



Uranium mineralization formed through multi-stage superposition: Case of the Qianjiadian deposit in Songliao Basin, China

Zhenyan Chen ^{a,1}, Qingchun Li ^{a,*}, Mingyi Liu ^{b,c}, Bairong Song ^a, Minqiang Cao ^a, Miao Wang ^b

^a Liaohe Petroleum Exploration Bureau, Panjin, Liaoning, 124010, China

^b Department of Geology, Northwest University, Xi'an, 710069, China

^c Xian Center of Geological Survey, CGS, Xian, 710054, China

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ABSTRACT

The Qianjiadian uranium deposit is located in the Kailu Depression of Songliao Basin. It is a large-scale in-situ leachable uranium deposit of sandstone type and provides a typical case of the uranium deposits in the Songliao Basin. Here we analyze the impact of oil and gas on reduction alteration, and factors including low grade mineralization. The Qianjiadian uranium deposit is characterized as a typical roll-type deposit with interlayers in oxidized zone. The occurrence of reductive low-permeability sandstone in uranium reservoirs controls the morphology of the uranium-rich orebody. We propose a metallogenic model involving multi-stage superposition characterized by "primary sediment enrichment - interlayer oxidation - superimposed transformation". Our model would be helpful for formulating guidelines in the exploration for sandstone-type uranium deposits in the Songliao Basin.

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1. Introduction

The Qianjiadian uranium deposit is located in the Qianjiadian Depression, a secondary structural unit in the Kailu Depression of Songliao Basin. Since the late 1990s, CNPC Liaohe Oilfield Company has discovered the large-super large Qianjiadian uranium deposit by implementing comprehensive prospecting strategy, providing an important model for searching in-situ leachable uranium deposits of sandstone type in the Songliao Basin. Many scholars have focused on researches of its structural evolution (Tian et al., 2008; Lei et al., 2018), sedimentary diagenetic characteristics of reservoir (Yin et al., 2000; Xu et al., 2017; Wang et al., 2014; Chen and Zhang,

2005; Chen et al., 2007; Li et al., 2018; Jia et al., 2018), hydrothermal process (F. Yan, 2018; Nie et al., 2017), metallogenic conditions (Tian, 2005; Xia et al., 2003), and the occurrence state of uranium (Zhang et al., 2004, 2005). The deposit has been generally considered as an interstratified oxidized-zone uranium deposit dominated by sandstone (Chen et al., 2008; Pang et al., 2010; Rong et al., 2016). However, its metallogenic model and orebody morphology are still under controversy. Some scholars deemed that uranium mineralization is dominated by the superimposition of water seepage and mineralization in oil field and subject to a three-stage hydrothermal transformation (Luo et al., 2007; Nie et al., 2017), whereas some other scholars thought that it should be a "four-stage" uranium mineralization model of sedimentation during later transformation (X. Yan, 2018). Nevertheless, the uranium orebodies are mainly tabular- or lens-shaped, and to date no uranium orebody of roll type has been reported. of the mechanism of formation of roll type ore body with interlayer oxidation zone feature is disputed. Based on exploration data, investigate metallogenic process and redefine the uranium metallogenic model and orebody morphology through a systematic analysis of macro, micro dimensions and geochemical characteristics. Our results have important bearing for blind exploration of deep uranium deposits in the Songliao Basin.

* Corresponding author.

E-mail addresses: liqch@petrochina.com.cn (Q. Li), 201921110@stumail.nwu.edu.cn (M. Wang).

¹ The first Author: Zhenyan Chen (1963.06–), male, mainly engaged in the research and exploration of multi-energy minerals such as petroleum, natural gas, and sandstone-type uranium deposits in sedimentary basins.



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2. Geological setting

The Qianjiadian Sag is located in the northeast Kailu Depression along the southwestern domain of the Songliao Basin, and distributed in an NNE belt (Fig. 1a). Its tectonic evolution is basically the same as that of the Songliao Basin. The Songliao Basin together with its internal Qianjiadian Sag has experienced a four-stage evolution: the Early Cretaceous fault depression, the uplift and denudation at the end of Early Cretaceous, the Late Cretaceous depression and tectonic inversion, and the uplift-exhumation at the end (Yin et al., 2000). Among them, the intense uplift at the end of the Late Cretaceous caused the uplift-exhumation in the south and southeast of the basin, which created conditions for the large-scale infiltration of groundwater. The tectonic inversion at the later stage made three denudation features distributed in the northeast of Qianjiadian Sag in the shape of a "pin", which played an important role in the uranium mineralization at the later period.

The Songliao Basin is underlain by Paleozoic metamorphic basement at a depth generally ranging from 1500 m to 3500 m. The basin covers are mainly the rift-related lacustrine deposits of the Lower Cretaceous Yixian, Jiufotang, Shahai and Fuxin Formations, as well as fluvial lacustrine deposits of the Upper Cretaceous Quantou, Qingshankou, Yaojia, Nenjiang, Sifangtai and Mingshui Formations, the tops of which are the loose deposits of the Neogene Da'an and Taikang Formations. The Quaternary and Mesozoic volcanic rocks, granite of the Carboniferous - Permian and Jurassic - Early Cretaceous as well as Paleozoic metamorphic rocks are mainly exposed in the erosion source area of the depression. The strata exposed in the study area from old to young are mainly the

Cretaceous Qingshankou Formation (K_2qn), Yaojia Formation (K_2y), Nenjiang Formation (K_2n) and the Cenozoic Taikang Formation (N_2t) (Fig. 1b).

Since the uranium mineralization usually occurs in sedimentary sand bodies, which can provide the storage space, the mineralization is not only restricted by interlayer oxidation zone, but also controlled by sedimentary sand bodies. That is, the formation of uranium deposits is controlled by specific sedimentary facies. The ore-bearing horizon of Qianjiadian uranium deposit is mainly the Upper Cretaceous Yaojia Formation (K_2y) stratum, which can be divided into the upper and lower sections with each section at ca.60m–80 m, and its lithologic type is mainly the gray, gray-white, red-brown, yellow-brown fine-grained sandstone with little purplish-red, gray mudstone, carbonaceous mudstone and silty mudstone (Fig. 1c). The occurrence of oil leaching and reduction can also be seen in the sandstone, with brown oil leaching belts and oil spots, gray rim of local purplish red gravel belt, and gray spots or strips in local purplish-red mudstone. The clastic composition mainly includes rock debris and quartz, accounting for 40%–50% and 35%–40% of the total amount respectively, as well as quartz, and feldspar debris constituting about 15%–20%. The detritus is mainly composed of volcanic rocks, mainly rhyolite, a small amount of coarse-grained andesite, and minor granite, granite porphyry and metamorphic rocks. The uranium content of the sandstone is $10\text{--}30 \times 10^{-6}$, and that of some mudstones can be up to more than 100×10^{-6} . Drilling data show that the Yaojia Formation is intruded by diabase dikes (Fig. 2), a product of magmatic activity during the tectonic inversion period at the end of Nenjiang movement.

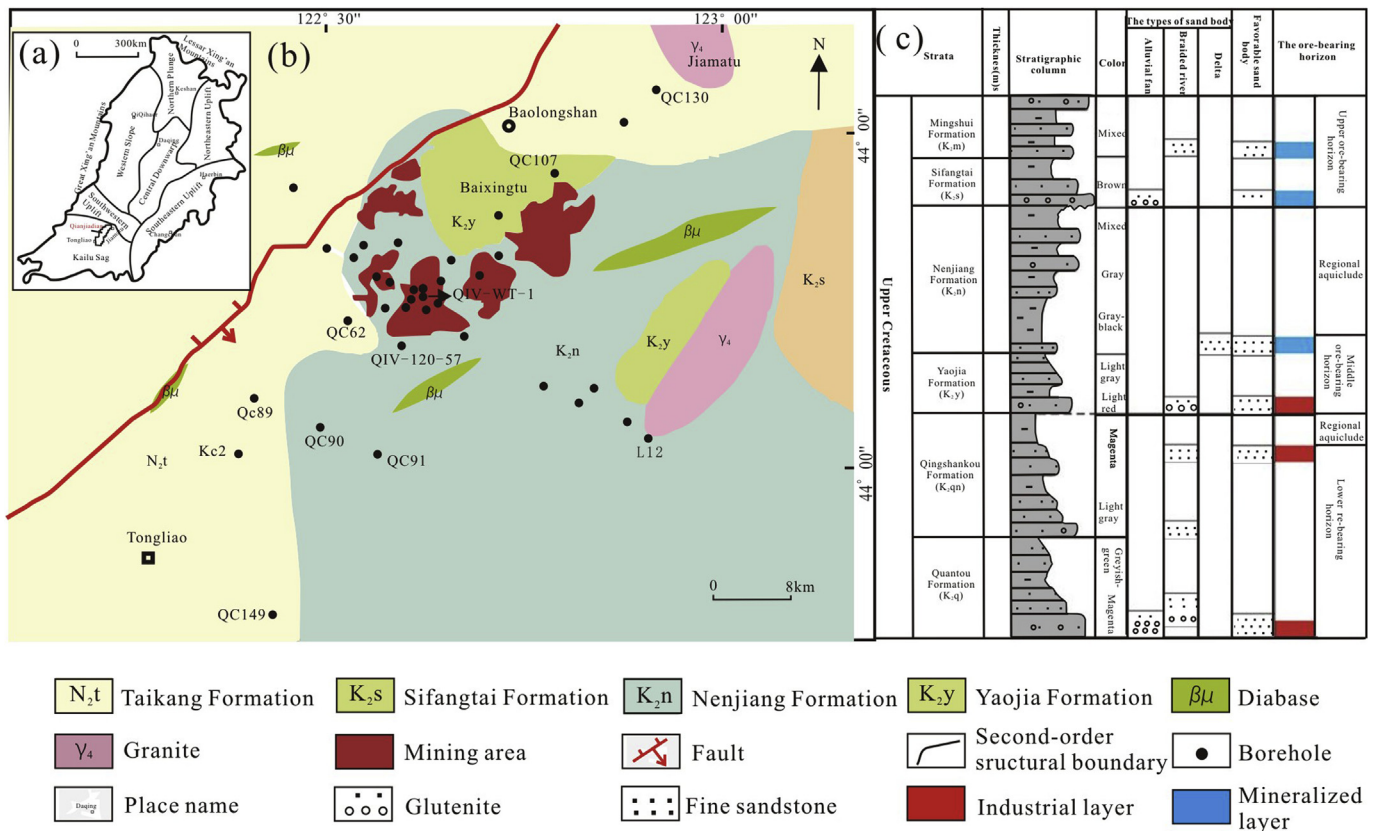


Fig. 1. (a) Tectonic units of the Songliao Basin (modified from Feng et al., 2010). (b) Geological map of Qianjiadian uranium deposit in the Songliao Basin. (c) Composite column chart of the mining stratum in Qianjiadian area (modified from Feng et al., 2010).

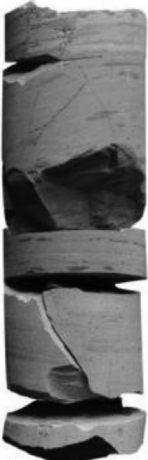
Core picture	Depth and length(m)	Core number and description	Core picture	Depth and length(m)	Core number and description
	382.80 — 0.3	113-1 Gray-white mudstone: relatively dense, massive and lamellar structure, local biological fossils, relatively pure composition. The compositions are mainly clay minerals, in which kaolinite is in the majority, and the secondary compositions are feldspathic clasts. The rock backed mudstone has strong fading and alteration kaolinization is obvious.		383.82 — 0.38	113-4
	— 0.38	113-2 The upper part is gray mudstone and the lower part is dark gray diabase. Diabase: diabase structure, crystal-embedded feldspar and massive structure. The crystallization is fine, which is the regional face of rock mass. Its main components are plagioclase, pyroxene and magnetite.		— 0.31	114-1 Gray-green diabase porphyrite: It is the rock mass central facies. The crystallization is relatively coarse, porphyritic and massive structure. Porphyry components are plagioclase and column pyroxene. The matrix has diabase structure and is mainly composed of plagioclase, pyroxene and magnetite.
	— 0.34 — 383.82	113-3 Dark gray diabase: diabase structure, crystal-embedded feldspar and massive structure. The crystallization is fine, which is the regional face of rock mass. The main components are plagioclase, pyroxene and magnetite. Occasionally fractures are completely filled with calcite.		— 0.33 — 384.89	114-2

Fig. 2. The characteristics of diabase dike and the upper and lower strata (taking ZKY2-1 as an example).

3. Characteristics of interlayer oxidation

The Upper Cretaceous Yaojia Formation (K_2y) of the Qianjiadian uranium deposit is of braided river delta facies. A pan connected sand body of moderate thickness and good percolation conditions is developed between the good roof and floor aquicludes, creating a necessary condition for large-scale interlayer oxidation in this area. Its components are complex, including purplish-red mudstone and mud gravel, light-yellow sandstone and gray sandstone and mudstone (Fig. 1c). The sedimentary bedding is characterized by oxidation with a complex mixture of components including purplish red mudstone and mudstone, red and light yellow sandstone, gray sandstone and mudstone, from which the characteristics of primary and epigenetic oxidation can be identified.

3.1. Identification marks of core and thin section

It can be seen from the core description that carbonaceous clasts are found in both gray and red and yellow sandstone (Fig. 3a), indicating that the sandstone was deposited in a relatively humid environment, and its primary color should be mainly gray. In the red sandstone with bedding, the oxidation color can reflect bedding development, and in the red sandstone with fractures and argillaceous fillings, the degree of oxidation is significantly different on both sides of fractures (Fig. 3b). This differential oxidation on the core scale indicates that the oxidized sandstone in Qianjiadian area is of epigenetic oxidation origin. The phreatic oxidation zone is mainly developed around the structural skylight in the north of the Qianjiadian deposit following the main interlayer oxidation

mineralization stage, and the core, mainly distributed downward along the denudation skylight, is shown as bright yellow (Fig. 3c). It can be seen from the micro section identification that the later iron oxide is relatively developed in the ore-bearing sandstone. The powdery and argillaceous iron oxide polymers fill in homogeneously at the edge or between grains or are adsorbed by argillaceous matrix (Fig. 3d).

3.2. Identification marks and planar features of well-tie section

The distribution characteristics of Qianjiadian uranium deposit are quite different from those of other typical hydric uranium deposits. First of all, the orebody does not show a curl head, and only the front line of interlayer oxidation zone on some sections is near the multi-layer industrial uranium orebody. According to the estimation on resources/reserves of uranium ore of the in-situ leachable sandstone type, the deposit can be compressed into an orebody. Secondly, the characteristics of the wing orebody are not remarkable, and the wings with complete upper and lower wings are rarely found on the section. Therefore, the orebodies of Qianjiadian uranium deposit are mostly plate-like and layered, and are markedly different from the typical "roll-like" orebodies of hydric uranium deposits. Furthermore, researchers have found that there are two main reasons for the particularity of uranium ore-body in the Qianjiadian uranium deposit, that is, the heterogeneity of ore-bearing aquifer and the low grade of uranium orebody. On one hand, the Yaojia Formation of the main ore-bearing layer is of braided river facies, but the mudstone interbeds are relatively developed, and more non-permeable layers block the migration

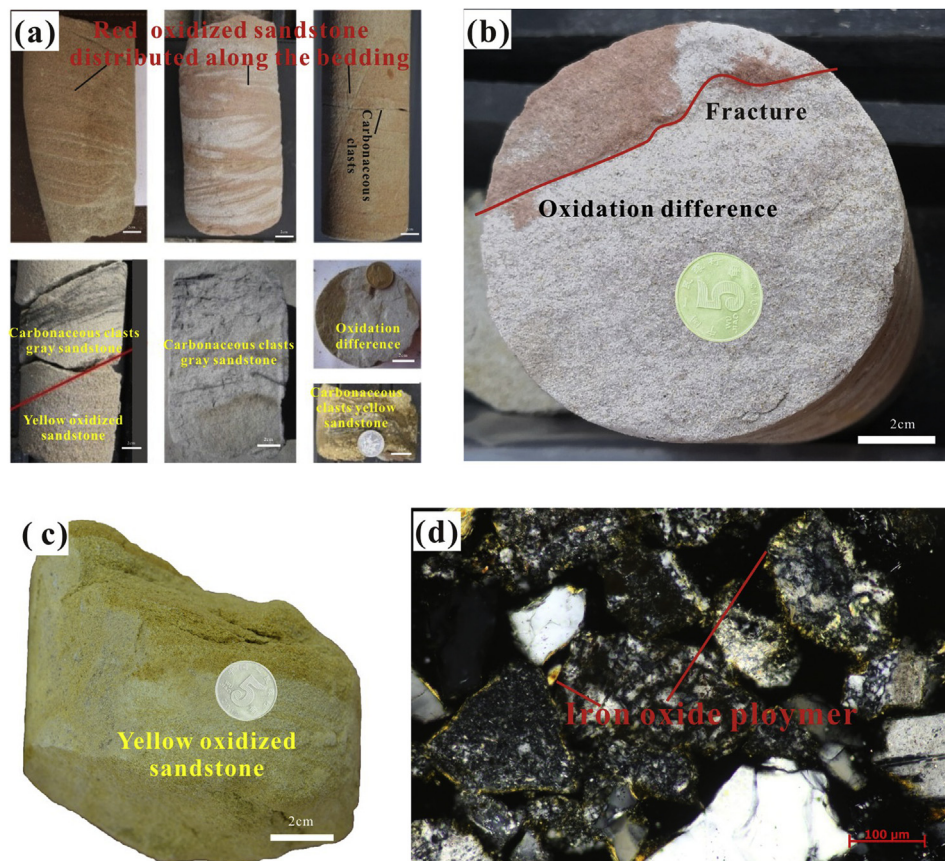


Fig. 3. The post-interlayer oxidation sandstone and primary red sandstone bleached and faded by oil-gas. (a) Red, gray, and yellow sandstone containing carbonaceous clasts; (b) Differential oxidation on both sides of the fracture. (c) Yellow sandstone in the interlayer oxidation zone. (d) The phenomenon of iron oxidation in the ore-bearing sandstone.

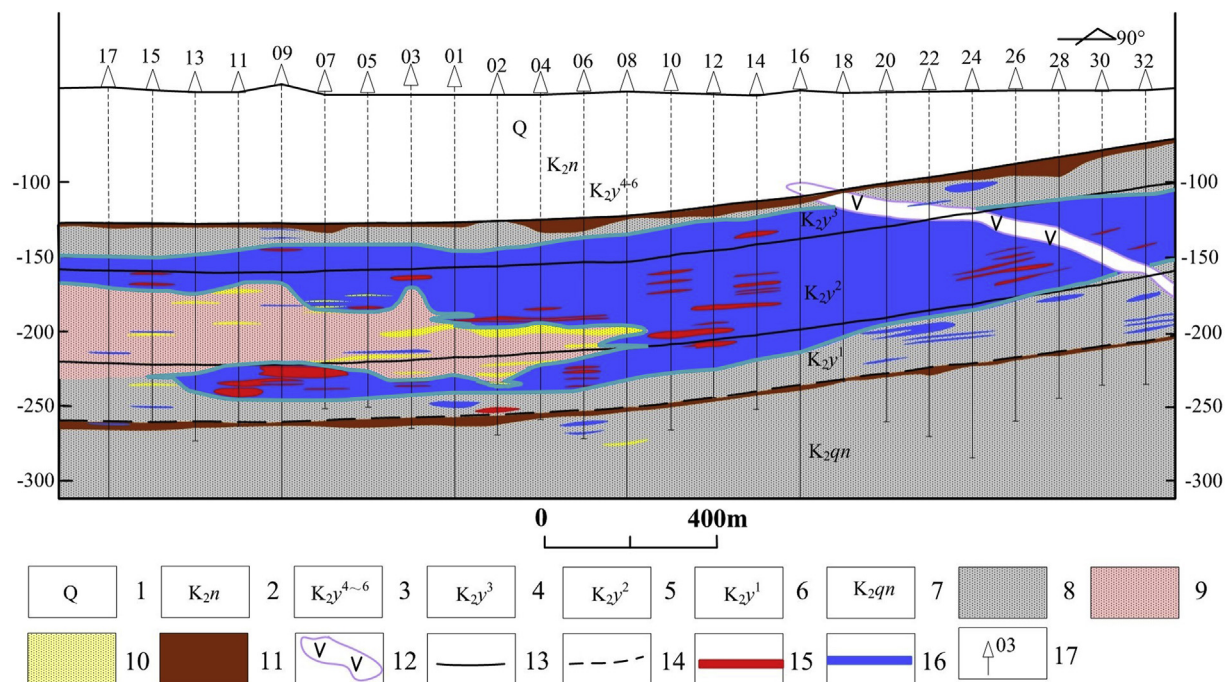


Fig. 4. Distribution of uranium orebody in No. 37 exploration line of Qian IV block of Qianjiadian uranium deposit (1. quaternary system; 2. Nenjiang Formation; 3. the 4th to 6th sections of Yaojia Formation; 4. the 3rd section of Yaojia Formation; 5. the 2nd section of Yaojia Formation; 6. the 1st section of Yaojia Formation; 7. Qingshankou Formation; 8. reduced sand body; 9. red oxidized sand body; 10. yellow oxidized sandstone; 11. mudstone; 12. diabase dike; 13. stratigraphic boundary; 14. angular unconformity boundary; 15. industrial uranium orebody; 16. uranium mineralized body; 17. low-grade uranium orebody; 17. borehole and numbering).

path of oxygen-bearing uranium water, resulting in the poor continuity of uranium orebody. Vertically, the uranium orebody has non-permeable layers interbedded with permeable ones. Therefore, the orebody continuity is poor. On the other, the content of organic matter, pyrite and other reducing media in the ore-bearing aquifer is low, and the ability to absorb uranium is poor, resulting in a uranium orebody of low grade as a whole with an average of 0.0250%, and the proportion of orebodies reaching industrial grade is low. When the low-grade uranium orebody (0.0050%) and industrial uranium orebody (0.0100%) are calculated simultaneously can it be interpreted as an orebody in the shape of a "roll" (Fig. 4).

The analysis of sedimentary facies of a single well in the target layer of the study area, comparison of the comprehensive geological conditions of consecutive wells, and interpretation of the core color of single well formation, show that the primary red bed is usually independent of lithology and lithofacies on a single well profile. Almost all sandstone and mudstone are uniformly red. The red in sandstone has no correlation with rock bedding, though it often cuts through rock bedding. The red layer generated with interlayer oxidation is stratum-bound despite the totally red area under strong oxidization. Based on log data, the sedimentary facies and rock geochemistry are projected onto the plane (Fig. 5), indicating that the interlayer oxidation occurs from north to south on the plane, and the primary and secondary oxidation zones superimposed in the tectonic skylight area overlap. Rong et al. (2016) also proposed that the ratios of oxidized/reduced sandstone in the Yaojia Formation decrease from southwest to northeast and the uranium mineralization occurs within reduced sandstones horizontally at the redox interface and vertically adjacent to mudstone

interlayers. The results just coincide with our outstanding.

3.3. Geochemical identification marks

The Th/U analysis of sandstone of different colors in Qianjiadian area (Table 1), shows that the ratio of red to yellow sandstones is greater than 1, and the content of $\text{Fe}^{3+}/\text{Fe}^{2+}$ in red and yellow sandstones is two to three times higher than that in gray sandstones, with the lowest figure shown in ore-bearing sandstones, indicating that both red and yellow sandstones have experienced oxidation and uranium migration. The S value can reflect the pyrite content in sandstones, and is in direct proportion to the u content of sandstone. The content of pyrite in red and yellow sandstones is very low, and that of the ore-bearing sandstone comes to the highest, indicating that pyrite is indeed conducive to uranium mineralization. The TOC value of sandstone in the mining area has no obvious regularity, so the possibility of reduction mineralization involved with organic matters in the Qianjiadian uranium deposit of sandstone type is relatively small (Table 1).

4. Fluid in characteristics of Qianjiadian uranium deposit

The fluid in the Qianjiadian uranium deposit mainly includes authigenic fluid released by sediment compaction and mineral phase transformation in the basin, atmospheric water supplied by the atmosphere at the edge of the basin and outside, deep-seated oil and gas fluid and magmatic hydrothermal fluid from diabase dike.

The Nenjiang tectonic inversion destroyed the deep-seated oil and gas reservoir, resulting in the deep oil and gas migrating

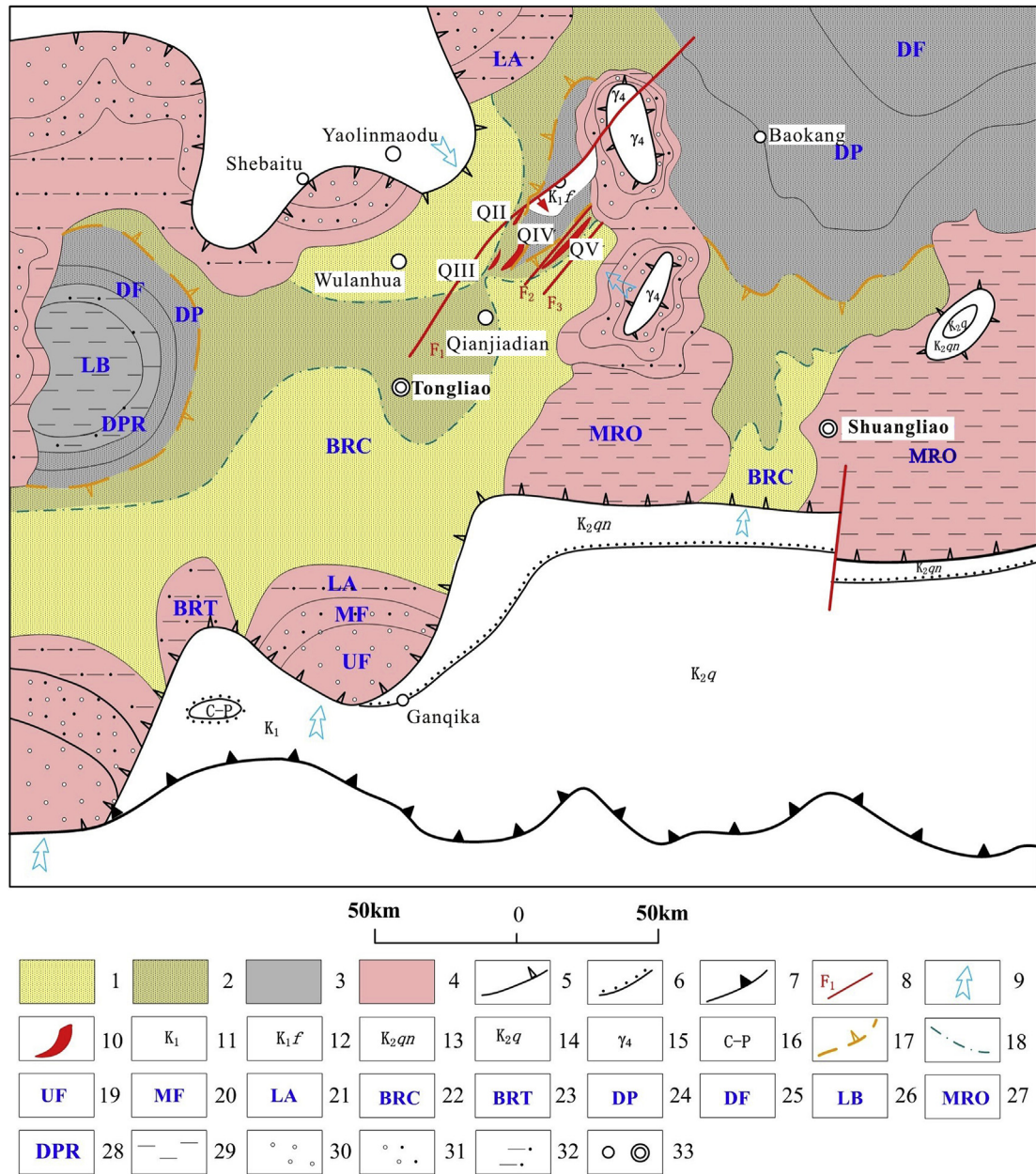


Fig. 5. Geochemical features of the Yaojia Formation in the Kailu Depression (1. epigenetic oxidation zone; 2. oxidation-reduction transition zone; 3. reduction zone; 4. primary oxidation zone; 5. denudation boundary; 6. unconformity boundary; 7. basin boundary; 8. fault and number; 9. provenance direction; 10. uranium deposit; 11. the Lower Cretaceous; 12. the Lower Cretaceous Fuxin Formation; 13. the Upper Cretaceous Qingshankou Formation; 14. the Upper Cretaceous Quantou Formation; 15. the Hercynian granite; 16. the Carboniferous Permian; 17. interlayer oxidation front line; 18. gray rock pinch-out line; 19. fan root; 20. fan middle; 21. fan end; 22. braided channel; 23. river alluvial island; 24. delta plain; 25. delta front; 26. shore shallow lake; 27. flood plain; 28. distributary channel; 29. mudstone; 30. conglomerate; 31. glutenite; 32. siltstone; 33. place name).

Table 1

Analytical results of U, Th, Fe, S and TOC contents in sandstones of different geochemical types in Qianjiadian uranium deposit (the Th/U value is quoted from [Chen, 2008](#)).

Lithology	U (10^{-6})		S (%)		TOC(%)		Th/U		Fe ³⁺ /Fe ²⁺	
	Sample number	average value	Sample number	average value	Sample number	average value	Sample number	average value	Sample number	average value
Red sandstone	6	2.73	29	0.01	28	0.11	10	1.2	27	3.43
Yellow sandstone	10	4.37	34	0.01	34	0.13	12	1.65	25	3.81
Gray-white sandstone	13	39.96	14	0.11	11	0.07	8	0.6	11	1.72
Gray-sandstone	8	9.23	8	0.03	8	0.18	5	0.16	8	1.37
Ore-bearing gray sandstone	4	264.78	4	0.14	4	0.16	11	0.03	4	1.09

Note: Th/U value is quoted from [Chen \(2008\)](#).

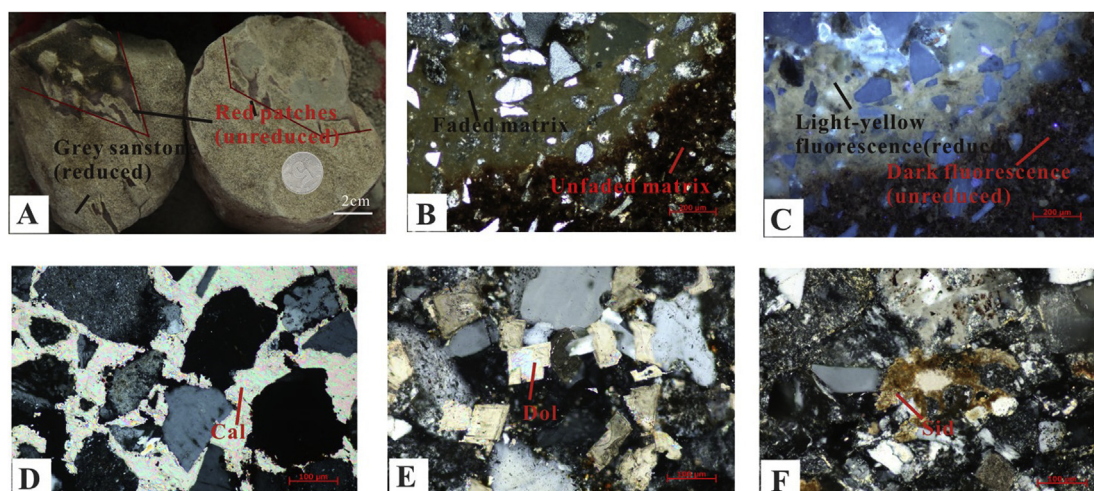


Fig. 6. Characteristics of oil-gas reduction and carbonation. A, B, C represent fine-grained lithic arkose.

upward along the fault to the shallow stratum. Oil stains, oil spots, oil mud marks or escape halos are common in the sand body, and the oil and gas are mainly composed of H_2S , CO_2 , hydrocarbon and H_2 . Acid alteration (Kaolinization) (Zhang et al., 2006) is produced by oil and gas escape, leading to originally oxidized sandstones getting reduced with a color change from red and others to gray and gray-white (Fig. 6A, B, C). The secondary oil-gas reduction after the mineralization makes the interlayer oxidation zone in the epoch of mineralization get reduced and transformed again, resulting in a frequently “interrupted” oxidation zone.

With the tectonic inversion, the intrusion of basic magma occurred in the mining area, forming a large area of basalt cover and diabase dike. Magma intrusion not only resulted in geothermal heating in the area, but also introduced abundant CO_2 , which can be the main mineralizer for uranium extraction (Tang et al., 2016). Carbonation alteration (Fig. 6D, E, F) is relatively developed in the mineralized section of the ore-bearing layer of the Yaojia Formation, especially in the gray green sandstone of the ore-bearing layer, and mainly produced by sparry calcite (Fig. 6D) with weak dolomitization (Fig. 6E) and occasionally by siderite (Fig. 6F).

5. The multi-stage metallogenic model

The fluid from both internal and external sources of the basin possibly contained the metallogenic components which served to provide the required metal source and physico-chemical conditions for uranium mineralization.

5.1. The syngenetic metallogenic stage in sedimentation

After the end of Qianjiadian fault subsidence, the strata are generally characterized by highs in the southwest and lows in the northeast with filling and leveling occurring in the Upper Cretaceous Qingshankou Formation. The sediments in the braided river depression of Yaojia Formation were mainly derived from uranium-rich acid volcanic rocks and granites (Chen et al., 2005), and formed in the paleoclimate environment with alternating dry heat and humid heat. The sediments are generally rich in plant detritus and gray, gray-black mudstone, peat interlayer, showing intense adsorption on uranium, so that the uranium anomaly in the Upper Cretaceous Yaojia Formation in Qianjiadian area is generally higher than that in other areas and other strata in this area. The abnormal deposition of uranium at this stage can be the source of uranium

enrichment and mineralization at a later stage.

5.2. The interlayer oxidation metallogenic stage

The Upper Cretaceous strata in Qianjiadian Sag inherit the characteristics of highs in the southwest and lows in the northeast during the whole period of subsidence. The south and southeast of Kailu Depression were uplifted and denuded at the end of Late Cretaceous, providing the necessary conditions for the interlayer oxidation of Yaojia Formation with both top and bottom strata and high-permeability sand body in Qianjiadian Sag. If the Qianjiadian low-grade orebody (i.e., with a quantitative gamma logging interpretation grade less than 0.01%, but greater than 0.005%) is connected with the industrial ore-body, we can still “restore” the roll-type orebody. It proves that the interlayer oxidation mineralization has indeed occurred and been key to the formation of the Qianjiadian uranium deposit.

5.3. The metallogenic stage of superimposed transformation

The tectonic inversion of Nenjiang movement also lifted some inner parts of the depression to form a denudation skylight. With the change of structural conditions, the denudation skylight also changed from the previous water release area to the water replenishment area, marking the end of the interlayer oxidation mineralization stage and the transition to a mineralization stage of phreatic oxidation transformation. It should be pointed out that with the development of Nenjiang movement, the faults generated communicated with the deep hydrothermal fluids, transferring a large amount of CO_2 and intruded into the ore-bearing strata of Yaojia Formation along the faults and fractures, extracting the uranium ions from the coiled orebody formed with the early interlayer oxidation. These uranium ions are in turn reduced and get enriched again by sandstones with stronger reduction ability, which is the main reason for the untypical development of industrial orebody of the roll type (the industrial orebody is mostly distributed in the form of plate and lens) and radium bias of the low-grade orebody in the present-day Qianjiadian uranium deposit (Luo et al., 2012). Meanwhile, the tectonic movement destroyed the deep oil-gas reservoirs, leading to the reducing agents from the oil-gas therein for uranium mineralization, and strong bleaching and fading effect on the strata (Ma et al., 2007). The phreatic oxidation zone is mainly developed around the tectonic skylight in the north

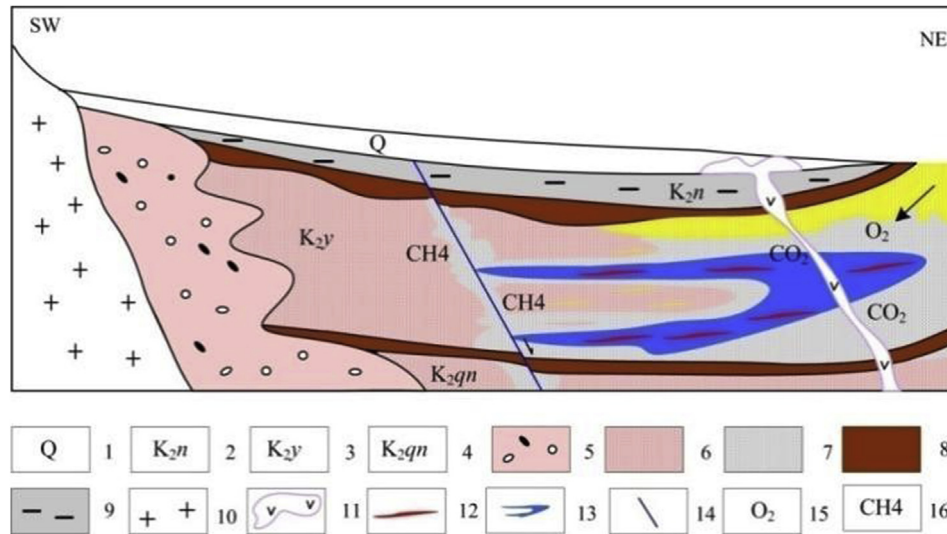


Fig. 7. Uranium metallogenic model of Qianjiadian deposit (1. Quaternary system; 2. Nenjiang Formation; 3. Yaojia Formation; 4. Qingshankou Formation; 5. alluvial fan; 6. oxidized sand body; 7. gray white sand body; 8. red mudstone; 9. gray mudstone; 10. granite; 11. diabase; 12. industrial uranium orebody; 13. uranium mineralized body; 14. fault; 15. Oxygen-bearing uranium water; 16. reducing gas).

of the Qianjiadian deposit during the main period of interlayer oxidation mineralization. The cores are almost bright-yellow, and mainly distributed downward along the denuded skylight.

6. Metallogenic model

The metallogenic characteristics of Qianjiadian uranium deposit are obviously different from those of uranium deposits of sandstone type in typical interlayer oxidation zones both at home and abroad. The Qianjiadian uranium deposit is a product of typical convoluted interlayer oxidation mineralization, and the early roll-type orebodies are reactivated by the hydrothermal fluid intrusion under tectonic movement and surface groundwater infiltration along the denudation skylight. Therefore, the occurrence of the reductive sand bodies of poor permeability in the uranium reservoir determines the shape of the current uranium-rich orebodies. Finally, the multi-stage convergence metallogenic model of "primitive sedimentary enrichment-interlayer oxidation-groundwater, oil-gas and hydrothermal superimposed transformation" is created (Fig. 7). It is characterized by comprehensive, superimposed, multi-period and multi-stage mineralization.

7. Conclusion

We analyzed the salient characteristics of Qianjiadian uranium deposit and propose a possible metallogenic model. The main conclusions from our study are as follows.

- (1) Qianjiadian uranium deposit in the Songliao Basin is a product of typical interlayer oxidation mineralization of roll type.
- (2) The occurrence of reductive sand bodies of poor permeability in the Qianjiadian uranium reservoir determines the shape of uranium-rich orebodies at present.
- (3) The Qianjiadian uranium deposit is characterized by the multi-stage superposition metallogenic model of "sedimentary preconcentration - interlayer oxidation - groundwater, oil-gas and hydrothermal superimposition transformation".

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

A is the core photo: is the lower part is reduced by oil and gas, and the upper part of brown red is patchy (unreduced); B is the micrograph in orthogonal polarized light, the upper left part is reduced by oil-gas, the matrix is faded, the lower right part is unreduced, and the matrix is not faded; C is the same field as B, fluorescence, upper left. The part is affected by oil and gas, with light yellow light between grains, and the lower right part is not affected by oil and gas, with extremely dark fluorescence. D, E, F are in orthogonal polarized light, and shows carbonation characteristics.

D shows bright calcite intergranular cementation, and local dissolution of debris and cement. E shows grains filled with rhombic dolomite. F shows intergranular mud and locally enriched silty siderite.

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