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Hydrodynamic regime as a major control on localization of uranium mineralization in sedimentary basins

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Uranium deposits in sedimentary basins can be formed at various depths, from near surface to the basement. While many factors may have played a role in controlling the location of mineralization, examination of various examples in the world, coupled with numerical modeling of fluid flow, indicates that the hydrodynamic regime of a basin may have exerted a major control on the localization of uranium deposits. If a basin is strongly overpressured, due to rapid sedimentation, abundance of low-permeability sediments or generation of hydrocarbons, fluid flow is dominantly upward and uranium mineralization is likely limited at shallow depths. If a basin is moderately overpressured, upward moving fluids carrying reducing agents may meet downward moving, oxidizing, uranium-bearing fluids in the middle of the basin, forming uranium deposits at moderate depths. If a basin is weakly or not overpressured, either due to slow sedimentation or dominance of high-permeability lithologies, minor topographic disturbance or density variation may drive oxidizing fluids to the bottom of the basin, leaching uranium either from the basin or the basement, forming unconformity-type uranium deposits. It is therefore important to analyze the hydrodynamic regime of a basin in order to predict the most likely type and location of uranium deposits in the basin.

uranium deposits, localization, sedimentary basins, hydrodynamic regime, fluid overpressure

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Uranium deposits can be found in various geologic environments, among which those in sedimentary basins are economically most important (Kyser and Cuney, 2008a). Uranium deposits in sedimentary basins mainly include the unconformity type, which occurs near the unconformity between basinal sedimentary rocks and the underlying basement rocks, and the sandstone type, which is developed at various levels within the basin (Nash et al., 1981; Kyser and Cuney, 2008b, 2008c; Dahlkamp, 2009).

When exploring for uranium deposits in a sedimentary basin, it is of strategic importance to predict what types of uranium deposits are potentially important, and what stratigraphic intervals or depths are favorable for mineralization. Much attention has been paid to local geologic conditions for localizing uranium ores, especially enrichment of reducing agents such as organic matter, graphite, ferrous lithologies, and sulfides (Nash et al., 1981; Reynolds and Goldhaber, 1983; Northrop and Goldhaber, 1990; Spirakis, 1996; Alexandre et al., 2005), which has been changing secularly with the geologic evolution of the earth (Cuney, 2010). On the other hand, several studies have focused on the fluid flow models related to different types of uranium deposits in sedimentary basins (Sanford, 1992; Raffensperger and Garven, 1995a; Xue et al., 2010, 2011; Cui et al., 2012), or coupled fluid flow and chemical reaction models (Raffensperger and Garven, 1995b). It is not the intention of this paper to discuss about the various geochemical pro-

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cesses involved in uranium extraction, transport and deposition, or the fluid flow models of individual types of uranium deposits. Instead, we focus on the differences in hydrodynamic regimes between different types of uranium deposits in sedimentary basins, based on examination of major uranium districts in the world, and try to explain how the hydrodynamic regime of a basin may have played a critical role in the localization of uranium deposits, through numerical modeling of fluid pressures in generic basins.

1 Uranium mineralization at different depths

Uranium deposits in sedimentary basins can be broadly divided into three categories based on the location of the deposits at the time of mineralization: Near the surface, at moderate depths, and at the base of the basin and immediately below the unconformity (Figure 1). The first two categories generally occur in roll or tabular forms, whereas the third one is in various forms and associated with faults (Figure 1) (Kyser and Cuney, 2008b, 2008c; Dahlkamp, 2009).

Uranium deposits formed near the surface are exemplified by those in the Colorado Plateau and Gulf Coast uranium provinces in the United States of America (Finch, 1996). The uranium deposits in the Colorado Plateau typically consist of tabular orebodies mainly hosted in Upper Jurassic sandstones of fluvial origin (Finch, 1996; Dahlkamp, 2009), and are interpreted to have formed at shallow depths shortly after the deposition of the host rocks (Fisher, 1970; Northrop and Goldhaber, 1990; Sanford, 1992; Spirakis, 1996). The mineralization has been related to a fresh water-basinal brine interface (Northrop and Goldhaber, 1990; Sanford, 1992), where either a change in pH (Northrop and Goldhaber, 1990) or accumulation of humate (Sanford, 1992; Spirakis, 1996) may have been responsible for precipitation of uranium minerals. The fresh water–basinal brine interface is maintained at shallow depth due to either strong compaction-driven, upward basinal fluid flow (Northrop and Goldhaber, 1990) or the interplay between deep, regional and shallow, local groundwater systems (Sanford, 1992). The uranium deposits in the Gulf Coast (southern Texas) are mainly of roll-front type, hosted in the Eocene to Pliocene marginal marine sandstones (Finch, 1996; Dahlkamp, 2009). Uranium mineralization took place during early diagenesis, when oxidizing uranium-bearing groundwater flowed down-dip from basin margin toward basin center and encountered reducing agents such as hydrocarbons and H_2S ascending from deeper parts of the basin along growth faults (Reynolds and Goldhaber, 1983; Finch, 1996; Dahlkamp, 2009). High fluid overpressures developed in the Gulf of Mexico basin (Harrison and Summa, 1991) are likely responsible for maintaining the oxidation-reduction front at shallow depths. The roll-front type uranium deposits hosted in the Cretaceous to Tertiary fluvial sandstones in continental basins related to the Laramide Orogeny (mainly in Wyoming) are interpreted to have formed from reactions between oxidizing uranium-bearing groundwater and detrital carbonaceous debris and H_2S derived from bacterial sulfate reduction (Fisher, 1970; Reynolds and Goldhaber, 1983; Spirakis, 1996). These deposits may have formed relatively long after the host rocks (Fisher, 1970), and the mineralization environment may have been slightly deeper than those of the Colorado Plateau and southern Texas uranium deposits.

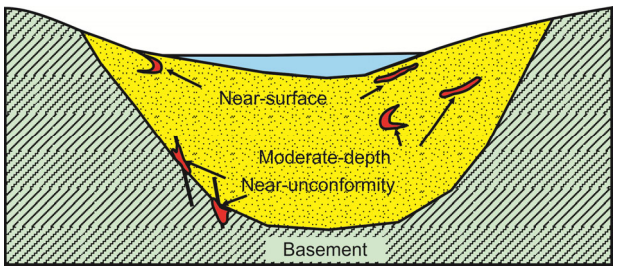

Figure 1 A sketch cross section of a sedimentary basin showing locations of uranium deposits at different depths.

Uranium deposits formed at moderate depths in sedimentary basins may be represented by those in the Chu-Saryssu and Syrdarya basins in Khazakstan and the Ordos Basin in China. The Khazakstan uranium deposits are of roll-front type and hosted mainly in Upper Cretaceous sandstones formed in continental to marginal marine environments, which are relatively poor in organic matter (Fyodorov, 1999; Jaireth et al., 2008; Dahlkamp, 2009). The host rocks of the uranium deposits are underlain by an oil- and gas-containing Paleozoic succession up to five kilometers thick (Bykadorov et al., 2003). The ages of mineralization range from the late Oligocene to Quaternary (Mikhailov and Petrov, 1998), and the current depths of orebodies range from 100 to more than 800 m, suggesting that mineralization took place at considerable depths. Uranium mineralization is interpreted to have resulted from basin-ward flow of oxidizing fluid driven by topographic relief related to the Tianshan Mountains more than 300 km to the southeast (Fyodorov, 1999); the uranium-bearing oxidizing fluids reacted with hydrocarbon and H_2S gases released from underlying hydrocarbon reservoirs, causing uranium precipitation (Fyodorov, 1999; Jaireth et al., 2008). The uranium deposits in the Ordos Basin are mainly of tabular shape and hosted in the Upper Jurassic fluvial sandstones, overlain by the Cretaceous rocks and underlain by the Paleozoic and lower Mesozoic successions containing abundant oil, gas and coal (Wei and Wang, 2004). The dominantly late Cretaceous U-Pb isotopic ages of uranium minerals (Xia et al., 2003) suggest that mineralization took place long after the deposition of the host rocks. Organic matter maturation studies indicate that as much as 1340 m of the Cretaceous sediments have been eroded above the currently preserved strata (Ren et al., 2006), which is consistent with fluid in-

clusion homogenization temperatures of 60°C to 180°C (Ling, 2007) indicating moderate burial and/or hydrothermal environment of mineralization. The uranium mineralization is interpreted to have resulted from interaction between downward flowing, oxidizing and uranium-bearing fluids and upward flowing, reducing and hydrocarbon-bearing fluids (Wei and Wang, 2004; Xue et al., 2010, 2011).

Uranium deposits formed at the base of basins or in the basement immediately below the unconformity (unconformity-type) are represented by those occurring in the late Paleoproterozoic to Mesoproterozoic Athabasca Basin in Canada and the Kombolgie Basin in Australia (Jefferson et al., 2007; Kyser and Cuney, 2008b). In the Athabasca Basin, uranium deposits are either hosted in sandstones of the Athabasca Group directly above or in the basement below the unconformity, both associated with faults that offset the unconformity, with or without graphite zones in the basement. Sediments were deposited from 1750 to <1541 Ma, whereas the primary uranium mineralization ages are mainly from ca. 1600 to 1500 Ma, although younger ages are abundant, possibly reflecting remobilization or younger mineralization events (Jefferson et al., 2007; Kyser and Cuney, 2008b). Fluid inclusions, stable isotopes, and clay mineralogy studies suggest that the ore-forming fluids were basinal brines with temperatures mainly from 180°C to 250°C (Pagel et al., 1980; Kyser et al., 2000), suggesting hydrothermal or diagenetic conditions with >5 km burial. The uranium deposits in the Kombolgie Basin are similar to those in the Athabasca Basin except that all the deposits in the former are hosted in the basement. Both the Athabasca and Kombolgie basins are characterized by paucity of mud in the sediments, partly due to the absence of sediment-stabilizing land plants (Kyser, 2007; Kyser and Cuney, 2008b), which may have facilitated basinal fluid circulation. Unconformity-type uranium mineralization is interpreted to have resulted from interaction of oxidizing basinal fluids and reducing agents derived from or located in the basement, with uranium being derived either from the basin (Kyser et al., 2000) or from the basement (Hecht and Cuney, 2000).

2 Numerical models of basin-scale hydrodynamic control on localization of the oxidation-reduction front

The above examples all indicate that uranium mineralization took place where oxidizing uranium-bearing fluids encountered reducing agents (either solid material such as organic matter or fluids such as hydrocarbons and H_2S). It can be shown that the hydrodynamic framework of the basin, specifically the fluid pressure regime, exerts a major control on the oxidation-reduction front, thus the localization of uranium mineralization. This concept is illustrated by numerical modeling of fluid overpressures and fluid flow patterns in different basin models, using the software Basin2 (Bethke et al., 1993). The basic assumption is that there is a disequilibrium compaction-driven, upward moving, reducing agent-carrying fluid system in the lower part of the basin, and a gravity-driven, downward moving, uranium-bearing oxidizing fluid system in the upper part of the basin, with the interface between the two systems representing the oxidation-reduction front (Figure 2). Note the basin is not entirely submerged in water (Figure 1), so the compaction-driven and gravity-driven systems can operate at the same time.

Three generic basins (cases), with different combinations of lithology, sedimentation rate, and topographic relief are modeled. The basin is 500 km wide and the thickness of sediments is 5 km for all three cases. The basin is considered to be symmetric, so only half of the basin is modeled. Although Basin2 can model basins with complex geometry and lithologic combinations, we deliberately choose simple physical models in order to highlight the basin-scale hydrodynamic features of general significance. In case 1, the basin is filled with sediments composed of 90% sand and 10% shale, the sedimentation took place over a period of 100 Ma, and a topographic relief of 0, 50 and 100 m is tested respectively. In case 2, the basin sediments are composed of 50% sand and 50% shale, and the duration of sedimentation is 30 Ma, whereas in case 3, the sediments consist of 30% sand and 70% shale, with a sedimentation duration of 10 Ma. A topographic relief of 0, 50 and 200 m is applied in case 2, and a set of values of 0, 100 and 500 m is used in case 3. Simulations were also carried out to evaluate fluid overpressure dissipation after the termination of sedimentation in case 3.

The model started with a thin layer of sediments at hydrostatic fluid pressure, and sediments were gradually added above it throughout the sedimentation history of the basin. The right and upper boundaries are open to fluid flow, and the bottom is impermeable. The left boundary is located at the center of a symmetrical basin, so the net horizontal fluid

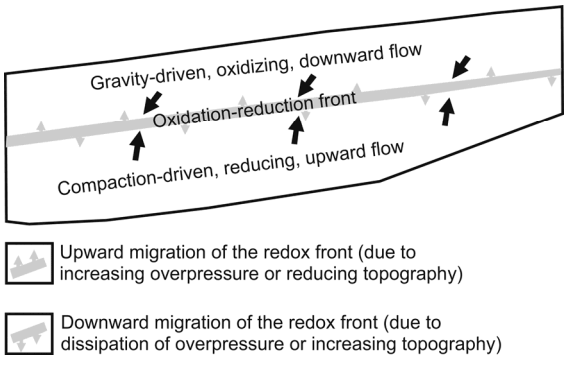

Figure 2 A conceptual model of two competing fluid flow systems in sedimentary basins. A compaction-driven, reducing, upward flow system in the lower part of the basin is overlain by a topography-driven, oxidizing, downward flow system in the upper part of the basin. The location of the interface between the two systems depends on the relative strength of fluid overpressure and topographic relief.

flow across this boundary is zero. The surface temperature is fixed at 20°C, and a heat flux of 1.5 HFU is supplied from the bottom. Equations governing disequilibrium compaction, fluid flow and heat transfer were solved with the finite difference method, as described in (Bethke, 1985; Bethke et al., 1993). The parameters related to fluid and solid properties, including those describing porosity-depth and porosity-permeability relationships, are adopted from the default values of Basin2, which are considered to be representative for general sedimentary basins (Bethke et al., 1993; Chi and Savard, 1998).

The modeling results (Figure 3) indicate that fluid overpressure (the difference between fluid pressure and hydrostatic values; Bethke, 1985), which is closely related to lithologies and sedimentation rate, strongly influences the location or depth of the interface between the compaction-driven upward-flowing system and the gravity-driven downward-flowing system. Sedimentary basins with abundant sandstone (or scarcity of mud components) and slow sedimentation rate, as represented by case 1 (models 1a–1c, Figure 3), are characterized by very low fluid overpressures (i.e., fluid pressures close to hydrostatic values). As a result, a slight topographic relief (50–100 m) on the surface will drive the oxidizing fluids to the lower part of the basin (model 1b, Figure 3) or the entire basin (model 1c, Figure 3). For sedimentary basins with moderate mud contents and sedimentation rates, represented by case 2 (models 2a–2c, Figure 3), fluid overpressures are moderate and the interface between the upward and downward flow systems is located at moderate depths with slight to moderate topographic relief (50–200 m) (models 2b and 2c, Figure 3). Sedimentary basins with high mud contents and rapid sedimentation rates are represented by case 3 (models 3a–3c, Figure 3), where fluid overpressures are strong, and the upward-downward

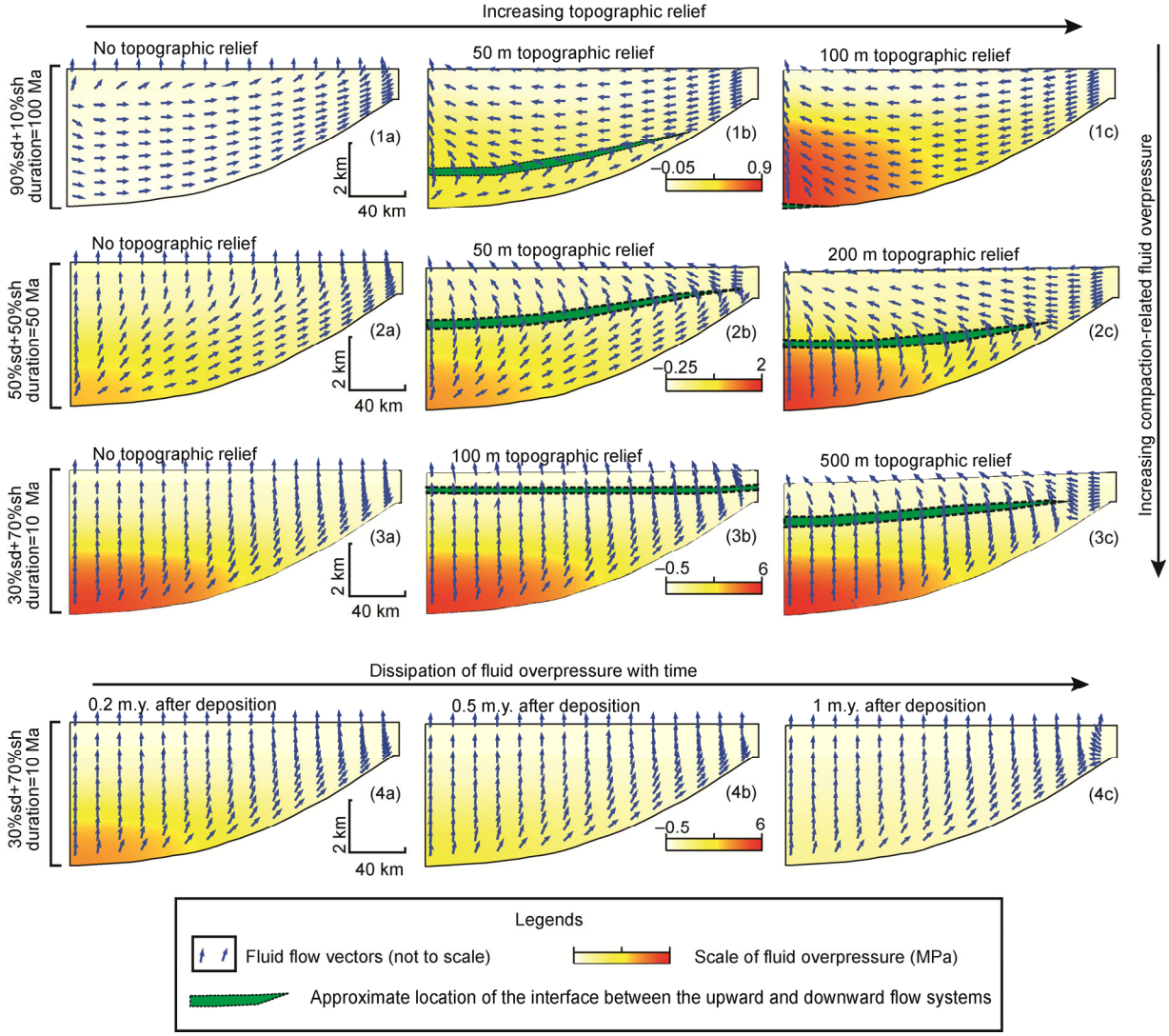

Figure 3 Numerical modeling results showing the distribution of fluid overpressures and fluid flow directions for generic sedimentary basins with different combination of lithologies, duration of sedimentation, and topographic relieves (models 1a–1c to 3a–3c) and dissipation of fluid overpressures after sedimentation (models 4a–4c). Note variation of the location of interface between the compaction driven flow system (upward and rightward) and gravity driven flow system (downward and leftward) depending on fluid pressure regimes in the basin.

flow interface is maintained at shallow depths even with significant topographic relief (500 m, model 3c, Figure 3). Strong fluid overpressures developed in model 3a of Figure 3 significantly diminished within 1 million years after cessation of sedimentation (models 4a–4c, Figure 3).

3 Discussion and conclusions

Uranium deposits within a basin are commonly preferentially concentrated in certain parts or stratigraphic intervals of the basin, which may be related to 1) the distribution of potential uranium source rocks, 2) the distribution of reducing agents required for uranium deposition, and 3) zones or stratigraphic intervals with enhanced permeabilities. Formation of uranium deposits requires a favorable combination of these factors, linked through a hydrodynamic framework. More specifically, uranium mineralization takes place when an oxidizing, uranium-carrying fluid meets reducing agents, where U^{6+} is reduced to U^{4+}. While reducing solid materials (e.g., graphite, organic matter-rich sediments, and ferrous minerals) are more or less "fixed" in space, both U-bearing, oxidizing fluids and reducing fluids can move, and thus the oxidation-reduction front, or the potential location of mineralization, is controlled largely by fluid flow systems.

The simulation results presented in this paper indicate that the interface between the two systems tends to be located at shallow depths when fluid overpressures are high, at moderate depths when fluid overpressures are moderate, and at the base of the basin when fluid overpressures are low. These results are broadly consistent with the observation that uranium mineralization tends to occur at shallow depth when the basin was strongly overpressured (e.g., the Gulf of Mexico basin) (Harrison and Summa, 1991), at moderate depths when the basin was moderately overpressured (e.g., the Chu-Saryssu and Ordos and basins) (Xue et al., 2010, 2011), and at the base of the basin or immediately below the unconformity when the basin was not or only weakly overpressured (e.g., the Athabasca and Kombolgie basins) (Chi et al., 2011, 2013). It may appear that the development of uranium deposits near the unconformity in the Athabasca and Kombolgie basins is controlled by the contrasting environments between the basin (oxidizing) and the basement (reducing), rather than by the hydrodynamic regime of the basins. However, the low fluid overpressure regimes in these basins may have been critical in allowing oxidizing fluids in the basin to circulate down to the base of the basin through topographic relief-driven flow or thermal gradient-related fluid convection (Raffensperger and Garven, 1995a; Cui et al., 2012); such fluid flow would have been difficult if there was significant overpressure development in the basin. Many unconformity-type uranium deposits in the Athabasca and Kombolgie basins are controlled by reactivated basement faults (Figure 1), and this may have been related to the alternating compressional and extensional stress regimes (Cui et al., 2012), or to the fault-valve mechanism (Sibson et al., 1988) operating in the transition zone between the near-hydrostatic pressure regime in the basin and the lithostatic pressure regime in the basement (Chi et al., 2013).

The formation of uranium deposits at the interface between the oxidizing and reducing fluid systems generally requires that the interface be maintained at a given stratigraphic level (or depth) for a prolonged period of time, although gradual downward movement of the interface, accompanied with re-mobilization and re-accumulation of uranium, may also be favorable for mineralization, such as in the formation of roll-front type deposits. The numerical simulation results demonstrate that increasing fluid overpressures tend to push the upward flowing-downward flowing interface upward, whereas increasing topographic relieves tend to lower the interface (Figure 3). Furthermore, dissipation of fluid overpressures (models 4a–4c, Figure 3) tends to lower the interface with time. If the tendency of downward migration of the interface due to gradual dissipation of fluid overpressure is balanced by that of upward migration due to gradual decreasing of topographic relief, the location of the interface will remain at a "fixed" position for a long period of time, favoring uranium mineralization (Figure 2).

In conclusion, although there are many factors influencing the localization of uranium deposits in sedimentary basins, the fluid pressure regime of the basin may have played a critical role. If the basin is filled with sediments with high mud contents and/or sedimentation rate is high, fluid overpressures will be strong; the basin is dominated by upward fluid flow, and the interface between the upper oxidizing and lower reducing fluid systems will be located at shallow depths, favoring uranium mineralization in shallow environments. In contrast, if the basin is filled with sediments with low mud contents and/or sedimentation rate is low, fluid overpressures will be weak; fluids in the basin may be dominated by oxidizing fluids driven by topographic relief or thermal gradient-related convection, favoring uranium mineralization at the base of the basin or in the basement immediately below the unconformity. In situations between these two extremities, uranium mineralization may be localized at moderate depths.

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