calculate the $\delta^{18}O_{H_2O}$ and δD values of ore fluids in equilibrium with their host mineral at the corresponding temperatures. In this study the authors used these two methods to analyze quartz fluid inclusions from the uranium ore sandstone and kaolinite from the foot-wall of the uranium deposit respectively for their H and O isotopes. The preparation of quartz inclusion samples for H and O isotopic analysis was effected by using the routine BrF_5 method. Under vacuum condition O_2 was extracted from 200 mg of pretreated quartz grains, the O_2 acquired was let to react with a hot carbon rod to convert to CO_2 for $^{18}O/^{16}O$ measurement. As for H isotopic analysis, the crushing method was used to open fluid inclusions so as to release water, and the water released was let to react with Zn at 400 ℃ to produce hydrogen gas (i.e., to produce hydrogen with the Zn method) for mass spectrometric isotopic analysis. Oxygen isotopic analysis of kaolinite was also effected by adopting the routine BrF_5 method while its hydrogen isotopic analysis was effected by using the decrepitating method (i.e., to produce hydrogen with the Zn method). At the same time, the 100% phosphate method was employed to analyze the $\delta^{18}O_{H2O}$ and δD values of calcite vein samples. The precisions of H and O isotopic measurement were ±0.2‰ and ±1‰, respectively. Both H and O isotopic analyses were accomplished on an MAT0251EM mass spectrometer at the Institute of Mineral Resources, Ministry of State Land Resources.

4 Analytical results and ore fluid sources

4.1 Results of fluid inclusion measurement and analysis of forming conditions of ore fluids

Shown in Fig. 2 are the results of homogenization temperature measurement of fluid inclusions from the Dongsheng uranium deposit. From this figure it can be seen that the homogenization temperatures of fluid inclusions from the Dongsheng uranium deposit have two peak values, one at 120–130℃ and the other at 150–160℃, with the high temperature peak value being dominant. The salinities of fluids acquired from ice-point temperatures of fluid inclusions are generally within the range of 0.80%–15.0%, those of vapor-liquid inclusions vary from 0.5% to 9.0%, and those of multi-phase inclusions in NaCl-bearing cubic daughter minerals are still higher with the maximum value up to 27.72% (the starting melting temperature is 70.2℃). The highest salinity of daughter mineral-bearing solid-liquid inclusions is 29%.

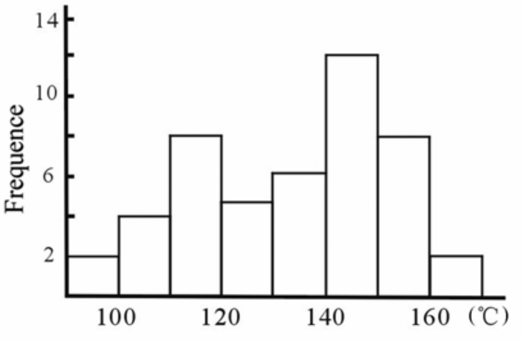

Fig. 2. Homogenization temperatures of fluid inclusions.

As viewed from the relationship between temperature and salinity (Fig. 3), it can be seen clearly that with increasing homogenization temperature, the salinity also tends to increase. At the time the temperature increases to 160℃, the salinity will increase rapidly. Microscopic observations revealed that although the homogenization temperatures of daughter mineral-bearing fluid inclusions and vapour/liquid inclusions coexisting in the contemporaneous fissures are both higher than 150℃, their salinities show a significant difference, indicating that the ore fluids experienced boiling, belonging to a kind of immiscible fluid inclusions.

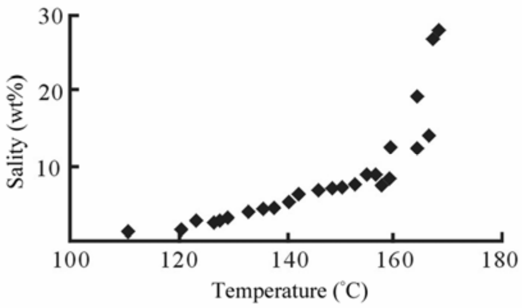

Fig. 3. Relationship between the temperature and salinity of fluid inclusions. ■ Stands for the projecting points.

On the basis of the T-ρ and T-W-ρ phase diagrams of the NaCl-H_2O system (Bodnar, 1983), the densities of fluid inclusions from the Dongsheng region were calculated to be 0.9–1.0 g/cm^3. At about 120–170℃ these fluids obviously belong to a kind of high-density fluids. From the above analytical results we can see that ore fluids of the Dongsheng uranium deposit belong to a kind of high-density immiscible fluids, whose forming temperatures are mainly within the range of 150–160℃.

4.2 Results of the He-Ar isotopic analysis of fluid inclusions and ore fluid sources

Listed in Table 1 are the results of He-Ar isotopic analysis of fluid inclusions from the Dongsheng uranium deposit. From this table we can see that the $^3He/^4He$ isotopic ratios of fluid inclusions are 0.02–1.00 R/Ra (Ra represents the $^3He/^4He$ ratio of the air), and their $^{40}Ar/^{36}Ar$ ratios range from 584 to 1970. $^3He/^4He$ ratios in the country rocks of the uranium deposit, i.e., red siltstone (sample No. D-1, containing as much U as 2.8 U/10^{-6}) are generally consistent with those of the atmosphere; $^3He/^4He$ isotopic ratios in uranium ores are generally about 0.5 R/Ra, but $^{40}Ar/^{36}Ar$ ratios tend to increase significantly with increasing U contents. From Fig. 4 it is seen that there is some relationship between the $^3He/^4He$ and $^{40}Ar/^{36}Ar$ ratios of fluid inclusions from the Dongsheng uranium deposit and the U contents of the

Table 1. Results of the He-Ar isotopic analysis of fluid inclusions from the Dongsheng uranium deposit

Sample No.	Rock type	U content ($U/10^{-6}$)	Analysis data					Radioactive ^4He ($\times10^{-8}$)	Primary fluid (R/Ra)
			$^3He/^4He$	R/Ra	$^{40}Ar/^{36}Ar$	^3He ($\times10^{-14}$)	^4He ($\times10^{-8}$)		
D-1	Red siltstone	2.8	$(1.40\pm0.69)\times10^{-6}$	1.00	600	17.13	12.21	0.59	1.05
D-2	Greyish-green siltstone	3.3	$(6.0\pm2.6)\times10^{-7}$	0.43	584	11.4	19.01	0.69	0.44
D-3	Grey course grit stone	169	$(3.31\pm0.24)\times10^{-8}$	0.02	1243	1.87	56.54	35.61	0.06
H-02	Grey course grit stone	216	$(8.8\pm1.3)\times10^{-7}$	0.63	1653	184.56	209.7	45.91	0.82
H-03	Grey course grit stone	369	$(0.59\pm2.71)\times10^{-7}$	0.42	1970	76.03	128.81	78.56	1.08
H-04	Grey course grit stone	224	$(1.134\pm0.13)\times10^{-7}$	0.81	1566	262.35	231.34	47.65	1.02

samples. This indicates that the $^3He/^4He$ and $^{40}Ar/^{36}Ar$ ratios of fluid inclusions from the Dongsheng uranium deposit are related to radioactive uranium. So it is necessary to probe into the factors affecting the He isotopic composition of fluid inclusions from the uranium ore deposit.

4.2.1 Factors affecting the He-Ar isotopic composition of fluid inclusions

The He-Ar isotopic composition of mineral fluid inclusions is controlled mainly by the following factors: 1) the diffusion-induced loss of He and its isotope fractionation; 2) deuterogenic geological processes; 3) ^3He resultant from cosmic rays; 4) radiogenic ^4He and ^{40}Ar produced from decay of the U, Th systems and K in the lattices of host minerals of fluid inclusions; and 5) He and Ar trapped in fluids at the time of formation of inclusions. However, He and Ar trapped in fluids only at the time of formation of inclusions (i.e., the primary He-Ar isotopic composition of inclusion fluids) can truly reflect the source of fluids and the He and Ar of other origins can not be the source of fluid inclusions, in return, they would influence their primary He-Ar isotopic composition.

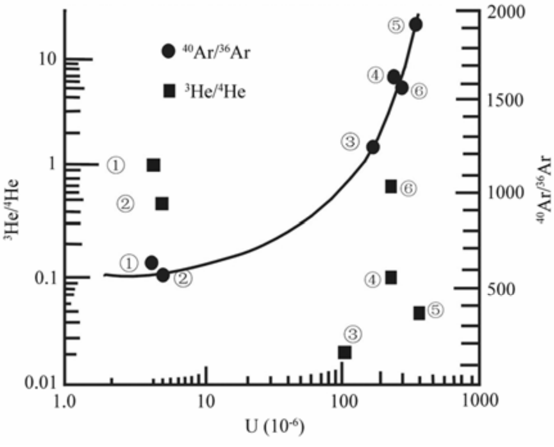

Fig. 4. Correlations between the contents of U and the ratios of $^3He/^4He$ and $^{40}Ar/^{36}Ar$ in fluid inclusions. ① Sample D-1; ② sample D-2; ③ sample D-3; ④ sample H-02; ⑤ sample H-03; ⑥ sample H-04.

Studies (Stuart et al., 1995; Hu Ruizhong and Bi Xianwu, 1999) have shown that different minerals possess varying capabilities of retaining He in fluid inclusions. As compared to metallic sulfide minerals such as pyrite, chalcopyrite and galena, although quartz fluid inclusions experienced obvious He loss in the long geological history, there would occur no remarkable He isotope fractionation in this process. Meanwhile, there would be no Ar loss. Microscopic observations showed that there had occurred neither significant He isotopic loss nor there was any sign of obvious late-stage transformation after the formation of fluid inclusions from the Dongsheng uranium deposit. Therefore, the post-ore geological processes had no significant influence on the He-Ar isotopic composition of fluid inclusions. As ^4He produced by cosmic rays is restricted on the Earth's surface at the depth of 1.5 m and the samples analyzed in this study were all drill core samples collected from a burial depth of about 500 m, the impact of cosmogenic He can be ruled out. From this it is known that the He and Ar isotopes in fluid inclusions from the Dongsheng uranium deposit are actually comprised predominantly of those trapped in fluids at the time the inclusions were formed and the radiogenic ^4He and ^{40}Ar resultant from decay of the U and Th systems and K in the lattices of minerals.

As Th is almost insoluble in hydrothermal fluids and the host minerals of fluid inclusions analyzed in this study are not K-bearing minerals such as quartz, He and Ar produced from radioactive decay of Th and K can be neglected. But, in consideration of the fact that fluid inclusions studied in this work are those from uranium ores, the samples contain a certain amount of radioactive U. Consequently, ^4He produced from radioactive decay of U would exert some influence on $^3He/^4He$ ratio. According to Craig and Lupton (1996), if the content of U in fluid inclusions was taken to be 3×10^{-6}, radiogenic ^{4}He produced after the formation of fluid inclusions can be neglected under the condition in which the ore-forming age of 50 Ma was taken as the boundary age. As the half life of U is relatively long (about 4500 million years), radiogenic ^{4}He in the two U-low samples analyzed in this study (D-1 and D-2 in Table 1) can also be neglected, as evidenced by actual calculations (Table 1). However, as for those U-high uranium ore samples (e.g. D-3 in Table 1), their U contents are estimated to be 169×10^{-6} and the amount of radiogenic ^{4}He since its formation was calculated to be 3.56×10^{-7} under the condition in which the forming time of 100 Ma for the Dongsheng uranium deposit was taken as the boundary age (the major ore-forming ages are within the range of 95–120 Ma; Zhang Ruliang, 2004). After deduction, the ^{3}He/^{4}He ratio of the primary inclusion fluids is 0.06 R, obviously higher than the measured value of 0.02 R. Listed in Table 1 are ^{3}He/^{4}He ratios in inclusion fluids after the radiogenic ^{4}He produced from radioactive decay of U in the uranium ores was deducted. It can be seen that the values are generally higher than the measured ones, indicating that the radiogenic ^{4}He contained in fluid inclusions from the Dongsheng uranium deposit could not be neglected.

It can be seen from the above analyses that radiogenic ^{4}He has a great influence only on ^{3}He/^{4}He ratios in fluid inclusions from U-high samples collected from the Dongsheng uranium deposit, but little influence on those of fluid inclusions from U-low samples collected from the same uranium deposit, generally reflecting the primary He-Ar isotopic composition at the time the fluid inclusions were formed.

4.2.2 The source of ore fluids of the Dongsheng uranium deposit

Studies have shown that ^{3}He/^{4}He ratios in crustal materials are 0.01–0.05 R/Ra and those in mantle materials are 6–9 R/Ra (Stuart et al., 1995). ^{3}He/^{4}He ratios in ore fluids from the Dongsheng uranium deposit are approximately 5–40 times higher than those of the crust, but obviously lower than those of the mantle. All this indicates that ore fluids from the Dongsheng uranium deposit contain not only crust-source He, but also mantle-source He. Fluid inclusions from the Dongsheng uranium deposit have high ^{40}Ar/^{36}Ar ratios (584–1970), obviously deviated from the isotopic composition of atmospheric argon (^{40}Ar/^{36}Ar=295.5; Stuart et al., 1995). Radiogenic argon and mantle-source argon are both characterized by high ^{40}Ar/^{36}Ar ratios. So merely on the basis of high ^{40}Ar/^{36}Ar ratios would it be impossible to distinguish radiogenic argon from mantle-source argon. However, high ^{40}Ar/^{36}Ar and ^{3}He/^{4}He ratios are the characteristic features of mantle-source fluids. As shown in the ^{40}Ar/^{36}Ar-^{3}He/^{4}He diagram (Fig. 5), the data points representing the results of fluid inclusion analysis for the Dongsheng uranium deposit all fall on both sides of the MORB-atmospherogenic crust fluid mixed line, implicating that ore fluids from the Dongsheng region have the compositional characteristics of binary (crust/mantle) fluid mixture. Although the analyzed inclusion samples from uranium ores have relatively low ^{3}He/^{4}He ratios (0.02–0.81 R/Ra), their primary ^{3}He/^{4}He ratios after the radiogenic ^{4}He was deducted are 0.06–1.08 R/Ra, also showing the compositional characteristics of crust/mantle mixed fluids.

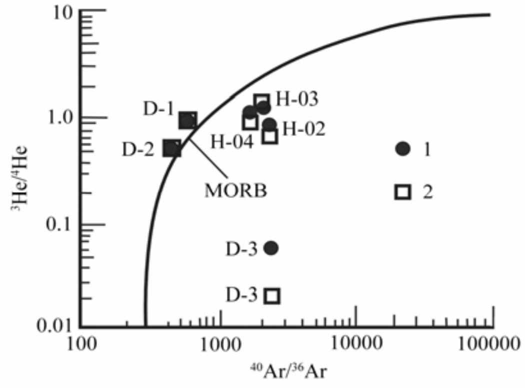

Fig. 5. ^{40}Ar/^{36}Ar-^{3}He/^{4}He diagram of fluid inclusions from the Dongsheng uranium deposit. 1. Original ratio; 2. results of experiments.

According to the simple binary mixing model, ^{3}He/^{4}He ratios can be used to impute the respective proportions of mantle- (Rm) and crust-source fluids (Rc) in the mixed fluid (Stuart et al., 1995). The proportion of mantle-source He can be calculated with the following formula: $[(R–Rc)/(Rm–Rc)]\times100\%$, where R is the He isotopic composition of inclusions, Rm and Rc represent the He isotopic composition of the mantle and that of the crust, respectively, with Rm=6–9 R/Ra and Rc=0.01–0.05 R/Ra. It is thus calculated that mantle-source fluids involved in metallogenesis account for 15%–17.5% of the ore fluids for the Dongsheng uranium deposit.

5 Comparisons of $\delta^{13}C_{V\text{-}PDB}$, $\delta^{18}O_{H_2O}$ and δD compositional characteristics of ore fluids

In order to further constrain the source of ore fluids for the Dongsheng uranium deposit, this study also dealt with the C, H and O isotopic composition of ore fluids for the Dongsheng uranium deposit (Table 2), including fluid inclusions in quartz sandstone, calcite veins and kaolinite in the basement of the uranium deposit. On the basis of the principle of H and O isotope fractionation, in the process of interaction between fluids and country rocks δD_{H_2O}

varies only slightly, but O isotopic fractionation and shift would occur with ore-forming temperature and water/rock interaction, and their controlling factors are mainly reaction temperature, time and water/rock ratio (Taylor, 1974). The oxygen isotopic composition ($\delta^{18}O_{H_2O}$) of ore fluids in isotopic exchange equilibrium with the host mineral (quartz) in quartz sandstone from the Dongsheng uranium deposit was calculated in terms of the oxygen isotopic composition of quartz ($\delta^{18}O_{quartz}$) with the formula $\delta^{18}O_{H_2O}=\delta^{18}O_{quartz}-3.38\times10^6T^{-2}+3.4$ (Clayton and O'Neil, 1972); the $\delta^{18}O_{H_2O}$ values of kaolinite fluids were calculated with the fractionation equation $\delta^{18}O_{H_2O}=\delta^{18}O_{kaolinite}-2.05\times10^6T^{-2}+3.68$ (Kulla, 1978); and those of calcite fluids were calculated with the fractionation equation $\delta^{18}O_{H_2O}=\delta^{18}O_{calcite}-2.78\times10^6T^{-2}+2.89$ (O'Neil and Clayton, 1969). *T* was worked out by conversion of the average homogenization temperature (150°C) of fluid inclusions (expressed as the absolute temperature, *K*) and the results are listed in Table 2.

The $\delta^{18}O_{H_2O}$ values of magmatic water are 7.0‰–9.0‰ and its δD values are -50‰– -80‰ (Rollinson, 1993). From Table 2 it is seen that the $\delta^{18}O_{H_2O}$ values of fluid inclusions in the quartz sandstone of the Dongsheng uranium deposit vary between -3.0‰ and -8.75‰, and the δD values are within the range of -55.8‰– -71.3‰. On the H-O isotopic composition diagram (Fig. 6), the $\delta^{18}O_{H_2O}$ and δD data points all fall in the area between meteoric water and magmatic water, showing that the ore fluids are characteristic of a mixed fluid of meteoric water and magmatic water.

Kaolinite in the foot-wall rocks of the Dongsheng uranium deposit has a $\delta^{18}O_{H_2O}$ value of 6.1‰ and a δD value of -77‰. As shown on the H-O isotopic composition diagram, all the data points fall in the area of magmatic water (Fig. 6), displaying the characteristics of magmatic water. But in common cases it is hard to distinguish magmatic water from mantle-source water; in most cases magmatic water and mantle-source water both have similar geochemical characteristics or they usually mix together in geological processes. One calcite vein formed at the metallogenic stage of the Dongsheng uranium deposit has a $\delta^{13}C_{V\text{-}PDB}$ value of -8.0‰ and a $\delta^{18}O_{H_2O}$ value of 5.76‰ (Table 2), equivalent or close to the isotopic ratios of mantle-derived carbon (-5‰±2‰) and oxygen (5.7‰±0.3‰) determined by Rollinson (1993), displaying the characteristics of mantle derivation. From the above analysis we can see that the compositional characteristics ($\delta^{13}C_{V\text{-}PDB}$, $\delta^{18}O_{H_2O}$ and δD ‰) of rock-forming minerals and the He-Ar isotopic composition characteristics of fluid inclusions from uranium ores at Dongsheng consistently indicate that the ore fluids of the Dongsheng uranium deposit seem to have come from the deep interior and possess the characteristics of crust/mantle mixed fluid.

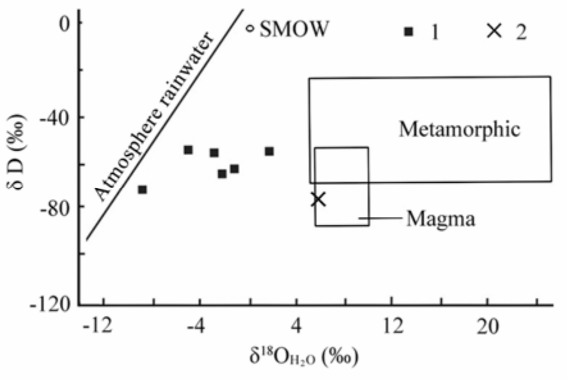

Fig. 6. Variations in $\delta^{18}O_{H_2O}$ and δD of fluid inclusions.
1. Fluid inclusions in quartz; 2. kaolinite.

6 Discussion and conclusions

Since the Cenozoic the Ordos Basin has experienced an integrated uplift stage, with a conversion from Late Paleozoic marine/continent transitional-facies coal series sedimentary strata to inland lake-facies and fluvial-facies sedimentation. According to the geophysics data available, the basement of the Yimeng uplift zone is composed of shallow-buried old metamorphic rocks and granites, the overlying Phanerozoic sedimentary cover strata are less than 3000 m in thickness (Zhang Hong et al., 1995). According to the analysis of stratigraphic

Table 2. Hydrogen and oxygen isotopic analyses of ore fluids from the Dongsheng uranium deposit (SMOW, ‰)

Sample No.	Rock type	Sample	$\delta^{18}O$	$\delta^{18}O_{H_2O}$	δD	$\delta^{13}C_{V\text{-}PDB}$
HD-1	Red siltstone	Quartz inclusion	6.73	-8.75	-71.3	
HD-2	Greyish-green muddy siltstone	Quartz inclusion	13.45	-2.03	-61.87	
HD-3	Grey grit stone	Quartz inclusion	12.48	-3.0	-55.8	
H-02	Grey grit stone	Quartz inclusion	15.26	-0.22	-68.64	
H-03	Grey grit stone	Quartz inclusion	11.04	-4.45	-57.53	
H-04	Grey grit stone	Quartz inclusion	17.83	2.34	-59.81	
HD-5	Kaolinite		13.7	6.1	-77	
HD-4	Calcite vein		18.4	5.76		-8.0

records, since the Early Paleozoic the Dongsheng region has experienced a number of important tectonic movements, leading to the denudation, thus deformity, of the Early Paleozoic, Late Paleozoic, Triassic, Jurassic and Cretaceous strata. The residual strata are variable in thickness. Paleomorphologically, the Ordos Basin was high in the north and low in the south during the Jurassic-Cretaceous period, where water flew in the direction from north to south and gathered in the center of the basin. Widespread both in the Daqingshan region on the northeastern margin of the basin and in the Northwest Shanxi uplift zone are metamorphic rocks and granites. These rocks had been weathering-denudated and their U-rich clastics in the source region were transported by meteoric waters to the Dongsheng region and were deposited as a suite of Middle Jurassic Zhiluo Formation sandstones composed of U-bearing clastic granules there.

Studies have shown that tectonothermal events were of extensive occurrence in the Ordos Basin during the Mesozoic period, giving rise to regional geothermal anomalies. Middle Triassic plaeo-geothermal gradient is so high as to be up to 5.7℃ /100 m (Zhao Mengwei, 1996) and Jurassic-Cretaceous paleo-geothermal gradient is about 4.0℃ /100 m (Ren Zhanli and Zhao Chongyuan, 1994). In the periphery and interior of the basin volcano-magmatic activities were frequent, giving rise to quite a number of igneous bodies, for example, the Early Cretaceous Zijinshan alkali massif (its isotopic age is 125 Ma; Ren Zhanli and Zhao Chongyuan, 1994) which, covering an area of 23 km^2, is distributed around Linxian County, Shanxi Province in the eastern part of the basin. In addition, in the Triassic strata exposed in some drilling wells of the basin are recognized basic volcanic dykes, and in the Triassic strata of some individual drilling wells are recognized diorite porphyrites as thick as 150 m (Sun Shaohua, 1997).

The Ordos Basin is generally considered to be one of the most stable basins in China, even in the whole Mid-Asia region. Nearly horizontal strata and gently monoclinal structures are the fundamental geological features (Zhang Hong et al., 1995; Zhang Ruliang, 2004). But in the Dongsheng region a large number of microfractures can be seen in the stratigraphic exposures in the field, which are measured at several centimeters to several tens of centimeters. In accordance with the aeromagnetic remote-sensing data, fault structures are extremely developed in the basement of this region, mainly including three sets of NW-, NE- and nearly EW-extending fault structures. These faults are characterized as being large in scale, deep in cutting and distant in extension, and they more or less control the distribution of the basement strata and sedimentary cover strata. The uranium deposit is located in the hanging wall of the NW-extending fault, microscopic observations revealed that quartz in the sandstone of the uranium deposit exhibited waved extinction, fractures resultant from breaking of feldspar and rock fragments were well developed, mica was of distortion, and sericitization and other phenomena of dynamic metamorphism were observed.

High-temperature anomalies which had appeared in the Ordos Basin during the Mesozoic speeded up the thermal evolution of coal, even its metamorphism, also speeded up the formation and migration of oil and gas (Zhang Ruliang, 2004). Tectonothermal events led to the formation of fractures in large numbers and the mobilization of deep giant faults, thus providing favorable channels for upwarping of deep fluids (including shallow-level oil and gas). It is these channels that made the deep-seated thermal fluids migrate upwards and fill in U-bearing fragmental sandstones to be involved in uranium metallogenesis. The major ore-forming age of the Dongsheng uranium deposit was determined to be 95–120 Ma, equivalent to late Early Cretaceous. It is at that time that Yanshanian tectonomagmatic thermal events occurred extensively, and it is also at that time that the formation of oil and gas reached its fastigium in the Ordos Basin (Zhang Ruliang, 2004; Zhao Mengwei, 1996). From the above it is known that there is some temporal relationship between magmatic activities in the deep interior and the formation of uranium ores.

To sum up, ore fluids responsible for the Dongsheng uranium deposit possess the characteristics of mixing of crust fluids and deep-interior fluids. During the Jurassic-Cretaceous the Ordos Basin was paleo-geomorphologically characterized as being high in the north and low in the south. Widespread both in the Daqingshan region on the northeastern margin of the basin and in the Northwest Shanxi uplift zone are metamorphic rocks and granites. These rocks had been weathering-denudated and their U-rich clastics in the source region were transported by meteoric waters to the Dongsheng region and were deposited as a suite of Middle Jurassic Zhiluo Formation sandstones composed of U-bearing clastic granules there. Mesozoic tectonothermal events in the Ordos Basin speeded up the formation of deep-seated fluids (including volcanomagmatic hydrothermal solutions) and shallow-level oil and gas and the mobilization of fractures and deep giant faults in large numbers, thus providing favorable channels for upwarping of deep-seated fluids and shallow-level oil and gas along the faults and deep giant fault zones. It is these channels that made the deep-seated thermal fluids migrate upwards and fill in U-bearing fragmental sandstones to be involved in uranium mobilization and metallogenesis. But He-Ar isotopic studies of fluid inclusions in this work have shown that the deep-seated fluids that were involved in metallogenesis account for 6.15%–17.5%. It is preliminarily considered that the most important role of the deep-seated fluids is to have provided a heat source for the mobilization and metallogenesis of uranium ores.

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