

Enhancement of organic matter maturation because of radiogenic heat from uranium: A case study from the Ordos Basin in China

Fan Zhang, Yangquan Jiao, Liqun Wu, Hui Rong, Jinhua Li, and Dun Wan

ABSTRACT

Carbonaceous debris (CD) within uranium-bearing strata has been studied in the Daying uranium deposit of the northern Ordos Basin, northern China. The influence of radiogenic heat from uranium on organic matter maturation was investigated through a series of tests including measurements of vitrinite reflectance (R_o), fission-track (FT) analysis in quartz grains, and the calculation of the radiogenic heat production rate of the samples. The results show that R_o in uranium-bearing strata generally increases as the burial depth increases, indicating that CD experienced normal burial coalification. However, R_o values of the samples rich in uranium are 0.062% R_o higher than those without uranium mineralization. Vitrinite reflectance bears a positive relationship with uranium content, and an inverse relationship with distance to the closest sandstone rich in uranium, indicating that uranium enrichment enhances organic matter maturation. The production of uranium decay makes FT observable in quartz grains, and the intensity of decay increases with proximity to the uranium ore body. The calculated radioactive heat production rate from the uranium ore body is $6.857 \times 10^{-5} \text{ W/m}^3$. During the long-term stable decay, as the uranium ore body theoretically results in an abnormal increase in temperature of 52°C without consideration of the loss of heat conduction, heat convection, and thermal radiation, this would yield a theoretical R_o increase of 0.209% R_o , reasonably greater than the observed. Therefore, the long-term stable radiogenic heat produced by uranium ore body can slightly enhance organic matter maturation, which is instructive in uranium prospecting.

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INTRODUCTION

Vitrinite reflectance (R_o) is the most commonly used indicator for evaluating thermal maturation of organic matter (International Committee for Coal and Organic Petrology, 1998; Carr, 2003). In addition to temperature and time, organic matter is influenced by other geological factors, for example, overpressure (Carr, 2003; Hao et al., 2007), liptinite content (Wenger and Baker, 1987; Lo, 1993; Huang, 1996), and lithologic boundaries (Mukhopadhyay 1992; Goodarzi et al., 1993). It has also been found that organic-inorganic interactions have a certain influence on thermal evolution of organic matter, for example, metal enhancement of organic matter maturation (Mango et al., 1994; Seewald et al., 2000).

Uranium, a radioactive metal, has a close relationship with organic matter in sedimentary basins (Gauthier-Lafaye and Weber, 1993; Cuney, 2010; Havelcová, et al., 2014). Uranium enhances organic matter maturation during thermal evolution while organic matter adsorbs, concentrates, and mineralizes uranium in sediments (Spirakis, 1996; Bordelet et al., 2013; Ortaboy and Atun, 2014). Sassen (1984) observed an increase in organic matter maturation and a disappearance in fluorescence through radiation over geologic time. Smieja-Król et al. (2009) found a positive relationship between maximum reflectance and uranium content in South Africa. Havelcová et al. (2014) observed that there was a decrease in H/C atomic ratio and an increase in O/C atomic ratio as uranium concentration increased within coals in the Sokolov Basin, Czech Republic. Similar phenomena are also found in bitumens associated with uranium mineralization (e.g., Gauthier-Lafaye and Weber, 1993; Nagy et al., 1993; Landais, 1996; Court et al., 2006; Jaraula et al., 2015).

Although enhanced influences of uranium on organic matter maturation have been reported in numerous papers, currently there is no published relationship between uranium content and organic matter maturation. The Daying uranium deposit is a superlarge ancient sandstone body in the Ordos Basin, China (Xie, 2016). Moreover, carbonaceous debris (CD) is abundant in uranium-bearing strata, and coexists spatially with uranium minerals (Cun et al., 2016; Xie, 2016), providing a favorable opportunity for investigating how uranium makes an impact on organic matter maturation.

In the present study, organic matter maturation of CD from uranium-bearing strata in the Daying uranium deposit, northern Ordos Basin, is investigated using R_o in an attempt to acquire information about the influence of uranium on organic matter maturation. More attention is paid to the quantitative influences of radiogenic heat from the entire uranium ore body on organic matter maturation.

GEOLOGIC SETTING

The Ordos Basin, with an area of approximately 250,000 km² (~96,526 mi²), is situated between the Inner Mongolia–Daxing'anling fold belt and the Qilian–Qinling fold belt in northern China. It developed as an intracontinental depression basin during the Mesozoic (Ren et al., 1994; Xue et al., 2010; Figure 1A). The Daying uranium deposit is located in the Yimeng uplift, northern Ordos Basin next to the Hetao graben (Figure 1A). Approximate east–west folds and local small normal faults are developed in the study area (Figure 1B). The strata from bottom to top are mainly the Jurassic Yan'an Formation (J_{2y}), Zhiluo Formation (J_{2z}), Anding Formation (J_{2a}), and Cretaceous strata (K). An unconformity exists between J_{2y} and J_{2z} (Jiao et al., 2016; Figure 2).

The main uranium-bearing unit with a thickness of approximately 130 m (~430 ft), J_{2z}, is divided into three members: lower member (J_{2z}¹), middle member (J_{2z}²), and upper member (J_{2z}³; Jiao et al., 2006). Primarily composed of gray sandstone and mudstone with a few local coals, J_{2z}¹ is classified as a lowstand system tract (LST). Mostly made of gray-green and purple-red mudstone, J_{2z}² is grouped into a lacustrine-expanding system tract (EST). Mostly formed by coarse sandstone, J_{2z}³ represents a highstand system tract (HST). The major uranium-bearing unit, J_{2z}¹, is further subdivided into the lower submember (J_{2z}¹⁻¹) and the upper submember (J_{2z}¹⁻²; Jiao et al., 2006). Fine-grained sedimentary rocks (e.g., coal and mudstone) underlay at the base of J_{2z}¹⁻² (Jiao et al., 2016; Figure 2).

The sandstone-type uranium deposit forms as uranium (VI)-bearing groundwater migrates to the edge of interlayer oxidation zone where precipitation takes place and then the uranium ore body develops by adsorption or reduction (Harshman, 1972). In the study area, the Yanshan movement and Hetao graben made the sandstone bodies undergo a secondary reduction event (Jiao et al., 2015). The sandstone bodies include oxidized sandstone bodies (i.e., paleo-oxidation zone) and gray reduced sandstone bodies without uranium mineralization (i.e., reduction zone; Figure 3). The paleo-oxidation zone in boreholes is primarily composed of green and gray-green sandstone (sporadic red sandstone) with minor CD (Li et al., 2007; Figure 3), whereas gray sandstone without uranium mineralization (reduction zone) is mostly formed of gray and light gray sandstone with abundant CD (Figures 3; 4A, B). Ore bodies occur in gray sandstones next to the paleo-oxidation zone, that is, gray sandstones rich in uranium occur in lenticular or tabular shapes with abundant CD (Figures 3; 4C, D). Uranium-rich sandstone is mostly made up of quartz and feldspar with minor uranium mineralization (Y. Jiao, L. Wu, H. Rong, Y. Liu, H. Wang, H. Xie, Q. Zhu, J. Li, M. Liu, L. Wang, Z. Wwang, L. Wan, D. Wan, F. Zhang, Z. Yan, and Q. Xiong, 2012, personal communication). Widely interspersed in sandstone

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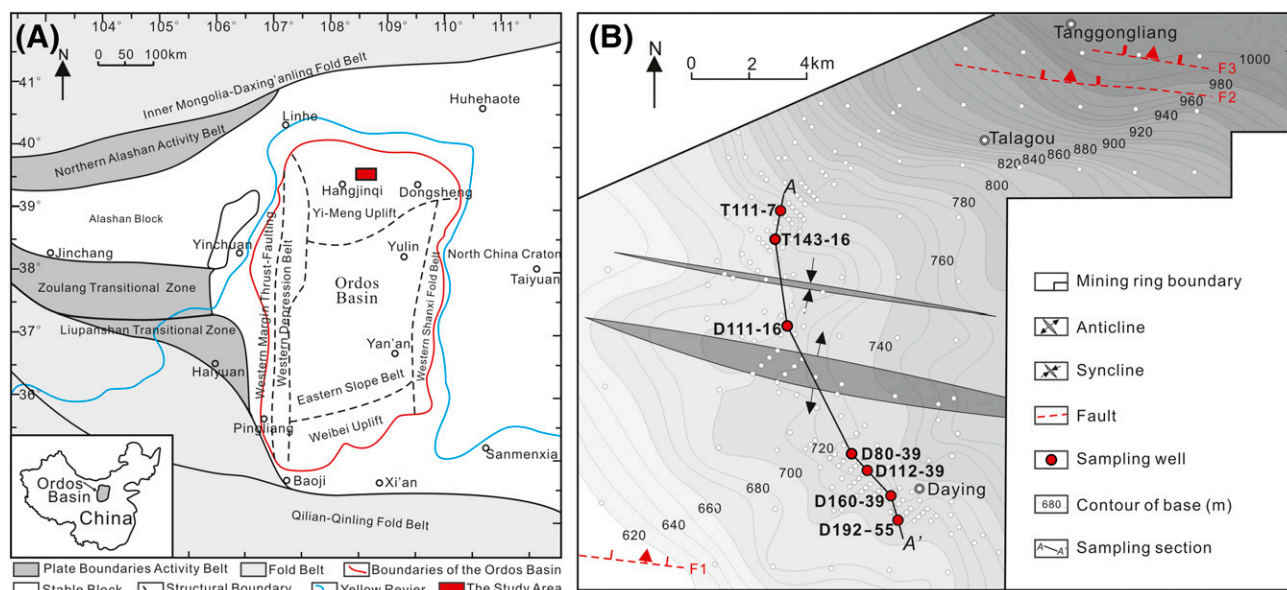


Figure 1. (A) Regional tectonic setting of the Ordos Basin. (Inset) The location of the Ordos Basin in China. (B) A structure map on the base of the Zhiluo Formation including locations of sampling wells in the Daying uranium deposit.

bodies in the forms of lumps and bands, ranging from 1- to 20-mm (0.04–0.79-in.) thickness (Figure 4A–C), CDs are mostly composed of vitrinite (type III kerogen). Moreover, CD is associated with uranium minerals (Figure 4D, E). The oldest age of uranium mineralization is 177–149 Ma (Middle–Late Jurassic), followed by several younger episodes, that is, 131–109 Ma (Early Cretaceous), 85–75 Ma (Late Cretaceous), 22–18 Ma (Miocene), and 9–7 Ma (Miocene). The main metallogenetic periods are Late Jurassic and Late Cretaceous (Xia et al., 2003; Xia and Liu, 2005; Jiao et al., 2015).

SAMPLING AND METHODS

Specific radioactivity (A , Bq/kg) is used to reflect uranium content (Dong, 1995). In the study area, the grade of uranium mineralization is divided into four groups by A : industrial uranium mineralization (IUM; >108 Bq/kg), boundary uranium mineralization (BUM; 36–108 Bq/kg), abnormal uranium mineralization (AUM; 18–36 Bq/kg), and no uranium mineralization (NUM; <18 Bq/kg; Dong, 1995; EJ/T 611-2005, Standard Navigation Network, 2005). Forty-six samples, that is, 2 coals and 44 samples of CD within sandstone, covering different zones (i.e., paleo-oxidation zone, uranium ore body, and reduction zone) and different mineralization grades, were systematically collected from uranium-bearing

strata from boreholes T111-7, T143-16, D111-16, D80-39, D112-39, D160-39, and D192-55. Among the collected samples, one is from J_{2y} . The other 45 samples from J_{2z}^1 , including 2 samples (i.e., D111-16-5 and D192-55-5) from coals in the reduction zone, 3 samples (i.e., D1143-16-3, D80-39-1, and D192-55-3) from gray-green sandstone in the paleo-oxidation zone influenced by a secondary reduction, 17 samples from gray sandstone rich in uranium, and 23 samples from gray sandstone without uranium mineralization in the reduction zone. The samples rich in uranium contain nine samples in IUM (i.e., D112-39-1 to D112-39-6, D80-39-2, D80-39-3, and D160-39-4), five samples in BUM (i.e., D111-16-2, T143-16-4, D80-39-4, D80-39-6, and D80-39-8), and three samples in AUM (i.e., D111-16-1, D111-16-3 and D192-55-1). The data of sampling locations, lithology, mineralization information, A , R_o , and distances from sampling points to the closest sandstone rich in uranium are shown in Figure 3 and Table 1.

Selected from sandstone by using tweezers, CD was crushed to less than 0.2 mm (0.08 in.), and 0.2 to 1 mm (0.08–0.04 in.), respectively. Then, CD with diameters less than 0.2 mm (0.08 in.) was mixed with the samples with diameters 0.2 to 1 mm (0.08–0.04 in.) by 1:4 or 1:5 to provide for better petrographic analysis. A representative pile of the mixed, air-dried CD was mixed with epoxy resin by a 5:1 ratio and solidified to form a cylindrical sample. The samples

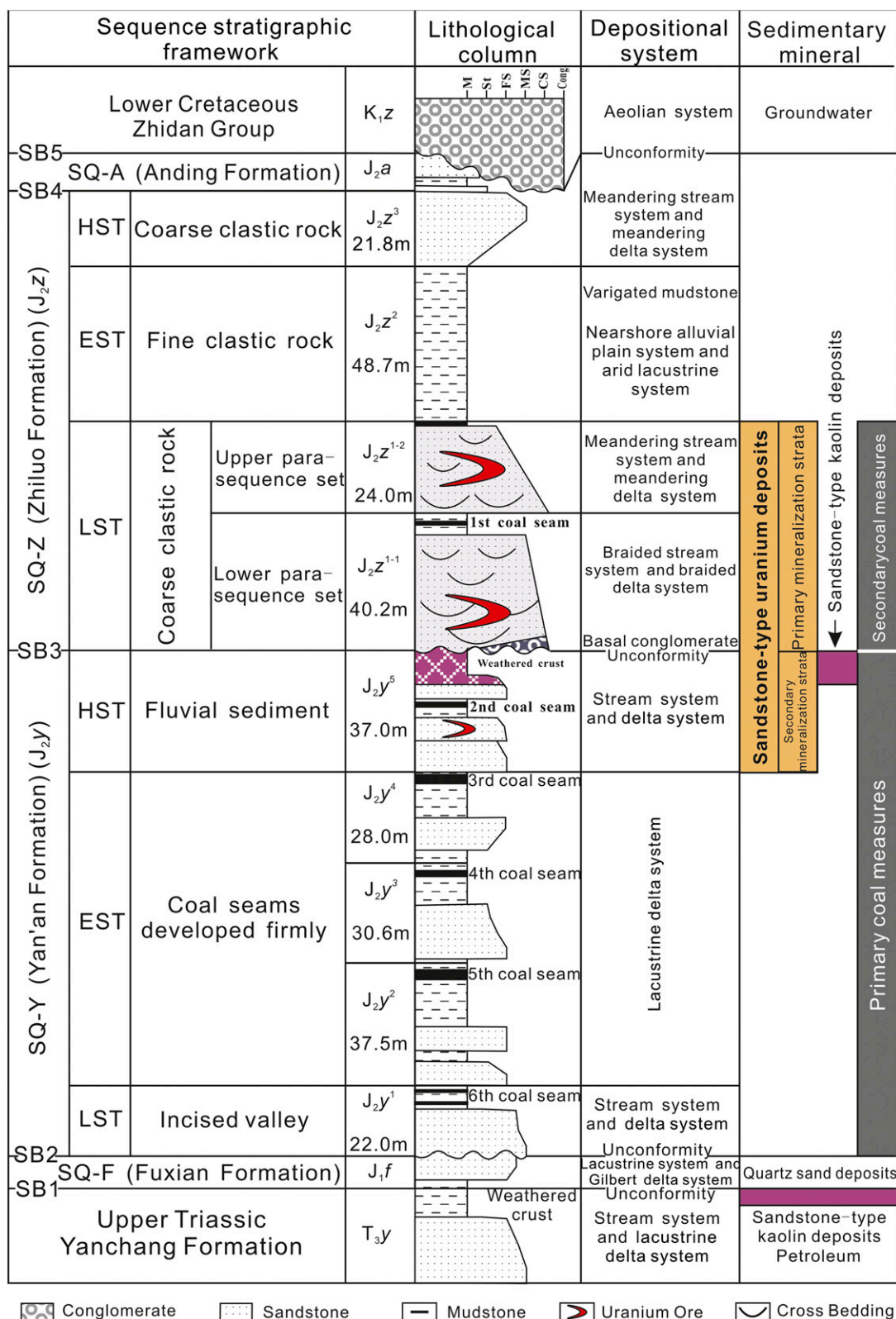


Figure 2. Stratigraphic column of the Daying uranium deposit, showing the distributions of the Yan'an Formation (J_{2y}), Zhiluo Formation (J_{2z}), Aning Formation (J_{2a}), and Cretaceous strata (K) (after Jiao et al., 2016). Cong = conglomerate; CS = coarse sandstone; EST = expanding systems tract; FS = fine sandstone; HST = highstand systems tract; LST = lowstand systems tract; M = mudstone; MS = medium sandstone; SB = sequence boundary; SQ = third-order sequence; St = siltstone.

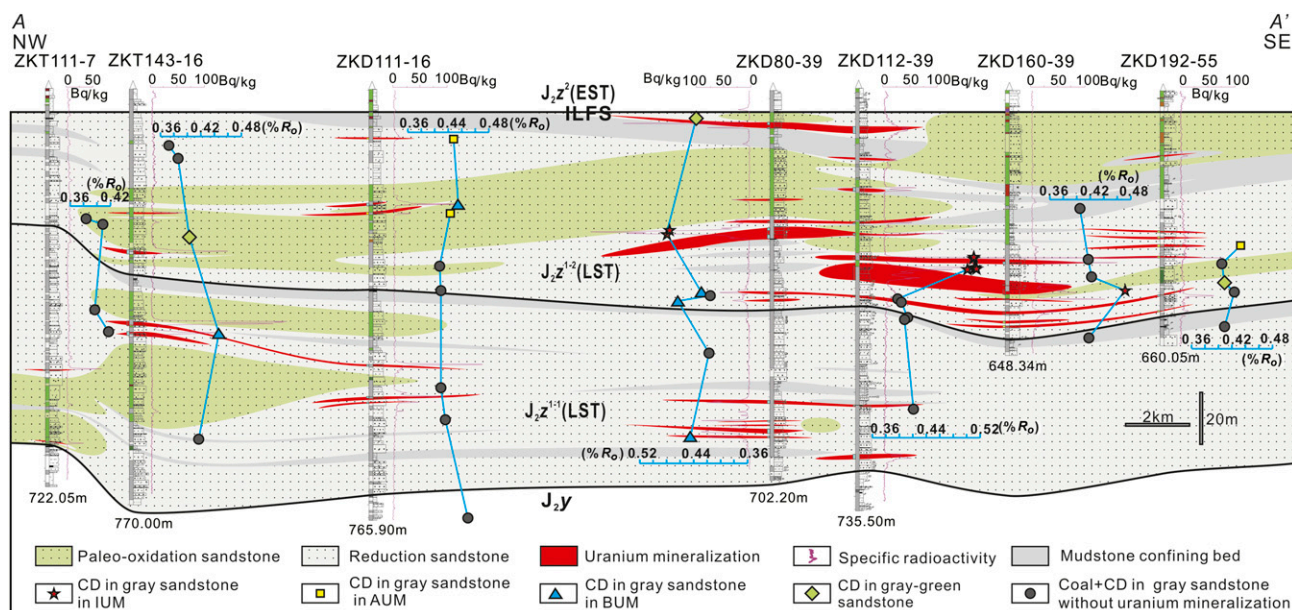


Figure 3. Regional lithologic geochemical profile in the Daying uranium deposit and sampling locations (the location of the sampling section is seen Figure 1B). AUM = abnormal uranium mineralization; BUM = boundary uranium mineralization; CD = carbonaceous debris; EST = expanding systems tract; ILFS = initial lake flooding surface; IUM = industrial uranium mineralization; J_2y = Yan'an Formation; J_2z = Zhiluo Formation; LST = lowstand systems tract; R_o = vitrinite reflectance.

were mechanically polished for R_o analysis, by first using 400 mesh, then followed by 600 mesh, and finally 2000 mesh polishing compounds (Stach et al., 1982).

The R_o analysis was performed at the China University of Geosciences (Wuhan), Key Laboratory of Tectonics and Petroleum Resources, Ministry of Education. The measurements of maximum R_o were made on nonoriented collotelinite grains using a Zeiss microscope equipped with a high-definition photometer with a plane-polarized monochromatic light ($\lambda = 546 \text{ nm}$), at a temperature of 23°C with an oil-immersion lens. During a complete 360° rotation on the platform, the maximum reflectance value (R) was recorded. Before measurements, the microscope was calibrated using sapphire ($R = 0.590\% R_o$), yttrium aluminum garnet ($R = 0.904\% R_o$), and gadolinium–gallium–garnet ($R = 1.719\% R_o$).

RESULTS

Measured R_o ranges from $0.372\% R_o$ to $0.514\% R_o$, with the average value of $0.432\% R_o$ (Figure 5; Table 1), indicating that the majority of CD in uranium-bearing strata is immature, whereas a small fraction is at a low-maturity stage.

An initial lake flooding surface (ILFS) exists between J_2z^1 (i.e., LST) and J_2z^2 (i.e., EST). Sampling depth does not represent maximum burial depth, so to actually obtain the variation of R_o in the study area, the burial depth is standardized using ILFS, which is a standard surface, thus providing a clearer perspective of evaluating the relationship between R_o and the burial depth. Moreover, gray-green sandstones and gray sandstones rich in uranium are removed to avoid the influences of uranium on R_o during the maturation process so as to actually obtain simply the burial effect on R_o . The results show that R_o generally increases as the burial depth increases, but there is a significant scatter [correlation coefficient $[R^2] = 0.475$; Figures 3, 5B), indicating that organic matter experienced normal burial coalification. However, R_o significantly increases in uranium-rich area (Figures 2, 5A), indicating that uranium may have influences on R_o .

DISCUSSION

As mentioned above, CD is a common sedimentary component in uranium-bearing strata. It has been documented that in basins, CD is a key factor in concentration, complexation, and reduction in uranium

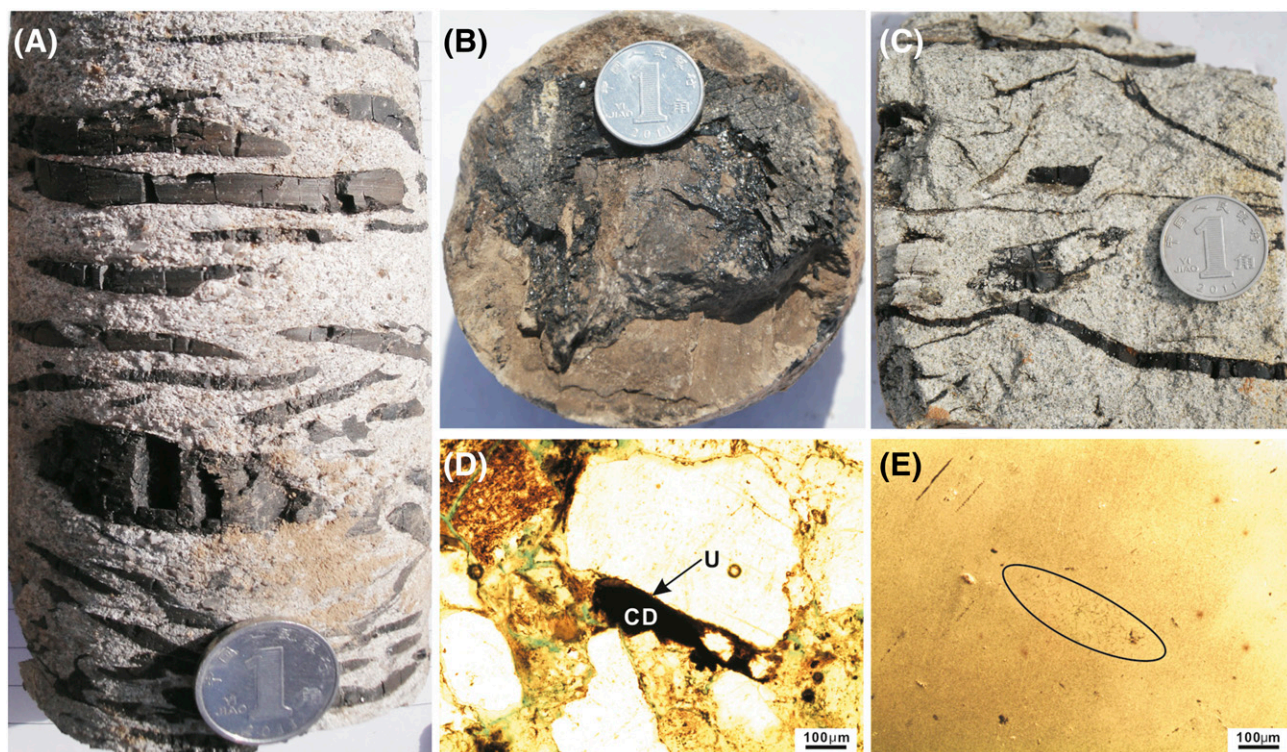


Figure 4. Petrologic photographs of carbonaceous debris (CD) in uranium (U)-bearing strata. (A) The CD within gray conglomeratic coarse sandstone in the reduction zone in the forms of lumps and bands, T63-0, 634.8 m (2083.0 ft). (B) The CD within gray fine sandstone in the reduction zone distributed along levels, ZKD160-39, 592 m (1942.3 ft). (C) The CD within gray fine sandstone rich in U in the forms of lumps and bands, ZKD111-16, 644.1 m (2113.2 ft). (D) The CD and U mineral in gray coarse U-rich sandstone, showing a spatial relationship between U and organic matter, T111-15, 706.5 m (2317.9 ft). (E) The same view as (D), showing the distributions of the fission track by the means of microradiography.

mineralization (Spirakis, 1996; Giordano, 2000; Douglas et al., 2011; Dai et al., 2015). However, little attention is paid to the influence of uranium enrichment on CD, especially thermal maturation, for example, R_o .

Influences of Uranium Enrichment on Vitrinite Reflectance

As mentioned in the Results section, the correlation coefficient between R_o and standardized burial is small ($R^2 = 0.475$). Whether CD is rich in uranium, burial is mostly in a range of 630–670 m (2070–2200 ft) (Figure 5A). Therefore, the samples are thought to be almost collected from the same burial depth, and the influence of burial on R_o variation can be neglected.

Compared to the averages of R_o of coals and CD from different zones, R_o is the highest in the uranium ore body (i.e., gray sandstone rich in uranium) whereas lowest in the reduction zone (i.e., gray

sandstone without uranium mineralization), and the former is 0.062% R_o higher than that of the latter (Figure 6). According to independent samples test, the variances of R_o between the reduction zone and uranium ore body are unequal (significance = $0 < 0.05$) by using Levene's test, and significance (two-tailed) of the t test for equality of means is 0.000 ($< \alpha = 0.05$) (Table 2), which indicates that the differences in R_o values between the reduction zone and uranium ore body are statistically significant. Furthermore, the increase of 0.062% R_o accounts for 14.35% of the average R_o value of the study area (i.e., 0.432% R_o). On the basis of the reconstruction of the burial thermal evolution history of the borehole D112-39 from the studied area (Figure 7), which is calibrated by using the observed R_o value of the sample without uranium mineralization (i.e., D112-39-9), the maximum burial temperature that CD without uranium mineralization experienced is 62°C, and theoretically, an increase of 0.062% R_o is correspondingly caused by an increase in the burial of 380 m (1247 ft)

Table 1. Vitrinite Reflectance Analysis of Coals and Carbonaceous Debris within Sandstones in the Daying Uranium Deposit, North Ordos Basin

Drill Holes	Sample Number	Depth, m (ft)	Lithology	Mineralization Information	A, Bq/kg	Vitrinite Reflectance, % R_o					H, m (ft)
						VR max', % R_o	VR min', % R_o	$\overline{VR}$ max, % R_o	n	SD	
D111-16	D111-16-1	618 (2027)	CD in gray medium sandstone	AUM	18.6	0.464	0.345	0.428	71	0.03	0 (0)
	D111-16-2	644.1 (2113.2)	CD in gray fine sandstone	BUM	106.8	0.582	0.367	0.435	50	0.036	0 (0)
	D111-16-3	647.8 (2125.3)	CD in gray fine sandstone	AUM	33.6	0.434	0.347	0.423	50	0.028	0 (0)
	D111-16-4	668.5 (2193.2)	CD in gray medium sandstone	NUM	2.9	0.464	0.351	0.407	52	0.029	23.71 (77.79)
	D111-16-5	677 (2221)	Coal	NUM	1.5	0.516	0.341	0.409	48	0.029	29.03 (95.24)
	D111-16-6	715 (2346)	CD in gray medium-coarse sandstone	NUM	1.3	0.465	0.344	0.409	57	0.025	3.49 (11.45)
	D111-16-7	727.5 (2386.8)	CD in gray medium-coarse sandstone	NUM	6.6	0.453	0.341	0.415	17	0.038	6.04 (19.82)
	D111-16-8	765.4 (2511.1)	CD in gray fine sandstone	NUM	1.9	0.469	0.400	0.449	6	0.029	44.38 (145.60)
T143-16	T143-16-1	631 (2070)	CD in gray coarse sandstone	NUM	5.1	0.405	0.305	0.372	45	0.029	25.96 (85.17)
	T143-16-2	636 (2087)	CD in gray medium sandstone	NUM	5.7	0.396	0.335	0.386	31	0.023	20.8 (68.24)
	T143-16-3	666.7 (2187.3)	CD in gray-green medium sandstone	NUM	8.3	0.429	0.303	0.403	58	0.03	5.51 (18.08)
	T143-16-4	703.8 (2309.0)	CD in gray coarse sandstone	BUM	72	0.492	0.375	0.447	48	0.029	0 (0)
	T143-16-5	744.8 (2443.5)	CD in gray medium-coarse sandstone	NUM	3	0.494	0.345	0.417	11	0.0414	41.12 (134.91)
D112-39	D112-39-1	638 (2093)	CD in gray coarse sandstone	IUM	216	0.583	0.453	0.51	41	0.029	0 (0)
	D112-39-2	638.55 (2095.00)	CD in gray medium-fine sandstone	IUM	210	0.521	0.466	0.51	25	0.025	0 (0)
	D112-39-3	641.97 (2106.18)	CD in gray medium-fine sandstone	IUM	192	0.522	0.474	0.512	35	0.025	0 (0)
	D112-39-4	642.02 (2106.34)	CD in gray medium sandstone	IUM	171.9	0.538	0.387	0.511	63	0.04	0 (0)
	D112-39-5	642.32 (2107.32)	CD in gray medium sandstone	IUM	170.4	0.532	0.388	0.514	36	0.025	0 (0)
	D112-39-6	642.91 (2109.26)	CD in gray medium sandstone	IUM	131.5	0.503	0.431	0.501	25	0.027	0 (0)
	D112-39-7	653.52 (2144.09)	CD in gray medium-coarse sandstone	NUM	15.8	0.524	0.3	0.392	93	0.048	6.65 (21.82)
	D112-39-8	655.12 (2149.32)	CD in gray fine sandstone	NUM	9.6	0.469	0.315	0.403	71	0.028	8.41 (27.59)
	D112-39-9	661.52 (2170.32)	CD in gray medium-coarse sandstone	NUM	8.3	0.488	0.332	0.413	98	0.048	14.78 (48.49)
	D112-39-10	662.32 (2172.94)	CD in gray medium-coarse sandstone	NUM	6.7	0.502	0.293	0.408	76	0.041	15.56 (51.05)
	D112-39-11	696.68 (2285.67)	CD in gray medium-coarse sandstone	NUM	4.5	0.448	0.347	0.421	11	0.032	2.19 (7.18)

(continued)

Table 1. Continued

Drill Holes	Sample Number	Depth, m (ft)	Lithology	Mineralization Information	A, Bq/kg	Vitrinite Reflectance, % R_o					
						VR max', % R_o	VR min', % R_o	$\overline{VR}$ max, % R_o	n	SD	H, m (ft)
D80-39	D80-39-1	563 (1847)	CD in gray green medium-fine sandstone	NUM	2.3	0.442	0.375	0.436	8	0.024	0.959 (3.146)
	D80-39-2	606.4 (1989.6)	CD in gray medium sandstone	IUM	296.9	0.508	0.3	0.476	64	0.034	0 (0)
	D80-39-3	607 (1991)	CD in gray medium sandstone	IUM	228.2	0.511	0.305	0.48	72	0.031	0 (0)
	D80-39-4	630 (2067)	CD in gray medium sandstone	BUM	61.2	0.462	0.354	0.428	52	0.029	0 (0)
	D80-39-5	630.8 (2069.5)	CD in gray medium sandstone	NUM	13.2	0.423	0.35	0.415	51	0.034	0.762 (2.500)
	D80-39-6	633 (2077)	CD in gray medium sandstone	BUM	39.6	0.474	0.38	0.463	17	0.034	0 (0)
	D80-39-7	653 (2142)	CD in gray medium sandstone	NUM	4	0.453	0.361	0.417	10	0.028	20.43 (67.03)
	D80-39-8	685.5 (2249.0)	CD in gray medium sandstone	BUM	86.4	0.478	0.385	0.445	39	0.028	0 (0)
D192-55	D192-55-1	621.8 (2040.0)	CD in gray coarse sandstone	AUM	24	0.445	0.367	0.432	26	0.025	0 (0)
	D192-55-2	629.15 (2064.12)	CD in gray medium sandstone	NUM	6.2	0.444	0.385	0.405	16	0.025	3.54 (11.61)
	D192-55-3	636.5 (2088.2)	CD in gray-green medium-coarse sandstone	NUM	8.4	0.475	0.323	0.409	80	0.035	4.385 (14.386)
	D192-55-4	639.5 (2098.1)	CD in gray fine sandstone	NUM	7.2	0.441	0.324	0.413	27	0.028	1.071 (3.514)
	D192-55-5	652.8 (2141.7)	Coal	NUM	2.5	0.437	0.317	0.409	13	0.024	5.679 (18.632)
D160-39	D160-39-1	592 (1942)	CD in gray fine sandstone	NUM	7.2	0.423	0.339	0.405	55	0.041	20.23 (66.37)
	D160-39-2	611.3 (2005.6)	CD in gray medium-coarse sandstone	NUM	9	0.445	0.369	0.417	30	0.03	0.912 (2.2992)
	D160-39-3	618 (2028)	CD in gray fine sandstone	NUM	2.4	0.461	0.332	0.422	21	0.036	3.031 (9.944)
	D160-39-4	623.8 (2046.6)	CD in gray coarse sandstone	IUM	132	0.532	0.373	0.472	68	0.037	0 (0)
	D160-39-5	641.25 (2013.8)	CD in gray coarse sandstone	NUM	4.7	0.447	0.343	0.418	52	0.028	3.815 (12.516)
T111-7	T111-7-1	618.75 (2030.0)	CD in gray medium-fine sandstone	NUM	6.8	0.414	0.332	0.384	24	0.027	58.932 (193.344)
	T111-7-2	621 (2037)	CD in gray medium-fine sandstone	NUM	9	0.416	0.364	0.409	11	0.025	55.922 (183.469)
	T111-7-3	653.9 (2145.3)	CD in gray medium sandstone	NUM	5.2	0.441	0.313	0.397	56	0.031	23.681 (77.693)
	T111-7-4	662.5 (2173.5)	CD in gray coarse sandstone	NUM	5	0.499	0.348	0.417	27	0.035	14.362 (47.119)

The locations of the sampling points are seen Figure 3.

Abbreviations: A = specific radioactivity; AUM = abnormal uranium mineralization, 18–36 Bq/kg; BUM = boundary uranium mineralization; CD = carbonaceous debris; H = distance from sampling point to the closest sandstone rich in uranium; IUM = boundary uranium mineralization, more than 108 Bq/kg; n = number of measured reflectance data per sample; NUM = no uranium mineralization, less than 18 Bq/kg; SD = standard deviation; R_o = vitrinite reflectance; $\overline{VR}$ max = mean maximum reflectance value; VRmax' = maximum values among measured maximum reflectance values per sample; VRmin' = minimum values among measured maximum reflectance values per sample.

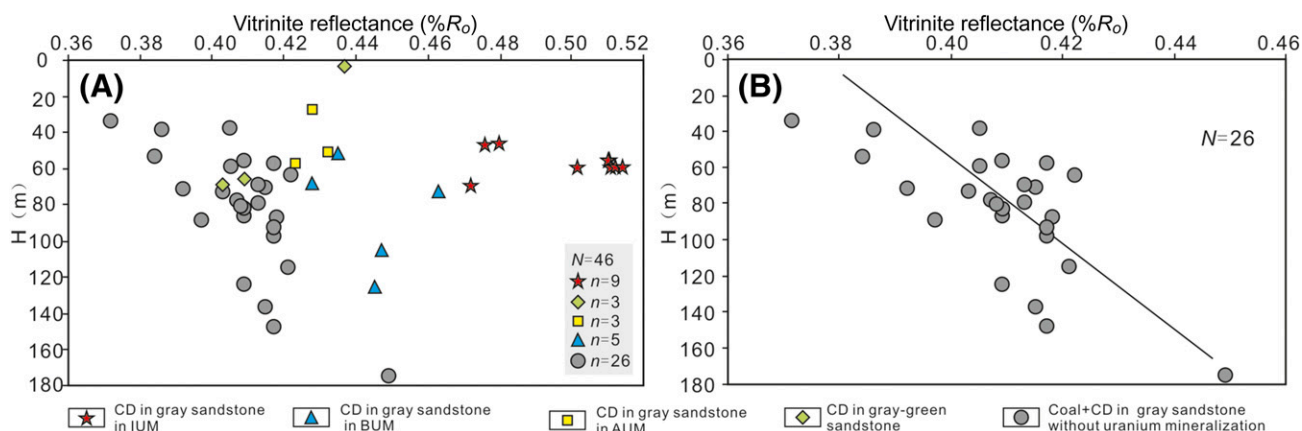


Figure 5. Vitrine reflectance (R_o) versus the distance from sampling point to initial lake flooding surface. (A) All samples. (B) All samples excluding gray sandstone rich in uranium and gray-green sandstone. AUM = abnormal uranium mineralization; BUM = boundary uranium mineralization; CD = carbonaceous debris; H = distance from the sampling point to the closest sandstone rich in uranium; IUM = industrial uranium mineralization; N and n = number of samplings.

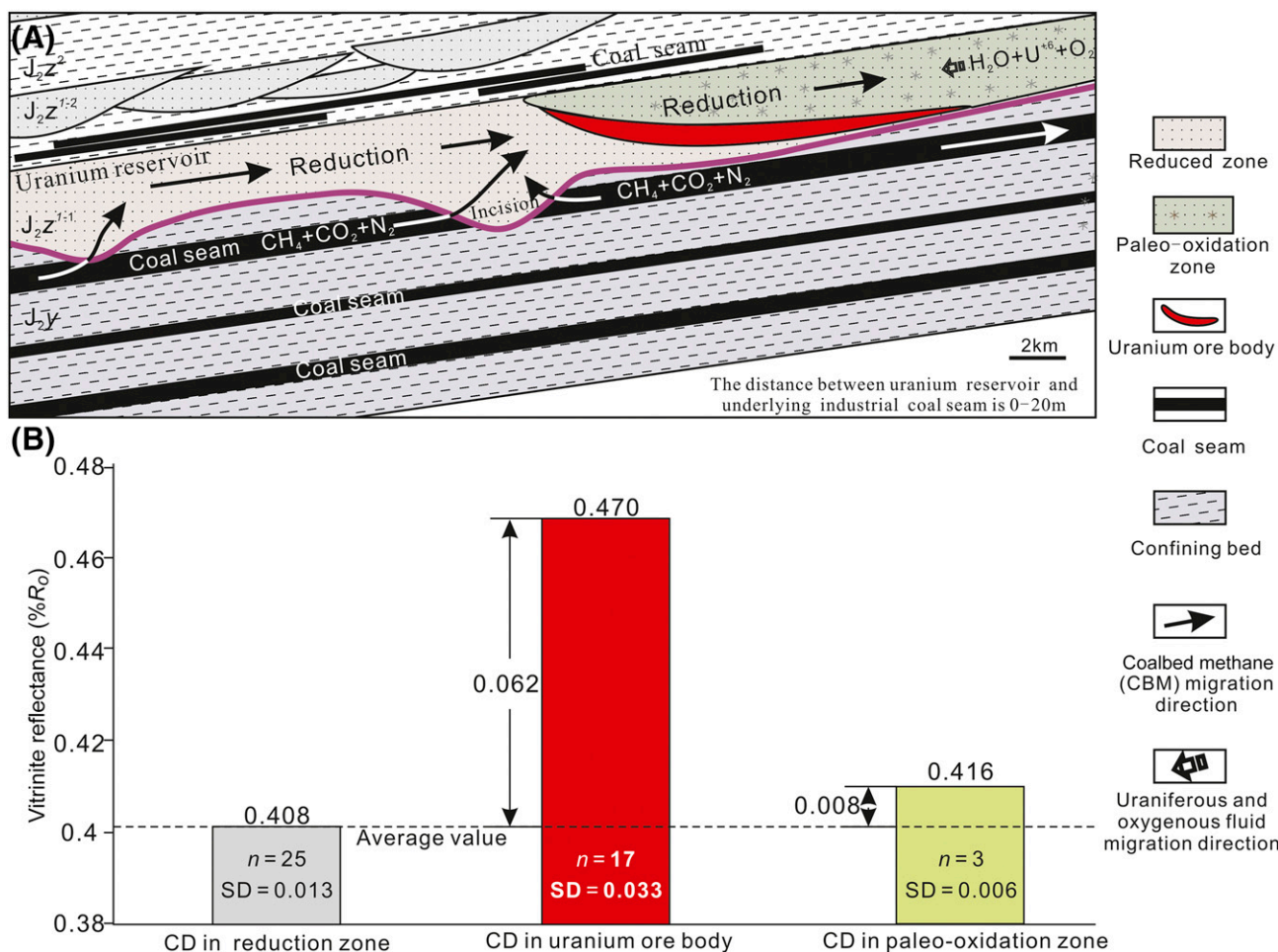


Figure 6. (A) The regional metallogenic mode of sandstone-type uranium deposits, northern Ordos Basin (after Jiao et al., 2016). (B) A histogram showing vitrine reflectance (R_o) in paleointerlayer oxidation zones from the studied area. CD = carbonaceous debris; J_2y = Yan'an Formation; J_2z = Zhiluo Formation; n = number of samplings; SD = standard deviation.

Table 2. The Student's t Test for Averages of Vitrinite Reflectance in Different Zones

Vitrinite Reflectance	Levene's Test for Equality of Variances			t Test for Equality of Means				
	F	Sig.	t	df	Sig. (Two-Tailed)	Mean Difference	Standard Error Difference	95% Confidence Interval of the Difference
Equal variances assumed	29.920	0.000	-8.083	40	0.000	-0.061864	0.007654	Lower -0.077333 Upper -0.046395
Equal variances not assumed			-6.992	19.358	0.000	-0.061864	0.008848	Lower -0.080360 Upper -0.043367

Abbreviations: df = degrees freedom; F = variance freedom; Sig. = significance.

as well as an increase in the temperature of 13°C. In the view of the burial thermal evolution history of the studied area, an increase in the burial of 380 m (1247 ft) accounts for 52.38% of the entire formation thickness from J_{2z} to K (i.e., 725.47 m [2380 ft]), and the increase in the temperature of 13°C accounts for 20.97% of the average maximum burial temperature of J_{2z} (i.e., 62°C), which indicates that the increase of 0.062% R_o is of great significance.

The R_o in the uranium enrichment area is higher, especially in high special radioactivity sections ($A > 220$ Bq/kg), the frequency distributions of R_o values are scattered (Figure 8A, B). For example, in borehole D80-39, R_o of CD in IUM at the lower burial (e.g., D80-39-2 and D80-39-3) is nearly 0.480% R_o , whereas it is just 0.417% R_o for that in NUM at the deeper burial (e.g., D80-39-7). Moreover, the maximum difference of individual reported R_o values of the sample rich in uranium (e.g., D80-39-2) is up to 0.208% R_o with a standard deviation up to 0.034, whereas generally less than 0.100% R_o with the standard deviation of 0.028 for that of free uranium (e.g., D80-39-7). This shows that the dispersion of R_o values of the samples rich in uranium are more remarkable than those uranium free, indicating that the heterogeneities of collotelinite from the samples rich in uranium are more significant. Additionally, R_o changes differently with different mineralization grades, that is, R_o of CD in IUM is the highest whereas lowest for that in AUM (Figure 8C), indicating that the abundance of uranium may result in different R_o . The R_o bears a positive relationship with uranium content and an inverse relationship with distance to the closest sandstone rich in uranium (Figure 9). Hence, comprehensive analyses show that uranium enrichment has an enhanced influence on R_o .

Radiogenic Heat of Uranium Ore Body

Radioactive uranium can spontaneously decay, accompanying with alpha, beta, and gamma rays with a certain amount of energy (Zhang, 1984). During the decay process, uranium may cause radiation-induced damage in quartz and other minerals (i.e., fission track [FT]; Botis et al., 2006, 2008) as well as the thermal abnormalities in surrounding rocks (Vidale, 1977; Zhang, 1984; Papadopoulos et al., 2013).

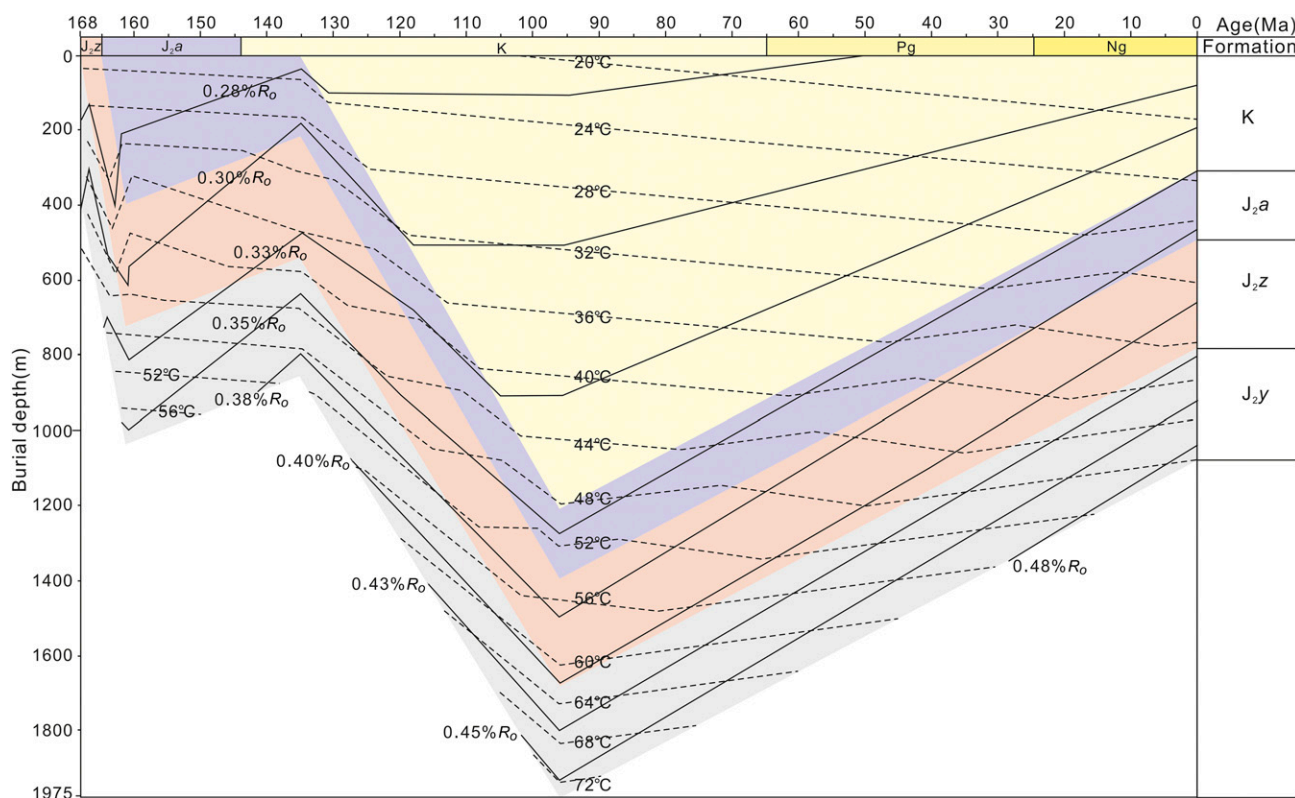


Figure 7. The reconstruction of the burial thermal evolution history of the borehole D112-39 from the studied area. J_2a = Anding Formation; J_2y = Yan'an Formation; J_2z = Zhiluo Formation; K = Cretaceous stratal; R_o = vitrinite reflectance.

Mineralogical Evidence of Uranium Decay

In the study area, FTs are randomly distributed in quartz grains within sandstones rich in uranium (Figure 10). Moreover, the density of FTs bears a negative relationship with distance to the uranium ore body, that is, the intensity of decay increases with proximity to the uranium ore body (Figure 11). The specific data are shown in Table 3.

Radiogenic Heat Production Rate of Uranium Ore Body

Radiogenic heat production by uranium, thorium, and ^{40}K is an important factor in geothermy (Vilá et al., 2010; Liu et al., 2015). The radiogenic heat production rate (Q_A , W/m^3) is the radiogenic heat that a given rock containing radioactive elements per unit volume (or unit weight) produces per unit time during the decay, and can be estimated as

$$Q_A (\text{W/m}^3) = 0.01 \rho (9.52C_U + 2.56C_{\text{Th}} + 3.48C_K) \times 10^{-6}$$

where ρ is the density of the rock in grams per cubic centimeter, C_U and C_{Th} are concentrations of uranium and thorium in parts per million, respectively, and C_K is the content of potassium in percentage (Zhang et al., 2010). As there is no radioactive thorium or potassium in the studied rocks (Y. Jiao, L. Wu, H. Rong, Y. Liu, H. Wang, H. Xie, Q. Zhu, J. Li, M. Liu, L. Wang, Z. Wwang, L. Wan, D. Wan, F. Zhang, Z. Yan, and Q. Xiong, 2012, personal communication), the radiogenic heat production rate is $Q_A (\text{W/m}^3) = 0.01\rho \times 9.52C_U \times 10^{-6}$.

According to the relevant report of the Daying uranium deposit (Y. Jiao, L. Wu, H. Rong, Y. Liu, H. Wang, H. Xie, Q. Zhu, J. Li, M. Liu, L. Wang, Z. Wwang, L. Wan, D. Wan, F. Zhang, Z. Yan, and Q. Xiong, 2012, personal communication), the Daying uranium deposit covers a total area of approximately 9.01 km^2 ($\sim 3.48 \text{ mi}^2$), with a thickness of industrial ore bodies of 5.26 m (17.26 ft). An average grade of uranium ore is 0.0343% with uranium content of 0.738 kg/m^3 and a density of uranium ore body of 2.10 g/cm^3 , and a density is up

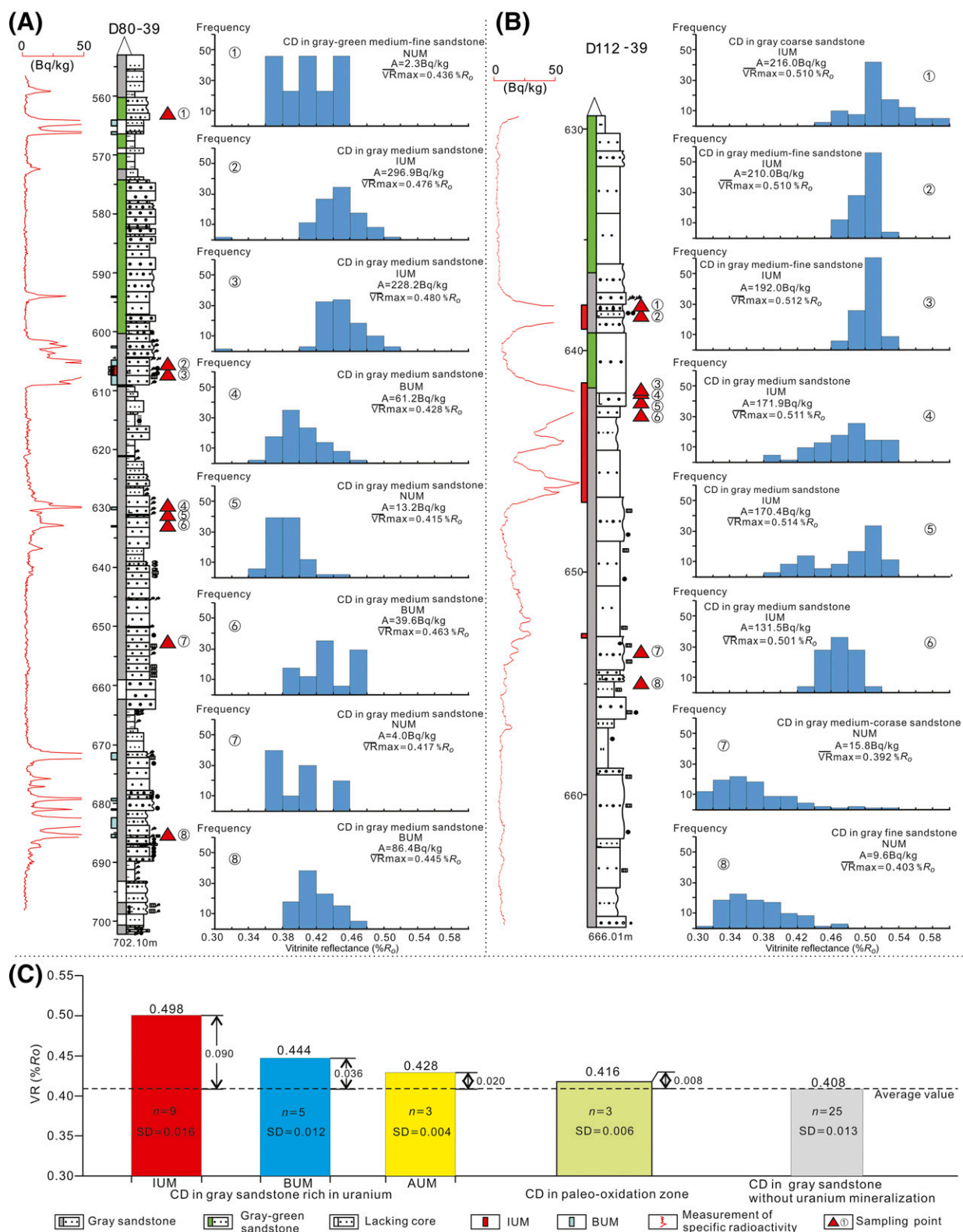


Figure 8. (A, B) Vertical profiles of frequency histograms of vitrinite reflectance (R_0) values in typical boreholes D80-39 and D112-39, respectively. (C) Histograms showing R_0 in different mineralization grades. A = specific radioactivity; AUM = abnormal uranium mineralization; BUM = boundary uranium mineralization; CD = carbonaceous debris; IUM = industrial uranium mineralization; n = number of samplings; NUM = no uranium mineralization; SD = standard deviation; $\overline{VR}_{max}$ = mean maximum reflectance.

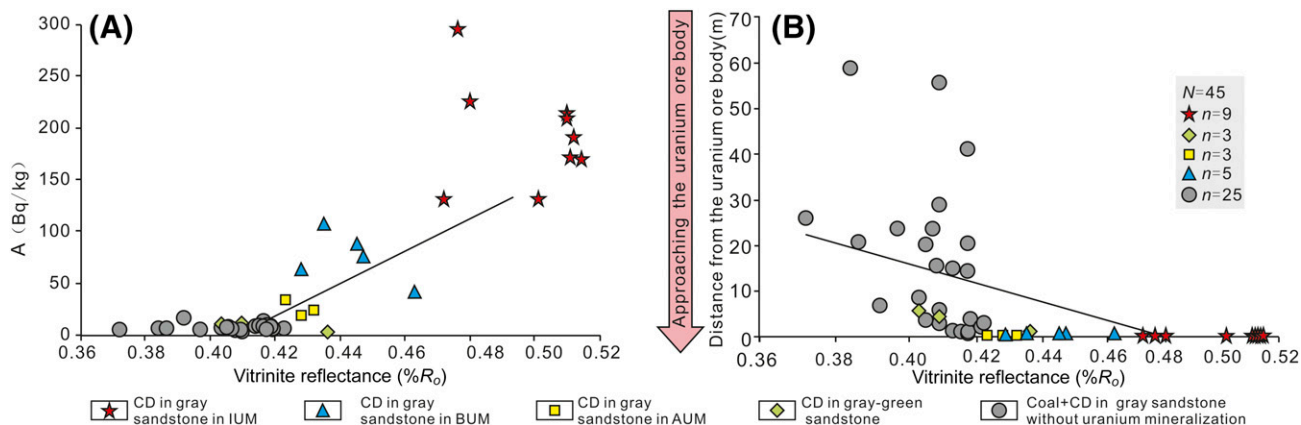


Figure 9. (A) Relationships between vitrinite reflectance (R_o) and specific radioactivity (A). (B) Relationships between R_o and distance from sampling point to the closest sandstone rich in uranium (data from Table 1). AUM = abnormal uranium mineralization; BUM = boundary uranium mineralization; CD = carbonaceous debris; IUM = industrial uranium mineralization; N and n = number of samplings.

to 2.40 g/cm^3 for the individual sandstones, which are extremely uranium rich. As sandstone bodies extremely rich in uranium account for less than 1% of all uranium-rich sandstone bodies (Y. Jiao, L. Wu, H. Rong, Y. Liu, H. Wang, H. Xie, Q. Zhu, J. Li, M. Liu, L. Wang, Z. Wwang, L. Wan, D. Wan, F. Zhang, Z. Yan, and Q. Xiong, 2012, personal communication),

the density of the entire uranium ore body is set as 2.10 g/cm^3 in this study. According to the relationship between uranium content and radiogenic heat production rate (i.e., $Q_A [\text{W/m}^3] = 0.01\rho \times 9.52C_U \times 10^{-6}$), the radiogenic heat production rate of the entire uranium ore body in the study area is $6.857 \times 10^{-5} \text{ W/m}^3$.

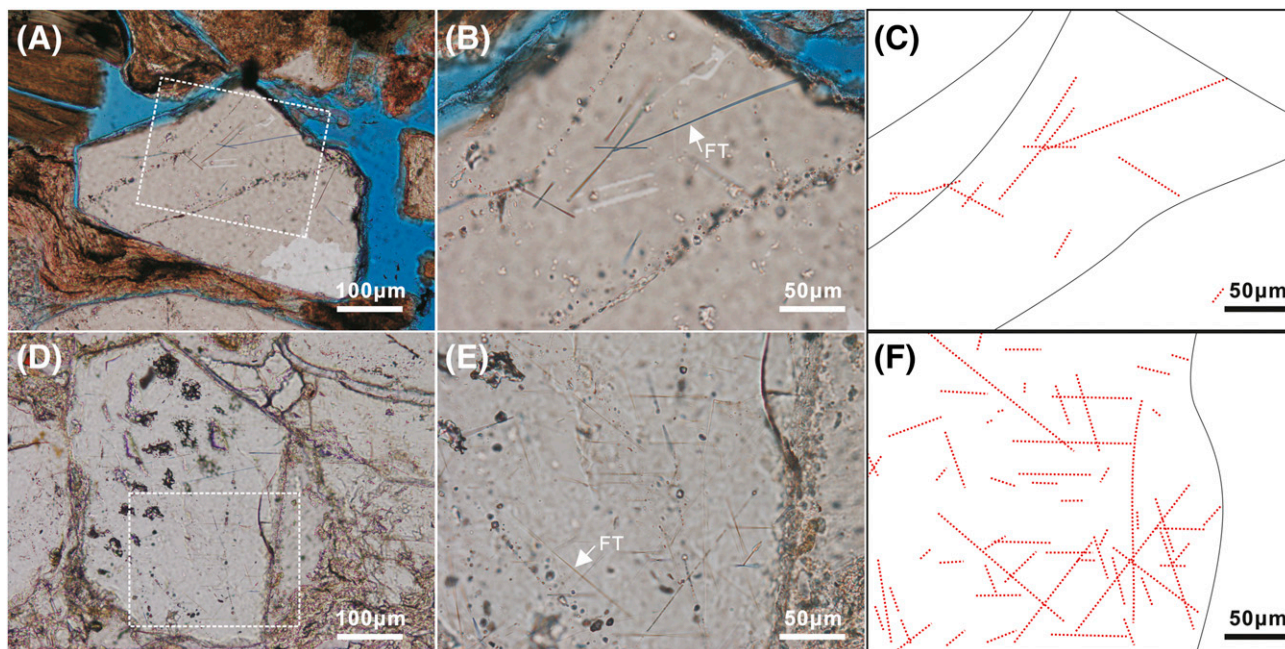


Figure 10. Distributions of fission track (FT) in quartz grains within sandstone rich in uranium from Zhiluo Formation in the Ordos Basin, China. (A–C) Gray coarse sandstone, T63-01, 592.3 m (1943.2 ft); (B) is the same view as the white dotted box in (A), and (C) is a sketch of (B) showing the distributions of FT. (D–F) Gray coarse sandstone, T191-16, 705.3 m (2314.0 ft); (E) is the same view as the white dotted box in (D), and (F) is a sketch of (E) showing the distributions of FT.

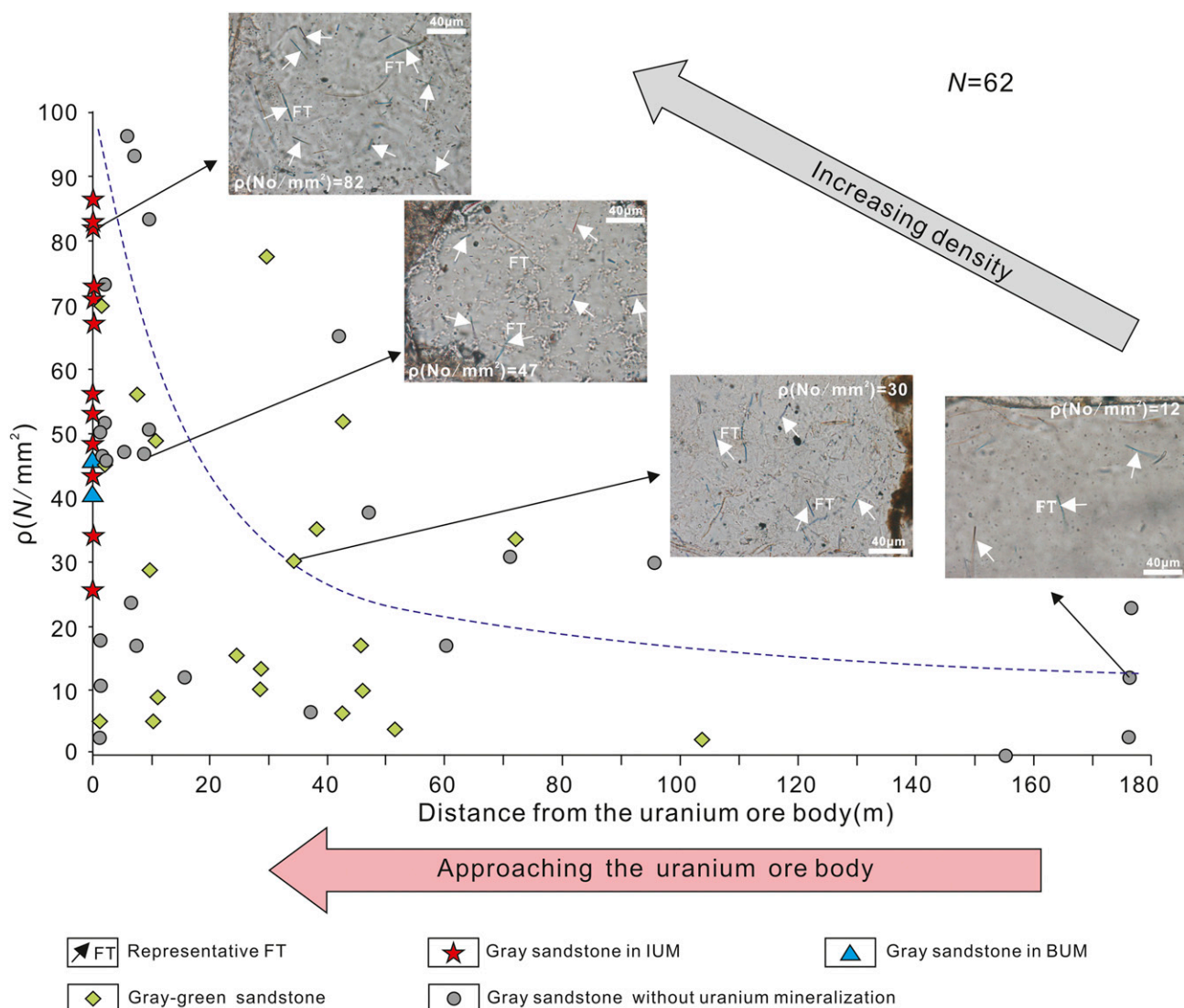


Figure 11. Schematic diagram showing the relationship between the density of fission track (FT) and distances from sampling points to the closest uranium ore body. ρ = density of FT; BUM = boundary uranium mineralization; IUM = industrial uranium mineralization; N = number of samplings.

Relationship between Vitrinite Reflectance and Radiogenic Heat of Uranium Decay

As mentioned in the Introduction section, radiogenic heat from uranium decay is the reason for R_o enhancement. For an ancient uranium ore body, radiogenic heat from uranium ore body decay can cause thermal abnormalities in surrounding space during the long-term stable decay (Cassou et al., 1975; Shlyakhter, 1983; Fujii et al., 2000; Papadopoulos et al., 2013). The Oklo uranium deposit in the Gabon Republic is an example of a well-known “natural reactor” (Cassou et al., 1975). The total amount of energy produced by the entire ore body, which can

be enough for a city like Budapest for about a year (Shlyakhter, 1983), causes an increase in temperature around its surroundings of up to 200°C (Vidale, 1977), resulting in an apparent increase in organic matter maturation (Cassou et al., 1975).

The radiogenic heat (Q_U , J) that the entire uranium ore body decay produces during Δt keeps the following function: $Q_U(J) = Q_A \times V \times \Delta t$, where V is the volume of the entire uranium ore body in cubic meters, and Δt is the time of the uranium ore body decay in seconds. Given the decay half-life of uranium (see Table 4), Δt is reasonably set as 50 k.y. (i.e., 1.58×10^{12} s) to maintain that the concentration of uranium is constant during Δt . Radiogenic heat

Table 3. The Density of Fission Track in Quartz Grains

Drill Holes	Depth, m (ft)	Lithology	Mineralization Information	ρ , n/mm ²	d, m (ft)
ZKB32-31	378.35 (141.29)	Gray-green medium sandstone	NUM	73	56.15 (184.22)
	387.4 (1271.0)	Gray-green fine sandstone	NUM	38	47.1 (154.5)
	441.55 (1448.64)	Gray medium sandstone	IUM	73	0 (0)
	451.25 (1448.64)	Gray coarse sandstone	NUM	83	9.55 (31.33)
	469.95 (1541.81)	Gray-green fine sandstone	NUM	10	28.25 (92.68)
	483.5 (1586.27)	Gray calcareous coarse sandstone	NUM	65	41.8 (137.1)
	501.6 (1645.65)	Gray coarse sandstone	NUM	17	59.9 (196.5)
Wtn-1	230.35 (755.73)	Gray calcareous coarse sandstone	NUM	23	176.765 (579.931)
	230.6 (756.6)	Gray calcareous medium sandstone	NUM	12	176.515 (579.110)
	230.95 (757.70)	Gray calcareous medium sandstone	NUM	3	176.165 (577.962)
	251.81 (826.14)	Gray coarse sandstone	NUM	0	155.305 (509.525)
	302.9 (993.8)	Gray-green fine sandstone	NUM	3	104.215 (341.909)
	311.2 (1021.0)	Gray siltstone	NUM	30	95.915 (314.678)
	335 (1099)	Gray-green medium sandstone	NUM	33	72.115 (236.595)
	364.5 (1195.9)	Gray-green medium sandstone	NUM	52	42.615 (139.811)
	407.15 (1335.78)	Gray-green medium sandstone	IUM	86	0 (0)
	408.65 (1340.70)	Gray coarse sandstone	IUM	82	0 (0)
	409.85 (1344.64)	Gray coarse sandstone	IUM	83	0 (0)
	391.05 (1282.96)	Green fine sandstone	NUM	5	0.95 (3.12)
	391.15 (1283.28)	Gray fine sandstone	NUM	3	0.85 (2.79)
ZKN3-7	393.3 (1290.3)	Green coarse sandstone	NUM	70	0.8 (2.6)
	394.45 (1294.11)	Gray coarse sandstone	IUM	26	0 (0)
	394.9 (1295.6)	Gray coarse sandstone	IUM	41	0 (0)
	395.95 (1299.03)	Gray coarse sandstone	IUM	34	0 (0)
	396.65 (1301.33)	Gray medium-coarse sandstone	IUM	71	0 (0)
	399.9 (1311.99)	Gray coarse sandstone	NUM	11	1.53 (5.02)
	414.12 (1358.64)	Gray medium-coarse sandstone	NUM	12	15.75 (51.67)
	436.8 (1433.1)	Gray-green coarse sandstone	NUM	35	38.43 (126.08)
	444.3 (1457.7)	Gray-green coarse sandstone	NUM	17	45.93 (150.69)
	717.95 (2355.45)	Green fine sandstone	NUM	4	51.425 (168.715)
ZKS0-31	735.09 (2411.68)	Gray-green medium sandstone	NUM	30	34.285 (112.482)
	758.3 (2487.8)	Green fine sandstone	NUM	9	11.075 (36.335)
	759.21 (2490.82)	Red-green fine sandstone	NUM	5	10.165 (33.349)
	762.02 (2500.04)	Gray-green medium-fine sandstone	NUM	56	7.355 (24.130)
	781.25 (2563.13)	Green fine sandstone	NUM	49	10.625 (34.859)
ZKT63-0	584.9 (2563.13)	Gray medium sandstone	NUM	24	6.5 (21.3)
	591.4 (1940.3)	Gray coarse sandstone	BUM	46	0 (0)
	592.2 (1942.9)	Gray coarse sandstone	BUM	42	0 (0)
T413-8	623.6 (1942.9)	Gray-green fine sandstone	NUM	15	24.4 (80.1)
	649.71 (2131.57)	Gray-green fine sandstone	NUM	45	1.71 (5.61)
	678.3 (2225.4)	Gray calcareous medium sandstone	NUM	51	9.7 (31.8)
	704.2 (2310.3)	Gray fine sandstone	NUM	46	2.2 (7.2)
	734.34 (2409.22)	Gray-green medium sandstone	NUM	13	28.34 (92.98)

(continued)

Table 3. Continued

Drill Holes	Depth, m (ft)	Lithology	Mineralization Information	ρ , n/mm ²	d, m (ft)
T191-16	595.33 (1953.16)	Gray medium sandstone	NUM	30	70.67 (231.85)
	660.31 (2166.35)	Gray fine sandstone	NUM	17	7.69 (25.23)
	699.5 (2294.92)	Gray-green medium sandstone	NUM	78	29.5 (96.8)
	701.8 (2302.47)	Gray coarse sandstone	NUM	51	1.2 (3.9)
	705.3 (2314.0)	Gray coarse sandstone	NUM	74	2.3 (7.5)
	713.67 (2341.41)	Gray medium sandstone	NUM	96	5.67 (18.60)
D95-16	641.2 (103.7)	Gray coarse sandstone	NUM	7	36.8 (120.7)
	690 (2264)	Gray medium sandstone	NUM	93	7 (23)
	706.2 (2316.9)	Gray coarse sandstone	NUM	47	5.2 (17.1)
D32-63	568.4 (1864.81)	Green medium-fine sandstone	NUM	29	9.6 (31.5)
	584 (1916)	Gray fine sandstone	IUM	53	0 (0)
	586.53 (1924.29)	Gray fine sandstone	IUM	48	0 (0)
D192-31	619.23 (2031.57)	Gray medium sandstone	IUM	67	0 (0)
	623 (2044)	Gray fine sandstone	IUM	41	0 (0)
	625 (2050)	Gray medium sandstone	IUM	56	0 (0)
D224-23	575.44 (1887.90)	Red calcareous fine sandstone	NUM	10	45.56 (149.47)
	578.64 (1898.40)	Gray-green calcareous medium sandstone	NUM	6	42.36 (138.097)
	612.39 (2009.13)	Gray calcareous fine sandstone	NUM	47	8.61 (28.25)
	640.3 (2100.70)	Gray medium-fine sandstone	NUM	18	0.3 (1.0)

Density of fission track (ρ) is that the number of fission track is divided by the area in the thin section analysis, and given in the number (n) of fission track per square millimeter. Abbreviations: BUM = boundary uranium mineralization; d = distance from sampling point to the closest sandstone rich in uranium; IUM = industrial uranium mineralization; NUM = no uranium mineralization.

(Qu, J) that the entire uranium deposit decay produces within 50 k.y. is 5.124×10^{15} J.

According to the regional formation mode of sandstone-type uranium deposits in the northern Ordos Basin (Jiao et al., 2016; Figure 6A), the roof and floor of uranium ore bodies are mostly confining beds, for example, coal and mudstone, which could be taken as insulating beds because of the very low thermal conductivities (Geothermal Research Team of Institute of Geology, Chinese Academy of Sciences, 1978; Qiu et al., 2004). Therefore, without consideration of the loss of heat convection, heat conduction, and thermal radiation during the decay, radiogenic heat that the

entire uranium ore body decay produces within 50 k.y. is totally absorbed by the entire uranium ore body itself, that is, $Q_U(J) = C \times m \times \Delta T$, where C is the specific heat capacity of sandstone rich in uranium in joules per kilogram Kelvin, m is the mass of the entire uranium ore body in kilograms (i.e., $m = \rho \times V$), and ΔT is the increased temperature of the entire uranium ore body in Kelvin. By using the specific heat capacity of sandstone in the Ordos Basin (i.e., 0.888 to 1.129×10^3 J/(kg.K); Gao, 2015), the specific heat capacity of sandstone rich in uranium is assumed to be the average value of specific heat capacity, that is, 1×10^3 J/(kg.K). Therefore, the temperature theoretically

Table 4. Geologic Constants and the Radiogenic Heat Production Rate of Natural Uranium Isotope

Isotope	Atomic Weight	Type of Radioactive		Heat Production Quantity, Heat Production Rate,		
		Decay	T_{α} , yr	T_f , yr	T_{α}/T_f	cal/g.yr W/m ³
²³⁸ U	238	α , f	4.51×10^9	8.0×10^{15}	1.8×10^6	4.3 0.251
²³⁵ U	235	α , f	7.1×10^8	1.9×10^{17}	2.7×10^8	0.71 1.509
Natural uranium						0.73 0.260

Abbreviations: T_{α} = alpha decay half-life; T_f = spontaneous fission half-life.

increases by 52°C. The increased temperature of uranium ore body bears a negative relationship with the specific heat capacity. Given the range of specific heat capacity of sandstone in the Ordos Basin (i.e., 0.888 to 1.129×10^3 J/[kg.K]; Gao, 2015), the increased temperature ranges theoretically from 46°C to 58°C, that is, the increased temperature is 52°C with a range of 6°C, indicating that the variation of specific heat capacity of rock within reasonable ranges has no obvious influence on the increased temperature.

Coal could achieve the same degree of coalification at high temperature during a relatively short time and low temperature for a long time (Coalfield Research Team of Wuhan Geological Institute, 1982, p. 97; Barker, 1989; Huang, 1996). As mentioned in the Geologic Setting section, the Daying uranium deposit is an ancient sandstone body (i.e., the earliest uranium mineralization is 177–149 Ma, and the latest is 20–8 Ma), and the temperature theoretically increases by 52°C within 50 k.y. without consideration of the loss of heat conduction, heat convection, and thermal radiation, which could influence R_o over the long geologic time. According to the calibrated burial thermal evolution history (Figure 7), the radiogenic heat from the entire uranium ore body theoretically yields a R_o increase of 0.219% R_o . As the heat yielding the R_o increase of 0.219% R_o is calculated under the assumption of an ideally closed system, where the radiogenic heat from the entire uranium ore body is totally absorbed by the entire uranium ore body itself without consideration of the loss of heat conduction, heat convection, and thermal radiation, the R_o increase of 0.219% R_o is reasonably greater than the actual increase of 0.062% R_o . No doubt, the long-term stable radiogenic heat of the ancient uranium ore body could enhance organic matter maturation in uranium-bearing strata to some extent.

CONCLUSIONS

1. The CD in the study area is immaturity to low maturity. The R_o generally increases as the buried depth increases. However, R_o of CD within uranium-rich sandstone is 0.062% R_o higher than that without uranium mineralization. The R_o bears a positive relationship with uranium content and an inverse relationship with distance from sampling point to the closest sandstone rich in uranium.

2. The intensity of the decay increases with proximity to the uranium ore body. The radioactive heat production rate of the uranium ore body is 6.857×10^{-5} W/m³. During the long-term stable decay (i.e., 50 k.y.), the ancient uranium ore body theoretically resulted in an abnormal increase in temperature of 52°C without consideration of the loss of heat conduction, heat convection, and thermal radiation, and R_o theoretically increased by 0.219% R_o (R_o actually increased by 0.062% R_o).

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Abstract

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Enhancement of organic matter maturation because of radiogenic heat from uranium: A case study from the Ordos Basin in China

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ABSTRACT

Carbonaceous debris (CD) within uranium-bearing strata has been studied in the Daying uranium deposit of the northern Ordos Basin, northern China. The influence of radiogenic heat from uranium on organic matter maturation was investigated through a series of tests including measurements of vitrinite reflectance (R_o), fission-track (FT) analysis in quartz grains, and the calculation of the radiogenic heat production rate of the samples. The results show that R_o in uranium-bearing strata generally increases as the burial depth increases, indicating that CD experienced normal burial coalification. However, R_o values of the samples rich in uranium are 0.062% R_o higher than those without uranium mineralization. Vitrinite reflectance bears a positive relationship with uranium content, and an inverse relationship with distance to the closest sandstone rich in uranium, indicating that uranium enrichment enhances organic matter maturation. The production of uranium decay makes FT observable in quartz grains, and the intensity of decay increases with proximity to the uranium ore body. The calculated radioactive heat production rate from the uranium ore body is $6.857 \times 10^{-5} \text{ W/m}^3$. During the long-term stable decay, as the uranium ore body theoretically results in an abnormal increase in temperature of 52°C without consideration of the loss of heat conduction, heat convection, and thermal radiation, this would yield a theoretical R_o increase of 0.209% R_o , reasonably greater than the observed. Therefore, the long-term stable radiogenic heat produced by uranium ore body can slightly enhance organic matter maturation, which is instructive in uranium prospecting.

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