A Study of the Accompanying Relationships between Uranium and Oil

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Abstract It is not occasional that uranium deposits and oil accumulation occur in the same depression in the Erlian basin, Inner Mongolia. Some evidences show certain relations between uranium and oil in origin. This paper discusses and analyses the evidence for the relations between uranium deposits and oil and gas accumulation in terms of spatial distribution, geology, hydrochemistry and geochemistry. The paper also deals with the mechanism of the formation of uranium deposits and points out that it is of significance to use uranium as an indicator to search for oil and vice versa.

Key word: Erlian basin, uranium deposit, oil accumulation, accompanying relationship

As is well known, uranium is a kind of metallic resources while hydrocarbon is a kind of nonmetallic resources. It seems that there is no relationship between them and it is unreasonable to study their relationships. In fact, there are close accompanying relationships between them in some geological environments. They were located in the same structural environment. U deposits are in the upper part of an oil and gas field and its peripheral areas. This can be seen in the Erlian basin, Tuha basin, Ordos basin, Songliao basin and Qaidam basin of China, and some regions in other countries (Russian and Canada for example). In this paper, U deposits and hydrocarbon reservoirs in the Erennuo'er sag of the Erlian basin are taken as an example to analyze and study their genetic mechanism and summarize the model of their relationships.

1 Geological Setting

The Erlian basin is a Ceno-Mesozoic continental basin developed on the basis of the Late Hercynian Xingmeng geosyncline fold belt. The Erennuo'er sag is located in the Ulan Qab depression, which is in the northeast Erlian basin, where only Proterozoic, Mesozoic and Cenozoic strata outcrop with active tectonism and magmatism. The Subeng and Nuheting uranium deposits and more than ten uranium occurrences have been found in the past few years. At the same time, the Jigesen and Bao'er oil-gas fields have been discovered, too. U deposits are mainly hosted in the Late Cretaceous Erliandabusu Formation and hydrocarbon reservoirs have been discovered in the A'ershan Formation and Tengger Formation of the Bayanhua Group of the Late Cretaceous.

2 Relations between Uranium Deposits and Hydrocarbon Reservoirs

2.1 Distribution and geological features

The Nuheting U deposit in the Erennuo'er sag lies above the Jigesen oil field, and Subeng U deposit is located on the northwest peripheries of the Jigesen and the Bao'er oil and gas accumulation (Fig. 1). Hydrocarbon reservoirs are distributed in the lower part of the fan-delta and braided stream fronts that consist of sandstone and sandy conglomerate and developed during the forming and expanding stage of the lake basin. U mineralization took place in the upper strata, mainly in braided channel sand, siltstone and shale siltstone in the fan front and the area between fans during the shrinking stage of the lake basin.

The Nuheting U deposit is distributed in a large area, about 9.32 km^2, but it is very thin (0.99 m thick on average) with a sheet form. The boundary of the deposit is quite straight on profile and the height difference of the deposit is only 15.31 m within a dis-

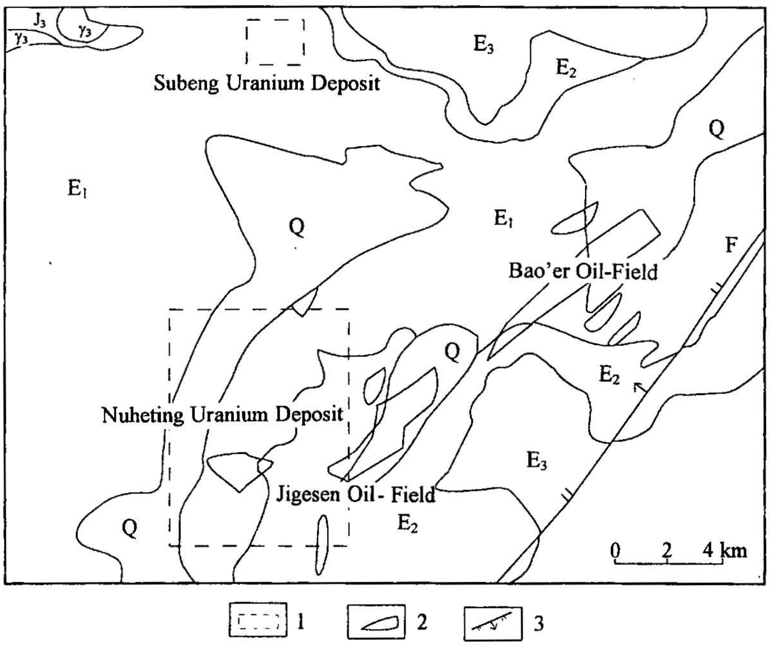

Fig. 1. Geological sketch map of the Erennuo'er Depression

1. Range of uranium deposit; 2. area of oil accumulation; 3. reverse fault and dip; Q–Quaternary alluvium; E_3–Oligocene sandstone and mudstone; E_2–Eocene sandstone, mudstone and conglomerate; E_1–Paleocene sandstone and conglomerate; J_3–Late Jurassic tuffaceous sandstone and conglomerate; γ_5–Late Yanshanian granite.

tance of 4 km, almost horizontal. The horizontal boundary of the deposit is more straight than the rock beds and intersecting of orebodies and rock beds are sometimes seen. The orebodies occur in argillaceous siltstone, silty mudstone, fine sandstone and medium-grained sandstone, showing the deposit has no close relationship with lithology. The unique feature of the U deposit shows that reducing gases joined the epidiagenesis. There must be a huge mudstone caprock 3-7 m away from the orebodies. The phenomenon that the main orebody is in the upper part and the minor bodies are in the lower part may be caused by gases dissolved in mineral liquids during the mineralizing period. Above it, CO_2 as an oxidation gas occupies a dominant position and so does the reducing hydrocarbon gas under it. The deposit is only formed in oxidation-reduction transition environment and the hydrocarbon reducing gas in ore-bearing beds is rich near the main orebody. The distribution feature of the orebodies also reflects that reducing gases joined the mineralization.

In the core, there are many vertical or steep fissures, some of them were filled with colloform pyrite. The rocks on two sides of the fissures are characterized by faded alteration. Around the U deposit there are some special grey sandstones that are proved to be oil-soaked sandstones by pyrolysis and the chloroform dissolve fluorescence method. Some of the ores contain a little hydrocarbon, and the thickness of the U orebody is 4.13 m and the uranium content is 1.13%. The phenomenon shows that oil and gas are very important to form U deposits. Two exploration wells have gas emanation in the Subeng U deposit. The black oil-bearing sandstone occurs at the core of one well and gives off asphalt smell. Film-like organisms can be seen on the surface of gravel at the core of the other well. It is noticeable that the mineralised beds around the gas-emanation well are very thick and have high grades of ore. The U-Ra balance deviates markedly from Ra. Obviously, sandstones and siltstones in the 12 oil exploration wells show gamma anomalies in the well logging curve. the radiant dose rate is 5–17.5 nc/kg·h, so do sandstones and siltstones in 4 oil wells in the Nuheting U deposit.

2.2 Hydrochemical characteristics

There are some relations between the groundwater in uranium deposits and the oil-field water. According to some research, the distribution of main cations (including Na^+, Ca^{2+}, Mg^{2+}) obeys a bond parameter regularity and their correlation with hydro-chemical energy is especially good. The contents of Na^+, Ca^{2+}, Mg^{2+} in fresh water do not obey a bond parameter regularity. The experiment of mixing oil-field water with fresh water shows that the correlative coefficient will gradually decrease with the increase of fresh water.

When oil-field water has a relationship with shallow water above it, the influence of the shallow water above oil reservoirs affected by the oil-field water can produce obvious regular anomalies. On the contrary, if the shallow water has evident regular anomalies, there

Table 1 Cation concentration in water and correlation coefficients with hydrochemical energy (modified from Zhang Ruliang et al., 1994)

Location	Water sample	Cation concentration (log Mi)			γ	a	$b\times10^{-2}$
		Na^+	Ca^{2+}	Mg^{2+}			
Jigesen oil field	Oil-field water	3.68	1.46	0.92	−0.99	4.406	−0.758
Well water	Fresh water	0.94	1.41	0.62	−0.159	1.093	−0.033
Mixed water (oil-field water: fresh water)	1:2	3.21	1.42	0.77	−0.99	3.863	−0.660
	1:4	2.99	1.42	0.73	−0.99	3.604	−0.604
	1:8	2.76	1.43	0.64	−0.99	3.346	−0.554
	1:16	2.48	1.51	0.64	−0.97	3.012	−0.468
Subeng uranium deposit	Ore bed water	2.79	2.16	1.93	−1.00	3.021	−0.233
	Sandstone water	2.93	2.09	2.05	−0.97	3.156	−0.255
Nuheting uranium deposit	Ore bed water	2.72	1.69	1.36	−0.99	3.088	−0.372
	Sandstone water	3.07	2.56	2.20	−0.98		−0.225
Subeng periphery	Spring water	3.28	3.17	1.30	−0.74	3.080	−0.430

Note: long Mi=a+bL (Mi=Na^+, Ca^{2+}, Mg^{2+}concentration, a=intercept, b=slope, γ=correlation coefficient of cation concentration in water with hydrochemical energy L)

Table 2 Comparison of soil gas with oil-gas in the Nuheting uranium deposit (modified from Zhang Ruliang et al., 1994)

SSPL	Sample depth (m)	MET	ETH	PRO	Butane		Pentane		k	Gas feature
					IS	NL	IS	NL		
		C_1%	C_2%	C_3%	IC_4%	NC_4%	IC_5%	NC_5%		
GF	500-600	89.17	1.79						49.82	SHG
	601-700	83.27	1.61	1.29	0.74	0.86	0.77	0.57	14.26	ADG
	701-800	81.89	1.82	0.92	0.32	0.43	0.19	0.20	21.11	ADG
	1000-1500	72.08	5.08	4.02	1.56	1.85	0.70	0.77	5.16	ADG
	1501-1500	68.03	4.75	6.07	1.24	3.23	1.12	1.30	3.84	ADG
	>2000	62.35	10.68	9.83	1.95	3.60	0.81	1.17	2.22	ADG
SB	0.4-1	81.01	10.47	3.84	3.10	1.94	0.54	0.24	4.70	ADG
NNH	0.4-1	76.13	13.24	7.84	0.43	1.67	0.29	0.12	3.66	ADG

Notes: SPL: sample location, JGF: Jigesen oil-field, SB: Subeng uranium deposit, NH: Nuheting uranium deposit, MET: methane, ETH: ethane, PRO: propane, IS: isomeric, NL: normal, SHG: shallow gas, ADG: associated gas, $K=C_1/(C_2+C_3+IC_4+NC_4+IC_5+NC_5)$.

are relationships between the shallow water and deep oil-field water. Based on this method, the correlation between the cations concentration of Na^+, Ca^{2+} and Mg^{2+} in the groundwater and the hydrochemical energy in the Nuheting and the Subeng U deposits was studied. The result is shown in Table.

The contents of Na^+, Ca^{2+} and Mg^{2+} in the groundwater of the U deposit show good relationships with hydrochemical energy, which shows that the groundwater of the uranium deposit is mixed with oil-field water, and has a connection with oil-field water.

The groundwater of the U deposit has a high content of H_2S, that is 2.21-25.55 g/m^3 in exploration wells. A lot of research results show that H_2S was generated by sulphate interacting with light hydrocarbon and microbiology:

$$SO_4^{2-}+CH_4 \xrightarrow{\text{bacterium}} H_2S+CO_3^{2-}+H_2O$$

2.3 Geochemical Characteristics

(1) Chemical exploration data collected from soil in the deep boreholes on the Nuheting and Subeng U deposits show that there are some hydrocarbon anomalies and gas anomalies of H_2S and CO_2 in the upper parts of both ore deposits. Like the ingredients of associated gases of deep-seated oil, the ingredient of the hydrocarbon above the U deposits is mainly

methane. The ratios of $C_1/(C_2+C_3+IC_4+NC_4+IC_5+NC_5)$ are generally 2.01–5.06 (Table 2). H_2S and a part of CO_2 may be the products of chemical reaction of the wall rocks and media with the accompanying gases of oil when the gases infiltrate the wall rocks.

(2) The results obtained by analyzing 11 rock-mineral samples taken from 2 boreholes in the Nuheting deposit show that the average content of total hydrocarbon in rocks and ores is 8253.12 μL/kg. Moreover, the highest content of hydrocarbon can reach 18217.0 μL/kg. In addition, the content of heavy hydrocarbon takes up 22% of the total hydrocarbon. All indexes and the scale of absorbed hydrocarbon in rocks and ores indicate that the hydrocarbon originates from oil because most samples were taken from the sandstone member of the Late Cretaceous Erliandabusu Formation, and the sandstone member is not an oil-generating stratum because it contains little kerogen and chloroform pitch A. As a result, the hydrocarbon gases in the sandstone member originated from deep oil and gas accumulation.

(3) Based on the results of the test for 111 rock samples of specific potential, the orebodies of the Nuheting U deposit show relative high ΔEh values which are about 65 mV. But in the barren zone, they are about 34–39 mV. This shows that there are quite abundant reducing media in the orebodies, which are consistent with the outer characteristics of grey rocks in the orebodies and red rocks in the barren zone, so the reducing media may be gas-oil materials causing the high ΔEh values.

(4) According to the identification and analysis of ore inclusions, the samples taken from the main body of the Nuheting U deposit contain a mass of organic gas-liquid inclusions ranging from organic gas inclusions to pure liquid (liquid hydrocarbon and petroleum) inclusions which have relatively small GLR. These inclusions give out brown-yellow fluorescence under the stimulation of ultraviolet. The inclusions have elongated or irregular forms with black ellipsoidal bubbles. In the experiment of static soaking of uranium ores with distilled water, , there are always black thick oily materials floating on the surface of the soaking liquor. Infrared spectroscopic analysis shows that they are aromatic hydrocarbon organic matter.

Although some new evidences may be found in further research, it has already been proved that the genesis of gas and oil is related to the uranium deposits in the research area.

3 Discussion on the Accompanying Relationships between Uranium Deposits and Oil-Gas Accumulation

When sediments were carried to depositional basins under warm and moist climate, they mostly formed dark grey or grey muddy and muddy sand strata that are rich in organism. The sedimentary strata are usually U-bearing formations because clay minerals and organisms absorb uranium quite a lot. Moreover, their organisms are also necessary reductants for forming hydrogenic uranium deposits. In addition, these sedimentary strata are fairly good source strata of oil and gas, so uranium and oil have the same source.

3.1 Formation of oil and gas reservoirs

At a certain temperature and pressure and under the action of bacteria, activators and radioactivity, sedimentary organisms can produce gas and oil, such as CO_2, NH_3, H_2S and CH_4, through the diagenetic, plutonic and quasi-metamorphic stages and oil and gas were accumulated in favourable traps.

(1) Diagenetic stage In the early stage of diagenesis, subjected by decomposition and hydrolysis of bacteria, biologic ploymers such as fat, protein, hydrocarbon and lignin can be transformed into biochemical monomer with low molecular weight (fat acid, amino acid, sugar and phenol) and produce CO_2, NH_3, H_2S, CH_4 and H_2O, etc. With the increase of the burial depth, bacteria action tends to end, and inorganic transformation is more important. Owing to polycondensation, biochemical monomers will become complex humus acids with high molecular weight, and then evolve into a geo-ploymer, i.e. kerogen, which will further become stable and insoluble organism because it is complexed by surrounding minerals.

(2) Plutonic stage It is the main stage for kerogen to turn into oil and gas. With the increase of the burial depth and temperature, a large amount of kerogen begin to degrade into petroleum at the critical points of depth and temperature and at last crack into light-crude and wet gas under the thermal action and cata-

lytic action of clay.

(3) Quasi-metamorphic stage In this stage, the oil-producing potential of kerogen becomes exhausted, and it transforms into high-temperature methane because kerogen is buried deeply where the geotherm is very high.. In addition, former light-crude and wet gas can also crack into the most stable methane in thermodynamics.

3.2 Formation of U deposits related to oil and gas fields

3.2.1 Accumulation and migration of source materials

(1) Formation of source layers U can move as hexavalent U in an oxidized condition and precipitate as quadravalent U in a reduced condition. As a result, in uraniferous layers (bodies) in denuded regions uranium is oxidized into a high valent state by superficial water and oxygenated groundwater and migrates into neighbouring restricted or semi-restricted drainage basins mainly in a form of solution. Then it is reduced and absorbed in sediments by reducing materials such as carbonate and organism and its derived matter and colloid hydrate. The sediments are consolidated to form rocks and source layer of uranium ore at last.

(2) Dissolution and migration of U in shallow ore source layers In the interstratified oxidizing milieu, when U is dissolved, the shallow ore source layer migrate nearly horizontally. U exists in groundwater as carbonate uranyl complex $[UO_2(CO_3)_2(H_2O)_2]^{2-}$, $[UO_2(CO_3)_3]^{4-}$, sulphate uranyl complex $[UO_2(SO)_2]^{2-}$, hydrate uranyl ionic complex $[UO_2OH]^+$. Uranly complex migrates from the runoff area to the region of outflow following oxygenated groundwater.

(3) Dissolution and migration of U in deep ore source layers Since oxygen is rare in deep layers and solutions always contain some reductants, so deep U is generally not easy to migrate. But when there are organisms and oil-gas materials, a small amount of uranium in the source layer can enter solutions, and then go through faults, fractures and holes, move from deep to shallow vertically as extracted U by petroleum hydrocarbon, bicarbonate uranyl complex and halogenide.

3.2.2 Formation of the reducing barrier of oil and gas

Low molecule hydrocarbon gases in the oil and gas accumulation percolate and move from the deep source to the shallow interstratified oxidizing milieu upwards faults, fractures and holes. They usually accumulate and form oil and gas reducing barrier if they are obstructed by a stable and thick cover of mudstone in a suitable structure position. That's to say, reducing gases like CH_4, CO, NH_3, H_2 accumulate in a large amount and lower the Eh value to form highly reducing environment. At the same time, CH_4 interact with sulfate in groundwater under the effect of microbe to originate the reductant of H_2S.

3.2.3 concentration and mineralization of U in oil and gas reducing barrier

The formation of a local oil and gas reducing barrier changed the physical and chemical conditions in the interstratified oxidizing milieu, which resulted in a number of oxidation-reducing reactions and other chemical reactions to accelerated uranium mineralization by means of reduction, precipitation and concentration. The possible geochemical expressions are as follows.

(1) Hydrocarbon is oxidized under bacterial action to form carbonate minerals.

$$CH_4+2O_2 \xrightarrow{\text{bacterium}} CO_2+2H_2O$$

$$CO_2+H_2O \longrightarrow H_2CO_3$$

$$H_2CO_3+M^{2+} \longrightarrow MCO_3 \text{ (dolomite and calcite)}$$

(2) U is reduced.

A: Under the action of reductants, hexavalent U in complexes is reduced to quadravalent U in alkaline media.

$$[UO_2(CO_3)_3]^{4-}+CO_3^{2-}+2H_2O+2e \xrightarrow{\text{reduction}} [U(CO_3)_4]^{4-}+4OH^-$$

$$[U(CO_3)_4]^{4-}+4OH^- \xrightarrow{\text{hydrolysis}} U(OH)_4\downarrow+4CO_3^{2-}$$

B: Acting with organisms, hexavalent U in complexes is reduced.

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

C: Acting with H_2S and HS^-, hexavalent U in complexes is reduced

$$Na_4[UO_2(CO_3)_3]+H_2S \longrightarrow UO_2\downarrow+S+2Na_2CO_3+H_2O+CO_2$$

$$4[UO_2(CO_3)_3]^{4-}+HS^-+15H^+ \longrightarrow 4UO_2\downarrow+SO_4^{2-}+12CO_2+8H_2O$$

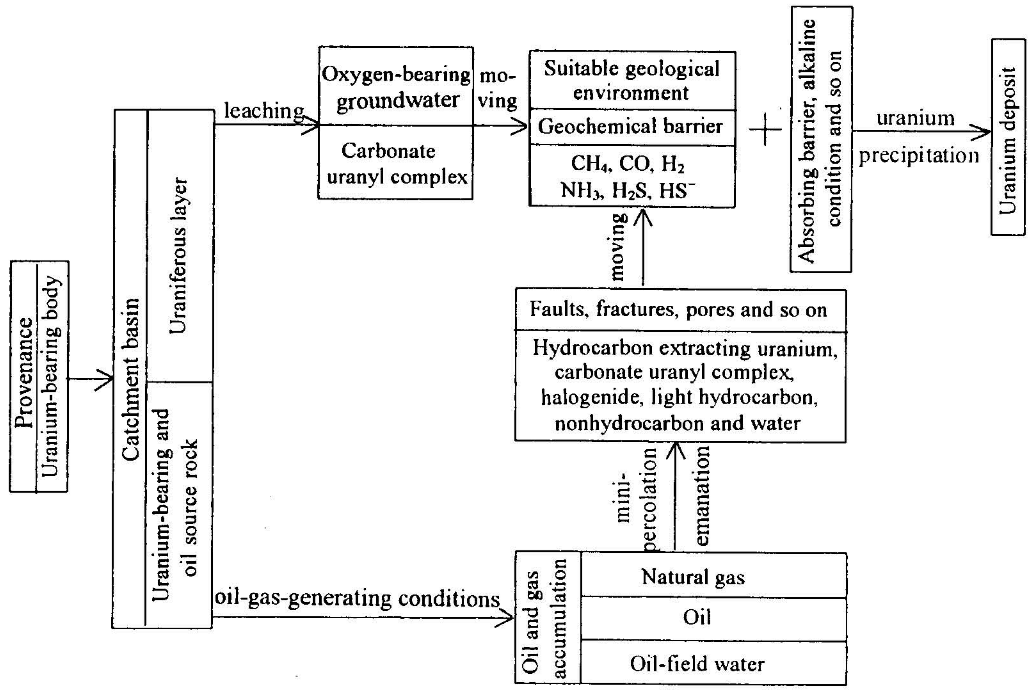

Fig. 2. Mechanism model of uranium deposits associated with oil accumulation.

(3) Under the action of reductants, Fe^{3+} is reduced to Fe^{2+} and SO_4^{2-} to S^{2-}, then Fe^{2+} is connected with S^{2-} to produce pyrite paragenerated with U.

In a kinetic oil and gas reducing barrier with spouts of outflow at suitable tectonic positions, U precipitates and concentrates continually to form U ore deposits due to secular interactions of the above geochemical reactions. The main orebody of the U deposit occur usually in the position where the migrating front of oil-gas water is obstructed by O-rich and uraniferous phreatic flows from eroded areas. Differing from the deposits of the interstratified oxidizing milieu in common sedimentary basins, geochemical mutation zones of the mineralizing area in this deposit occurred under the condition that phreatic flows, which came from eroded areas and flowed along the pervious beds in mudstone shield, were mixed in the migration front of oil-gas. However, U deposits in the interstratified oxidizing milieu were formed like this: uncombined oxygen was exhausted by reducing medium when O-rich and uraniferous phreatic flows moved slowly to deeper positions from eroded areas along permeable beds in screening beds of mudstone. And the frontal position of the phreatic flow became a redox zone. At the same time hexavalent U was reduced and precipitated to form curved or tongue-shaped U bodies.

3.3 Mechanism model of U ore deposits associated with oil accumulation

A sketch diagram of the mechanism model of U ore deposits associated with oil accumulation (Fig.2) was proposed on the basis of the discussion on the genetic mechanism of U ore deposits (mineralization) and oil and gas accumulation (Weart, 1981). It shows visually the close relationships between the formation of U ore deposits and the genesis of oil and gas accumulation.

4 Conclusions

Uranium deposits on the top or marginal parts of oil and gas fields are generally formed when reducing gases (such as organic gases, CO and H_2S) permeate hydrocarbon traps through faults, fractures and pores, where there were sufficient hexavalent U and suitable geological environment for mineralization. This is a very important concept which can be applied to "prospect for uranium by oil or to prospect for oil by uranium".

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