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Role of hydrodynamic factors in controlling the formation and location of unconformity-related uranium deposits: insights from reactive-flow modeling

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Abstract The role of hydrodynamic factors in controlling the formation and location of unconformity-related uranium (URU) deposits in sedimentary basins during tectonically quiet periods is investigated. A number of reactive-flow modeling experiments at the deposit scale were carried out by assigning different dip angles and directions to a fault and various permeabilities to hydrostratigraphic units). The results show that the fault dip angle and direction, and permeability of the hydrostratigraphic units govern the convection pattern, temperature distribution, and uranium mineralization. A vertical fault results in uranium mineralization at the bottom of the fault within the basement, while a dipping fault leads to precipitation of uraninite below the unconformity either away from or along the plane of the fault, depending on the fault permeability. A more permeable fault causes uraninite precipitates along the fault plane, whereas a less permeable one gives rise to the precipitation of uraninite away from it. No economic ore mineralization can form when either very low or very high permeabilities are assigned to the sandstone or basement suggesting that these units seem to have an optimal window of permeability for the formation of uranium deposits. Physicochemical parameters also exert an additional control in both the location and grade of URU deposits. These results indicate that the difference in size and grade of different URU deposits may result from variation in fluid flow pattern and physicochemical conditions, caused by the change in

structural features and hydraulic properties of the stratigraphic units involved.

Keywords Unconformity-related uranium · Conceptual models · Reactive-flow modeling · Thermal convection · Sedimentary rocks

Introduction

Unconformity-related uranium (URU) deposits, hosted by Paleoproterozoic sedimentary basins in Canada and Australia, have been formed by large-scale circulation of diagenetic brines that percolated between basal sandstone and underlying basement rocks (Kyser and Cuney 2009; Boiron et al. 2010; Morichon et al. 2010; Richard et al. 2012). In addition to interaction of oxidizing uranium-bearing brines with basement-derived reducing fluids or reducing minerals in the basement, favorable physicochemical conditions (e.g. temperature, pressure, pH, and oxygen fugacity) are required to form URU deposits in the Athabasca Basin (northeastern Canada) over a period of 0.1–1 million years (Raffensperger and Garven 1995b; Richard et al. 2012; Aghbelagh and Yang 2014, and unpublished data).

Numerical modeling has provided valuable insights into the formation of URU deposits. Thermal convection has been proposed, for example, by Raffensperger and Garven (1995a) and Cui et al. (2010) as a driving force for circulating fluids in the sandstone and leaching uranium from the source rocks. Cui et al. (2012a) investigated the effect of tectonic deformation on the fluid flow and uranium ore mineralization, and suggested that thermal convection is a dominant driving mechanism for fluid flow in the absence of tectonic deformation. Others (Boiron et al. 2010; Mercadier et al. 2010; Richard et al. 2010; Cui et al. 2012b; Li et al. 2015) have shown that convective flow

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not only circulates within the sandstone unit, but also percolates into the underlying basement to some depth (a couple of hundred meters below the unconformity). Location of the faults in the basement and fault spacing also influence the flow convection pattern within the basal sandstone (Li et al. 2015).

Understanding the factors controlling the formation and location of URU deposits in sedimentary basins is a major scientific challenge. Hydrodynamic modeling (Cui et al. 2012a) showed that during compressive deformation, fluids migrate upward along faults and form sandstone-hosted deposits (egress type), while during extensional deformation, fluids migrate downward along faults and form basement-hosted deposits (ingress type). In weakly overpressured or zero overpressure regimes, the uranium mineralization occurs near the unconformity interface (Chi et al. 2011, 2013; Chi and Xue 2014; Li et al. 2015). According to these studies, uraninite can precipitate only along fault zones either in the basement or sandstone, but not in other parts of basin. However, recent geological records (e.g. Thomas et al. 2000; Jefferson et al. 2007; Alexandre and Kyser 2012; Alexandre et al. 2012) show that uranium mineralization can also occur away from fault zones. This is confirmed by recent numerical modeling studies (e.g. Aghbelagh and Yang 2014, and unpublished data). Aghbelagh and Yang (2014), assuming methane as a reducing agent in precipitation of uraninite, showed that uraninite could precipitate away from fault zones and below the unconformity, whereas Aghbelagh and Yang (unpublished data) confirmed that ferrous iron, released by destruction of Fe-rich chlorite, could be an efficient reducing agent in the precipitation of uraninite. They also discovered that the permeability of fault zones has a controlling role in the location of precipitated uraninite, and a uranium deposit can form in the basement either away or along fault zones, depending on the fault permeability.

In the Athabasca Basin (Fig. 1), the dip angle and direction of faults differ from one area to another (Finch 1996; Hajnal et al. 2005). For example, in the McArthur River deposit, the major fault is a northeast-trending, southeast dipping, reverse fault (McGill 1999; Derome et al. 2005); whereas in the Dragon Lake area, the major fault is a left-lateral strike-slips structure

(Hoeve and Sibbald 1978), and in the Rabbit Lake deposit, the fault is a thrust type with a low dip angle (Hoeve and Sibbald 1978). Additionally, faults may become more permeable after reactivation (Lorilleux et al. 2003; Dieng et al. 2013), which is a common process in the Athabasca Basin.

The permeability of various hydrostratigraphic units in sedimentary basins is another primary factor controlling the fluid flow dynamics, and consequently the thermal regime (Hitchon 1969a, b; Senger and Fogg 1987; Belitz and Bredehoeft 1988), and it can be altered through different processes. Seismic reactivation may rejuvenate the permeability periodically (Lorilleux et al. 2003; Rutqvist et al. 2013). Active deformation concurrent with hydrothermal activity also regenerates the permeability following mineral precipitation (Lonergan and Wilkinson 2000; Vajdova et al. 2004; Kwon et al. 2005). In addition, geochemical reactions leading to the dissolution or precipitation of minerals can increase or decrease the permeability of the hydrostratigraphic unit (Kuhn et al. 2004; Lai et al. 2012; Nogues et al. 2013).

Previous numerical studies have not yet conducted systematic investigation into the effect of the afore-stated parameters on URU mineralization. This paper therefore aims to fill the gap, in particular addressing the role of a variety of hydrodynamic factors, including the fault dip angle, the fault dip direction and the permeability of various hydrostratigraphic units, in controlling the formation and location of uranium deposits. The previous study by Aghbelagh and Yang (2014) aimed to investigate the effect of a faulted graphite zone in the formation of URU deposits, with consideration of a fixed dip angle, dip direction, and permeability for the fault and the hydrostratigraphic units involved. In the present study, however, these parameters are variable corresponding to different scenarios. It should be noted that the present work only addresses the cases during tectonically quiet periods, without taking into account the control of tectonic deformation.

Recent studies (Pascal et al. 2015; Potter and Wright 2015; Wang et al. 2015) confirm that graphite disaggregates and depletes in proximity to the uranium mineralization (i.e. at Dufferin Lake zone and Phoenix deposit); thus, methane,

Fig. 1 The location of Athabasca and Thelon Basins in Canada (modified from Aben Resources Ltd 2015)

produced from dissolution of graphite, could be a reductant in the formation of URU deposits. Based on these findings and previous studies (Hoeve and Sibbald 1978; Bray et al. 1988; Kyser et al. 1989; Raffensperger and Garven 1995b; Aghbelagh and Yang 2014), the present work assumes a reducing condition is provided via hydrothermal alteration of graphite as follows.

Methane is first produced by the dissolution of graphite (Hoeve and Sibbald, 1978; Kyser et al. 1989; Raffensperger and Garven, 1995b) at temperatures of typical URU ore-forming brines (120–200 °C):

This then acts as a reductant for the reduction of uraninite

and is a valid redox mechanism for precipitation of uraninite in the presence of the graphite mineral, which is considered to be concentrated primarily along fault zones (Kotzer and Kyser 1995; McGill 1999; De Veslud et al. 2009).

Two-dimensional conceptual model

Although the complete geologic cross-section of the Athabasca Basin is not available due to significant erosion, this basin is characterized by the following features: (1) primary mineralization of URU deposits began when the fluid temperature reached about 200 °C (Kotzer and Kyser 1995; Renac et al. 2002; Derome et al. 2005; De Veslud et al. 2009; Richard et al. 2012), depicting that the sedimentary fill must have been 6–7 km thick assuming a typical intracontinental geothermal gradient of 30 °C/km (Fridleifsson et al. 2008); (2) the bedrock consists of Archean and Paleoproterozoic gneisses overlain unconformably by approximately 1,500 m of flat-lying, unmetamorphosed sandstones and conglomerates of the mid-Proterozoic age; (3) the permeable sandstones and conglomerates are covered by relatively less permeable shallow marine sedimentary facies; (4) graphite is primarily concentrated along the fault zones (Kotzer and Kyser 1995; McGill 1999; De Veslud et al. 2009); and (5) the faults occur predominantly in the basement rocks but often extend several meters up into the Athabasca Group (McGill 1999; Derome et al. 2005). These faults were reactivated after filling the basin (Hoeve and Sibbald 1978; Kotzer and Kyser 1995), and have remained active until recent times (Hoeve and Quirt 1984). By considering these features and based on previous studies (Raffensperger and Garven 1995b; Cui et al. 2012a; Aghbelagh and Yang 2014, and unpublished data), a sandwich-like conceptual model is developed which includes four major hydrostratigraphic units (Fig. 2): a vertical fault, a

lower basement layer, an intermediate sandstone layer, and an upper confining layer. The fault, which is rooted in the basement and extends into sandstone, is assumed to straddle the unconformity in the central part of the model, with a dimension of 300 m × 1,200 m.

The unconformity surface, which separates the sandstone from underlying basement, is set up to be positioned 6 km below the surface. This is because previous studies (Kotzer and Kyser 1995; Renac et al. 2002; Derome et al. 2005; De Veslud et al. 2009; Richard et al. 2012) show that primary mineralization of URU deposits began when the fluid temperature reached about 200 °C, depicting that the sedimentary fill must have been 6–7 km thick assuming a typical intracontinental geothermal gradient of 30 °C/km (Fridleifsson et al. 2008).

The basement is composed of Archean to Paleoproterozoic rocks that were metamorphosed during the Trans-Hudson Orogeny (ca. 1800 Ma). The overlying sedimentary basin fill represents the Athabasca Group sedimentary rocks (2 km thick), which mainly comprises quartz-rich sandstone and conglomerate from alluvial, fluvial and upper-shore sedimentary environments, and is covered by a relatively low-permeability layer that represents the shallow marine sedimentary rocks.

The model area is discretized into 80 × 80 quadrilateral integral finite difference (IFD) elements. Various rock properties assigned for each unit (Table 1) are based on the data used in similar numerical modeling studies and published compilations (e.g. Freeze and Cherry 1979; Garven and Freeze 1984; Raffensperger and Garven 1995a, b; Mclellan et al. 2004; Oliver et al. 2006; Cui et al. 2012a; Aghbelagh and Yang 2014, and unpublished data). The fault dip angle and direction, and the permeability shown in Table 1 for different hydrostratigraphic units, will be adjusted in the following numerical case studies.

Fig. 2 Conceptual model used in the simulation of URU deposits. After Cui et al. (2012a) and Aghbelagh and Yang (2014, and unpublished data)

Table 1 Major physical properties of various hydrostratigraphic units (based on previous studies on URU deposits by Raffensperger and Garven 1995b and Cui et al. 2012a)

Property	Confining unit	Sandstone unit	Basement unit	Fault zone
Density [kg/m ³]	2,400	2,500	2,650	2,400
Porosity	0.15	0.2	0.1	0.2
Permeability [m ²]	1×10^{-15}	3×10^{-13a}	3×10^{-16a}	1×10^{-12a}
Heat conductivity [W/(m·°C)]	2.5	3.5	2.5	4
Specific heat capacity [J/(kg·°C)]	803	803	803	803
Pore fluid compressibility [1/Pa]	3.5714×10^{-11}	3.125×10^{-11}	2.0202×10^{-11}	4.2918×10^{-11}
Pore fluid expansivity [1/°C]	8.0×10^{-6}	1.0×10^{-5}	8.0×10^{-6}	1.0×10^{-5}

^a The permeability of sandstone, basement, and fault zone is varied in different scenarios

The upper boundary is assigned a fixed pore pressure of 30 MPa (assuming hydrostatic condition based on previous studies, e.g. Cui 2012; Cui et al. 2012a; Aghbelagh and Yang 2014, and unpublished data). The side and bottom boundaries are assumed to be impermeable to fluid flow. For heat transport, the top and bottom boundaries are assigned temperatures of 90 and 240 °C, respectively, corresponding to a geothermal gradient of 30 °C/km. The side boundaries are assumed to be insulated to heat transport. For aqueous component concentrations and mineral volume fractions at the top and bottom boundaries, a first-type boundary condition is assigned; that is, aqueous component concentrations and mineral volume fractions are assumed to be constant, which are the same as those of their respective units (Tables 2, 3, 4, and 5). For the side boundaries, a second-type boundary condition is employed with the normal gradient of the aqueous component concentrations and mineral volume fractions equal to zero.

For the initial conditions, a hydrostatic pressure gradient of 10 MPa/km and a thermal gradient of 30 °C/km are applied (Cui et al. 2012a; Aghbelagh and Yang 2014, and unpublished data). Initial chemical compositions for each hydrostratigraphic unit are tabulated in Tables 2, 3, 4, 5, 6, 7, 8, and 9. These tables, which summarize the aqueous compositions and volume fractions of minerals for each hydrostratigraphic unit, are based on previous mineralogical studies in the Athabasca basin (e.g. Alexandre et al. 2005) and hydrochemical modeling of URU deposits (e.g. Raffensperger and Garven 1995b; Aghbelagh and Yang 2014, and unpublished data). The basement unit is mainly composed of quartz, muscovite, and K-feldspar with accessory anhydrite, chlorite, hematite, pyrite, and kaolinite (Raffensperger and Garven 1995b; Hoeve and Quirt 1984; Fayek and Kyser 1997; Adlakha et al. 2014). It is assumed that this unit has initially moderately reducing ($\log fO_2 = -46.834$) and acidic (pH = 4.541) conditions. The fault is mainly composed of quartz, muscovite, and graphite, with accessory pyrite, kaolinite, and chlorite (Raffensperger and Garven 1995b). This unit is assumed to be in more reducing ($\log fO_2 = -51.285$) and slightly more acidic (pH = 4.094) conditions compared to the basement unit. The sandstone unit is predominantly composed of quartz and hematite with accessory muscovite, anhydrite,

chlorite, K-feldspar, and kaolinite (Tremblay 1982; Fayek and Kyser 1997; Hiatt et al. 2007; Alexandre et al. 2009; Adlakha et al. 2014). For this unit, oxidizing ($\log fO_2 = -22.825$) and acidic (pH = 5.131) conditions are assigned. The selection of such high oxygen fugacity is based on the fact that the uranium ore-forming fluids must have had high oxygen fugacities to transport appreciable amounts of uranium (Komninou and Sverjensky 1996; Raffensperger and Garven 1995b). The thermodynamic study by Komninou and Sverjensky (1996) showed that $\log fO_2$ must have been above -24 at 200 °C to account for dissolved U(VI) (as uranyl complexes), well above the hematite-magnetite buffer ($\log fO_2 = -39.5$ at 200 °C). It is believed that aqueous uranium is leached from uranium bearing minerals within sandstone unit by basinal brines (Wilson and Kyser, 1987; Kotzer and Kyser 1995; Fayek and Kyser, 1997); therefore, based on previous studies (e.g., Raffensperger and Garven 1995b; Richard et al. 2012), the sandstone unit is assumed to be a uranium source by assigning an initial fluid composition of 1×10^{-4} mol/L (≈ 27 ppm) total uranium. The upper confining unit is a sequence of less permeable marine sandstones, siltstone, and

Table 2 Initial condition of the aqueous phase associated with the basement unit (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Component species	Concentration (mol/L)
AlO ₂ ⁻	0.1
Ca ²⁺	0.853
CH ₄ (aq)	1.0
Cl ⁻	2.7
Fe ²⁺	0.002
H ⁺	2.8774×10^{-5}
H ₂ O	1.0
HCO ₃ ⁻	0.049
K ⁺	0.163
Mg ²⁺	0.365
Na ⁺	0.1
O ₂ (aq)	1.4655×10^{-47}
SiO ₂ (aq)	1.0
SO ₄ ²⁻	5.6×10^{-4}
UO ₂ ²⁺	8.2×10^{-11}

Table 3 Initial mineral volume fractions, possible secondary mineral phases, and their kinetic properties associated with the basement unit (K_{25} kinetic rate constant at 25 °C, E_a activation energy and A_m reactive surface area; based on previous study on URU deposits by Raffensperger and Garven 1995b)

Mineral	Composition	Volume fraction	K_{25} (mol/m ² s)	E_a (kJ/mol)	A_m (cm ² /g)
Primary:					
Anhydrite ^a	CaSO ₄	0.0032	At equilibrium	At equilibrium	At equilibrium
Chlorite	(Mg _{2.5} Fe _{2.5})Al ₂ Si ₃ O ₁₀ (OH) ₈	0.0056	1×10^{-13}	62.76	151.6
Hematite	Fe ₂ O ₃	0.0005	2.514×10^{-15}	66.2	12.87
K-feldspar	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	0.0494	1×10^{-12}	67.83	9.8
Muscovite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	0.3263	1×10^{-13}	62.76	151.6
Pyrite	FeS ₂	0.0001	1×10^{-11}	62.76	12.87
Quartz	SiO ₂	0.5148	1.258×10^{-14}	87.5	9.8
Secondary:					
Albite	NaAlSi ₃ O ₈	0	1×10^{-12}	67.83	9.8
Anorthite	CaAl ₂ Si ₂ O ₈	0	1×10^{-12}	67.83	9.8
Calcite ^a	CaCO ₃	0	At equilibrium	At equilibrium	At equilibrium
Dolomite	CaMg(CO ₃) ₂	0	2.951×10^{-8}	52.2	9.8
Graphite	C	0	1×10^{-12}	67.83	9.8
Halite	NaCl	0	1×10^{-13}	67.83	9.8
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	0	1×10^{-13}	62.76	151.6
Magnesite	MgCO ₃	0	4.571×10^{-10}	23.5	9.8
Magnetite	Fe ₃ O ₄	0	1×10^{-13}	67.83	12.87
Siderite	FeCO ₃	0	1.26×10^{-9}	62.76	9.8
Sylvite	KCl	0	1×10^{-12}	67.83	9.8
Rutherfordine	UO ₂ CO ₃	0	1×10^{-13}	67.83	9.8
Uraninite	UO ₂	0	1×10^{-13}	67.83	9.8
Porosity	—	0.1	—	—	—

^a Anhydrite and calcite were assumed to react with aqueous species at equilibrium because their reaction rate is typically quite rapid (Xu et al. 2004). Dissolution and precipitation of other minerals are kinetically controlled

mudstone (Ramaekers 1990; Kotzer and Kyser 1995). It is mainly composed of calcite, quartz, and kaolinite along with other minerals such as dolomite, muscovite, anhydrite, and hematite (Aghbelagh and Yang 2014, and unpublished data). In comparison with the intermediate sandstone layer, this unit is in a more oxidizing state ($\log f_{O_2} = -14.763$) but with a similar acidic (pH = 5.290) condition.

Numerical modeling scheme

Numerical simulations are performed using the TOUGHREACT code which is applicable to chemically reactive nonisothermal flows of fluids in porous and fractured media (Xu et al. 2004). Physical and chemical process capabilities and solution techniques of the code have been discussed in detail by Xu and Pruess (2001) and Xu et al. (2004). The governing equations and integral finite differences (IFD) space discretization method (Narasimhan and Witherspoon 1976) are presented in electronic supplementary material (ESM).

Aqueous complexation, acid-base, and redox reactions proceed under the local equilibrium assumption. For dissolution and precipitation of the minerals (except for anhydrite and calcite), a kinetic approach is adopted. The selection of the equilibrium approach for anhydrite and calcite is based on the fact that they have a typically quite rapid reaction rate (Xu et al. 2004) when they react with aqueous species. For kinetically controlled mineral dissolution and precipitation, a general form of the rate law (Steefel and Lasaga 1994) is used:

$$r_m = \pm k_m A_m \left[\left(\frac{Q_m}{K_m} \right)^\mu - 1 \right]^n \quad (3)$$

where m is the mineral index, r_m is the dissolution/precipitation rate (positive values indicate dissolution, and negative values correspond to precipitation), A_m is the specific reactive surface area per kg of H₂O, k_m is the rate constant (moles per unit mineral surface area and per unit time) which is temperature dependent, K_m

Table 4 Initial condition of the aqueous phase associated with the confining unit (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Component species	Concentration (mol/L)
AlO_2^-	0.1
Ca^{2+}	2.8×10^{-2}
$\text{CH}_4^0(\text{aq})$	0.1
Cl^-	0.1
Fe^{2+}	7.3×10^{-15}
H^+	5.1286×10^{-6}
H_2O	1.0
HCO_3^-	0.396
K^+	0.022
Mg^{2+}	0.0026
Na^+	0.1
$\text{O}_2(\text{aq})$	1.7258×10^{-15}
$\text{SiO}_2(\text{aq})$	1.0
SO_4^{2-}	1.5×10^{-2}
UO_2^{2+}	1×10^{-5}

Table 6 Initial condition of the aqueous phase associated with the fault zone (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Component species	Concentration (mol/L)
AlO_2^-	0.1
Ca^{2+}	0.1
$\text{CH}_4^0(\text{aq})$	4.0
Cl^-	3
Fe^{2+}	0.0005
H^+	8.0538×10^{-5}
H_2O	1.0
HCO_3^-	1.2×10^{-4}
K^+	0.038
Mg^{2+}	0.88
Na^+	1.0
$\text{O}_2(\text{aq})$	5.188×10^{-52}
$\text{SiO}_2(\text{aq})$	0.5
SO_4^{2-}	0.0010
UO_2^{2+}	1.6×10^{-6}

Table 5 Initial mineral volume fractions, possible secondary mineral phases, and their kinetic properties associated with the confining unit (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Mineral	Composition	Volume fraction	K_{25} (mol/m ² s)	E_a (kJ/mol)	A_m (cm ² /g)
Primary:					
Calcite	CaCO_3	0.4414	At equilibrium	At equilibrium	At equilibrium
Dolomite	$\text{CaMg}(\text{CO}_3)_2$	0.0129	2.951×10^{-8}	52.2	9.8
Hematite	Fe_2O_3	0.0003	2.514×10^{-15}	66.2	12.87
Anhydrite	CaSO_4	0.0024	At equilibrium	At equilibrium	At equilibrium
Kaolinite	$\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$	0.2967	1×10^{-13}	62.76	151.6
Muscovite	$\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$	0.0011	1×10^{-13}	62.76	151.6
Quartz	SiO_2	0.0952	1.258×10^{-14}	87.5	9.8
Secondary:					
Albite	$\text{NaAlSi}_3\text{O}_8$	0	1×10^{-12}	67.83	9.8
Graphite	C	0	1×10^{-12}	67.83	9.8
Chlorite	$(\text{Mg}_{2.5}\text{Fe}_{2.5})\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_8$	0	1×10^{-13}	62.76	151.6
Anorthite	$\text{CaAl}_2\text{Si}_2\text{O}_8$	0	1×10^{-12}	67.83	9.8
K-feldspar	KAlSi_3O_8	0	1×10^{-12}	67.83	9.8
Pyrite	FeS_2	0	1×10^{-11}	62.76	12.87
Halite	NaCl	0	1×10^{-13}	67.83	9.8
Magnesite	MgCO_3	0	4.571×10^{-10}	23.5	9.8
Magnetite	Fe_3O_4	0	1×10^{-13}	67.83	12.87
Siderite	FeCO_3	0	1.26×10^{-9}	62.76	9.8
Sylvite	KCl	0	1×10^{-12}	67.83	9.8
Rutherfordine	UO_2CO_3	0	1×10^{-13}	67.83	9.8
Uraninite	UO_2	0	1×10^{-13}	67.83	9.8
Porosity	—	0.15	—	—	—

Table 7 Initial mineral volume fractions, possible secondary mineral phases, and their kinetic properties associated with the fault zone (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Mineral	Composition	Volume fraction	K_{25} (mol/m ² s)	E_a (kJ/mol)	A_m (cm ² /g)
Primary:					
Graphite	C	0.0848	1×10^{-12}	67.83	9.8
Chlorite	(Mg _{2.5} Fe _{2.5})Al ₂ Si ₃ O ₁₀ (OH) ₈	0.0011	1×10^{-13}	62.76	151.6
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	0.0007	1×10^{-13}	62.76	151.6
Muscovite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	0.4227	1×10^{-13}	62.76	151.6
Pyrite	FeS ₂	0.001	1×10^{-11}	62.76	12.87
Quartz	SiO ₂	0.2897	1.258×10^{-14}	87.5	9.8
Secondary:					
Anhydrite	CaSO ₄	0	At equilibrium	At equilibrium	At equilibrium
Albite	NaAlSi ₃ O ₈	0	1×10^{-12}	67.83	9.8
K-feldspar	KAlSi ₃ O ₈	0	1×10^{-12}	67.83	9.8
Anorthite	CaAl ₂ Si ₂ O ₈	0	1×10^{-12}	67.83	9.8
Calcite	CaCO ₃	0	At equilibrium	At equilibrium	At equilibrium
Dolomite	CaMg(CO ₃) ₂	0	2.951×10^{-8}	52.2	9.8
Hematite	Fe ₂ O ₃	0	2.514×10^{-15}	66.2	12.87
Halite	NaCl	0	1×10^{-13}	67.83	9.8
Magnesite	MgCO ₃	0	4.571×10^{-10}	23.5	9.8
Magnetite	Fe ₃ O ₄	0	1×10^{-13}	67.83	12.87
Siderite	FeCO ₃	0	1.26×10^{-9}	62.76	9.8
Sylvite	KCl	0	1×10^{-12}	67.83	9.8
Rutherfordine	UO ₂ CO ₃	0	1×10^{-13}	67.83	9.8
Uraninite	UO ₂	0	1×10^{-13}	67.83	9.8
Porosity	—	0.2	—	—	—

Table 8 Initial condition of the aqueous phase associated with the sandstone unit (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Component species	Concentration (mol/L)
AlO ₂ ⁻	0.1
Ca ²⁺	1.502
CH ₄ ⁰ (aq)	1.0
Cl ⁻	5
Fe ²⁺	2.6×10^{-11}
H ⁺	7.3961×10^{-6}
H ₂ O	1.0
HCO ₃ ⁻	1×10^{-4}
K ⁺	0.045
Mg ²⁺	0.477
Na ⁺	1.0
O ₂ (aq)	1.4962×10^{-23}
SiO ₂ (aq)	5.0
SO ₄ ⁻²	1.5×10^{-3}
UO ₂ ²⁺	1.0×10^{-4}

is the equilibrium constant for the mineral-water reaction written for the destruction of one mole of mineral m , Q_m is the ion activity product, and the exponents μ and n are two positive numbers normally determined by experiments, and are usually, but not always, taken equal to one (as in the present work).

The reaction rate constant (k_m) is considered to be a function of temperature (Lasaga, 1984; Steefel and Lasaga 1994) using the Arrhenius equation:

$$k_m = k_{25} \exp \left[\frac{-E_a}{R} \left(\frac{1}{T} - \frac{1}{298.5} \right) \right] \quad (4)$$

where E_a is the activation energy, k_{25} is the rate constant at 25 °C, R is the gas constant, and T is the absolute temperature. The geochemical subsystem (see the ESM) includes 14 primary species, 46 secondary species, and 20 minerals. H⁺ is used as a primary species for variation in fluid pH associated with every layer.

Table 9 Initial mineral volume fractions, possible secondary mineral phases, and their kinetic properties associated with the sandstone unit (based on previous study on URU deposits by Raffensperger and Garven 1995b)

Mineral	Composition	Volume fraction	K_{25} (mol/m ² s)	E_a (kJ/mol)	A_m (cm ² /g)
Primary:					
Hematite	Fe ₂ O ₃	0.0114	2.514×10^{-15}	66.2	12.87
Anhydrite	CaSO ₄	0.0058	At equilibrium	At equilibrium	At equilibrium
K-feldspar	KAlSi ₃ O ₈	0.01	1×10^{-12}	67.83	9.8
Chlorite	(Mg _{2.5} Fe _{2.5})Al ₂ Si ₃ O ₁₀ (OH) ₈	0.0003	1×10^{-13}	62.76	151.6
Muscovite	KAl ₂ (AlSi ₃ O ₁₀)(OH) ₂	0.0183	1×10^{-13}	62.76	151.6
Quartz	SiO ₂	0.7542	1.258×10^{-14}	87.5	9.8
Secondary:					
Albite	NaAlSi ₃ O ₈	0	1×10^{-12}	67.83	9.8
Graphite	C	0	1×10^{-12}	67.83	9.8
Kaolinite	Al ₂ Si ₂ O ₅ (OH) ₄	0	1×10^{-13}	62.76	151.6
Anorthite	CaAl ₂ Si ₂ O ₈	0	1×10^{-12}	67.83	9.8
Calcite	CaCO ₃	0	At equilibrium	At equilibrium	At equilibrium
Dolomite	CaMg(CO ₃) ₂	0	2.951×10^{-8}	52.2	9.8
Pyrite	FeS ₂	0	1×10^{-11}	62.76	12.87
Halite	NaCl	0	1×10^{-13}	67.83	9.8
Magnesite	MgCO ₃	0	4.571×10^{-10}	23.5	9.8
Magnetite	Fe ₃ O ₄	0	1×10^{-13}	67.83	12.87
Siderite	FeCO ₃	0	1.26×10^{-9}	62.76	9.8
Sylvite	KCl	0	1×10^{-12}	67.83	9.8
Rutherfordine	UO ₂ CO ₃	0	1×10^{-13}	67.83	9.8
Uraninite	UO ₂	0	1×10^{-13}	67.83	9.8
Porosity	—	0.2	—	—	—

The thermodynamic database in the code is originally based on the equilibrium constants for aqueous species and minerals given in the EQ3/6 V7.2b database (Wolery 1992), containing component species data, reaction stoichiometries and log (K) data entries. The EQ3/6 database is one of the most commonly used thermodynamic databases for geochemical modeling; however, it does not incorporate any uranium aqueous species and uranium minerals. In order to simulate uranium mineralization, a number of uranium aqueous species—e.g. U³⁺, U⁴⁺, UCl³⁺, UCl₂²⁺, UHCO₃³⁺, U(HCO₃)₂²⁺, UO₂⁺, UO₂Cl(aq), UO₂Cl₂[−], UO₂HCO₃(aq), UO₂(HCO₃)₂[−], UO₂(CO₃)₂^{−2}, UO₂Cl⁺, UO₂Cl₂(aq), UO₂CO₃(aq), UO₂HCO₃⁺, UO₂(HCO₃)₂(aq), UO₂HSO₄⁺, and UO₂SO₄(aq)—and uranium minerals (e.g. Uraninite, Rutherfordine) are added to the existing primary thermodynamic database (EQ3/6 V7.2b database; Wolery 1992) in TOUGHREACT. The selection of these complexes is based on previous studies on URU deposits (Kojima et al. 1994; Raffensperger and Garven 1995b) showing that uranyl chloride (e.g. UO₂Cl⁺, UO₂Cl₂(aq)), uranyl carbonate (e.g. UO₂CO₃(aq), UO₂(CO₃)₂^{−2}), and uranyl sulfates—e.g. UO₂SO₄(aq)—are predominant uranium complexes in the Athabasca Basin. Refer to recent publications Aghbelagh

and Yang (2014, and unpublished data) for further details of the modeling approach.

Results

Effect of fault dip angle and direction

Figure 3a shows the fluid flow pattern and flow rate at 500,000 years around the fault zone. The high permeability of the fault zone leads to a maximum flow rate of 9.95×10^{-8} m/s (about 3.1 m/year) within it. This zone acts as a pathway for the downward migration of basinal brine into the basement, and also for the upward migration of basal fluids into the basinal sandstone so as to facilitate the interaction between the sandstone and basement fluids. The fluid flow pattern throughout the model area at 500,000 years is depicted in the Fig. 3b. At the unconformity interface the flow direction is nearly horizontal, except for the intersection of this interface with the fault and for the regions where convection cells attempt to interact with the basement lithology via the downwelling and upwelling flow. Two equally sized fluid convection cells develop within this unit, which is due to the

fluid density variation as a result of the thermal gradient (Kuhn and Gessner 2009). These cells control the transport of the aqueous species within the sandstone unit, and are responsible for leaching the uranium from the sandstone and transporting it down into the basement. The redox condition for precipitation of uraninite is provided through the interaction of uranium-bearing brines with aqueous methane in the system. The downwelling parts of the convection cells allow the basal brines to penetrate into the basement; while the upwelling parts of the convection cells enable the basal fluids to be brought into the sandstone. This confirms the previous findings that basal brines can penetrate into the underlying basement to some depth (a couple of hundred meters below the unconformity; Boiron et al. 2010; Richard et al. 2010; Cui et al. 2012b), and also highlights the importance of the convection cells in transferring the mass between the sandstone and underlying metamorphic basement. The flow rate in the basal sandstone is moderate (about 1.6 m/year), while in the basement it is much less (about 4.7×10^{-4} m/year) due to its very low permeability compared with that of the other units (refer to Table 1). The thickness and permeability of the sandstone, and the thermal gradient are determining parameters for the flow rate within the sandstone (Hanor 1987; Evans and Nunn 1989; Garven 1995; Raffensperger and Garven 1995a, b).

Temperature distribution throughout the model area at 500,000 years is also depicted in Fig. 3b. Fluid convection results in a temperature range of 120–150 °C within the sandstone unit. Temperature is one of the most important factors that affect the uranium mineralization (Kotzer and Kyser 1995; Renac et al. 2002; Derome et al. 2005; De Veslud et al. 2009; Richard et al. 2012; Aghbelagh and Yang 2014, and unpublished data). The degree to which metals become concentrated and transported in ore-forming fluids is highly dependent on temperature (Barnes 1997).

Figure 3c shows the uranium mineralization at 500,000 years. Uraninite precipitates with a maximum volume fraction of 0.0118 in a small area at the bottom of the fault in the basement. The high flow rate along the fault zone allows this zone to act as a structural trap, and causes the uranium mineralization therein. This volume fraction is equivalent to a uranium grade of 0.48 %, comparable with the average grade of the Rabbit Lake uranium deposit (0.45 %; Hoeve and Sibbald 1978; Raffensperger and Garven 1995b), Cluff Lake OP zone (0.425 %), and West Bear uranium deposit (0.44 %; Jefferson et al. 2007) in the Athabasca Basin.

Figure 3d shows the fluid flow pattern and rate at 500,000 years around the fault zone, when the fault dips 45° to the right. The flow rate along the fault is lower than that of the vertical fault scenario (compare part a with part d in Fig. 3). Figure 3e depicts the fluid flow pattern and temperature distribution throughout the model area at 500,000 years. One major and four minor convection cells develop within the

sandstone unit, with the major convection cell located in the left. Temperature within this unit ranges from 120 to 150 °C, similar to that of the previous case. At 500,000 years (Fig. 3f), uraninite precipitates mainly away from the fault (in its footwall) and below the unconformity interface, with a maximum volume fraction of 0.0118. In addition to these major orebodies in the left of the solution domain, a minor orebody also forms beside the fault (in its hanging wall), but with a very small volume fraction (about 0.03).

Aghbelagh and Yang (2014) previously considered a fault that dips 45° to the left; the result is illustrated in Fig. 3g–i for comparison. It can be seen that Fig. 3g–i are nearly a reflection image of Fig. 3d–f. The subtle difference between the two cases is probably due to the difference in numerical approximations of the integral terms in the mass and energy conservation equations (refer to Xu et al. 2004). Again at 500,000 years, major uraninite (Fig. 3i) precipitation occurs away from the fault (also in its footwall) and below the unconformity interface, but now in the right part of the solution domain. The major uranium orebodies also have a maximum volume fraction of 0.0118.

Diagenetic changes during uranium mineralization generate alteration halos associated with uranium deposits. Minerals associated with the alteration halos for some of the URU deposits in the Athabasca Basin are presented in Table 10, showing that the altered minerals differ from one deposit to the other. In spite of the difference, common altered minerals include muscovite, chlorite, hematite, quartz, and pyrite (Raffensperger and Garven 1995b). The simulated alteration minerals when the fault is vertical are presented in Fig. 4. Muscovite (Fig. 4a) shows several episodes of precipitation and dissolution below the unconformity interface and around the precipitated uraninite. Dissolution of muscovite is prevalent in the cover and along the fault zone; precipitation mainly occurs below the unconformity interface and around the uranium deposit. Precipitation of muscovite is a consequence of k-feldspar dissociation (Fig. 4b) below the unconformity interface which increases the concentration of K^+ , $SiO_2(aq)$ and AlO_2^- species in the basement, and leads to precipitation of muscovite through the following reactions.

K-feldspar dissociation:

Precipitation of muscovite:

Hematite (Fig. 4c) precipitation follows a trend more or less similar to muscovite, precipitating below the unconformity interface and around the uranium deposit. Pyrite alteration (Fig. 4d) mainly consists of dissolution below the

Fig. 3 Fluid flow pattern and rate around the fault, fluid flow pattern, temperature distribution, and precipitated uraninite throughout the model area at 500,000 years: **a–c** when the fault is vertical, **d–f** when the fault dips 45° to the right, **g–i** when the fault dips 45° to the left

unconformity interface. Dissolution of this mineral releases Fe^{2+} into solution below the unconformity, and results in precipitation of hematite as follows:

Dissolution of pyrite:

Hematite precipitation:

In addition to hematite, magnetite (Fig. 4e) also precipitates below the unconformity because of pyrite dissolution through the following reaction:

At 500,000 years, chlorite (Fig. 4f) mainly dissolves below the unconformity interface. It is observed from Fig. 4a and f that the alteration zones associated with the uranium deposit are similar to those of the basement-hosted Millennium deposit in the Athabasca Basin, consisting of muscovite rather than the chlorite and muscovite (Cloutier et al. 2009). Kaolinite (Fig. 4g) precipitates along the unconformity and within the fault zone, forming an alteration halo around the

Table 10 Altered minerals associated with URU deposits in the Athabasca Basin

Deposit	Alteration minerals
Cigar Lake	Illite, chlorite, hematite (Bray et al. 1987)
Key Lake	Kaolinite, chlorite, quartz, siderite, calcite (Dahlkamp 1978)
Midwest Lake	Chlorite, sericite, muscovite (Ayres et al. 1983)
Collins Bay	Illite, chlorite, hematite (Tremblay 1982)
Rabbit Lake	Chlorite, dolomite, quartz, tourmaline (Hoeve and Sibbald 1978; Alexandre et al. 2005)
Dawn Lake	Illite, chlorite, hematite (Tremblay 1982; Quirt and Wasyluk 1997)
McClellan Lake	Illite, chlorite, hematite (Bray et al. 1987)
McArthur River (sandstone part)	Quartz, kaolinite, chlorite, dravite (McGill et al. 1993)
McArthur River (basement part)	Illite, chlorite, and dravite, with local apatite and carbonate (McGill et al. 1993)

Fig. 4 Alteration minerals at 500,000 years when the fault is vertical. **a** muscovite, **b** k-feldspar, **c** hematite, **d** pyrite, **e** magnetite, **f** chlorite, **g** kaolinite, and **h** graphite. *Positive values* for the volume fraction refer to precipitation of the mineral and *negative values* refer to dissolution of the mineral

precipitated uraninite as a result of k-feldspar dissociation below the unconformity:

Graphite (Fig. 4h) dissolution, which occurs mainly along the fault zone, releases aqueous methane into solution (refer to reactions 1 and 2). It should be noted that albite and anorthite do not precipitate in the modeled area, which is in agreement with the field observations in the Athabasca Basin (J. Cloutier, University of St Andrews, 2015, personal communication).

Figure 5 shows the alteration minerals at 500,000 years for the case when the fault dips 45° to the right. Muscovite (Fig. 5a) precipitation is pervasive in the basement and surrounding the uranium deposit, as a result of K-feldspar dissociation (Fig. 5b) below the unconformity interface. Hematite

(Fig. 5c) precipitates below the unconformity on both sides of the fault zone, which is due to the dissolution of pyrite (Fig. 5d). In addition to the hematite, some magnetite is also formed below the unconformity as a consequence of pyrite dissolution. Chlorite (Fig. 5f) is mainly subject to dissolution below the unconformity. Kaolinite (Fig. 5g) predominantly precipitates in the cover unit and on the unconformity interface, and graphite (Fig. 5h) dissolves along the fault zone and within the sandstone unit. When the fault dips 45° to the left, the simulated alteration minerals have a pattern (not shown) as a reflection image of Fig. 5.

It should be pointed out that regardless of the fault dip angle and direction, dissolution of the graphite produces aqueous methane (Fig. 6a,e,i) that reduces the oxidized uranium (Fig. 6b,f,j) in the solution and leads to the precipitation of uraninite (Fig. 3c,f,i). In addition, the uranium-bearing brines

Fig. 5 Alteration minerals when the fault dips 45° to the right at 500,000 years: **a** muscovite, **b** k-feldspar, **c** hematite, **d** pyrite, **e** magnetite, **f** chlorite, **g** kaolinite, and **h** graphite. Positive values for the volume fraction refer to precipitation of the mineral and negative values refer to dissolution of the mineral

also have physicochemical conditions that are in favor of the formation of URU deposits, including a pH range of about 4–4.5 (Fig. 6c,g,k), a temperature of about 180–200 °C (Fig. 3b,e,h), and a reduction of the oxygen fugacity (Fig. 6d,h,l), as confirmed in previous studies (Aghbelagh and Yang 2014, and unpublished data).

The study also considered a non-fault scenario (results not shown here). Free convection cells still develop within the sandstone unit in the absence of the fault. However, no significant uraninite precipitation occurs, which is likely due to the lack of effective interaction of the basinal brines with the basement lithology.

Effect of the permeability of the hydrostratigraphic units

Here, the case considered is when the fault dips 45° to the left. Figure 7a shows the fluid flow pattern and flow rate around the

fault zone at 500,000 years when the fault permeability is reduced one order in magnitude (from 1×10^{-12} to 1×10^{-13} m²). Following this permeability reduction, the flow rate drops substantially from 5.53×10^{-8} m/s (1.7 m/year; refer to Fig. 3g) to 7.38×10^{-19} m/s (2.33×10^{-11} m/year). Figure 7b illustrates the fluid flow and temperature distribution throughout the model area. The maximum flow rate 4.37×10^{-8} (1.4 m/year) now occurs within the sandstone unit rather than in the fault zone. This is because the fault is now less permeable than the sandstone unit. One major and three minor convection cells develop within the sandstone unit, with the major cell localized in the right. In comparison with the case when the fault permeability is equal to 1×10^{-12} m², the major convection cell is shifted to the edge of the solution domain (compare Figs. 7b and 3h). Temperature distribution (Fig. 7b) is modified as well, following the change in flow pattern. At 500,000 years, uraninite (Fig. 7c) precipitates below the unconformity interface and

Fig. 6 Concentration of $\text{CH}_4(\text{aq})$, UO_2^{2+} , pH regime, and variation of oxygen fugacity at 500,000 years: **a–d** when the fault is vertical, **e–h** when the fault dips 45° to the right, and **i–l** when the fault dips 45° to the left

Fig. 7 Fluid flow pattern and rate around the fault, fluid flow pattern and temperature distribution, precipitated uraninite, and variation of oxygen fugacity throughout the model area at 500,000 years: **a** subject to a

permeability of 10^{-13} m^2 for the fault, **e** subject to a permeability of 10^{-14} m^2 for the fault, **i** subject to a permeability of 10^{-11} m^2 for the fault, **m** subject to a permeability of 10^{-10} m^2 for the fault

away from the fault zone. In accordance with the major convection cell shift, uraninite now precipitates closer to the right edge as well. The maximum volume fraction of precipitated uraninite (0.00333) is less than that of the case with the fault permeability of $1 \times 10^{-12} \text{ m}^2$, which is likely due to the lower flow rate along the fault zone and the resultant weakened interaction of the oxidized basal brines with the basement lithology. Further reduction of the fault permeability to $1 \times 10^{-14} \text{ m}^2$ leads to a smaller flow rate of $4.31 \times 10^{-19} \text{ m/s}$ ($1.36 \times 10^{-11} \text{ m/year}$) along this zone (Fig. 7e), that now serves as a barrier for mass transport between the sandstone and basement. One major and five minor convection cells develop within the sandstone, with the major cell positioned immediately above the fault zone. The maximum flow rate is 1.1 m/year, again occurring in the sandstone unit but less than that of the previous case. Temperature distribution within this unit (Fig. 7f) is modified, following the further reduction of flow rate. No uraninite (Fig. 7g) precipitates in the model area. This is mainly due to the fact that the less permeable fault zone prevents mass transport from the basal

sandstone into the basement, and the basal brines mainly interact with the basement via the convection cells but at a much lower flow rate (1.1 m/year) than that in the previous case. It should be noted that oxygen fugacity (Fig. 7h) shows no reduction below and around the unconformity interface; moreover, no graphite is dissolved along the fault zone (results not shown here), implying that no methane is provided to reduce the oxidized uranium in the solution. This is also another reason why no uraninite precipitates in the model area.

Aghbelagh and Yang (unpublished data), with consideration of ferrous iron as a reducing agent, conducted a sensitivity analysis to investigate the role of the fault permeability in the precipitation of uraninite. Their results showed that enhancing the fault permeability leads to a higher flow rate along the fault and the development of two equally sized convection cells within the sandstone. In the present work, a similar study is implemented by assigning higher permeabilities (10^{-11} and 10^{-10} m^2) to the fault zone, but considering methane as a reductant. The results show that the flow rates along

Fig. 8 Fluid flow pattern and temperature distribution, precipitated uraninite, and variation of oxygen fugacity throughout the model area at 500,000 years: **a** and **c** subject to a permeability of $3 \times 10^{-14} \text{ m}^2$ for the

sandstone, **b** subject to a permeability of $3 \times 10^{-15} \text{ m}^2$ for the sandstone, **d–f** subject to a permeability of $3 \times 10^{-12} \text{ m}^2$ for the sandstone, **g–i** subject to a permeability of $3 \times 10^{-11} \text{ m}^2$ for the sandstone

Fig. 9 Fluid flow pattern and temperature distribution, precipitated uraninite, and variation of oxygen fugacity throughout the model area at 500,000 years: **a–c** subject to a permeability of $3 \times 10^{-17} \text{ m}^2$ for the basement, **d–f** subject to a permeability of $3 \times 10^{-18} \text{ m}^2$ for the basement

the fault zone are increased to $2.2 \times 10^{-7} \text{ m/s}$ (about 7 m/year) and $7.3 \times 10^{-7} \text{ m/s}$ (23 m/year), respectively (Fig. 7i,m), and two symmetric convection cells develop within the sandstone (Fig. 7j,n). Uranium mineralization occurs along the fault in the basement (Fig. 7k,o). The simulation results are similar to those of Aghbelagh and Yang (unpublished data), implying that location of the precipitated uraninite is not a function of the reducing agent, but is controlled by the hydraulic properties of the fault zone. It should be noted that when the fault permeability is equal to 10^{-10} m^2 , the volume fraction of the precipitated uraninite (Fig. 7o) is slightly higher than that when the permeability is equal to 10^{-11} m^2 (Fig. 7k). It is also

observed from Fig. 7d,l,p that uraninite precipitates in the areas experiencing a reduction of the oxygen fugacity.

The sandstone unit, as a prime medium accommodating basal fluid circulation that leaches uranium from the source rocks, plays an important role in the formation of URU deposits. One order reduction of its permeability (from 3×10^{-13} to $3 \times 10^{-14} \text{ m}^2$), while other properties remain the same, leads to no deposition of uranium in the solution domain. Only a single convection cell (Fig. 8a) develops, and the temperature distribution (Fig. 8a) is modified accordingly. The less permeable sandstone leads to a low flow rate (about $9.5 \times 10^{-4} \text{ m/year}$), thus the basal brines (including aqueous oxidized uranium) cannot

Fig. 10 Fluid flow pattern and temperature distribution, precipitated uraninite, and variation of oxygen fugacity throughout the model area at 500,000 years: **a–c** subject to a permeability of $3 \times 10^{-15} \text{ m}^2$ for the basement, **d–f** subject to a permeability of $3 \times 10^{-14} \text{ m}^2$ for the basement

easily flow within this unit and penetrate downwards to interact with the basement lithology. As a result, the aqueous uranium oxide cannot be efficiently delivered to proper sites for precipitation of uraninite. Additionally, oxygen fugacity (Fig. 8c) is also not reduced in the solution domain. Consequently, no uraninite precipitates in the model area. Similarly, with two orders of reduction in permeability of the sandstone unit (from 3×10^{-13} to 3×10^{-15} m²) a single convection cell (Fig. 8b) develops and the flow rate becomes even smaller (4.7×10^{-4} m/year). Since the permeability of the sandstone unit is now close to that of the cover (1×10^{-15} m²), the convection cell extends to the cover unit. Temperature distribution (Fig. 8b) is similar to that in the previous case, and once again no uraninite precipitates in the modeled area.

One order of magnitude increase in permeability of the sandstone (from 3×10^{-13} to 3×10^{-12} m²) leads to the development of one major and three minor convection cells (Fig. 8d). These cells are confined in the sandstone unit, and the strongest flow occurs within the sandstone rather than in the fault zone (with a maximum rate of 7.2 m/year) since the sandstone is now more permeable. Thermally induced

convective flow results in a temperature (Fig. 8d) range of 120–150 °C within the sandstone. Similar to the case when the sandstone permeability is 3×10^{-13} m², the downwelling and upwelling parts of the convection cells contribute to the interaction between the basal brines and the basal fluids, in addition to the mass transport along the fault zone (compare Figs. 8d and 3h). Uraninite (Fig. 8e) precipitates away from the fault zone and below the unconformity interface where the dominant downwelling flow interacts with the basement lithology. Additionally, it is observed from Fig. 8f that oxygen fugacity is reduced in that area; however, it should be pointed out, that the volume fraction of the precipitated uraninite (maximum 0.000828) is now very small compared with that when the sandstone permeability is 3×10^{-13} m² (compare Figs. 8e and 3i). This volume fraction is equivalent to a uranium ore grade of 0.03 %, which is much lower than the world average uranium grade (0.2 %; Aben Resources Ltd 2015) and is not economical. Further increasing the permeability from 3×10^{-13} to 3×10^{-11} m² leads to the development of two convection cells within the sandstone unit (Fig. 8g). The flow rate (maximum 15 m/year) is even higher than that of the

Fig. 11 Concentration of UO_2^{2+} and $\text{CH}_4(\text{aq})$ at 500,000 years: **a–b** subject to a permeability of 3×10^{-17} m² for the basement, **c–d** subject to a permeability of 3×10^{-18} m² for the basement, **e–f** subject to a permeability of 3×10^{-15} m² for the basement, **g–h** subject to a permeability of 3×10^{-14} m² for the basement

previous case. No uraninite precipitates near the unconformity interface. Uranium mineralization occurs mainly on the boundary between the cover and the sandstone units (Fig. 8h) with a negligible volume fraction (maximum 2.00×10^{-6}) which is due to the reduction of oxygen fugacity in that boundary (Fig. 8i). The flow rate within the sandstone unit is now so high that the circulating flow in the vicinity of the unconformity interface is almost parallel to this interface, reducing the penetration of basinal brines into the basement. This may explain why no uraninite precipitates near the unconformity interface.

The permeability of the metamorphic basement also has an important role in the formation of URU deposits since it influences the interplay between the basal fluids (the fluids circulating within the basement) and the basinal brines (the fluids circulating within sandstone). Figure 9a presents the flow pattern at 500,000 years when the permeability of the basement is decreased one order in magnitude (from 3×10^{-16} to 3×10^{-17} m²), while other parameters remain unchanged. Two major and two minor convection cells now develop within the sandstone, and the flow rate in the basement is reduced. These cells are positioned symmetrically within the sandstone unit. No uraninite precipitates close to the unconformity interface, except for a negligible volume fraction (maximum 2.03×10^{-6}) along the sandstone-cover interface (Fig. 9b) where the oxygen fugacity (Fig. 9c) is reduced. With a two orders of magnitude reduction in its permeability (from 3×10^{-16} to 3×10^{-18} m²), three convection cells develop in the sandstone (Fig. 9d). Once again no significant uranium mineralization occurs (Fig. 9e), and oxygen fugacity distribution has a pattern (Fig. 9f) similar to that shown in Fig. 9c.

For the case when the permeability of the basement is increased one order of magnitude (from 3×10^{-16} to 3×10^{-15} m²), the fluid flow and temperature distribution at 500,000 years are presented in Fig. 10a. Following this permeability enhancement, the basal fluids also circulate in the basement, which is in agreement with previous studies (e.g. Kuhn et al. 2004; Kuhn and Gessner 2006) that thermally driven free convection may occur in the low permeable strata, in addition to the convective flow within the basinal sandstone. Two equally sized convection cells now develop in the sandstone and basement units. Temperature is greatly affected by the horizontal and vertical flow in the basement. Uranium mineralization occurs along the sandstone-cover boundary (Fig. 10b), but with a negligible volume fraction (maximum 1.89×10^{-6}), in which ore-forming fluids experience a reduction in oxygen fugacity (Fig. 10c). Further increasing the basement permeability to 3×10^{-14} m² leads to a more or less similar fluid flow pattern, temperature distribution, oxygen fugacity regime, and uranium mineralization pattern (Fig. 10d–f).

It should be noted that the less permeable basement unit does not allow the basinal brines to easily interact with it. In other words, the aqueous uranium (UO_2^{2+}) assigned to the sandstone (1.0×10^{-4} mol/L) hardly flows into the basement via the convection cells or along the fault (Fig. 11a,c). This results in an extremely low concentration for aqueous uranium in the basement (about 8.9×10^{-40} mol/L) which is not sufficient for precipitation of uraninite (compare Fig. 11a,c with Fig. 6j). Similarly, no methane (Fig. 11b,d) can easily move within the basement, and the concentration of this species remains almost unchanged. The low concentration of aqueous uranium in the basement explains why no uranium precipitates in the vicinity of the unconformity. On the other hand, the more permeable basement allows brines to circulate in both the sandstone and the basement (refer to Fig. 10a,d). This makes the aqueous uranium (Fig. 11e,g) and methane (Fig. 11f,h) evenly distributed within these units. The concentration of aqueous uranium (3.25×10^{-5}) is high enough, but the concentration of methane (0.575 mol/L) is not sufficient for precipitation of uraninite (compare Figs. 11f,h and 6i). This may explain why no uranium deposit can be formed when the basement is too permeable.

Discussion

The numerical results show that the fault dip angle and direction control the basinal fluid convection and temperature distribution within the sandstone unit, which in turn determines the location of precipitated uraninite. When the fault is vertical, uraninite always precipitates at the bottom of the fault zone (Fig. 3c); when the fault is inclined, uraninite precipitates below the unconformity interface either away from or along the fault zone, dependent upon the fault permeability. Regardless of the reducing agent, hydraulically more permeable fault zones tend to lead to precipitation of uraninite in the basement along the fault zones, while less permeable faults induce that uraninite precipitation occurs away from the fault zones and in the footwall (refer to Figs. 3f, i and 7c,k,o). Exploration companies currently take advantage of fault zones as an indicator of URU deposits. This research suggests that the structural and hydraulic analysis of fault zones (i.e. determining their dip angle, dip direction, and hydraulic properties) would also be beneficial in locating the orebodies.

Uraninite tends to precipitate (refer to Fig. 3b,c,e,f,h,i) in the locales where dominant downwelling basinal brines interact with the basement lithology. This study along with the previous studies (e.g. Wilde et al. 1985, 1989; Walshe 1986; Kotzer and Kyser 1995; Komninou and Sverjensky 1996; Kojima et al. 1994; Cuney 2009; Richard et al. 2010, 2012; Aghbelagh and Yang 2014, and unpublished data) also confirms that these locales have physicochemical conditions that are in favor of uranium mineralization, including a pH of 4–

4.5 (Fig. 6c,g,k), a temperature of about 180–200 °C (Fig. 3b,e,h), and a reduction of the oxygen fugacity (Fig. 6d,h,l). These parameters collectively determine whether the aqueous uranium mineralize to form uraninite, or remain in the fluids to be transported elsewhere (Peiffert et al. 1994, 1996; Bali 2012).

The permeability of different hydrostratigraphic units also plays an important role in uranium ore mineralization. When the sandstone is less permeable, a single convection cell (refer to Fig. 8a,b) develops and spreads to the cover unit. However, no uranium mineralization occurs in the solution domain. This is likely due to the very low flow rate within the sandstone that is unfavorable for mass transport between the sandstone and basement. When the sandstone permeability is increased to $3 \times 10^{-12} \text{ m}^2$, four convection cells (refer to Fig. 8d) develop within the sandstone unit, but the precipitated uraninite has a very small volume fraction (not economic) and is located away from the fault zone below the unconformity interface (Fig. 8e). Further increasing the sandstone permeability to $3 \times 10^{-11} \text{ m}^2$ leads to a higher flow rate and the development of two convection cells within the basinal sandstone (Fig. 8g). Again, no economic URU deposit is formed in the basement, and uranium mineralization occurs mainly along the sandstone-cover interface (Fig. 8h) with a negligible volume fraction.

Reducing the permeability of the basement also modifies the fluid flow pattern and temperature distribution (Fig. 9a,d). No uranium mineralization occurs around the unconformity when the basement permeability is reduced. Uraninite precipitation tends to occur along the sandstone-cover (Fig. 9b,e), but has no economic value—the grade of precipitated uraninite is much less than world average grade (0.2). Increasing the permeability of the basement leads to fluid circulation in both the basement and the sandstone unit (Fig. 10a,d); however, once again, uraninite precipitates only in the cover unit with a negligible volume fraction (Fig. 10b,e).

For the cases leading to uranium mineralization along the sandstone-cover interface, the grade of the precipitated uraninite is much less than that of the cases leading to the formation of URU deposits in the basement (compare Figs. 9b and 3i). This is mainly due to the fact that the reduction of oxygen fugacity along the sandstone-cover boundary is several orders less than that in other cases (compare Figs. 9c with 6l). Additionally, the temperature along the sandstone-cover interface is less than that in the basement; therefore, physicochemical parameters not only control the location of the precipitated uraninite, but also affect the grade of orebodies.

Although the Athabasca and Thelon basins (Fig. 1) in Canada have a similar sedimentology and evolutionary record (Miller 1995; Renac et al. 2002), the size and grade of precipitated uraninite are different in these basins. The Athabasca Basin accommodates the world's highest grade URU deposits such as McArthur River and Cigar Lake with an average grade of 22.28 and 15.4 %, respectively (Jefferson et al. 2007). This

basin also hosts some lower grade URU deposits such as McClean Lake (with an average ore grade of 2.78 %), Millennium deposit (2.304 %), and Rabbit Lake mine (0.27 %; Jefferson et al. 2007). In contrast, the Thelon Basin seems like it does not host high grade URU deposits (Renac et al. 2002) with the Boomerang Lake prospect (0.5 %) and the Kiggavik deposit (0.4 %) at the eastern and western margin, respectively (Jefferson et al. 2007). The numerical results indicate that the variation in size and grade of the uranium deposits in these two basins may be due to the difference in fluid flow patterns and physicochemical conditions, which likely result from the change in structural features (e.g., fault dip angle, fault direction, and fault permeability) and hydraulic properties (e.g. permeability) of the stratigraphic units involved.

Conclusion

Reactive flow modeling has been conducted to address the role of hydrodynamic factors in controlling the formation and location of URU deposits. The simulation results show that the fault dip angle and direction, and the permeability of the stratigraphic units control the fluid flow pattern and rate in the fault, the basinal sandstone and the basement. This in turn influences the mass and energy exchange between the sandstone and the basement, and consequently the location of precipitated uraninite. For a vertical fault, uranium mineralization occurs at its bottom in the basement, whereas for a dipping fault, uranium mineralization occurs either away from or along the fault zone, depending on the fault permeability. A more permeable, dipping fault tends to lead to uranium mineralization along this zone, while a less permeable fault likely results in the precipitation of uraninite away from it. A sandstone unit with either a very low or a very high permeability seems to be unfavorable for the formation of an economic URU deposit, and so does a basement unit with such a hydraulic condition, which suggests that both the sandstone and the basement seem to have an optimal window of permeability ($3 \times 10^{-13} \text{ m}^2$ for the sandstone, and $3 \times 10^{-16} \text{ m}^2$ for the basement) in order for an economic deposit to be formed. This study also reveals that the variation in size and grade of URU deposits in sedimentary basins is likely due to the difference in fluid flow patterns and physicochemical conditions caused by the change in structural features (e.g. fault dip angle, fault direction, and fault permeability) and hydraulic properties of the stratigraphic units (e.g. permeability) involved. From an exploration point of view, this research highlights the importance of the fault dip angle and direction, and the permeability of various hydrostratigraphic units in determining the location of the uranium orebodies.

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