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Alteration Mineralogy and Stable Isotope Geochemistry of Paleoproterozoic Basement-Hosted Unconformity-Type Uranium Deposits in the Athabasca Basin, Canada

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Abstract

Unconformity-type uranium deposits are characterized by mineralization developed along the contact between younger sandstone cover and underlying crystalline basement rocks. Mineralization may extend up to 400 m into the underlying basement rocks. Whereas sandstone-hosted unconformity-type deposits have been well studied, deposits hosted primarily in the basement have not. This study examines the deposits at Rabbit Lake, Dawn Lake, and McArthur River, in the Athabasca basin of Canada, which are hosted by the metamorphic Archean and Early Paleoproterozoic rocks forming the basement to younger Late Paleoproterozoic sandstones. Alteration is similar in the three deposits and is characterized by three distinct paragenetic stages: (1) preore alteration involving illitization of plagioclase and amphibole, followed by chloritization of biotite and illite, which formed at ca. 230°C; (2) ore-stage alteration, characterized by uraninite and coarse-grained illite, which formed at ca. 240°C; (3) postore alteration comprising spherulitic dravite, vein chlorite, quartz, calcite, and Fe, Cu, Co, and Pb sulfides, which formed at ca. 135°C. Fluid circulation associated with emplacement of later Mackenzie dikes initiated partial recrystallization of uraninite. A later stage of alteration includes kaolinite and iron hydroxide precipitation formed at much lower temperatures of ca. 50°C.

Stable isotope compositions of the alteration minerals in conjunction with their paragenesis indicate that oxidized basinal fluids ($\delta D = -43$ to $-21‰$, $\delta^{18}O = 3–8‰$) were derived primarily from evolved seawater and leached uranium from the overlying sandstones of the Athabasca Formation and transported it into the basement via infiltration along fracture zones associated with reverse faults. Graphitic units in the basement and preore alteration served as both physical (fractured zones) and chemical (reductants) traps for the uranium mineralization. The basinal fluids were responsible for the preore illite-chlorite, synore uraninite-illite, and the early postore alteration events; this differs from many other sandstone-hosted deposits, where both oxidized basinal and reduced basement-derived fluids were responsible for uranium precipitation.

Introduction

PALEOPROTEROZOIC unconformity-type uranium deposits in Canada account for about 35 percent of the global stock of uranium (McGill et al., 1993). Most of these orebodies occur in the Athabasca basin, where they are hosted primarily in the sandstone of the Athabasca Group immediately above the basement-sandstone unconformity. The mineralogy of the deposits and associated suite of metals is complex and the ore grades are high, typically 5 to 15 percent U_3O_8 (e.g., Fryer and Taylor, 1987; Quirt et al., 1991). Basement rocks below the unconformity host another group of deposits, in which uranium mineralization is mineralogically less complex (Thomas et al., 1998). The spatial association of the basement-hosted mineralization with the more complex unconformity-type uranium mineralization in many deposits suggests that the former is a variant of the sediment-hosted uranium deposits (Thomas et al., 1998). Known basement-hosted deposits occur mostly in Paleoproterozoic basins in Australia and Canada, and their combined tonnage represents a significant proportion of all uranium deposits.

Unlike their sediment-hosted counterparts, for which various models have been proposed (e.g., Little, 1974, Wilson and Kyser, 1987), the basement-hosted deposits have not been well documented and their origin and character are less well understood, in particular the role of the basement in the ore-forming process (e.g., Fayek and Kyser, 1997; Thomas et al., 1998). The Rabbit Lake deposit is a notable exception, being the best-documented basement-hosted deposit to date (Knipping, 1974; Hoeve and Sibbald, 1978; Heine, 1986). Here, Knipping (1974) first defined the controversy about basement-hosted deposits in terms of the origin of the uranium (i.e., sediment- versus basement-derived). Little (1974) and Darnley (1981) suggested a deep (crustal?) origin for the uranium in unconformity-type deposits. According to this model, uranium-transporting fluids originate from the basement and precipitate uranium where they reach the unconformity. The major criticism of this model is that the deep basement fluids are reducing and therefore cannot transport the uranium (Thomas et al., 1998). Hoeve and Sibbald (1978), for the Rabbit Lake deposit, and Wallis et al. (1983), for the McClean Lake deposit, argued that uranium originating in the sandstones was transported by basinal fluids to the unconformity where part of the fluids were reduced in contact with graphitic basement lithological units that served as a reductant for uranium precipitation. Wilson and Kyser (1987) modified this model, based on stable isotope data that indicated the precipitation of uranium was caused by mixing with reduced fluids of basement origin. More recently, Hecht and

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Cuney (2000) and Derome et al. (2003b) proposed a model in which the uranium originated from the basement metamorphic rocks, leached from uranium-bearing minerals such as zircon and monazite by "aggressive" and oxidizing basinal fluids, and was transported to and precipitated at or under the unconformity. A model for the basement-hosted unconformity deposits, supported by stable isotope data, was proposed by Fayek and Kyser (1997), which involved the derivation of uranium from sediment and its transport by oxidizing basinal fluids. In this case, fluids penetrated the basement along a fracture zone, where they were reduced by graphitic lithological units promoting uranium precipitation. In a recent review of the Athabasca unconformity-type deposits, Thomas et al. (1998) support the basinal origin of uranium and the models of Hoeve and Sibbald (1978) and Fayek and Kyser (1997). Derome et al. (2003a) proposed a basement origin for the uranium in the McArthur River deposit, based on a fluid inclusions study showing that fluids with basement chemistry (Ca and Mg rich) could have transported and deposited the uranium. For the Australian basement-hosted uranium deposits, recent fluid inclusion and geochemical studies suggest both a basement origin (basement chemical signature of the fluids; Derome et al., 2003b) and a sediment origin (uranium-carrying fluids with a basin signature and reducing fluids with a basement signature; Polito et al., 2004) of uranium.

The present paper documents the role that basement rocks play in the formation of three major basement-hosted uranium deposits at Rabbit Lake, Dawn Lake, and McArthur River in the eastern part of the Athabasca basin. The character of uranium mineralization and alteration events in the basement, the different fluids involved in the ore-forming process, and the physical and chemical conditions that prevailed during the time of ore formation are used to develop a model for the petrogenesis of the deposits.

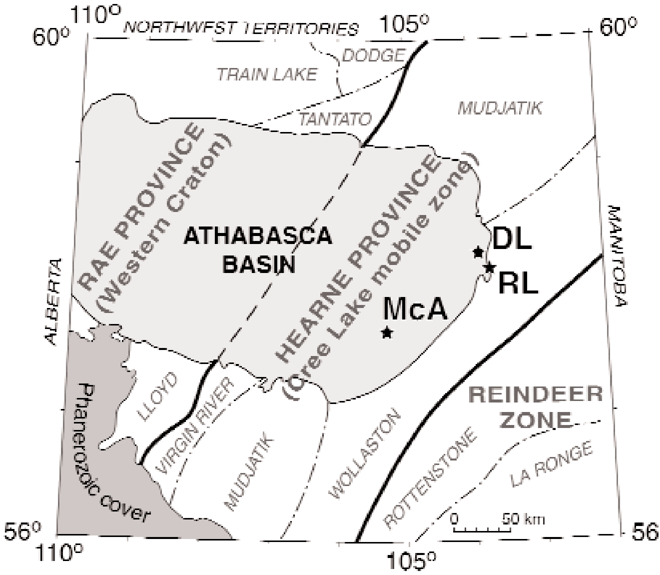

FIG. 1. Simplified geologic map of the Athabasca basin in northern Saskatchewan, Canada, and the major lithotectonic units of its basement. Also shown is the position of the three basement-hosted deposits of this study (DL = Dawn Lake, McA = McArthur River, RL = Rabbit Lake). Modified after Hoeve and Sibbald (1978), Sibbald and Quirt (1987), Thomas et al. (1998), and Stern et al. (2003).

Geologic Setting

The deposits studied are situated on the eastern margin of the Paleoproterozoic Athabasca basin and are aligned along a trend with an orientation of ca. N40°, parallel to the lithostructural trend of the underlying basement rocks (Fig. 1). The deposits at Dawn Lake and Rabbit Lake are situated on the easternmost part of the basin, whereas the deposit at McArthur River is situated in the southeast part of the basin.

The deposits are contained in Archean and Paleoproterozoic rocks of the Wollaston domain, which lies along the eastern margin of the Hearne province (Hoffman, 1988; Fig. 1). The Hearne province consists of Archean rocks of the Hearne craton and the overlying supracrustal rocks that were intensely reworked during formation of the 1.8 to 1.9 Ga Trans-Hudson orogen (Lewry and Sibbald, 1980). The Wollaston domain is part of the larger Cree Lake mobile zone, which is situated between the Western craton and the Reindeer zone and records a Paleoproterozoic thermal and structural reworking (Tran et al., 2003; Fig. 1).

The lower part of the Wollaston domain is composed of various Archean granitic, granodioritic, and tonalitic orthogneisses and subordinate upper amphibolite to granulite facies metamorphic rocks. These are uncomfortably overlain by the metasediments of the Wollaston Group, which is divided into the Lower and Upper subgroups (Delaney et al., 1995; Yeo, 1998; Tran, 2001). The Lower subgroup consists of various rock types with a distinctive spatial distribution. In the western part of the Wollaston domain, graphite-rich rocks and silicate-facies iron formations form the basal part of this subgroup. They are overlain by thick, fine-grained siliciclastic rocks, including turbiditic deposits. The Upper subgroup includes mostly molasses-type sedimentary rocks, ranging from talus (fanglomerate and conglomerate) to arkose to carbonate and evaporite deposits. The nonmarine talus deposits form a wedge-shaped, upward-coarsening succession, which reaches more than 1,000 m in thickness in the east and rapidly wedges out westward (Delaney et al., 1995). Geochronological studies suggest that the Wollaston Group was deposited between ca. 2100 Ma (the age of a feldspar porphyry in the Needle Falls Group: Ansdell et al., 2000) and 1860 Ma (the average emplacement age of the Wathaman batholith: Van Schmus et al., 1987; Meyer et al., 1992). It was then complexly deformed and metamorphosed by the Trans-Hudson orogen, which reached peak metamorphism at ca. 1800 to 1820 Ma (Lewry and Sibbald, 1980; Kyser at al., 2000; Stern et al., 2003). Subsequent rapid uplift began at ca. 1750 Ma, as recorded by K-Ar and ^{40}Ar/^{39}Ar cooling ages (Burwash et al., 1962; Kyser at al., 2000), leading to significant erosion followed by the initiation of the Athabasca basin (Rainer et al., 2003).

The Athabasca basin formed after ca. 1750 Ma (Armstrong and Ramaekers, 1985; Rayner et al., 2003), and probably around 1730 Ma (Kyser at al., 2000), as a series of northeast-southwest–oriented subbasins. Basin fill consists of thick clastic sequences of Paleoproterozoic age resulting from the rapid exhumation of the Trans-Hudson orogenic belt (Ramaekers, 1990). The basin is subdivided into three subbasins, with most of the uranium deposits located in the easternmost, Cree subbasin. These basins contain sequences of polycyclic, mature, fluvial to marine quartz sandstone, collectively referred to as the Athabasca Group. These were deposited in a near-shore, shallow shelf environment (Ramaekers, 1976; Ramaekers and Dunn, 1977; Ramaekers et al., 2001; Jefferson et al., 2003). The basal sequence of the Athabasca Group, the Manitou Falls and Fair Point Formations, consists of coarse- to fine-grained hematite-rich conglomerates and silty sandstones that filled the subbasins and combine to form the full extent of the Athabasca basin (Ramaekers, 1990). In the Manitou Falls Formation, hematite occurs along thin stratigraphic horizons, indicating oxidation of heavy mineral layers. The basal sandstones are overlain by an arkosic succession of less permeable marine sandstones and minor fluorapatite siltstones and phosphatic mudstones (the Lazenby Lake, Wolverine Point, Locker Lake, Otherside and Tuma Lake Formations, respectively), which are in turn overlain by a series of shales (Douglas Formation) and stromatolic dolomites (Carswell Formation). The present thickness of sedimentary rocks in the Athabasca basin is 1 to 2 km; however, temperature estimates from fluid inclusions indicate that the sedimentary sequence may have reached a thickness of 5 to 6 km during the Mesoproterozoic (Pagel et al., 1980).

The Athabasca Group sedimentary rocks and the basement complex are cut by a series of northwesterly trending mafic dikes, related to the Mckenzie dike swarms (Cumming and Kristic, 1992). The dike trend corresponds to tensional directions associated with left-lateral movement along ancient Hudsonian faults (Hoeve and Sibbald, 1978; Sibbald and Quirt, 1987). These intrusions range in size from one meter to several hundred meters wide and have a U-Pb age of 1267 ± 2 Ma (LeCheminant and Heaman, 1989).

Rabbit Lake deposit

The Rabbit Lake deposit, situated on the eastern margin of the Athabasca basin (Fig. 1), was discovered in 1968 in ground follow-up to an airborne radiometric survey (Hoeve and Sibbald, 1976). Subsequent work showed the deposit to be characterized by a conductivity anomaly, low magnetic and gravity signatures, and by geochemically anomalous lake bottom sediment. Stripping began in 1974, following a development drilling program and mining started in 1975. Mining was completed in 1984 with total production of 40 Mlbs at an average grade of 1.55 percent U_3O_8 (Hoeve and Sibbald, 1976).

The deposit is hosted by the Paleoproterozoic Wollaston Group, comprising interlayered meta-arkoses and calc-silicate rocks in the hanging wall (upper gneisses), massive meta-arkose in the core of the ore zone, and variably graphitic plagioclasite, calc-silicate rocks and dolomitic marbles in the footwall (Fig. 2; Wallis, 1971; Knipping, 1974; Chandler, 1978). Subconcordant sheets of anatectic pink granite pegmatite cut the hanging-wall gneisses. The massive meta-arkose is cut by one or more sheets of gray microgranite (Hoeve and Sibbald, 1978). The gneisses exposed in the open pit define an east-northeast–trending southeast-inclined synformal structure with a moderately northeast plunging fold axis (Hoeve and Sibbald, 1976; Ward, 1989). Evidence of faulting is widespread, and two major structures are recognized. The younger structure, the Rabbit Lake fault, underlies the deposit, dips 30° SSE, and downthrows the Athabasca Group sediments to the north at least 75 m against the crystalline basement (Fig. 2). An older north-northeast–trending, subvertical fault bounds the ore zone to the east (Sibbald, 1978).

Host-rock alteration pervades and surrounds the orebody and is typified by Mg chloritization, dolomitization, silicification, tourmalinization, and hematization. This alteration is more complex than that observed in the paleoweathered basement rocks under the Athabasca Group in the downthrown block of the Rabbit Lake fault (Pagel, 1977).

The ore zone has a flattened cylindrical shape plunging moderately northeast and inclined to the west and is about 550 m long, up to 250 m wide, and extends up to 200 m below the present-day surface. A high-grade core is surrounded by a lower grade envelope of mineralization (Hoeve and Sibbald, 1978). Mineralization comprises several generations of uraninite and coffinite, each accompanied by a gangue assemblage that may include euhedral quartz, dolomite, calcite, hematite, chlorite, and sulfides.

Dawn Lake deposit

The Dawn Lake deposit consists of four zones, 11, 11A, 11B, and 14, with the two most important being 11A (Fig. 2) and 11B, which are the focus of this study. Each zone consists of several northeast-southwest–elongated, cigar-shaped orebodies varying between 100 and 190 m long and 20 to 45 m wide. Zones 11A and 11B contain four and three distinct mineralized lenses, respectively. The combined reserves for zones 11A and 11B are 12.94 Mlbs U_3O_8 at an average grade of 1.69 percent U_3O_8. (D. Chan, et al., 2000, Geology and uranium resources of the Dawn Lake deposit: unpublished report of Cameco Corporation).

The Dawn Lake mineralization is hosted by metapelites, calc-silicate rocks, biotite gneiss, and pegmatite of the Wollaston Group (Chan et al., 2000, Fig. 2). Alteration of the basement rocks around the deposit consists mainly of chlorite and illite (Quirt, 1997). Although a gradual drop in the unconformity elevation is apparent, there is no significant vertical displacement of the unconformity in the vicinity of the mineralization, despite the presence of significant shearing in both the sandstone and basement rocks. The main ore-controlling structure is a steeply west dipping strike-slip fault system with an apparent horizontal displacement (Chan et al., 2000). This structure makes the Dawn Lake deposit atypical of basement-hosted deposits in the eastern Athabasca basin, the shape and position of which are normally controlled by major north-northeast or south-southeast high-angle reverse fault systems (Thomas et al., 1998).

McArthur River deposit

The McArthur River deposit, discovered in 1988 and put into production in 1999, includes both sediment- and basement-hosted orebodies. It is the world's largest, high-grade uranium deposit with total combined sediment- and basement-hosted reserves of 416.5 Mlbs U_3O_8 at an average ore grade of 14.36 percent U_3O_8 (Thomas et al., 1998). Four orebodies are present, three of which are situated in the sandstone wedge of the footwall (Fig. 2). The zone 2 orebody, also referred to as Pod 2, is predominantly basement hosted and occurs largely in the footwall of the P2 reverse fault (Fig. 2).

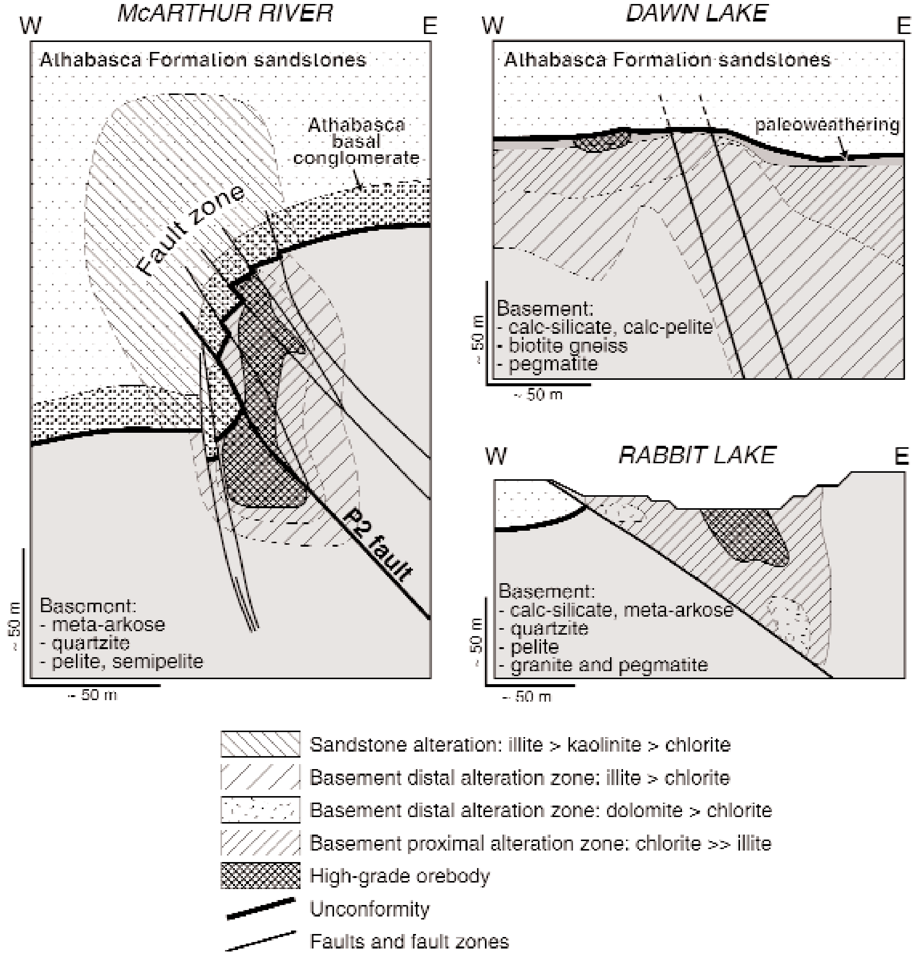

FIG. 2. Simplified schematic representation of lithology, structure, clay mineral alteration halos, and orebodies of the basement-hosted McArthur River (showing the P2 fault zone and the Pod 2 orebody), Dawn Lake (11A ore zone composite section), and Rabbit Lake (main zone) deposits. After Hoeve and Sibbald (1978), McGill et al. (1993), and Anneslay et al. (1997).

It contains half of the uranium reserves in the deposit (McGill et al., 1993).

The Paleoproterozoic Athabasca Group in the McArthur River area consists of members A, C, and D of the Manitou Falls Formation (Ramaekers, 1990). The sandstone section has a total thickness varying from 480 to 560 m above the hanging wall and footwall of the P2 fault, respectively. The sub-Athabasca basement consists of two distinct metasedimentary sequences: a hanging-wall pelitic sequence of cordierite- and graphite-bearing pelitic and psammopelitic gneiss with minor meta-arkosse and calc-silicate gneisses, and a sequence consisting of metaquartzite and silicified meta-arkose and rare pelitic gneisses (McGill et al., 1993).

The major structural feature of the deposit is the P2 northeast-southwest (N 45°) reverse fault with a vertical displacement ranging between 60 and 80 m and dipping 40° to 45° to the southeast (McGill et al., 1993). A series of steeply dipping, east-southeast–west-northwest transcurrent faults (100°–110°) are also present. Alteration minerals in the sandstone are mostly quartz, kaolinite, chlorite, and dravite (McGill et al., 1993), whereas basement alteration includes illite, chlorite, and dravite, with local apatite and carbonate. The unusual characteristics of this mineralization include an abrupt transition from weakly altered basement host rock to intense chlorite alteration and monomineralic high-grade uranium mineralization over distances of less than a meter (McGill et al., 1993). Two uranium-rich whole-rock samples have been dated by the U/Pb method and gave upper intercept discordia ages of 1348 ± 16 and 1521 ± 8 Ma, the older being interpreted as the age of the primary uranium mineralization and the younger as the age of a remobilization event (McGill et al., 1993).

Methodology

More than 700 diamond drill core specimens representing unaltered host rock, distal, and proximal alteration zones, and uranium mineralization were collected from the three deposits. At the McArthur River deposit, basement samples were taken from 15 surface exploration and delineation drill holes generally disposed in a northeast-southwest direction, as well as from eight underground delineation holes from three northwest-southeast sections; sampling does not extend laterally beyond the distal alteration zone (Fig. 2). At the Rabbit Lake deposit, basement samples were taken from 11 open-pit delineation drill holes, mostly disposed in a west-southwest—east-northeast direction and covering the extent of the open pit (Fig. 2). At Dawn Lake, basement samples were taken from 12 surface regional drill holes and five surface delineation holes, with no particular orientation, extending beyond the alteration shown in Figure 2. Clay mineralogy was determined using a Portable Infrared Mineral Analyzer (PIMA) and was used to select about one third of the samples for further petrological and geochemical studies. Polished thin sections were examined using conventional transmitted and reflected light microscopy to determine the mineral paragenesis. Chemical compositions of the minerals were determined by electron microprobe analyses using an ARL-SEMQ instrument with an energy dispersive spectrometer (EDS) at Queen's University, Kingston, Canada, and a Camebax MBX electron microprobe equipped with four wavelength dispersive X-ray spectrometers (WDX) at Carleton University, Ottawa, Canada. The crystal chemistry of chlorite and illite was used to estimate the crystallization temperatures of these minerals. For illite, tetrahedral site occupancy was used (Cathelineau and Izquierdo, 1988), whereas for chlorite, both oxide composition (Cathelineau and Nieva, 1985) and site occupancy (Cathelineau, 1988; Walshe, 1986) were used. The error on the calculated temperatures is typically ±10 percent relative, based on the statistical variation of multiple analyses (between 7 and 33 analyses per phase; Table 1). Similar temperatures and errors were obtained using the clay crystallinity (Arkai, 1991; Kubler and Goy-Eggenberger, 2001); however these are not reported nor used here, as the results are incomplete.

Clay minerals were extracted by ultrasound disintegration and centrifuge separation of clay-size fractions (10–5, 5–2, and 2–1 μm) and analyzed by X-ray diffraction (XRD) using a Siemens X-pert installation at Queen's University. Monomineralic fractions, typically >95 percent pure, were used for stable isotope studies. Oxygen isotope compositions of illite, chlorite, and kaolinite were measured using the BrF_5 method of Clayton and Mayeda (1963). Hydrogen isotope compositions of these minerals were determined using the methods of Kyser and O'Neil (1984). Stable isotope measurements were made using a dual inlet Finigan MAT 252 isotopic ratio mass spectrometer and are reported in the δ notation in units of per mil relative to the standard V-SMOW. Replicate $\delta^{18}O$ analyses were reproducible to ±0.2 per mil, and δD values were reproducible to ±3 per mil. Oxygen isotope fractionation factors used throughout this paper are those proposed by Eslinger and Savin (1973) for water-illite, Land and Dutton (1978) for water-kaolinite, and Yeh and Savin (1976) for water-chlorite. Hydrogen isotope fractionation factors used are those proposed by Yeh (1980) for water-illite, Lambert and Epstein (1980) for water-kaolinite, and Graham et al. (1987) for water-chlorite.

Paragenesis of Alteration Minerals

Although there are some minor differences in alteration minerals among the three deposits studied, the alteration assemblages are significantly similar, enabling one paragenetic sequence to adequately describe all of the deposits studied (Fig. 3).

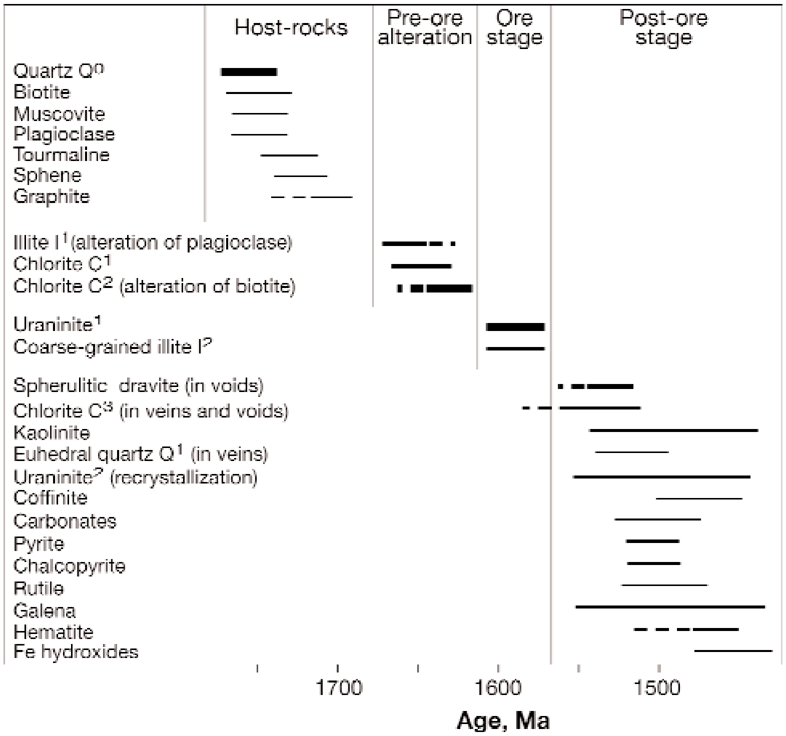

FIG. 3. General mineral paragenesis of the McArthur River, Dawn Lake, and Rabbit Lake deposits. Three main alteration stages are recognized as pre-, syn-, and postore alteration. The thickness of the lines indicates the relative abundance; dashed lines indicate uncertainty in the position. Also shown are the ages of the different events (Alexandre and Kyser, 2003).

TABLE 1. Average Chemical Composition (in wt % and 1s), Average Temperatures of Formation, and Average Structural Formulas of Various Illite and Chlorite Phases in Dawn Lake, Rabbit Lake, and McArthur River Basement-Hosted Deposits

	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	SUM	Temp. (°C)	Average structural formula
Preore alteration illite (I^1)													
Dawn Lake (*n* = 20)	51.6 ± 1.1	<DL	31.0 ± 2.5	<DL	1.4 ± 0.7	<DL	2.5 ± 0.8	0.3 ± 0.3	<DL	8.2 ± 1.1	94.9	225	$K_{1.4}Al_4(Si_{6.7}Al_{0.8}Mg_{0.5}Fe_{0.1})O_{20}OH_4$
McArthur River (*n* = 7)	50.5 ± 1.0	0.1 ± 0.1	31.8 ± 0.6	<DL	2.2 ± 0.5	<DL	2.0 ± 0.4	0.2 ± 0.1	<DL	8.0 ± 0.5	94.7	225	$K_{1.3}Al_4(Si_{6.6}Al_{0.9}Mg_{0.4}Fe_{0.2})O_{20}OH_4$
Preore alteration chlorite													
C^1													
Dawn Lake (*n* = 12)	32.8 ± 3.7	<DL	25.6 ± 3.9	<DL	11.2 ± 7.9	0.1 ± 0.1	17.5 ± 1.7	0.1 ± 0.1	<DL	0.2 ± 0.2	87.5	185	$(Mg_{5.0}Fe_{1.8}Al_{3.9})(Si_{6.2}Al_{1.8})O_{20}(OH)_{16}$
Rabbit Lake (*n* = 33)	31.5 ± 2.6	0.1 ± 0.5	17.2 ± 1.7	<DL	11.5 ± 8.6	0.1 ± 0.1	26.1 ± 6.2	0.1 ± 0.0	<DL	<DL	86.7	235	$(Mg_{7.6}Fe_{1.9}Al_{2.2})(Si_{6.2}Al_{1.8})O_{20}(OH)_{16}$
McArthur River (*n* = 17)	28.3 ± 1.7	0.1 ± 0.1	21.8 ± 0.6	0.1 ± 0.1	17.4 ± 6.0	0.2 ± 0.1	19.0 ± 3.9	<DL	<DL	0.1 ± 0.1	87.0	290	$(Mg_{5.7}Fe_{3.0}Al_{2.9})(Si_{5.7}Al_{2.3})O_{20}(OH)_{16}$
C^2													
Rabbit Lake (*n* = 29)	29.5 ± 2.7	<DL	18.2 ± 1.8	n.a.	15.6 ± 5.5	0.1 ± 0.1	22.3 ± 4.6	<DL	<DL	<DL	85.8	245	$(Mg_{6.5}Fe_{2.5}Al_{2.0})(Si_{5.8}Al_{2.2})O_{20}(OH)_{16}$
Synore illite (I^2)													
Dawn Lake (*n* = 31)	51.0 ± 2.3	0.1 ± 0.2	32.6 ± 2.6	<DL	0.9 ± 0.7	<DL	2.4 ± 1.6	0.2 ± 0.1	0.1 ± 0.1	7.7 ± 1.3	95.0	230	$K_{1.4}Al_4(Si_{6.6}Al_{1.0}Mg_{0.5}Fe_{0.1})O_{20}OH_4$
Rabbit Lake (*n* = 9)	48.1 ± 0.8	<DL	32.9 ± 1.7	<DL	0.7 ± 0.4	<DL	1.4 ± 0.6	0.1 ± 0.1	<DL	9.2 ± 0.5	92.6	250	$K_{1.6}Al_4(Si_{6.5}Al_{1.2}Mg_{0.3}Fe_{0.1})O_{20}OH_4$
McArthur River (*n* = 20)	48.0 ± 0.8	0.1 ± 0.1	33.1 ± 2.3	<DL	1.7 ± 0.9	<DL	2.1 ± 0.7	0.1 ± 0.1	0.1 ± 0.1	8.2 ± 1.3	93.4	240	$K_{1.4}Al_4(Si_{6.4}Al_{1.2}Mg_{0.4}Fe_{0.2})O_{20}OH_4$
Postore vein chlorite (C^3)													
Rabbit Lake (*n* = 15)	35.2 ± 1.6	<DL	14.8 ± 2.7	<DL	3.0 ± 0.5	<DL	33.3 ± 1.3	0.1 ± 0.0	<DL	<DL	86.5	135	$(Mg_{9.3}Fe_{0.5}Al_{1.9})(Si_{6.6}Al_{1.4})O_{20}(OH)_{16}$

Notes: Paragenesis of all phases analyzed is shown in Figure 2; the variation of the calculated temperatures is ca ±10% relative; see text for more discussion; n indicates the number of individual analyses on which the average was calculated; n.a. = non analyzed; <DL = concentration lower than the determination limit

Host rock

The principal minerals in the metasedimentary and igneous host rocks of the deposits are medium-grained (1–5 mm) anhedral quartz, biotite, plagioclase, potassium feldspar, amphibole, muscovite, and dolomite, with accessory tourmaline, sphene, and graphite (Figs. 3, 4A, B). Dolomite is abundant only at the Rabbit Lake deposit. In the ore zone, these host-rock minerals, with the exception of quartz, are extensively altered, obscuring the nature of the protolith, particularly at the McArthur River deposit, where they are totally replaced by chlorite and uraninite.

Preore alteration

The earliest alteration involved illitization (I^1) of feldspar and amphibole, but biotite was unaffected (Figs. 3, 4A). This illitic alteration is preserved in the distal alteration zone, which extends from ~30 to ~100 m from the ore zone; beyond this, the host rocks are relatively to quite fresh. Relatively minor amounts of coarse-grained (up to 30 μm) and irregularly shaped, purple Fe- and green Mg-rich chlorite (C^1) occur in voids created by illitization (I^1). This chloritization (C^1) is particularly evident at Rabbit Lake. It is spatially associated, and thus considered to have been coeval with, alteration of biotite to a second variety of chlorite (C^2) that is typically blue under polarized light (Fig. 4B). With progressive chloritization, chlorite (C^2) was also pseudomorphous after illite. Closer to the orebody, chlorite alteration is more intense, including extensive replacement of illite (I^1) and any other minerals present, except quartz. The spatial distribution reflects an illite-dominated (I^1) distal halo, whereas C^1 and C^2 chlorite defines a proximal halo up to 30 m surrounding the orebody. At McArthur River, I^1 illite is absent in the orebody. Fine-grained hematite is present in minor amounts in the proximal alteration halo.

Main ore stage

Uraninite (U^1; Figs. 3, 4C) is the only uranium mineral to form at this stage. It is typically massive at the McArthur River deposit, where it is rimmed by and closely associated with coarse-grained (30–50 μm), euhedral illite (I^2; Figs. 3, 4D). This illite is not found elsewhere. Generally, and particularly at Dawn Lake, uraninite precipitated in voids and open fractures created by preore alteration, whereas at Rabbit Lake it is finer grained and more disseminated. Uraninite crystals vary from euhedral (Fig. 4C) to subhedral or colloform, particularly at the McArthur River deposit, or have a more irregular shape as at the Dawn Lake and the Rabbit Lake deposits (Fig. 4E). Significantly, uraninite and graphite are never found together, and there is even a zone of a few to a few dozens of meters between them in which neither occurs.

The alteration halo at the McArthur River deposit exhibits a sharp transition from variably illitized host rock to a narrow intense chlorite alteration zone within 10 m of the orebody. In contrast, the Rabbit Lake and Dawn Lake deposits, which have considerably lower uranium grades than McArthur River (0.4–5% compared to ca. 15%), show a more progressive transition between the distal illite-dominated halo and the proximal predominantly chlorite-dominated envelope surrounding the uraninite orebody.

Postore stage

The U^1 uraninite is typically zoned in reflected light (Fig. 4E, F), where zones of lower reflectivity or porous zones indicate partial to complete recrystallization to uraninite U^2 or coffinite (Fig. 3), particularly from the outside of the grains (Fig. 4E). In some samples, microscopic U^2 veins cut through U^1 uraninite (Fig. 4F). Uranium recrystallization was accompanied by the release of radiogenic lead, which has precipitated as microscopic (1–10 μm across) grains of galena in uraninite U^1.

Spherulitic dravite is abundant in the postore stage, particularly at the McArthur River deposit, where it has precipitated in voids created by preore alteration but not filled by uraninite (Fig. 4G). The postore alteration assemblage also includes chlorite (C^3) and rare kaolinite (Fig. 3). The C^3 chlorite is blue or purple (Fe rich) and green (Mg rich) and occurs as thin veinlets or seams less than 50 μm across (Fig. 4H), cutting the preore alteration phases and uraninite U^1. Euhedral quartz veins (Q^1) and dolomite or rare calcite veins locally crosscut this postore assemblage. This latter relationship is particularly well developed at the Rabbit Lake deposit. Variable amounts of drusy quartz veins and veinlets cut the pre- and synore alteration assemblages indicating they postdate the main ore stage. Crosscutting relationships indicate that precipitation of druzy quartz preceded the late sulfide event, which is characterized by various Fe, Cu, and Pb sulfides such as pyrite, chalcopyrite, and galena (Fig. 3). Therefore fluid inclusion data derived from druzy quartz cannot be used to characterize the conditions of uraninite (U^1) precipitation. Pyrite, rutile, and rare chalcopyrite, bornite, pentlandite, cobaltite, and magnetite precipitated late as disseminated, euhedral to subeuhedral grains, up to 100 μm across, which crosscut all earlier minerals (Fig. 3). They were followed by locally abundant fine-grained hematite, whereas iron oxides and hydroxides correspond to the youngest events.

The alteration observed in these three basement-hosted uranium deposits is very similar to that of the basement-hosted Nabarlek deposit in Australia, which also shows preore illite and chlorite alteration, ore-stage uraninite and illite, and postore vein chlorite and sulfides (Ewers et al., 1983; Polito et al., 2004).

Crystal Chemistry of Alteration Clay Minerals

Preore illite (I^1) and synore coarse-grained illite (I^2)

The composition of preore alteration illite (I^1) is similar at the McArthur River and Dawn Lake deposits with 51 wt percent SiO_2, 32 wt percent Al_2O_3, and 8 wt percent K_2O (Table 1). The only notable difference in the composition of I^1 illite between the two deposits is in the FeO content, which is 1.4 ± 0.7 and 2.2 ± 0.8 wt percent, respectively (Table 1, Fig. 5A). Synore, coarse-grained illite (I^2) from the three deposits is more compositionally variable than the preore illite (I^1; Table 1, Fig. 5A), with a range to lower SiO_2 (48–51 wt %) and FeO (1–2 wt %) contents, higher Al_2O_3 (up to 33 wt %), more variable K_2O contents (8–9 wt %), and higher MgO contents (1.4–2.5 wt %). Despite these small differences, the molar proportions expressed as Fe + Mg + Mn/Si + Ti – 3 are similar (Fig. 5A). The average structural formula of I^1 and I^2 is $K_{1.4}\,Al_4\,[Si_{6.6}\,Al_{1.1}\,Mg_{0.5}\,Fe_{0.1}\,O_{20}]\,OH_4$.

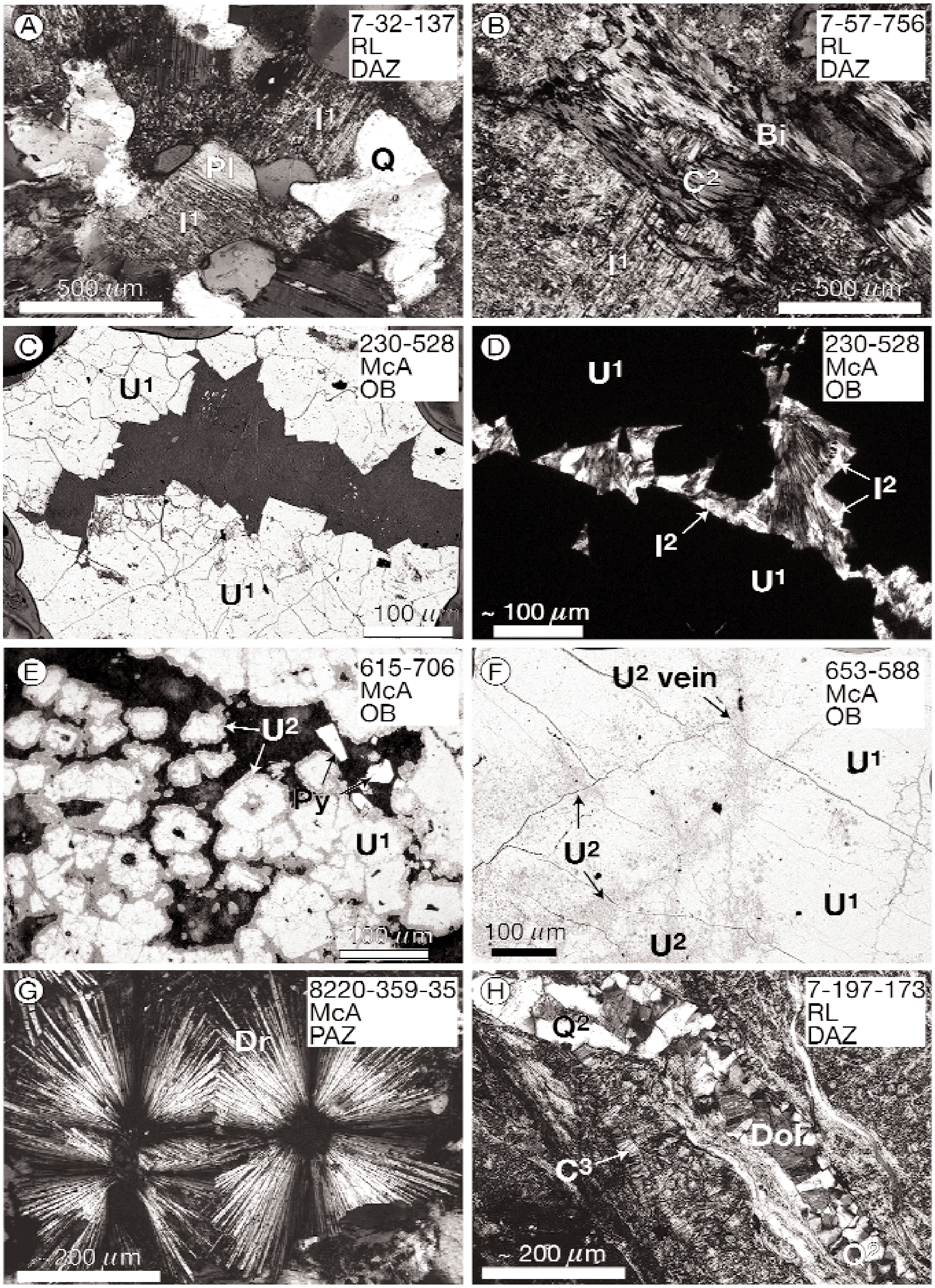

FIG. 4. Photomicrographs of typical mineral assemblages from the preore alteration in the distal and the proximal alteration zones, uraninite mineralization stage in the orebody, and the postore alteration of the proximal alteration zone. A. Preore illitization (I^1) of plagioclase (Pl); biotite is not yet altered (transmitted light). B. Preore chloritization (C^2) of biotite (Bi), all other minerals are illitized (I^1) (transmitted light). C. Scanning electron microscope (SEM) image of euhedral and visually homogeneous uraninite (U^1). D. Synore coarse-grained illite (I^2) accompanying uraninite (U^1) (transmitted light). E. Euhedral uraninite (U^1) with recrystallized rims (U^2) and postore pyrite (reflected light); F. Massive uraninite (U^1) crosscut by diffuse elongated zones of recrystallized uraninite (U^2 vein) (reflected light). G. Postore spherulitic dravite (Dr) (transmitted light). H. Postore vein quartz (Q^1), dolomite (Dol), and chlorite (C^3) (transmitted light). Also shown are the sample number, the deposit, and the location relative to the ore. Abbreviations: DAZ = distal alteration zone, DL = Dawn Lake, McA = McArthur River, OB = orebody, PAZ = proximal alteration zone, RL = Rabbit Lake.

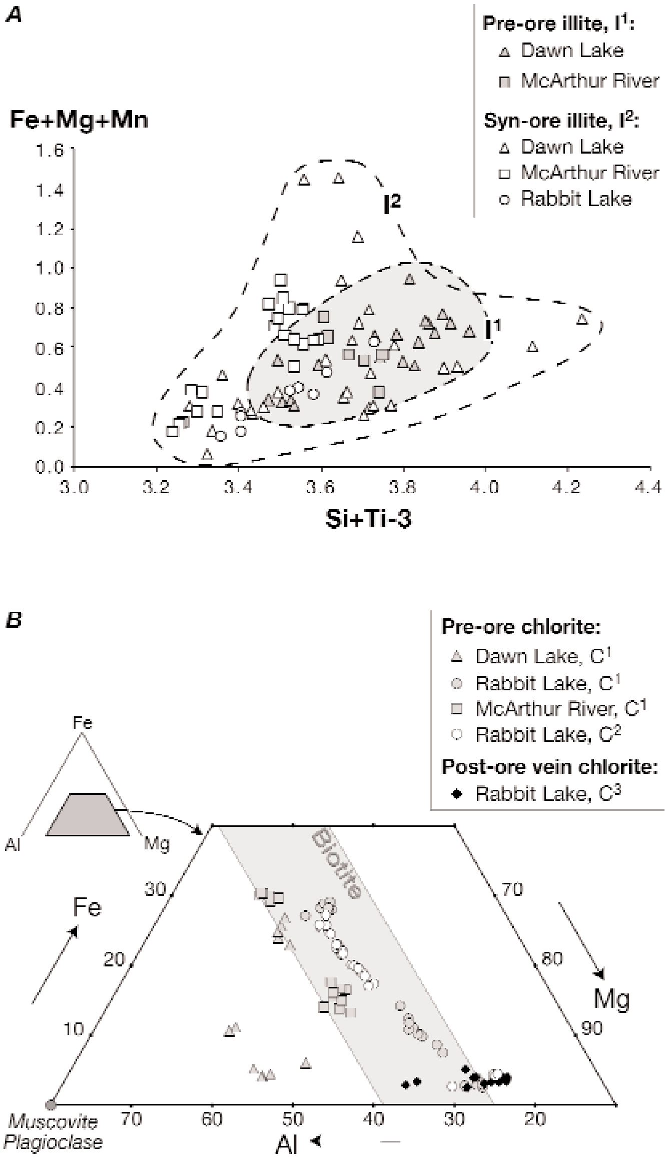

FIG. 5. Chemical compositions of alteration minerals from the McArthur River, Dawn Lake, and Rabbit Lake deposits. A. Pre-ore I^1 and synore I^2 illite compositions, in relationship to molar proportion Fe + Mg + Mn and Si + Ti – 3. The shaded area indicates the range of compositions of the preore illite I^1, which is more restricted than that of the synore illite I^2. B. Preore C^1 and C^2 and postore vein C^3 chlorite compositions, plotted as a function of the molar proportions of Al-Mg-Fe. The shaded area indicates the compositional field of common biotite (Deer et al., 1992). Also shown is the composition of muscovite and plagioclase.

The temperature of formation of these two generations of illite was estimated on the basis of their crystal chemistry using the technique of Cathelineau and Izquierdo (1988). Preore illite (I^1) formed at ca. 225°C, whereas the synore illite (I^2) formed at 230°C at Dawn Lake and 250°C at Rabbit Lake (Table 1).

The water content of the I^1 illite is lower at Rabbit Lake than at McArthur River, with averages of 4.5 and 8 wt percent H_2O, respectively (Table 2). The high water content at McArthur River can be attributed to incorporation of interlamellar water during retrograde alteration of the illite (Graham, 1981; Wilson et al., 1987).

Preore chlorite (C^1 and C^2) and postore vein chlorite (C^3)

Preore chlorites (C^1 and C^2) are chemically variable, with SiO_2 from 28 to 33 wt percent, Al_2O_3 from 17 to 25 wt percent, and Mg/Fe wt percent ratios from 2 to 9 (Fig. 5B, Table 1). When plotted on a molar proportion Al/Mg/Fe triangular diagram (Fig. 5B), most of C^1 and C^2 chlorites fall within the compositional range of typical biotite. Some C^1 chlorite, particularly from Dawn Lake, has higher Al_2O_3 contents (Fig. 5B), which likely indicates that other precursor minerals, such as hornblende, muscovite, and plagioclase have been replaced by this chlorite.

The chemical composition of postore vein C^3 chlorite is less variable, with average SiO_2 contents of 35.2 ± 1.6 wt percent, Al_2O_3 contents of 14.8 ± 2.7 wt percent, and an average Mg/Fe ratio of 11 (Table 1, Fig. 5B).

The formation temperatures of the various generations of chlorite were estimated using their crystal chemistry (Cathelineau and Nieva, 1985; Walshe, 1986; Cathelineau, 1988) and indicates that preore alteration chlorite (C^1) formed at ca. 185°C at Dawn Lake and ca. 290°C at McArthur River. Similar temperatures of ca. 245°C are obtained from the preore C^2 chlorite at Rabbit Lake (Table 1). The crystal chemistry of postore vein chlorite (C^3) indicates that it formed at significantly lower temperatures, between 80° and 180°C (Table 1).

The temperatures of formation of the preore illite (I^1) and chlorite (C^1 and C^2) and synore illite (I^2) indicate that the preore alteration event and the mineralization event occurred between ca. 230° and 250°C. This is similar to, although slightly higher than, the temperature of ca. 200°C that was proposed earlier for the formation of the basement deposits in the Athabasca basin (Fayek and Kyser, 1997) and the temperature of 200°C for the basement-hosted Nabarlek deposit in Australia (Polito et al., 2004). The temperature of the postore alteration, calculated from the composition of the postore vein chlorite C^3, is much lower, averaging 135°C.

The water content of the preore alteration chlorite is ca. 12 wt percent H_2O, which is a typical value for chlorites (Deer et al., 1992).

Isotopic Composition of Alteration Fluids

Stable isotope compositions were determined for preore illite I^1 and C^1 and C^2 chlorites from Rabbit Lake and McArthur River, U^1 uraninite and postore C^3 chlorite from the McArthur River deposit, and kaolinite from McArthur River and Dawn Lake. The results are given in Table 2 and Figure 6.

Preore illite

Measured $\delta^{18}O$ values for preore illite I^1 vary from 7.1 to 10.3 per mil (Table 2) and are slightly lower at Rabbit Lake than at McArthur River, with average values of 7.9 ± 0.8 and 9.7 ± 0.8 per mil, respectively. The measured δD values for illite I^1 vary from –81 to –53 per mil, with most analyses at ca. –55 per mil (Table 2). Using the formation temperature for illite I^1 of 225°C (Table 1), the calculated $\delta^{18}O$ values for the

TABLE 2. Measured Mineral δ^{18}O and δD and Calculated δ^{18}O and δD Values for Fluids in Equilibrium with Alteration Clay Minerals from the Rabbit Lake, Dawn Lake, and McArthur River Deposits

	Mineral		Temperature	Water		
Sample	δ^{18}O	δD	(°C)	(wt %)	δ^{18}O	δD
Preore alteration illite (I^{1})						
Rabbit Lake						
7-197-234	8.7	–56	226	4.5	7.2	–41
7-197-235	8.1	–57	226	4.4	6.6	–42
7-197-244	7.1	–57	226	4.5	5.6	–42
McArthur River						
677-13.5	10.3	–81	225	7.8	6.0	–61
593-65.5	9.1	–53	225	8.3	4.3	–38
Preore alteration chlorite (C^{1}, C^{2})						
Rabit Lake						
7-57-443	3.1	–73	234	11.8	2.7	–41
7-57-443	4.4	–72	234	14.7	4.0	–40
7-57-470	5.1	–62	234	15.2	4.7	–30
7-57-470	4.4	–61	234	12.5	4.0	–29
7-57-470	4.8	–60	234	11.8	4.4	–28
7-32-908	4.6	–61	234	14.1	4.2	–29
7-32-908	4.4	–61	234	14.4	4.0	–29
7-124-150	4.0	–61	234	13.8	3.6	–29
7-32-647	3.8	–61	234	12.1	3.4	–29
7-32-670	4.2	–65	234	11.8	3.8	–33
7-32-893	4.4	–53	234	12.1	4.0	–21
7-32-923	4.3	–57	234	10.3	3.9	–25
7-32-949	3.5	–59	234	11.5	3.1	–27
7-32-981	4.0	–59	234	12.1	3.6	–27
7-32-1017	5.3	–55	234	11.2	4.9	–23
7-57-139	7.5	–86	234	11.2	7.1	–54
7-57-511	4.4	–59	234	12.2	4.0	–27
7-124-150	5.3	–59	234	12.0	4.9	–27
7-125-151	7.2	–102	234	13.2	6.8	–70
7-125-229	5.2	–64	234	12.3	4.8	–32
McArthur River						
323-56.3	9.1	–133	289	12.2	8.7	–101
250-530	10.4	–75	289	9.5	10.0	–43
677-58.5	10.1	–134	289	12.8	9.7	–102
271-98.5	11.8	–115	289	8.4	11.4	–83
329-497.5	10.6	–61	289	9.6	10.1	–31
Dawn Lake						
11A-112-162	6.4	–58	183	11.5	6.0	–27
11A-112-162	5.6	–58	183	12.9	5.2	–27
Uraninite (U^{1})						
McArthur River						
353-50.2	7.8		225		19.2	
653-58.8	–11.0		225		0.4	
387-91.5	–2.3		225		9.1	
615-71.4	–16.9		225		–5.5	
653-55	–13.6		225		–2.2	
Postore vein chlorite (C^{3})						
McArthur River						
8220-387-55	6.2	–62	143	11.4	5.8	–31
8220-387-55	6.4	–94	143	12.0	6.0	–63
8220-267-52	6.4	–111	143	14.3	6.0	–80
Postore kaolinite						
McArthur River						
329-493	10.5	–79	50	10.7	–11.5	–57
329-493	11.3	–81	50	10.4	–10.7	–59
615-70.6	8.1	–151	50	14.0	–13.9	–129
Dawn Lake						
11B-605-103	11.5	–63	50	11.9	1.2	–53

Notes: The temperatures used to calculate the fluid values are derived from the crystal chemistry of the clay minerals; the variation of the individual O and H analyses are ±0.2 and ±3 per mil; see the text for more details

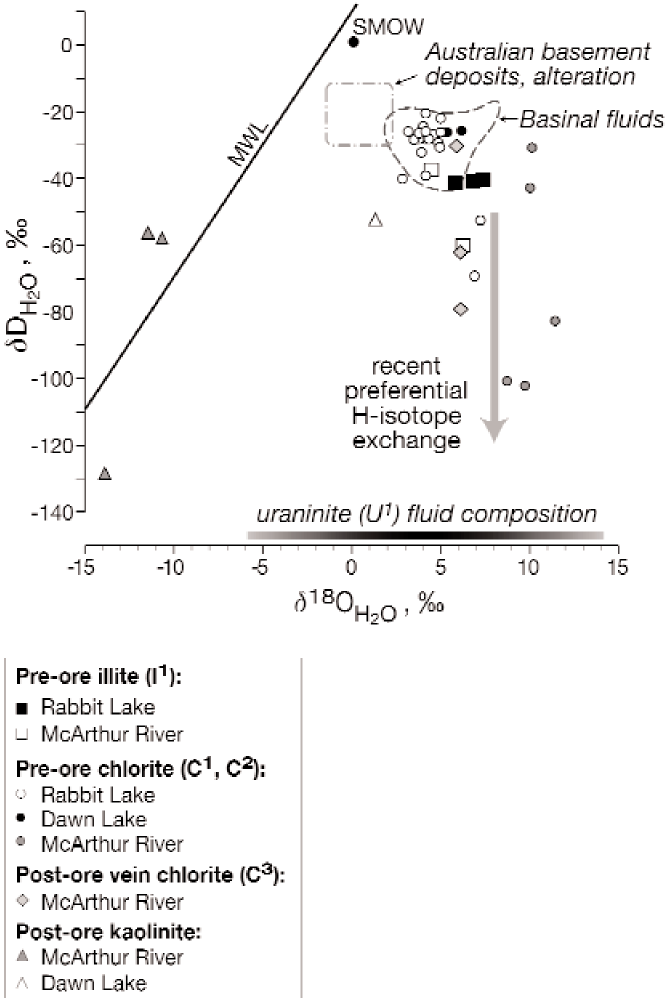

FIG. 6. Calculated δD and $\delta^{18}O$ values of fluids in equilibrium with clay minerals from various alteration stages from the McArthur River, Dawn Lake, and Rabbit Lake basement-hosted deposits. The temperatures used for the calculation of the fluid values are those obtained from the crystal chemistry of the corresponding clay minerals (Table 2). The field corresponding to basin fluids from the Athabasca basin is indicated as are the effects of recent preferential hydrogen isotope exchange (Wilson and Kyser, 1987; Kotzer and Kyser, 1995). Also shown are the meteoric water line (MWL) and the isotopic composition of standard modern ocean water (SMOW).

fluid in equilibrium with this illite range from 4.3 to 7.2 per mil, with an average of 6.0 ± 1.1 per mil, and the δD fluid values range from –61 to –42 per mil, with most near –41 per mil (Table 2). These values are very similar to but slightly heavier than those for fluids that produced the illite alteration in the gneissic host rock of the Key Lake basement-hosted uranium deposit in the southwest Athabasca basin (Wilson and Kyser, 1987), where $\delta^{18}O_{FLUID}$ ranged from –1 to 6 per mil, and δD_{FLUID} was from –55 to –30 per mil.

Preore chlorite

Measured $\delta^{18}O$ values for preore chlorite C^1 and C^2 vary from 3.1 to 11.8 per mil (Table 2) and are higher at McArthur River than at Rabbit Lake and Dawn Lake, with average values of 10.4 ± 1.0 per mil (McArthur River) and 4.8 ± 1.1 per mil (Dawn Lake and Rabbit Lake). The measured δD values for chlorite C^1 and C^2 vary from –134 to –53 per mil, with most near –63 per mil (Table 2). The $\delta^{18}O$ value of the fluid in equilibrium with the preore chlorite, calculated using formation temperatures from 180° to 290°C (Table 1), vary from 2.7 to 11.4 per mil (Table 2) and are higher at McArthur River than at Rabbit Lake and Dawn Lake, with averages of 10 ± 1 (McArthur River) and 4.4 ± 1.1 per mil (Dawn Lake and Rabbit Lake). The calculated δD fluid values range from –102 to –21 per mil (Table 2), with most near –28 per mil. The $\delta^{18}O$ fluid values are similar to those of fluids that produced chlorite alteration in the Key Lake basement-hosted deposit (Wilson and Kyser, 1987), whereas the δD fluid values are slightly lower than those at Key Lake. The unusually low δD fluid values for a few of the analyzed chlorites, down to ca. –100 per mil (Table 2, Fig. 6), are interpreted to result from hydrogen isotope exchange with relatively modern meteoric water having low δD values, as has been observed at the Key Lake deposit (Wilson and Kyser, 1987).

Uraninite

Measured $\delta^{18}O$ values for uraninite U^1 from McArthur River range from –16.9 to 7.8 per mil (Table 2). Assuming a formation temperature similar to that of the preore alteration (i.e., ca. 225°C, Table 1) and similar to what has been estimated for other deposits in the eastern Athabasca basin (Wilson and Kyser, 1987) and in Australia (Polito et al., 2004), the calculated $\delta^{18}O$ values of a fluid in equilibrium with the uraninite range from –5.5 to 19.2 per mil (Table 2). This large variation and the negative values are interpreted to result from variable degrees of coffinitization and recrystallization of uraninite by reduced fluids in the early postore stages producing higher $\delta^{18}O$ values and its subsequent alteration by much later supergene fluids having low $\delta^{18}O$ values (Kotzer and Kyser, 1993). Using the measured and the theoretical oxygen yields and the $\delta^{18}O$ values of the most chemically pristine uraninites (Kotzer and Kyser, 1993), we estimate that the $\delta^{18}O$ values for the fluid in equilibrium with the initial U^1 uraninite were ca. 3 per mil, marginally lower than that of the preore alteration fluids.

Postore chlorite

The $\delta^{18}O$ values for postore C^3 vein chlorite vary from 6.2 to 6.4 per mil (Table 2), averaging 6.3 per mil. The measured δD values range between –111 and –62 per mil (Table 2), with an average of –90 per mil. The $\delta^{18}O$ and δD values for the fluid in equilibrium with this chlorite at 135°C vary from 5.8 to 8 and from –80 to –31 per mil, respectively (Table 2). Whereas the $\delta^{18}O$ fluid values of C^3 chlorite are quite similar to that of the preore C^1 and C^2 chlorite, the δD fluid values are lower (Fig. 6).

Postore kaolinite

Measured $\delta^{18}O$ values for postore kaolinite vary from 8.1 to 11.5 per mil, with an average of 11 per mil, and the measured δD values range between –151 and –63 per mil (Table 2), with an average of –93 per mil. The formation temperature of this late kaolinite based on its poor crystallinity is ca. 50°C, the same as kaolinite in equilibrium with late fluids in the Athabasca basin (Kotzer and Kyser, 1995). The calculated $\delta^{18}O$ fluid values range from –13.9

per mil at Dawn Lake to 1.2 per mil at McArthur River (Table 2), and the δD fluid values vary from –129 to –53 per mil (Table 2). These values are situated near the modern meteoric water line (Fig. 6).

Discussion

Origin of the fluids responsible for alteration and mineralization

The calculated $\delta^{18}O$ value of the fluids that produced the preore illite (I^1) in all of the deposits varies from 4.9 to 7.2 per mil (Table 2), which overlaps with that of the fluids that produced the preore chlorite (C^1 and C^2) at Rabbit Lake and Dawn Lake but is several per mil lower than the values for the fluids that produced the preore chlorite at McArthur River (Table 2). The fluids responsible for uraninite (U^1) mineralization had an estimated $\delta^{18}O$ value of ca. 3 per mil. These $\delta^{18}O$ values are similar to those reported for mineralizing fluids in the sandstone-hosted deposits in the eastern Athabasca basin ($\delta^{18}O$ from 1–9‰: Wilson and Kyser, 1987; Kotzer and Kyser, 1995), which may indicate a basin origin for the fluids that produced alteration and mineralization in the three basement-hosted deposits studied here.

A more definitive indication for the origin of the fluids responsible for alteration and mineralization is given by their hydrogen isotope composition because it will not be buffered by interaction with the crystalline basement host rock, the amount of water and therefore of hydrogen in the basement being low. The calculated δD values of fluids in equilibrium with preore illite (I^1) and chlorite (C^1 and C^2) range from –100 to –20 per mil, although most of them are ca. –28 per mil (Table 2, Fig. 6). The lower δD fluid values, ranging between –100 and –50, correspond to interaction with relatively recent meteoric water having low δD (Fig. 6), as has been documented for the sandstone-hosted unconformity-type uranium deposits (Wilson and Kyser, 1987; Kotzer and Kyser, 1995). On the other hand, the higher δD fluid values, ranging from –40 to –20 (Fig. 6), most likely correspond to the fluid responsible for preore alteration (basinal fluids in Fig. 6). They are also similar to the values determined for the Australian Nabarlek uranium deposit, which ranged from –35 to –15 per mil, and are also interpreted to represent basinal brines (Polito et al., 2004). The δD values are lower than and distinct from basement-derived fluids associated with some sandstone-hosted deposits, which have δD values higher than –15 per mil (Wilson et al, 1987; Bray et al., 1988; Kotzer and Kyser, 1995). Thus, the isotopic compositions of the preore alteration minerals are consistent with derivation from basin-derived brines, similar to what has been proposed previously for pre- and synore alteration in basement-hosted deposits in the Athabasca basin (e.g., Fayek and Kyser, 1997) or in Australia (Polito et al., 2004). These diagenetically evolved fluids originated from mixing between evaporated seawater and low-latitude meteoric waters (Kotzer and Kyser, 1995).

Model reactions during alteration and mineralization

Petrographic study reveals the presence of three distinct stages of alteration that are common to all three deposits: a preore stage with illite I^1 and chlorite C^1 and C^2, a synore stage with uraninite U^1 and illite I^2, and a postore stage with vein chlorite C^3, quartz, and calcite, disseminated sulfides, and late hematite and kaolinite. Chlorite was not contemporaneous with uraninite precipitation as suggested by previous studies (e.g., Hoeve and Sibbald, 1978).

The first reaction between the basinal fluids and the basement rocks was illitization of plagioclase and hornblende. In this case, the fluids needed to contain only potassium, as iron and magnesium required for the illitization of plagioclase would have been made available from the hornblende:

Hornblende (theoretical) → Illite (I^1, as probed), (1)

$$1.3(Na_{0.5}K_{0.2})Ca_2(Mg_{1.6}Fe_{1.6}Al_{1.5})(Al_2Si_6)O_{22}(OH)_2 + 1.1K^+ + 11.8H^+ \rightarrow K_{1.4}Al_{3.4}Mg_{0.5}Fe_{0.2}(Si_{6.8}Al_{1.2}O_{20})(OH)_4 + 0.7Na^+ + 2.6Ca^{2+} + 1.9Fe^{2+} + 1.6Mg^{2+} + SiO_2 + 5.2H_2O$$

and

Plagioclase (theoretical) → Illite (I^1, as probed), (2)

$$4.6NaAlSi_3O_8 + 1.1H_2O + 1.4K^+ + 0.5Mg^{2+} + 0.2Fe^{2+} + 1.8H^+ \rightarrow K_{1.4}Al_{3.4}Mg_{0.5}Fe_{0.2}(Si_{6.8}Al_{1.2}O_{20})(OH)_4 + 7SiO_2 + 4.6Na^+.$$

Two other reactions that occured during this alteration stage were the illitization of muscovite and potassium feldspar, which would have released silica to the fluid.

During the second alteration phase, chlorite replaced biotite according to the reaction:

Biotite (theoretical) → Chlorite (C^2, as probed) (3)

$$1.5K_{1.7}Mg_4Fe_{1.5}(Si_{5.5}Al_3O_{20})(OH)_4 + H^+ + 4.5H_2O + Mg^{2+} \rightarrow Mg_7Fe_2Al_{2.6}(Si_6Al_{1.9})O_{20}(OH)_{16} + 2.6K^+ + 2.3SiO_2 + 0.2Fe^{2+}.$$

Only water, magnesium, and hydrogen were needed for this reaction to proceed, liberating potassium, silica, and some iron, which would have been added to the fluid. Minor chloritization of hornblende also occured at this stage. These two reactions were followed by the chloritization of illite in the ore zone:

Illite (I^1, as probed) → Chlorite (C^2, as probed), (4)

$$K_{1.4}Al_{3.4}Mg_{0.5}Fe_{0.2}(Si_{6.8}Al_{1.2}O_{20})(OH)_4 + 6.5Mg^{2+} + 1.8Fe^{2+} + 13.6H_2O \rightarrow Mg_7Fe_2Al_{2.6}(Si_6Al_{1.9})O_{20}(OH)_{16} + 0.8SiO_2 + 1.4K^+ + 15.2H^+.$$

This reaction consumes significant amounts of magnesium, iron, and water, and therefore likely depended on the preceding reactions to provide these elements. The silica that was released remained in solution until the fluid entered a quartz-rich environment such as the sandstone, where silicification would have occurred because of silica oversaturation.

The ore-forming event was characterized by the precipitation of uraninite from the basinal fluid and by the formation of euhedral illite I^2. The uranium carried by the basinal fluids from the sandstones (e.g., Kotzer and Kyser, 1995) precipitated in the reducing environment of the basement where fluids encountered graphitic lithologic units and the Fe-rich chlorite alteration zone. The corresponding precipitation reactions would have been:

$$U^{6+} + 3H_2O + \tfrac{1}{2}C \rightarrow UO_2 + \tfrac{1}{2}CO_2 + 6H^+, \qquad (5)$$

and

$$U^{6+} + 5H_2O + 2Fe^{2+} \rightarrow UO_2 + Fe_2O_3 + 10H^+. \qquad (6)$$

The precipitation of the synore illite requires potassium, magnesium, and iron that would have been made available by replacement of biotite and hornblende by chlorite.

Conceptual model of volume changes related to preore alteration

Based on these reactions, variations in pore volume as well as the chemical evolution of the rock and the fluids during alteration can be examined. Volume changes can be estimated from the molar volume of the minerals involved in the reactions and the results are presented in Figure 7. For example, if the host rock contains 30 percent plagioclase (molar vol of 100.3 Å^3) and half of this plagioclase is altered to illite (molar vol of 283 Å^3), the resulting rock will contain 9 vol percent illite and will have a void space of 6 vol percent, based on the molar proportions in reaction 2.

Illitization of plagioclase and hornblende thus resulted in the creation of void space in the basement rocks in the distal

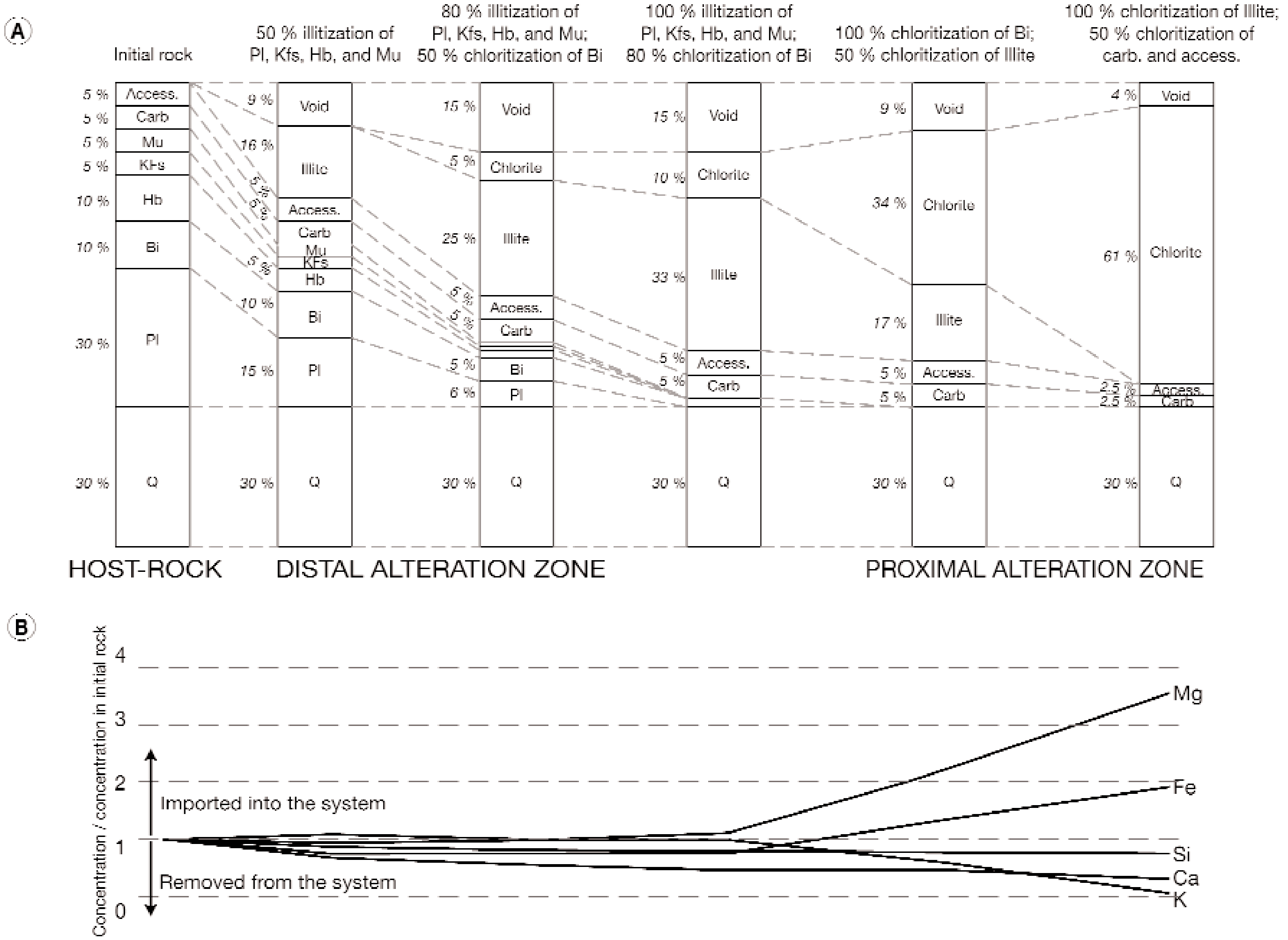

FIG. 7. Results of the volume and chemical variation calculations. A. Model volume changes during various alteration stages of basement rocks, based on the major alteration reactions (see text) and the average molar volume of the various minerals (Deer et al., 1992). The initial host rock corresponds to an average mineralogical composition of the basement host rock of the three deposits. Five alteration steps between fresh host rock and strongly chloritized basement are defined following petrographic observations (see text), with illitization occurring mostly in the distal alteration zone and chloritization in the proximal alteration zone. Note that illitization creates void space, whereas chloritization reduces void space, as the molar volume of chlorite is much higher than that of illite, 430 Å^3 and 283 Å^3, respectively (Deer et al., 1992). Abbreviations: Access = various accessory minerals, Bi = biotite, Carb = carbonates, Hb = hornblende, KFs = potassic feldspar, Mu = muscovite, Pl = plagioclase, Q = quartz. B. Chemical variation of some of the major elements of interest. Mg and Fe are removed from the distal, illite-dominated alteration zone and added to the proximal, chlorite-dominated alteration zone, whereas Ca, Si, and K are removed from the system. See text for further explanations.

alteration zone, assuming that all the SiO_2 released was removed by the hydrothermal fluid. The estimated volume decrease after half of these minerals have been illitized is ca. 9 vol percent (Fig. 7). Further illitization, as observed proximal to the thrust fault zone, would have increased the void space to 15 vol percent (Fig. 7). However, chloritization of biotite, and to a lesser degree hornblende, in the proximal alteration zone would have resulted in an increase in rock volume. Initially, with low amounts of chloritization, this reaction involved no major volume change as the space created during illitization would have been filled by chlorite. More extensive chloritization, characterizing the proximal alteration zone, would have resulted in reduction of void space to ca. 9 vol percent. The ore zone is characterized by strong chloritization, which would have further reduced the void space (Fig. 7).

Several important implications result from these calculations.

1. Illitization creates space whereas chloritization consumes space, as the molar volume of chlorite is much higher than that of illite. Initial illitization and fracturing of the host rock therefore created porosity and promoted flow of basinal fluids into the basement. This is supported by the observed paragenetic sequence, which indicates that illite was the first and locally the only alteration mineral formed.
2. Illitization liberated iron and magnesium from the basement rocks and these elements were consumed during chloritization in the ore zone. Further, the volume of rock that was illitized had to be much larger than the volume of rock that was chloritized because more magnesium and iron are necessary for chlorite alteration than would have been liberated by an equivalent volume of illitization.
3. In the ore zone, chloritization consumed more magnesium than iron. Thus the extra iron could have acted as a reductant for uranium precipitation together with graphite in the basement rocks. Graphite, which is present in the alteration zones, is noticeably absent from the ore zone, as it was consumed during uraninite precipitation.
4. Part of the potassium liberated during chloritization of biotite likely precipitated in the ore zone as coarse-grained illite (I^2). The rest of the potassium and most of the silica released are exported from the system toward the Athabasca Formation sandstones, where they contribute to the silica and illite alteration halos.

Estimates of permeability modification based only on the mineralogical composition of the alteration zones do not account for the structure-induced modifications of fluid flow (e.g., by fracturing and offset of the unconformity). Fracturing would not only initiate fluid flow but also would control it to a great extent (e.g., Lorilleux et al., 2002).

Timing of alteration and mineralization events

Reported mineralization ages vary from 900 to 1400 Ma from U/Pb dating of uraninite and pitchblende from the Key Lake (Carl et al., 1991), Cigar Lake (Philippe et al., 1993), and Midwest deposits (Badsgaard et al., 1984; Worden et al., 1985), and from ^{40}Ar/^{39}Ar and K-Ar dating of illite associated with mineralization at the McClean Lake deposit (Bray et al., 1988). More recent U-Pb ages of uraninite using an ion probe have upper intercept ages of 1598 ± 25 Ma for the McArthur River sandstone-hosted deposit, 1543 ± 8 Ma for the Sue C zone deposit (M. Fayek, pers. commun.), and 1467 ± 47 Ma for the Cigar Lake deposit (Fayek et al., 2002). Recent laser ablation ICP-MS U-Pb dating of uraninite from the basement-hosted deposits in this study indicate upper intercepts of 1540 ± 38, 1247 ± 88, and 952 ± 27 Ma (Alexandre and Kyser, 2003). The highest age has been interpreted as the mineralization age, and the two lower ages have been interpreted to reflect recrystallization of the uraninite as a result of fluid circulation induced by intrusion of the mafic dikes at 1267 Ma (LeCheminant and Heaman, 1989), the last stages of the Grenvillian orogeny and the assembly of Rodinia at ca. 975 Ma (Rennac et al., 2002; Wingate et al., 2002). These later events can explain the lower U-Pb ages of 900 to 1400 Ma reported for uraninite in various deposits (Badsgaard et al., 1984; Bray et al., 1988; Carl et al., 1991; Philippe et al., 1993). ^{40}Ar/^{39}Ar dating of samples from the basement-hosted Dawn Lake, Rabbit Lake, and McArthur River deposits indicates that the preore alteration illite (I^1) has an age of ca. 1667 Ma, higher than the synore illite (I^2) ^{40}Ar/^{39}Ar age of ca. 1591 Ma (Alexandre and Kyser, 2003).

Model for basement-hosted deposits

Stable isotope analyses and temperatures deduced from crystal chemistry of clay minerals indicate that the same diagenetic fluids were responsible for the pre- and synore alteration in the basement. There is no isotopic evidence for the presence of basement-derived fluids in the ore zones or mixing between two fluids, as suggested for the sediment-hosted unconformity-type deposits (Wilson and Kyser, 1987; Fayek and Kyser, 1997). Diagenetic basinal fluids were responsible for the bulk of the alteration and also transported the uranium from the sediments where it was leached to the fractured basement where it was precipitated (Fig. 8). Enhanced permeability and a reducing environment were required for uranium precipitation. Along with faulting, the illite alteration increased the permeability thereby channeling larger volumes

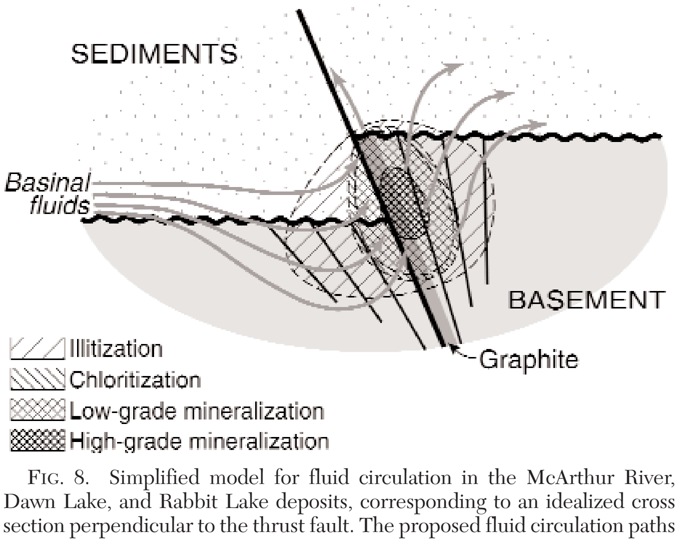

FIG. 8. Simplified model for fluid circulation in the McArthur River, Dawn Lake, and Rabbit Lake deposits, corresponding to an idealized cross section perpendicular to the thrust fault. The proposed fluid circulation paths and the alteration and mineralization zones are indicated.

of fluid into the fault system. Reaction with graphite in the basement lithological units and, to a lesser extent, the iron liberated during chloritization, caused precipitation of uranium. The latter likely accounts for the strong spatial relationship between the uranium orebody and the chloritic alteration, particularly at the McArthur River deposit (McGill et al., 1993).

In the model proposed by Fayek and Kyser (1997) for the Sue deposit, the fluids penetrate the basement via a reverse fault, resulting in planar orebodies contained in the fault system. However, the orebodies in the three deposits studied here are cigar shaped (Dawn Lake) or isometric (Rabbit Lake and particularly McArthur River), possibly reflecting the permeability evolution during alteration.

The model proposed here differs from that of Hecht and Cuney (2000), in which uranium is leached from uranium-bearing phases such as monazite and zircon present in the basement rocks. This model does not take into account the amount of basement rock that would have to be altered to produce the uranium, the much lower permeability of the basement, or its reduced nature, which would have inhibited the transport of uranium.

Comparison between sandstone- and basement-hosted deposits

The major differences between sandstone- and basement-hosted uranium deposits result from the differences in the chemistry and the permeability of the host rocks. The permeability and the fluid/rock ratio in the sandstones are much higher than in the basement so that fluid composition largely controls the alteration clay mineral compositions, whereas in the basement the mineralogy and host-rock chemistry are the main control on the composition of the alteration assemblage. Thus, the composition of chlorite in the basement is inherited from biotite and is less aluminous and more iron and magnesium rich than in the sandstone (Wilson and Kyser, 1987). The ore-associated alteration in sandstone-hosted deposits is dominantly illitic, whereas alteration in the basement-hosted deposits is mostly chloritic. This is explained by the fact that in the sandstones, the fluids are potassium rich and illite forms readily, whereas formation of chlorite is more difficult because of the lack of iron and magnesium. Abundant iron and magnesium are available in the basement-hosted systems, and chlorite forms readily without addition from an external source.

From an exploration point of view, the difference in porosity and permeability between the sandstone and basement also may be fundamental in determining the size and uranium concentration of the orebodies. Basement-hosted deposits tend to be smaller and have higher grades, such as the McArthur River zone 2 orebody. Sandstone-hosted deposits are volumetrically larger and have lower grades (Thomas et al., 1998). The amount of altered country rock is also lower in the basement-hosted deposits where the uranium-bearing fluids were possibly more focused at the precipitation site.

Conclusions

The main conclusion of this work is that basinal fluids were responsible for both the preore alteration and the subsequent mineralization in basement-hosted uranium deposits. The basement played both a passive role as a physical trap, allowing basinal fluids to infiltrate into strongly fractured zones, and an active role as a chemical (reducing) trap for uranium precipitation. The observed chlorite alteration is the product of the Mg and Fe redistribution within the basement during illitization and chloritization. The only elements added in the basement alteration system (distal, proximal, and ore zones) are potassium, necessary for the formation of illite and originating from illite-saturated basinal fluids, and uranium leached from uranium-bearing phases such as zircon, monazite, and apatite in the sandstone.

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