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Uranium enrichment in lacustrine oil source rocks of the Chang 7 member of the Yanchang Formation, Erdos Basin, China

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ABSTRACT

The oil source rocks of the Chang 7 member of the Yanchang Formation in the Erdos Basin were deposited during maximum lake extension during the Late Triassic and show a remarkable positive uranium anomaly, with an average uranium content as high as 51.1 μg/g. Uranium is enriched together with organic matter and elements such as Fe, S, Cu, V and Mo in the rocks. The detailed biological markers determined in the Chang 7 member indicate that the lake water column was oxidizing during deposition of the Chang 7 member. However, redox indicators for sediments such as S^{2-} content, V/Sc and V/(V + Ni) ratios demonstrate that it was a typical anoxic diagenetic setting. The contrasted redox conditions between the water column and the sediment with a very high content of organic matter provided favorable physical and chemical conditions for syngenetic uranium enrichment in the oil source rocks of the Chang 7 member. Possible uranium sources may be the extensive U-rich volcanic ash that resulted from contemporaneous volcanic eruption and uranium material transported by hydrothermal conduits into the basin. The uranium from terrestrial clastics was unlike because uranium concentration was not higher in the margin area of basin where the terrestrial material input was high. As indicated by correlative analysis, the oil source rocks of the Chang 7 member show high gamma-ray values for radioactive well log data that reflect a positive uranium anomaly and are characterized by high resistance, low electric potential and low density. As a result, well log data can be used to identify positive uranium anomalies and spatial distribution of the oil source rocks in the Erdos Basin. The estimation of the total uranium reserves in the Chang 7 member attain 0.8×10^8 t.

1. Introduction

Black shale generally contains high concentrations of uranium (Swanson, 1956; Disnar and Sureau, 1990). For example, the uranium concentration in Pennsylvanian black shale is closely associated with organic matter in the shales and a reducing depositional environment (Doveton and Merriam, 2004; Coveney and Martin, 1983; Coveney et al., 1987; Coveney et al., 1989). The maximum uranium concentration in those shales reaches 320 μg/g. Uranium in Devonian shale in the Appalachian Basin of the eastern USA, in the Parkhouse marine shale from the Late Carboniferous in the UK and in the Timahdit black shale in Morocco are all abundant, with concentrations of 2–36 μg/g, >8 μg/g and 41 μg/g, respectively (Lev and Filer, 2004; Lev et al., 2008; Fisher, 2001; Galindo et al., 2007). In China, the best-known uranium-rich strata are Lower Silurian and Lower Cambrian black shales that occur in the Yangtze Platform with a uranium concentration >10 μg/g. Some samples contain up to 80 μg/g of uranium (Liu, 1992). The close relationship between organic matter and uranium in black shale is explained by the properties of organic matter acting in the processes of uranium concentration. Organic matter may serve as sorbent, reducing and complexing agent. The geological settings of uranium in black shales support the view that enrichment of uranium is performed at the syngenetic stage (references within Disnar and Sureau, 1990).

Even though numerous reports have been published on uranium enrichment in black shales, the identification of uranium sources were tentative, and the discussions about uranium supplies were quite general. Available data indicate that uranium can be derived from weathering of a granite landmass, with subsequent basin entry via river or groundwater flows followed by concentration (Zhang, 1992; Li, 2007). In other cases, uranium could be derived from global rift belts, such as the oceanic rift in the Red Sea, with oceanic entry via volcanism or hydrothermal activity

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(Zhang, 1992). Uranium concentrations in black shale in South China were considered to have originated from a hydrothermal reworking of uranium-rich rocks (Min, 1995), whereas uranium concentrations in polymetallic hydrothermal deposits in the Red Sea were considered to be a result of underwater hydrothermal events (Zhang, 1992).

The most cases of uranium enrichment in black shale were reported in marine deposition. The lacustrine oil source rock generally do not contain a high uranium content because the uranium sources in lacustrine sediments are usually insufficient. In recent years, lacustrine rocks, characterized by remarkable positive radioactive gamma-ray values, and showing extensive distribution areas and varying thicknesses, were detected at the base of the Chang 7 member in the Erdos Basin. Organic geochemists subsequently verified that this occurrence was a series of organic-rich oil source rocks (Yang and Zhang, 2005), which contain high trace elements such as U, Mo, Cu and V. In the present paper, we characterize this uranium anomaly and discuss the factors affecting uranium enrichment. This study should help to identify the mechanism of uranium enrichment in lacustrine sediment and shed light on the possible origin of these oil source rocks.

2. Geological background

As part of the North China Craton, the Erdos Basin developed as a terrestrial lake with a maximum water area of ca. 1×10^5 km^2 in the Late Triassic. During this period, the North China landmass collided and subsequently integrated with the Yangtze landmass, which led to closure of the relict of Youjiang and Qinling troughs and formation of the Qinling Mountains (Yang, 2002). As a result, evolution of the Erdos terrestrial lake basin in the Late Triassic possibly was controlled mainly by the orogeny that occurred at the Qinling tectonic belt (Zhang et al., 2006). Intense regional tectonic movement may have resulted in a maximum lake area at an early stage of formation of the Chang 7 member and large-scale deposition of organic-rich source rocks in deep to semi-deep water of the lake (Yang and Zhang, 2005).

3. Sample collection and analyses

Samples were collected from drillholes, representing a full set of cores from the Chang 7 member, and from field outcrop profiles of oil shales of the Chang 7 exposed in Tongchuan in the southeast part of the basin (Fig. 1). Oil shale samples of the Chang 7 were collected from cores of the Li 57, Li 68, Zheng 8 and Ning 36 wells. The samples were taken generally at an interval of 0.5–1.0 m, depending on the core length and rock type. Field outcrop samples were taken at an interval of 1 m. Samples were rinsed with distilled water and dried before further sampling for testing. Each test sample was approximately 10 mm thick and weighed approximately 10 g. In total, 107 test samples were taken, including 68 for oil shales from Chang 7, 12 for black argillaceous rocks from Chang 7, and 27 for dark-colored argillaceous rocks from the Chang 6, Chang 8 and Chang 9 members, respectively.

Major and trace elements in the samples were analyzed at the Analytical Center of the No. 203 Research Institute of the Ministry for the Nuclear Industry in China. Testing was carried out according to China national standard GB/T16399-1996 for SiO_2 and to China national standard GB/T14506-1993 for Al_2O_3, Fe_2O_3, CaO, MgO, TiO_2, P, K_2O and Na_2O using Shimadzu UV-2401PC and AA-6800 instruments. Trace elements were tested according to China national standard DZ/T0223-2001 by inductively coupled plasma mass spectrometry (ICP-MS; Finnigan MAT). Representative samples were sent to the Testing Center of the Exploration

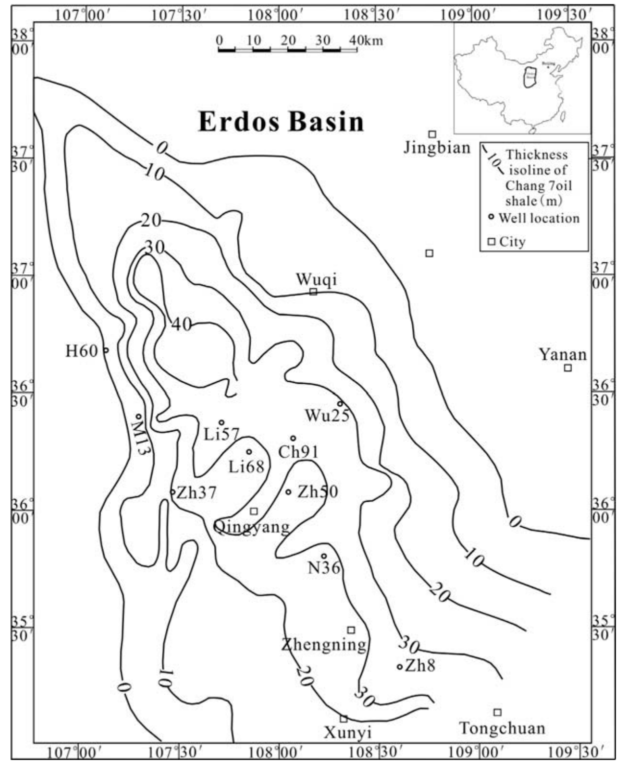

Fig. 1. Sketch map showing the sampling locations. The isolines are the thickness isolines of the Chang 7 member oil shale: ○ well; □ city; △ inset, showing the location of Erdos Basin.

and Development Research Institute, Changqing Oilfield, PetroChina for C and S testing (using a CS-400 carbon and sulfur analyzer, Leco Corp., USA), scanning electron microscopy coupled to energy-dispersive spectroscopy (SEM-EDS, model S-3200 SEM and LINK ISIS 300 X-ray EDS), microscopic observation and mineral identification.

To extract the bitumen, samples were pulverized and extracted in a Soxhlet apparatus using dichloromethane/methanol (93:7 v/v) for 72 h. The concentrated extracts were subjected to asphaltene precipitation by adding a 40-fold volume of *n*-hexane, followed by column chromatography on silica gel/Al_2O_3. *n*-Hexane and dichloromethane were used as the elution solvents for the saturated and aromatic hydrocarbons. Biological markers in the saturated and aromatic hydrocarbons were analyzed on a Finnigan Voyager mass spectrometer interfaced with a CE GC8000^{top} gas chromatograph. A fused silica column (DB5-ms, 30 m × 0.32 mm i.d., film thickness 0.25 μm; J&W) was used for chromatography. The oven temperature was held at 50 °C for 5 min, then increased at 3 °C min^{-1} to 300 °C and isothermal at 300 °C for 15 min.

4. Results and discussion

Because a great number of samples were analyzed, only analytical data for some representative core samples are listed in Table 1. The statistical elemental contents and elemental ratios for different horizons and different rock types are listed in Table 2. The data indicate that the uranium content in oil source rock from the Chang 7 member varies from 5.4 μg/g to 140.0 μg/g, for an average of 51.1 μg/g, which is remarkably higher than for oil source rocks in other members such as the Chang 9, Chang 8 and Chang 6 (Table 2).