

Significance of Alteration Assemblages for the Origin and Evolution of the Proterozoic Nabarlek Unconformity-Related Uranium Deposit, Northern Territory, Australia

PAUL A. POLITO,[†] T. KURT KYSER,

Department of Geological Sciences and Geological Engineering, Queen's University, Kingston, Ontario, Canada K7L 3N6

JIM MARLATT,

Cameco Corporation, 2121 11th Street W., Saskatoon, Saskatchewan, Canada S7M 1J3

PAUL ALEXANDRE,

Department of Geological Sciences and Geological Engineering, Queen's University, Kingston, Ontario, Canada K7L 3N6

ZIA BAJWAH,

Northern Territory Geological Survey, GPO Box 3000, Darwin, Northern Territory, Australia 0801

AND GARTH DREVER

Cameco Corporation, 2121 11th Street W., Saskatoon, Saskatchewan, Canada S7M 1J3

Abstract

The Proterozoic Nabarlek unconformity-related uranium deposit in the Alligator Rivers uranium field is hosted by Paleoproterozoic amphibolite-grade, metamorphosed semipelitic sedimentary rocks and amphibolite schist. High-grade ore is confined to the Nabarlek fault, a reverse fault/shear zone that crosscuts a series of interbedded muscovite-quartz-biotite schists and amphibolite. Petrographic studies on polished thin sections combined with electron microprobe analyses, X-ray diffraction, fluid inclusion data, O-H and U-Pb isotope values, as well as ⁴⁰Ar/³⁹Ar dating have identified up to eight significant fluid events beginning with the precipitation of early quartz veins during uplift of the Myra Falls Metamorphics at 1830 Ma and ending with limited uraninite recrystallization during reactivation of the Nabarlek fault between 1380 and 750 Ma.

Quartz veins that likely formed toward the end of the Top End orogeny represent the earliest recorded fluid event. Fluid inclusion data and $\delta^{18}\text{O}$ and δD values indicate that these veins formed from basement-derived fluids that may have been heated by a cooling Nabarlek Granite during circulation through reverse faults/shear zones. The next fluid event, represented by fine-grained sericite and chlorite occurred when fluids passed into these faults and altered the metamorphic minerals following the exhumation of the basement and deposition of the Kombolgie Subgroup at ca. 1760 Ma. The intrusion of the Oenpelli Dolerite at ca. 1720 Ma resulted in the local remobilization of silica and the precipitation of quartz associated with minor pyrite and dolomite around the reverse faults.

Uranium mineralization is associated with an inner and outer alteration halo that extends as far as 1 km from the Nabarlek fault. Alteration in the outer halo began as early as 1700 Ma and is dominated by chlorite and sericite, which formed when a 200°C fluid flowed into the Nabarlek fault from the overlying Kombolgie Subgroup. U-Pb and ²⁰⁷Pb/²⁰⁶Pb dating reveals that massive uraninite precipitated at ca. 1640 Ma and formed together with illite and hematite at ca. 200°C. Chlorite was not coeval with uraninite precipitation. Stable isotope values indicate that the pre- and synore alteration assemblage formed from basinal brines with $\delta^{18}\text{O}_{\text{fluid}}$ and $\delta\text{D}_{\text{fluid}}$ values of 2 ± 2 and -25 ± 10 per mil, respectively.

Reactivation of the Nabarlek fault at ca. 1360, 1100, and 900 Ma is indicated by U-Pb and ²⁰⁷Pb/²⁰⁶Pb dating of uraninite. These ages correlate with the intrusion of the Maningkorri phonolitic dikes and the Derim Derim Dolerite at ca. 1316 ± 40 and 1324 ± 4 Ma, respectively, the amalgamation of Australia and Laurentia during the Grenville orogen at ca. 1140 Ma, and the breakup of Rodinia between 1000 and 750 Ma. Fluid incursions associated with these events precipitated much of the chlorite that has previously been related to uraninite precipitation. Drusy quartz veins that host high-salinity fluid inclusions and sulfides, particularly galena, also formed after the initial uraninite-forming event. Finally, erosion of the Kombolgie Subgroup and subsequent weathering of the deposit resulted in the recent formation of kaolinite and numerous secondary uranium minerals. These data constrain individual events more precisely than previous studies and thus advance the current genetic model to a level that takes into account the multiple stages of fluid overprinting that occurred over a period of at least 800 m.y.

Introduction

THE NABARLEK unconformity-related uranium deposit is located in the Alligator Rivers uranium field within the Pine

Creek geosyncline, approximately 270 km due east of Darwin, Australia (Fig. 1). Nabarlek is small when compared to the nearby Jabiluka and Ranger deposits (Fig. 1), but intersections of almost pure uraninite up to 1 m in length (Anthony, 1975) make Nabarlek the highest grade uranium

[†] Corresponding author: e-mail, polito@geol.queensu.ca

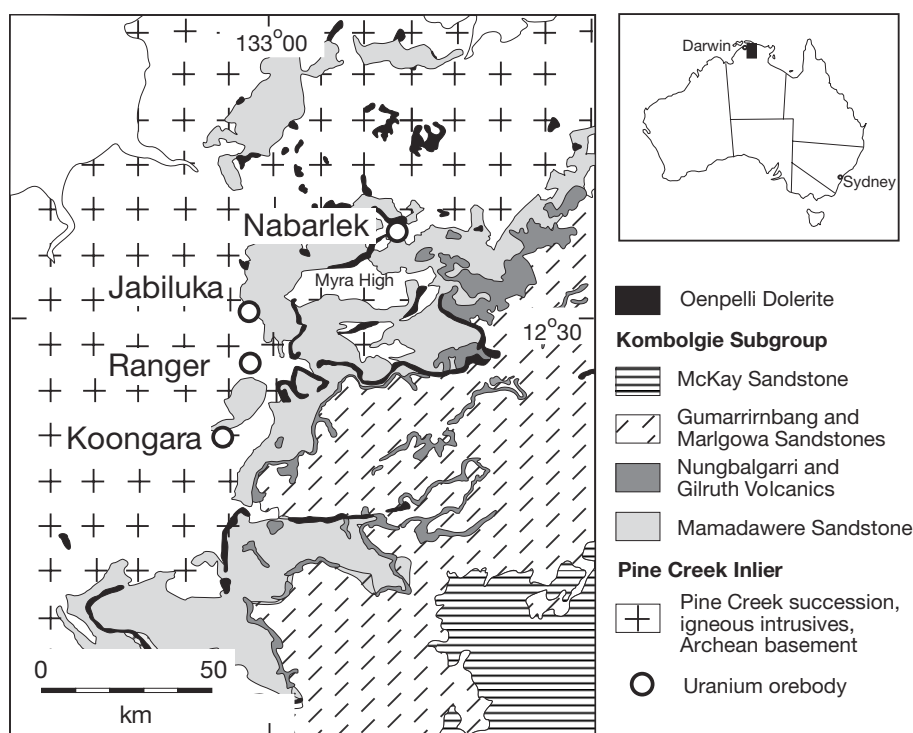


FIG. 1. Regional geology of the east Alligator Rivers uranium field. Modified after Needham (1988).

deposit discovered to date in Australia. The premining reserve estimate was ca. 1.82 million pounds (Mlbs) of recoverable uranium (Anthony, 1975). Open-pit mining in 1979 recovered 564,437 t of ore at 1.86 percent U_3O_8 and 157,000 t at 0.05 percent U_3O_8 (Wilde and Noakes, 1990).

Nabarlek is hosted in amphibolite-grade, metamorphosed semipelitic sedimentary rocks and amphibolite schist belonging to the Myra Falls Metamorphics (Anthony, 1975; Wilde and Wall, 1987). Regional peak metamorphism was attained during the Barramundi and Top End orogenies between 1890 and 1800 Ma (Ferguson and Winer, 1980; Page et al., 1980; Needham, 1988). By 1800 Ma, the Alligator Rivers uranium field had experienced significant uplift and erosion (Sweet et al., 1999b), which resulted in the formation of a paleotopographic high known as the Myra high (Fig. 1). Recent sequence stratigraphic mapping (Hiatt and Kyser, 2002) has established that the Myra high was present prior to the deposition of the Komolgie Subgroup. It is along this high that the Nabarlek uranium deposit is found.

High-grade ore ($>1.0\%$ U) is confined to within a breccia zone of the Nabarlek reverse fault/shear zone (Fig. 2; Johnston, 1984). Primary mineralization is particularly focussed within the Footwall Amphibolite and the Hanging-Wall schist (Fig. 3). The mineralized Footwall Amphibolite and the entire rock package at Nabarlek are devoid of massive bedded dolomite or magnesite and only traces of carbonaceous material occur in unaltered schist units (Ewers and Ferguson, 1980; Wilde and Wall, 1987). The absence of graphite and carbonate in the main host sequence at Nabarlek contrasts to other uranium deposits in the Alligator Rivers uranium field, including Koongarra, Jabiluka, and Ranger (Fig. 1).

A number of publications document the Nabarlek uranium deposit. However, there are many inconsistencies in the literature and considerable diversity of opinion between authors. Anthony (1975) briefly outlined the local geology and mineralization of the orebody. Ewers and Ferguson (1980), Ewers et al. (1983), and Wilde and Wall (1987) published detailed petrological and geochemical investigations pertaining to the mineralization. Johnston (1984) produced a detailed structural analysis of the deposit, and Ypma and Fuzikawa (1980) and Wilde et al. (1989) reported on fluid inclusions hosted within veins in and around the deposit. Ypma and Fuzikawa (1980) also presented limited oxygen isotope data. Initial dating of the deposit indicated a formation age of 920 Ma (Hills and Richards, 1976). However, this was interpreted to be a remobilization event by Page et al. (1980) and Maas (1989), who reported Rb-Sr and Sm-Nd ages of $1,616 \pm 50$ Ma for primary ore.

Few papers have presented a model that can explain all of the alteration and mineralization assemblages owing to limited paragenetic investigations. In this paper, we use detailed petrography, stable and radiogenic isotope systematics, fluid inclusion microthermometry, and electron microprobe analyses to improve existing models for the deposit and to constrain the multiple stages of fluid overprinting that have been recognized.

Local Geology

The oldest lithologies in the mine area are the Paleoproterozoic Myra Falls Metamorphics. The lowest unit of the Myra Falls Metamorphics is a ca. 200-m-thick sequence of muscovite-quartz-biotite schist interlayered with thin intervals of

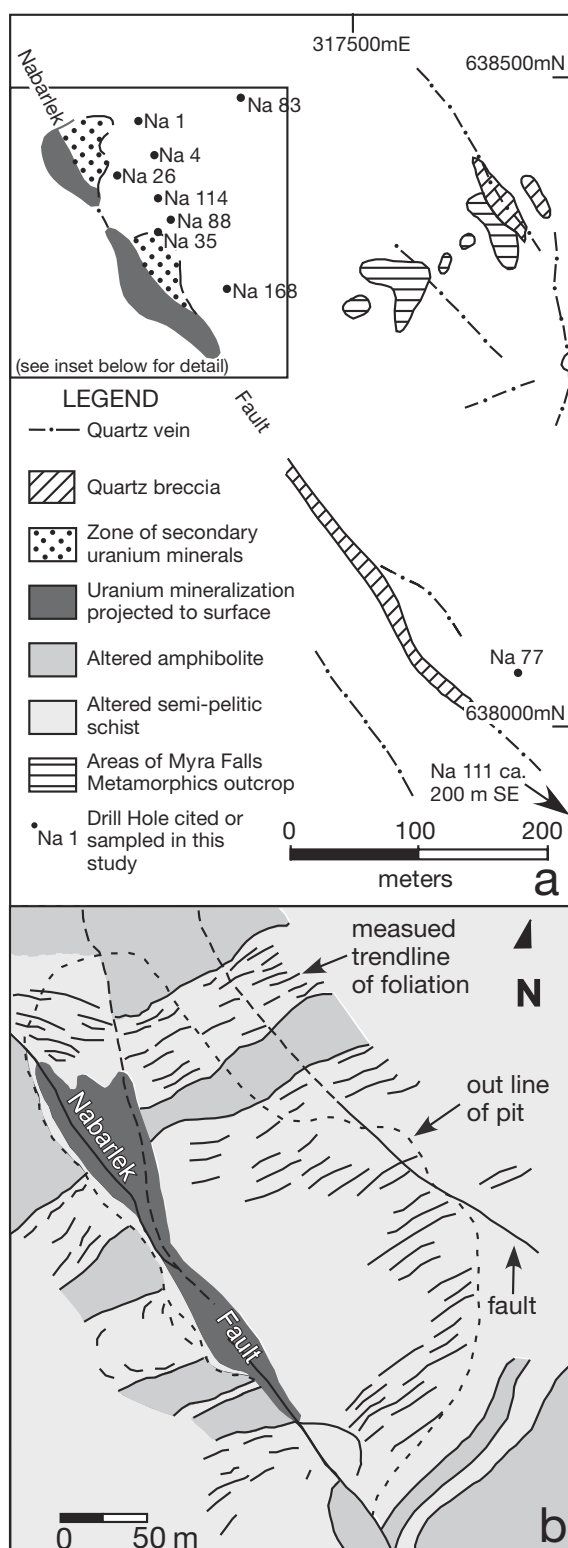


FIG. 2. a. Local outcrop geology of the Nabarlek deposit as it occurred prior to mining operations, showing drill holes sampled, analyzed, or cited in this paper. Open rectangle represents area of enlargement shown in (b), which details the geology exposed during mining of the deposit. Modified after Ypma and Fuzikawa (1980) and Wilde and Wall (1987). The basement geology was only exposed during the lifetime of the mine. Flatlying sandstones belonging to the Kombolgie Subgroup surround the deposit and limit mapping of the regional basement geology.

hornblende-plagioclase-biotite-clinopyroxene amphibolite and referred to as the Lower Schist unit (Fig. 3). The oldest recorded quartz veins at Nabarlek occur in the Lower Schist unit and are herein referred to as Q^1 veins (Fig. 4). They are commonly subparallel to foliation and are massive in texture, with anhedral, interlocking quartz grains. Ypma and Fuzikawa (1980) characterized these veins as metamorphogenic quartz veins.

Structurally overlying the Lower Schist unit is ca. 100 m of interlayered amphibolite and schist, collectively referred to as the Footwall Amphibolite (Fig. 3). The Footwall Amphibolite hosts most of the known mineralization at Nabarlek (Wilde and Wall, 1987). Graphite and garnet are rare and only occur south of the deposit (Wilde and Noakes, 1990). The amphibolite precursor comprised clinopyroxene, hornblende, plagioclase, biotite $\pm$ K-feldspar $\pm$ ilmenite $\pm$ bravoite $\pm$ prehnite $\pm$ pyrite $\pm$ euhedral apatite (Wilde and Wall, 1987).

Overlying the Footwall Amphibolite is a biotite-muscovite-quartz-feldspar schist referred to as the Hanging-Wall schist (Fig. 3). The Hanging-Wall schist is at least 75 m thick in the mine area and is the only other host to known mineralization. Pit mapping reveals that it has been faulted over the Footwall Amphibolite (Johnston, 1984; Wilde and Wall, 1987). The Hanging-Wall schist is locally distinguished by layers of quartz-rich psammitic schist, up to 60 cm thick, alternating with more muscovite-rich pelitic to semipelitic layers that have been interpreted as sedimentary layering (Wilde and Wall, 1987). Garnet has been noted but is rare. Thin quartzite layers are relatively common and contain coarse muscovite flakes. Several thin amphibolite bands are intercalated in the Hanging-Wall schist.

Uncommon, isoclinally folded pegmatite veins that have not been dated and the Nabarlek Granite intrude the Myra Falls Metamorphics (Wilde and Wall, 1987). The Nabarlek Granite is considered to be part of the Jim Jim Suite of granites that include the 1838 ± 7 Ma Jim Jim Granite and the 1820 ± 8 Ma Malone Creek Granite (Edgecombe et al., 2002).

A 220- to 250-m-thick sheet of Oenpelli Dolerite intrudes the Myra Falls Metamorphics and separates the Lower Schist unit from the Footwall Amphibolite (Fig. 3). The upper and lower contacts of the Oenpelli Dolerite are characterized by coarsely porphyritic to pegmatitic facies, whereas the central part is composed of sparse tabular phenocrysts of plagioclase in a groundmass of randomly orientated plagioclase laths enclosed by ophitic pale-green augite and dark-green hornblende (Wilde and Wall, 1987). A 20-m-thick granophyric zone occurs close to the upper contact of the Oenpelli Dolerite. The Oenpelli Dolerite has been dated by $^{40}\text{Ar}/^{39}\text{Ar}$ and U-Pb methods, giving an age of 1723 ± 6 Ma (Kyser et al., 2000; Edgecombe et al., 2002). Regionally, the Oenpelli Dolerite is known to intrude to the Paleoproterozoic sedimentary rocks of the Kombolgie Subgroup (Needham, 1988; Carson et al., 1999).

Unconformably overlying these older units are the unmetamorphosed, shallowly dipping sandstones, conglomerates, and volcanic rocks belonging to the Katherine River Group. At the base of the Katherine River Group is the Kombolgie Subgroup, with an age between 1822 and 1720 Ma, as defined by the underlying Plum Tree Creek Volcanics and the

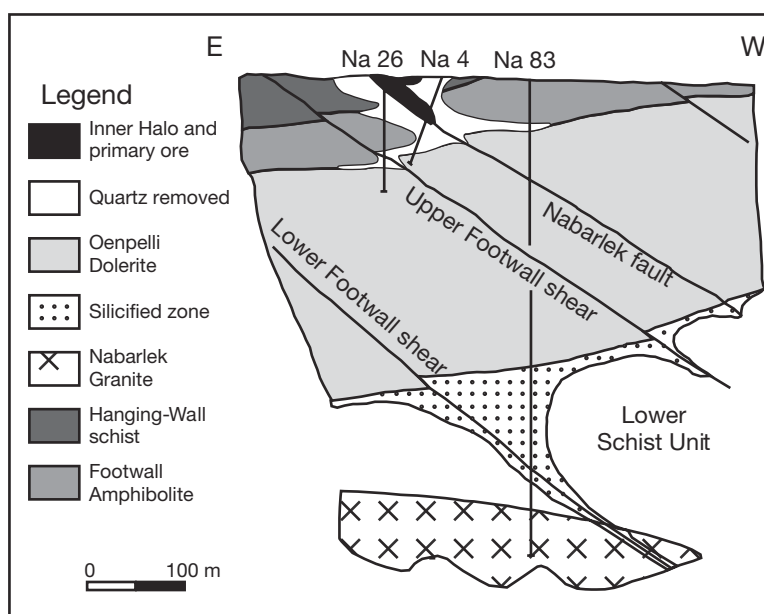


FIG. 3. Cross section through the Nabarlek orebody, showing the inner alteration halo and the area identified to be devoid of quartz in relationship to the Nabarlek fault. Modified after Wilde and Wall (1987) and Wilde and Noakes (1990).

intruding Jimbu Microgranite, respectively (Sweet et al., 1999a). The Kombolgie Subgroup is composed of at least three stratigraphic sequences that are separated by volcanic flows belonging to the Nungbagarri Formation and the Gilruth Member. The Kombolgie Subgroup is known to be at

least 1,350 m thick at the southern margins of the Alligator Rivers uranium field (Holk et al., 2003) and may have been as thick as 1,810 m in the Mount Marumba area east of Nabarlek (Sweet et al., 1999b). Although Nabarlek sits on the Myra high, at least 1,980 m of sandstone and volcanic rocks

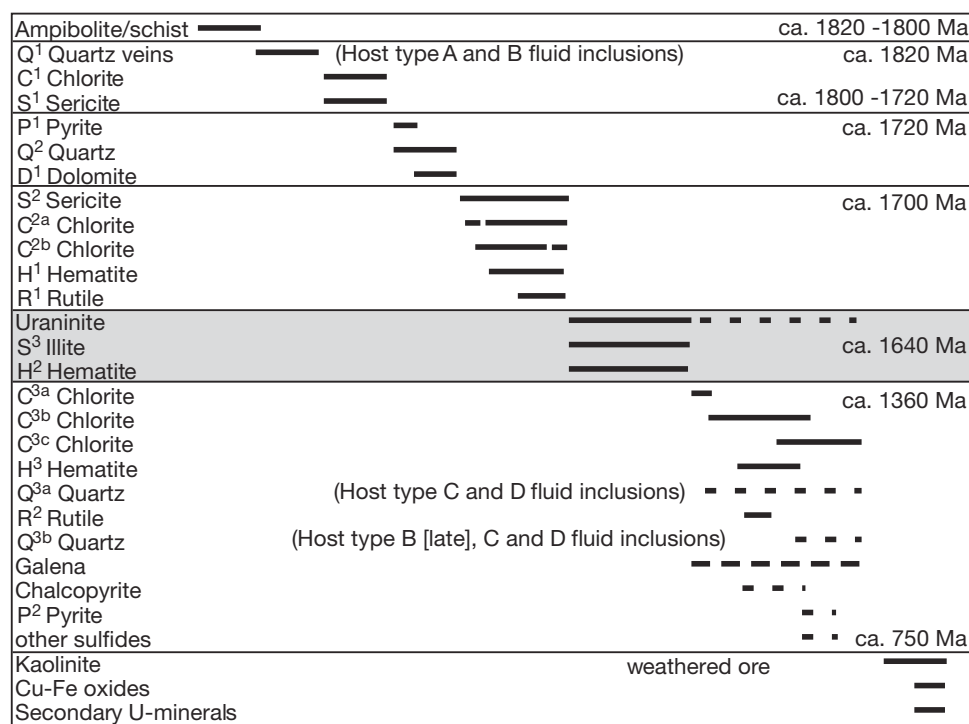


FIG. 4. Paragenetic diagram summarizing the timing of alteration assemblages around the Nabarlek deposit. The ages presented for each event are based on crosscutting relationships that have been constrained by K/Ar ages for the Q¹ quartz veins (Page et al., 1980), ⁴⁰Ar/³⁹Ar and U/Pb ages for the Q² quartz vein event (Kyser et al., 2000; Edgecombe et al., 2002), and ⁴⁰Ar/³⁹Ar and ²⁰⁷Pb/²⁰⁶Pb ages (this study).

belonging to the Katherine River Group were present above the deposit by 1705 Ma (Rawlings, 1999; Sweet et al., 1999b), and a minimum of 2,780 m of strata, up to and including the overlying Roper Group, would have been present by ca. 1500 Ma. Koul et al. (1988) suggested that approximately 5 km of Lower Proterozoic sedimentary rocks were in place in the Nabarlek area by 1600 Ma.

The mineralized Nabarlek fault is a reverse fault/shear zone with a net displacement of up to 20 m (Johnston, 1984). In the mine area, the Nabarlek fault parallels two unmineralized reverse structures, the Upper Footwall shear and the Lower Footwall shear (Fig. 3). Breccia zones up to 15 m wide, intense veining, and hydraulic fracture textures define these individual faults (Johnston, 1984). Small breccia zones (2–3 cm wide) up to 10 m away from the main shear structures are reported as splays off these shear zones.

Methods

One hundred and five diamond drill core specimens were collected from the core storage facility at the Northern Territory Geological Survey. Polished thin sections were prepared from all samples. The Northern Territory Geological Survey supplied additional polished thin sections cut from diamond drill core and open-pit material. The thin sections were examined using transmitted and reflected light to determine the mineral paragenesis. Electron microprobe analyses were performed on 22 carefully selected polished thin sections, using a Cambax MBX electron microprobe equipped with 4 WDX X-ray spectrometers at Carleton University, Ottawa, Canada.

Microthermometric measurements on doubly polished thick sections were performed using a Linkam TH600 heating-cooling stage by standard techniques (Shepherd et al., 1985). Samples from five quartz veins from within and below the Oenpelli Dolerite sill (drill hole Na 83), one from the Hanging-Wall schist (Na 114), and one sample each from drill holes Na 77 and Na 111 at the southern end of the deposit (Fig. 2) were analyzed.

Quartz veins, uraninite, and phyllosilicate minerals from the outer and inner alteration halos were extracted from a crushed and washed fraction of sample for stable isotope analysis. All samples were analyzed by XRD using the method of Mellinger (1979) to ensure purity. In most cases, only pure separates of each phase were analyzed. Some separates containing up to 5 percent contamination by another phase were analyzed. Oxygen isotope compositions of quartz, illite, chlorite, kaolinite, and uraninite were measured using the BrF_5 method of Clayton and Mayeda (1963). Hydrogen isotope compositions of quartz-hosted fluid inclusions, sericite, chlorite, illite, and kaolinite were determined using the methods of Kyser and O'Neil (1984). Stable isotope measurements were made using a Finnigan MAT 252 mass spectrometer and are reported in the δ notation in units of per mil relative to the standard V-SMOW. Replicate $\delta^{18}\text{O}$ analyses were reproducible to ± 0.2 per mil and δD values to ± 3 per mil. Uraninite $\delta^{18}\text{O}$ in two high-grade samples was determined using the high spatial-resolution, CAMECA IMS 1270 ion microprobe at the University of Tennessee. The technique followed the method of Fayek et al. (2000) and reproducibility was within 0.9 per mil. Oxygen isotope fractionation factors used throughout this paper are those suggested by Eslinger and Savin (1973) for water-illite,

Land and Dutton (1978) for water-kaolinite, Yeh and Savin (1976) for water-chlorite, O'Neil and Taylor (1969) for water-muscovite, Clayton et al. (1972) for water-quartz, and Fayek and Kyser (2000) for water-uraninite. Hydrogen isotope fractionation factors used are those suggested by Suzuoki and Epstein (1976) for water-muscovite, Yeh (1980) for water-illite, Lambert and Epstein (1980) for water-kaolinite, and Graham et al. (1984, 1987) for water-chlorite.

Sericite, muscovite, and illite separates were dated using the $^{40}\text{Ar}/^{39}\text{Ar}$ laser-heating technique of Lee et al. (1990). U-Pb and Pb-Pb analyses were performed using laser ablation (Mercantek® LUV213 with Nd-YAG) coupled with an online Finnigan MAT Element® high-resolution inductively coupled plasma mass spectrometer (LA-HR-ICPMS). The analyses were performed on polished thin sections in spot mode using the technique of Kyser et al. (2003). The isotopes measured were ^{201}Hg , ^{202}Hg (used for correction of interferences by ^{204}Hg on ^{204}Pb), ^{204}Pb (used for common Pb correction), ^{206}Pb , ^{207}Pb , ^{208}Pb , ^{235}U , and ^{238}U .

Alteration around the Nabarlek Deposit

Wilde and Wall (1987) divided alteration at Nabarlek into an inner and outer halo. The outer alteration zone can be traced as either chlorite-hematite schist, chlorite-muscovite $\pm$ hematite schist, or chlorite-sericite schist up to 1 km from the Nabarlek fault (Wilde and Wall, 1987).

Outer alteration halo

The earliest alteration (Fig. 4) is represented by partial to complete replacement of metamorphic biotite, muscovite, and hornblende by Fe chlorite (C^1 chlorite) and minor fine-grained sericite (S^1 sericite). S^1 sericite also replaces plagioclase. Metamorphic biotite, plagioclase, and hornblende are preserved in an extensive zone of silicification that occurs extensively within the Lower Schist unit and up to 21 m above the Oenpelli Dolerite in the Footwall Amphibolite adjacent to the Lower Footwall shear, the Upper Footwall shear, and possibly the Nabarlek fault (Fig. 3). Paragenetic observations suggest that at some point, probably coincident with the intrusion of the Oenpelli Dolerite, the weakly altered metamorphic assemblage was overprinted by silica and dolomite veinlets. The quartz cement (Q^2) comprises 10- μm to 1-mm interlocking quartz grains, commonly with pyramidal termination points that locally replace the metamorphic assemblage (Fig. 4). Elsewhere, this generation of quartz consists of 0.1- to 1-cm-wide veins of comb-textured quartz that crosscut the altered metamorphic assemblage (Fig. 5a). This quartz corresponds to the silicified zone of Wilde and Wall (1987). The silica most likely originated during the alteration of plagioclase to sericite. Euhedral pyrite (P^1) appears to have been contemporaneous with the earliest stages of silica remobilization (Fig. 5b), but crosscutting relationships with dolomite and Q^2 quartz veinlets indicate that it was later brecciated and annealed during the introduction of these minerals (Fig. 5b). Quartz is, however, relatively rare in the outer alteration zone of the Footwall Amphibolite and the Hanging-Wall schist, having been removed during the next phase of alteration.

Unaltered amphibole, plagioclase, muscovite, and biotite, Q^1 quartz, C^1 chlorite, S^1 sericite, P^1 pyrite, dolomite, and Q^2 quartz were replaced or cut by coarse-grained (30–300 μm),

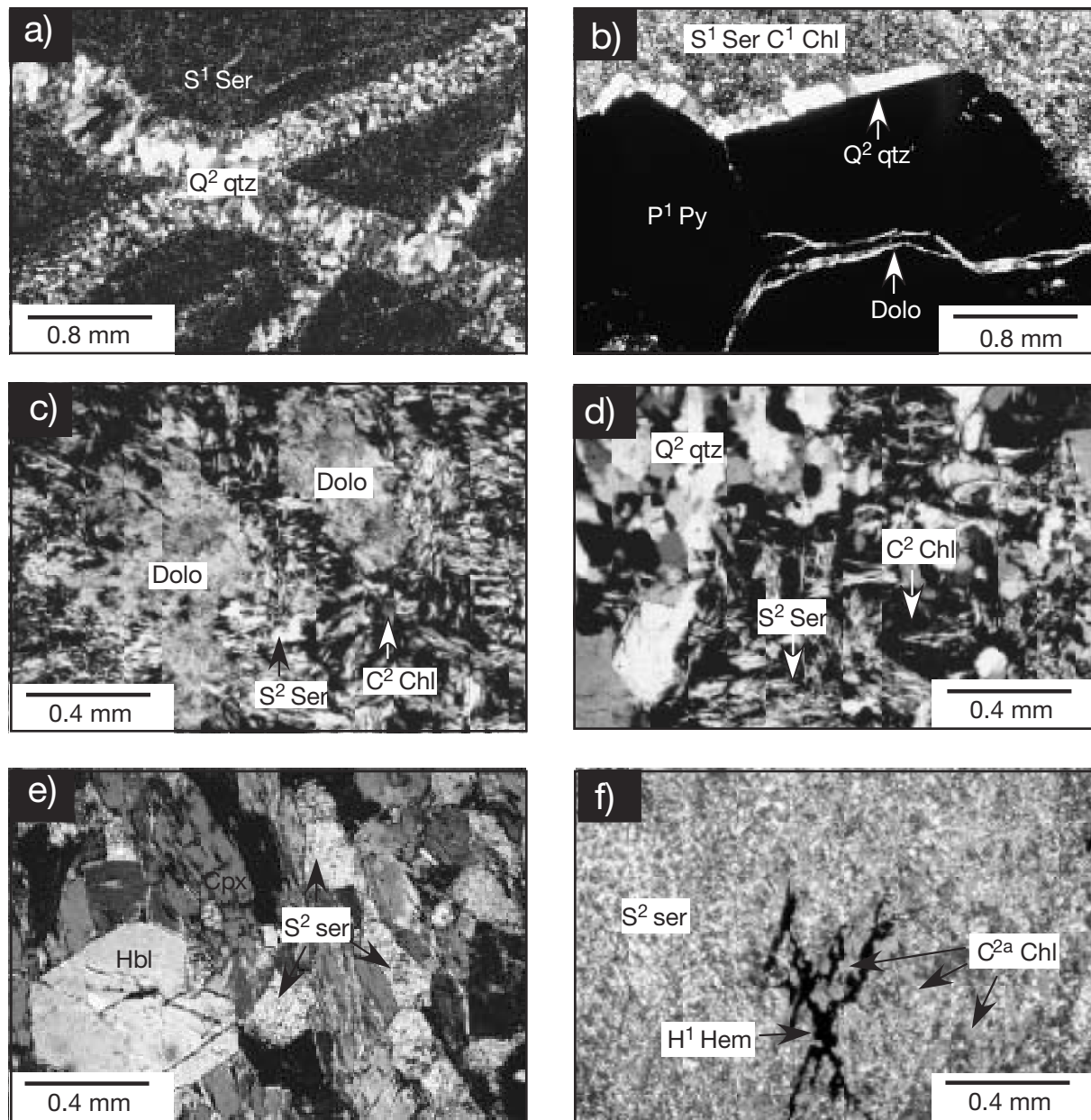


FIG. 5. Alteration textures around the Nabarlek fault. a. Comb-textured Q² quartz veins crosscutting fine-grained S¹ sericite and associated with silicification below the Oenpelli Dolerite (sample Na83 414.0m, cross-polarized light). b. Euhehedral P¹ pyrite with a thin rim of Q² quartz growing into fine-grained S¹ sericite and C¹ chlorite. Pyrite is crosscut by anastomosing veinlets of dolomite (sample Na83 368.5m, cross-polarized light). c. Coarse-grained C² chlorite and S² sericite replacing dolomite in the Lower Schist unit (sample Na83 364.2m, cross-polarized light). d. Coarse-grained C² chlorite and S² sericite replacing Q² quartz in the Lower Schist unit (sample Na83 316.1m, cross-polarized light). e. Plagioclase crystals replaced by S² sericite in the presence of pristine hornblende, minor clinopyroxene, and weakly altered biotite in schistose amphibolite (sample Na83 336.8m, cross-polarized light). f. S² sericite pseudomorphous after hornblende, C^{2a} chlorite and H¹ hematite distal to the Nabarlek fault (sample Na26 80.4m, cross-polarized light). Abbreviations: C² Chl = C² chlorite, Cpx = clinopyroxene, Dolo = dolomite, Hbl = hornblende, P¹ Py = P¹ pyrite, Q² qtz = Q² quartz, S² Ser = S² sericite.

locally radially textured C² chlorite and S² sericite laths (Fig. 5c-d). Their size and texture indicate growth into open pore spaces that were possibly created by brittle deformation along shear zones or mineral dissolution. Petrographic evidence shows that plagioclase was replaced by S² sericite before hornblende and biotite were altered to C² chlorite (Fig. 5e). Closer to the

Nabarlek fault, S² sericite, C² chlorite, and H¹ hematite dominate the assemblage and appear to be contemporaneous (Figs. 4–5f). Here, the precursor assemblage is impossible to determine, with the exception of coarse-grained, metamorphic muscovite laths that commonly appear unaltered as close as 20 m from the inner alteration zone. However, similarly

described muscovite from the Koongarra U deposit (Fig. 1) has highly variable compositions indicative of retrograde alteration (Kominou and Sverjensky, 1995a).

Two C^2 chlorite varieties are recognized at Nabarlek. The most common variety, C^{2a} chlorite, ranges from green or pale brown to transparent in plain-transmitted light and has first-order gray birefringence in cross-polarized light. It generally occurs as fine- to coarse-grained (<10 – $300\ \mu\text{m}$), matrix-filling material and rarely as <1 -mm-wide veinlets that grew in open spaces. Coarse-grained C^{2a} chlorite with no preferred orientation is most common close to the inner alteration halo, whereas fine-grained ($<10\ \mu\text{m}$) C^{2a} chlorite is typical in the outer alteration halo where it primarily replaces hornblende and minor amounts of muscovite. The second variety of chlorite (C^{2b}) identified in the outer alteration halo is coarse-grained, strongly pleochroic, dark-green to brown chlorite with blue to purple and pale-green to yellow birefringence. Biotite laths are typically pseudomorphous after C^{2b} chlorite.

Rutile needles (R^0) are uncommon and restricted to biotite laths that are replaced by C^{2b} chlorite. Progressive replacement of the C^{2b} chlorite by C^{2a} chlorite and S^2 sericite toward the inner alteration zone results in the formation of small, brown euhedral R^1 rutile crystals that are randomly distributed throughout the alteration assemblage and contemporaneous with the coarse-grained phyllosilicates (Fig. 4).

Quartz typically comprises <1 percent of the whole-rock assemblage near the inner alteration zone but is locally common in quartz-mica-schist layers that occur in the Hanging-Wall schist above the inner alteration zone. Here the quartz occurs in two forms: corroded quartz grains that are remnants of Q^1 and Q^2 quartz, and Q^3 quartz that postdates mineralization and occurs as veins that cut foliation and the alteration assemblage.

Inner alteration halo

Large illite fans, hematite, Fe chlorite, and the complete absence of quartz mark alteration of the inner halo. The inner alteration halo is distinguished from the outer alteration halo by the abundance of finely disseminated hematite (H^2) that gives the host rock a red to purple color (Figs. 4, 6a–b). The distribution of this hematite is patchy, but it comprises up to 20 percent of the assemblage close to the orebody. Within 20 m of the ore zone, pale-green and colorless C^{2a} chlorite and illite (S^3) occur, with slightly more chlorite than illite (Wilde and Wall, 1987). Illite abundance continues to increase toward the ore zone as C^{2a} and C^{2b} chlorite are replaced.

Ore zone

Within the ore zone, S^3 illite locally forms a massive, monomineralic rock that has obliterated all earlier structures (Johnston, 1984; Wilde and Wall, 1987; this study). Elsewhere it occurs with H^2 hematite intergrowths. The S^3 illite is commonly clear to pale-blue in plain-transmitted light and typically forms large (ca. 3 mm) radial fans that indicate growth into open space and fractures or as randomly orientated, elongated laths (Fig. 6a–b). The filamentous terminations of the illite commonly interfinger with massive and vein-type uraninite (Fig. 6b).

Uraninite occurs as fine disseminations and reticulate or anastomosing veins and veinlets within the Nabarlek fault

(Wilde and Wall, 1987; this study). The uraninite may be massive and homogeneous (Fig. 6c) or colloform with concentric banding and radial shrinkage cracks (Ewers and Ferguson, 1980). Brannerite is intergrown with uraninite (Fig. 6d) but comprises <5 percent of the ore (Wilde and Wall, 1987; this study). Filamentous veins and veinlets often display delicate interfingering between uraninite and S^3 illite (Fig. 6b), indicating that the two minerals are coeval.

Sulfides are common in the inner alteration halo. Reflected light petrography and SEM images show that chalcopyrite and galena are the two most common sulfides present. Galena is commonly observed as finely disseminated cubes and veinlets within and crosscutting uraninite and S^3 illite in the inner alteration halo (Figs. 4, 6d). Anhedral to blebby chalcopyrite is relatively common within the S^3 illite matrix and can be observed crosscutting uraninite (Wilde and Wall, 1987). It is commonly seen at the core of or overgrowing galena (Ewers et al., 1983; Wilde and Wall, 1987). Chalcocite, cobaltite, P^2 pyrite, rare bornite, and arsenopyrite are present but subordinate to galena and chalcopyrite.

C^3 chlorite is common in the inner alteration zone at Nabarlek, as are kaolinite, carbonate, late-stage hematite, and a range of sulfide minerals including galena, chalcopyrite, and cobaltite. Petrographic evidence indicates that these minerals postdate the uraninite- S^3 illite- H^2 hematite assemblage.

Postore alteration

Three distinct generations of chlorite have been observed crosscutting the uraninite-illite-hematite assemblage at Nabarlek (Fig. 4). In hand specimens and thin section, the dominant chlorite phases give the ore a characteristic green color, which in the past led to the interpretation that chlorite and uraninite were contemporaneous (Ewers and Ferguson, 1980).

The earliest and least common generation of postmineralization chlorite (C^{3a} chlorite) is clear to transparent in transmitted light. It forms radial fans and veinlets that cut uraninite and synore illite (Fig. 6e). The next generation of chlorite (C^{3b}) has a characteristic green color in hand specimens and in plane transmitted light. It is typically textureless, but lath-textured specimens have been observed. In this study, C^{3b} chlorite replaces illite (Fig. 7a–b) and is commonly observed as veins that cut the uraninite-illite-hematite assemblage and the C^{3a} chlorite (Fig. 6e–f). In its massive form, it surrounds and replaces the uraninite (Fig. 6f). Fine-grained, disseminated, black H^3 hematite and large, clear euhedral rutile (R^2) crystals occasionally occur with the C^{3b} chlorite (Fig. 4). The H^3 hematite and R^2 rutile clearly overprint uraninite (Fig. 6f). It is most likely that the rutile laths originate from the remobilization of Ti from the C^{2b} chlorite that replaced biotite. The third generation of postore chlorite (C^{3c}) is massive in appearance (Fig. 4). C^{3c} chlorite has been observed replacing illite, $C^{2a/b}$ chlorite, and C^{3b} chlorite. It appears to be less common than the C^{3b} chlorite but can only be positively distinguished when crosscutting relationships occur.

Postore quartz veins within the inner alteration halo are rare (Fig. 4). These quartz veins are clear to white and dominated by massive early quartz (Q^{3a}), which may be overgrown by euhedral quartz crystals with clearly defined growth zones (Q^{3b}). Both quartz generations crosscut and cement brecciated chlorite fragments and uraninite.

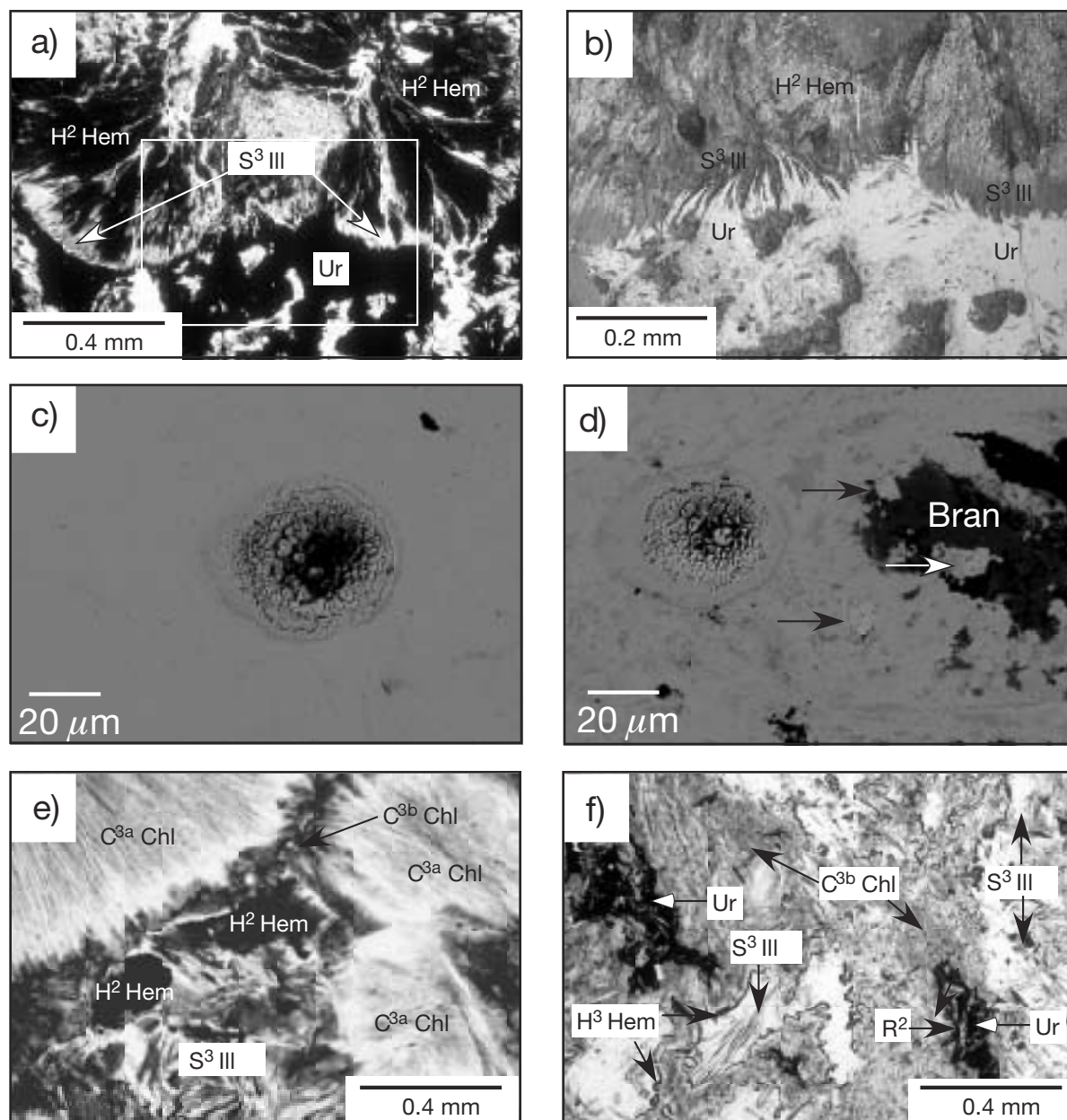


FIG. 6. a. Finely disseminated H² hematite within large, radial fans of clear to pale blue S³ illite, indicating growth into open space. Open rectangle represents area of enlargement shown in (b) (sample Na4 40.5m, plain-transmitted light). b. An enlargement of (a) taken in reflected light, showing in detail the finely disseminated hematite within illite and the filamentous terminations of the illite interfingering with uraninite. (c) Backscattered electron image of homogeneous uraninite with no obvious alteration in a 1.5-cm-thick uraninite vein. The ablation pit was created during analysis by LA-HR-ICPMS for U/Pb ratios, which revealed a ²⁰⁷Pb/²⁰⁶Pb age of 1642 Ma (sample Na4 40m). d. Backscattered electron image of brannerite (dark gray) and contemporaneous uraninite (light gray), displaying variable alteration to younger uraninite (intermediate gray) and galena cubes (arrows; sample Na4 40m). e. Large radial fans of transparent C^{3a} chlorite replacing S³ illite and H² hematite crosscut by dark green C^{3b} chlorite (sample Na1 36.1m, plain-transmitted light). f. C^{3b} chlorite and fine-grained, disseminated black H³ hematite replacing S³ illite and uraninite. Large, euhedral rutile crystals can be seen overprinting uraninite (sample Na4 43.1m, plain-polarized transmitted light). Abbreviations: Bran = brannerite, C^{3a} Chl = C^{3a} chlorite, C^{3b} Chl = C^{3b} chlorite, H² hem = H² hematite, H³ hem = H³ hematite, R² = R² rutile, S³ Ill = S³ illite, Ur = uraninite.

Kaolinite, anatase, hematite-goethite, digenite, covellite, native copper, marcasite, and disseminated yellow uranium minerals, including sklodowskite, rutherfordine, kasolite, and curite, occur in weathered ores (Wilde and Wall, 1987; Fig. 4). Fractures filled with torbernite and autunite represent the most recent postore mineralization at Nabarlek.

Mineral Chemistry

Electron microprobe analyses of chlorite, sericite, and illite reveal two chemically distinct chlorite varieties in the outer alteration halo, three chlorite varieties in the inner alteration halo, and an Fe-, Mg-enriched illite within the ore zone

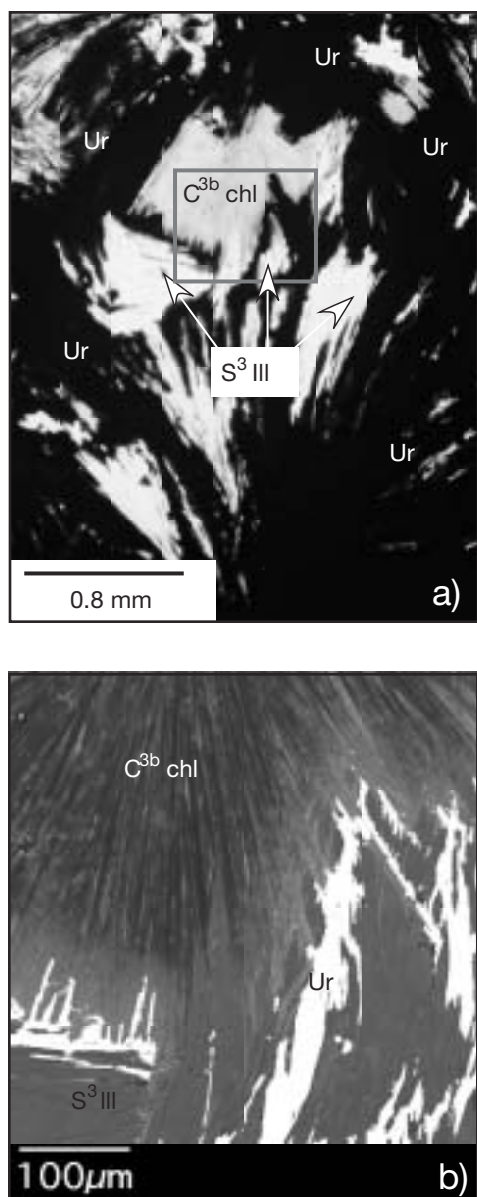


FIG. 7. a. Uraninite with fine intergrowths of C^{3b} chlorite. Although C^{3b} chlorite and S^3 illite have previously been interpreted as contemporaneous, S^3 illite is pseudomorphous after chlorite (sample Na1 36.1m, plain-transmitted light). b. Backscattered electron image of the area outlined in (a) showing the replacement of S^3 illite by C^{3b} chlorite. Abbreviations: C^{3b} Chl = C^{3b} chlorite, S^3 Ill = S^3 illite, Ur = uraninite.

(Tables 1–2, Fig. 8). The most common chlorite variety in the outer alteration halo, C^{2a} chlorite (Fig. 4), has a homogeneous, low iron content despite its optical and textural diversity (Table 1, Fig. 8). It also has an unusually low total octahedral occupancy, which may be as low as 10.5 per unit formula (Table 1). These low total octahedral contents are commonly observed in chlorite associated with alteration around unconformity-related U deposits (Ewers and Ferguson, 1980; Wilde and Wall, 1987; Nutt, 1989) and are ascribed to interlayering between trioctahedral and ditrioctahedral chlorite (Kominou and Sverjensky, 1995b). Temperature estimates calculated

from this chlorite phase using the methods of Cathelineau and Nieva (1985) and Cathelineau (1988) indicate that C^{2a} chlorite formed at ca. 200°C (Table 1).

C^{2b} chlorite is enriched in iron compared to the C^{2a} chlorite and more closely resembles unaltered biotite in terms of Fe/Al and Mg/Al ratios (Fig. 8). Unlike the homogeneous C^{2a} chlorite, C^{2b} chlorite is chemically heterogeneous and includes both Fe- and Mg-dominated chlorite end members (Fig. 8). Temperature estimates calculated from this chlorite phase are not reliable (e.g., temperatures range from 189°–312°C within a single chlorite lath).

Electron microprobe analyses indicate that most S^2 sericite grains are relatively pure, although anomalous FeO and MgO concentrations up to 3.5 and 4.4 wt percent, respectively, are recorded in some grains (Table 2; Fig. 9a). Similar chemical compositions are reported for sericite from Koongarra where it is proposed that high Fe and Mg contents and octahedral occupancy exceeding 4 per formula unit (pfu) are due to mixtures of S^2 sericite and C^{2a} chlorite (Kominou and Sverjensky, 1996). This is supported by some of our microprobe results (Fig. 9a–b), but high K^+ and low stoichiometric H_2O contents of some S^2 sericite indicate that chlorite-sericite mixtures are unlikely to account for all of the anomalous values observed.

S^3 illite within the ore zone has FeO and MgO contents up to 2.85 and 4.74 wt percent, respectively (Table 2). The Si/Al ratio of this illite is less than 3:1, but the overall composition has previously been referred to as phengite (Wilde and Wall, 1987). All S^3 illite samples have H_2O values that exceed the ideal value of ca. 6 wt percent, indicating that pure S^3 illite is rare (Kotzer and Kyser, 1991; Table 2, Fig. 9a–b). The most altered S^3 illite has FeO and MgO contents up to 6.34 and 7.96, respectively, as well as abnormally low K_2O (Fig. 9a–b). These values are most likely the result of retrograde alteration to C^3 chlorite or kaolinite. Temperature estimates calculated from X-ray diffraction (XRD) patterns using the scheme developed by Kubler (1967) and Arkai (1991) on samples that contain the most pristine S^3 illite indicate formation at ca. 180° ± 25°C, with one sample indicating formation at ca. 230° ± 30°C.

Electron microprobe analysis shows that C^{3a} chlorite in the inner alteration halo has a homogeneous, low Fe composition (Fig. 8) that formed at ca. 310°C (Cathelineau and Nieva, 1985; Cathelineau, 1988; Table 1). C^{3b} chlorite has a relatively homogeneous Fe-rich composition (Fig. 8, Table 1) and formed at approximately 280°C. C^{3c} chlorite has a lower Fe composition than C^{3b} chlorite but a higher Mg composition than C^{3a} chlorite and formed at ca. 210°C (Cathelineau and Nieva, 1985; Cathelineau, 1988; Table 1). In thin section, C^{3c} chlorite appears to be less common than C^{3b} chlorite but electron microprobe data for some chlorite samples optically identified as C^{3b} chlorite are similar to results for chlorite positively identified as C^{3c} chlorite. This discrepancy highlights the difficulty in distinguishing these two chlorites (Fig. 8).

Fluid Inclusions

Previous fluid inclusion studies at Nabarlek (Ypma and Fuzikawa, 1980; Wilde et al., 1989) demonstrated the existence of a number of different fluid inclusion generations,

TABLE 1. Electron Microprobe Analyses of Representative Nabarlek Chlorite Phases, Including Formation Temperatures Calculated Using the Method of Cathelineau and Nieva (1985) and Cathelineau (1988)

Sample	1	2	3	4	5	6	7
Oxide (wt %)							
SiO ₂	34.80	32.65	32.57	26.18	30.33	28.15	30.78
Al ₂ O ₃	28.16	27.43	18.69	20.31	25.01	18.69	20.28
FeO	6.60	3.86	8.27	26.49	4.56	24.86	12.26
MnO	<0.01	0.12	0.13	0.33	<0.01	0.03	0.09
MgO	17.76	20.48	26.83	13.29	27.00	15.34	19.66
CaO	0.06	0.19	0.08	<0.01	0.03	0.04	0.12
Na ₂ O	<0.01	<0.01	<0.01	<0.01	0.02	<0.01	0.04
K ₂ O	<0.01	0.13	0.06	<0.01	0.03	<0.01	0.13
Total	87.38	84.86	86.63	86.60	86.98	87.11	83.36
Calculated on the basis of 28 oxygens							
Tetrahedral sites							
Si	6.43	6.18	6.28	5.62	5.70	5.92	6.28
Al ^{IV}	1.57	1.82	1.72	2.38	2.30	2.08	1.72
S	8	8	8	8	8	8	8
Octahedral sites							
Al ^{VI}	4.57	4.30	2.52	2.75	3.24	2.56	3.15
Fe ²⁺	1.02	0.61	1.33	4.75	0.72	4.37	2.09
Mn	—	0.02	0.02	0.06	0.00	0.01	0.02
Mg	4.90	5.78	7.71	4.25	7.56	4.81	5.98
S	10.5	10.7	11.6	11.8	11.5	11.8	11.2
Interlayer sites							
K	—	0.03	0.01	—	0.01	—	0.03
Ca	0.01	0.04	0.02	—	0.01	0.01	0.03
Na	—	0.00	—	—	0.01	—	0.02
S	0.01	0.07	0.03	—	0.02	0.01	0.08
Al ^{IV} /2	0.78	0.91	0.86	1.19	1.15	1.04	0.86
Est. temp	190	231	216	322	309	272	215

Notes: Chlorite types with drill hole number and depth in parenthesis: 1 = C^{2a} chlorite (Na26 95.5m); 2 = C^{2a} chlorite (Na4 41.3m); 3 = C^{2b} chlorite (Na26 80.4m); 4 = C^{2b} chlorite (Na26 95.5m); 5 = C^{3a} chlorite (Na1 36.1m); 6 = C^{3b} chlorite (Na1 36.1m); 7 = C^{3c} chlorite (Na1 36.1m); location of samples is shown in Figures 2 and 3; — = no data

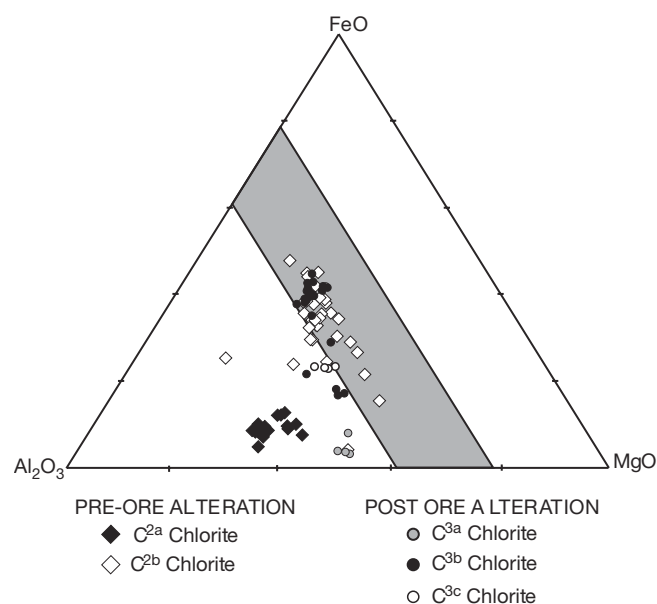


FIG. 8. Ternary diagram showing the chemical variation of different chlorite types (in wt %) around the Nabarlek deposit. Shaded area represents the field occupied by pristine biotite assuming the molecular formula for the most common biotite species presented in Deer et al. (1992).

including single-, two-, and multiphase fluid inclusions. The conclusions of these studies were that uranium mineralization formed from complex $\leq 200^\circ\text{C}$ brines.

Description of fluid inclusions

In this study, we describe four fluid inclusion types. Type A fluid inclusions contain >40 vol percent vapor and no daughter minerals (Fig. 10). Type B fluid inclusions contain <40 vol percent vapor and at least two daughter minerals or trapped solids including halite. The solids may constitute up to 80 percent of the fluid inclusion volume. Daughter minerals and trapped solids found in various studies of fluid inclusions at Nabarlek include dolomite, chlorite, white mica, calcite, hematite, and anhydrite (Fig. 10; Ypma and Fuzikawa, 1980; Wilde et al., 1989). Type C fluid inclusions contain <20 vol percent vapor and no daughter minerals (Fig. 10). Type D fluid inclusions contain <20 vol percent vapor and only halite daughter minerals.

Fluid inclusion types A and B occur as randomly distributed three-dimensional clusters in Q¹ quartz and are interpreted as primary (Fig. 10). They are also observed in corroded, metamorphic quartz grains in the outer alteration halo in the Hanging-Wall schist, thereby suggesting that these fluid inclusions were present prior to desilicification that

TABLE 2. Electron Microprobe Analyses of Representative S² Sericite and S³ Illite Displaying Variable Alteration to C³ Chlorite or Kaolinite

Sample	1	2	3	4	5	6	7	8
Oxide (wt %)								
SiO ₂	46.9	47.5	49.6	49.2	48.0	46.2	47.6	49.3
Al ₂ O ₃	34.7	33.9	29.4	30.1	29.4	23.5	27.8	21.2
FeO	2.5	1.1	3.1	1.3	2.0	6.3	1.9	0.3
MnO	0.1	0.2	0.2	<0.01	<0.01	0.3	<0.01	<0.01
MgO	0.5	0.7	3.0	3.2	3.6	2.8	2.5	8.0
CaO	0.1	0.1	0.1	0.1	0.1	0.3	0.2	<0.01
Na ₂ O	<0.01	0.1	<0.01	0.2	0.3	<0.01	<0.01	<0.01
K ₂ O	11.0	11.0	7.7	8.7	8.3	4.9	7.4	7.4
H ₂ O	4.2	5.5	7.0	6.8	8.0	15.6	12.7	9.6
Total	100.0	100.0	100.0	99.7	99.7	99.9	99.9	95.8
Calculated on the basis of 22 oxygens								
Tetrahedral sites								
Si	6.2	6.4	6.7	6.6	6.6	6.9	6.8	7.1
Al ^{IV}	1.8	1.6	1.6	1.6	1.7	1.6	1.4	1.3
S	8.1	8.0	8.3	8.2	8.3	8.5	8.2	8.4
Octahedral sites								
Al ^{VI}	3.6	3.7	3.0	3.2	3.0	2.5	3.3	2.3
Fe ²⁺	0.3	0.1	0.4	0.2	0.2	0.8	0.2	—
Mn	—	—	—	—	—	—	—	—
Mg	0.1	0.1	0.6	0.7	0.7	0.6	0.5	1.7
S	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
K ⁺	1.9	1.9	1.3	1.5	1.5	0.9	1.3	1.3
Ca ²⁺	—	—	—	—	—	—	—	—
Na ⁺	—	—	—	0.1	0.1	—	—	—
S	1.9	1.9	1.3	1.6	1.5	1.0	1.4	1.3

Notes: Illite types with drill hole number and depth in parenthesis: 1 = S² sericite (Na26 95.5m); 2 = S² sericite (Na26 95.5m); 3 = altered S² sericite (Na83 52.6m); 4 = weakly altered S³ illite (Na1 36.1m); 5 = weakly altered S³ illite (Na1 36.1m); 6 = strongly altered S³ illite (Na4 40.5m); 7 = strongly altered S³ illite (Na4 40.5m); 8 = moderately altered S³ illite (Na4 43.1m); — = no data; location of samples is shown in Figures 2 and 3

accompanied mineralization. Type A and B fluid inclusions occasionally occur along secondary trails in Q¹ quartz and were possibly trapped within an active fault or shear zone (cf. Boullier and Robert, 1992). Therefore, type A and B fluid inclusions may be the product of immiscibility rather than necking down and fracturing as suggested by Ypma and Fuzikawa (1980). In other well-documented fluid inclusion investigations, end-member fluids can partition into different sets of healed microfractures during phase separation owing to different wetting properties (Watson and Brenan, 1987; Boullier and Robert, 1995).

Fluid inclusion types C and D are generally found in randomly distributed three-dimensional clusters in Q^{3a} quartz and along primary growth zones in late, clear, euhedral Q^{3b} quartz (Fig. 10). They are primary fluid inclusions in Q^{3a} and Q^{3b} veins (cf. Roedder, 1984) but occur along secondary trails in the preore Q¹ quartz veins (Fig. 10). Type D fluid inclusions formed later than the type C fluid inclusions. In one sample, Na 83 93.3m, a later generation of type B fluid inclusions was observed along primary growth planes together with type C and D fluid inclusions in clear euhedral quartz.

Microthermometry

Type A fluid inclusions have eutectic temperatures indicating a pure NaCl solution (Table 3). Some type A fluid inclusions contain a relatively pure CO₂ vapor phase (Table 3). CO₂ clathrates melt between 9.2° and 10.6°C, indicating

salinities of ≤1 wt percent NaCl equiv. This is in agreement with final ice melting temperatures (T_m) that also indicate low salinities (Fig. 11). Type A fluid inclusions homogenize into the liquid or vapor phase between 320° and 392°C and occasionally exhibit critical behavior by fading of the meniscus at ca. 377°C (Table 3). Type B fluid inclusions that coexist with type A fluid inclusions infrequently form hydrohalite that begins to melt close to the NaCl eutectic temperature and persists up to 25.9°C. Many type B fluid inclusions decrepitate during heating, but when complete homogenization was achieved, temperatures ranged between 326° and 392°C (Fig. 11; Table 3). Salinity estimates calculated from the homogenization temperature of the halite daughter minerals range from 27.4 to 43.3 wt percent NaCl equiv.

Type C fluid inclusions have eutectic temperatures from −69.1° to −21.4°C (Table 3). Final melting temperatures indicate salinities ranging from 2.2 to 34.1 wt percent NaCl equiv (Fig. 11). Two populations are evident. An older type C generation occurs close to the vein walls in Q^{3a} quartz, has low salinities, and is dominated by MgCl₂-NaCl brines. The younger type C generation primarily occurs in Q^{3b} quartz and has final melting temperatures that indicate highly saline brines dominated by CaCl₂-MgCl₂-KCl-NaCl. Homogenization temperatures for the MgCl₂-NaCl fluid inclusions mostly cluster around 165° ± 25°C. Using the pressure correction curves of Potter (1977) and the lithostatic pressures estimated using the thickness for the overlying sediments ranging

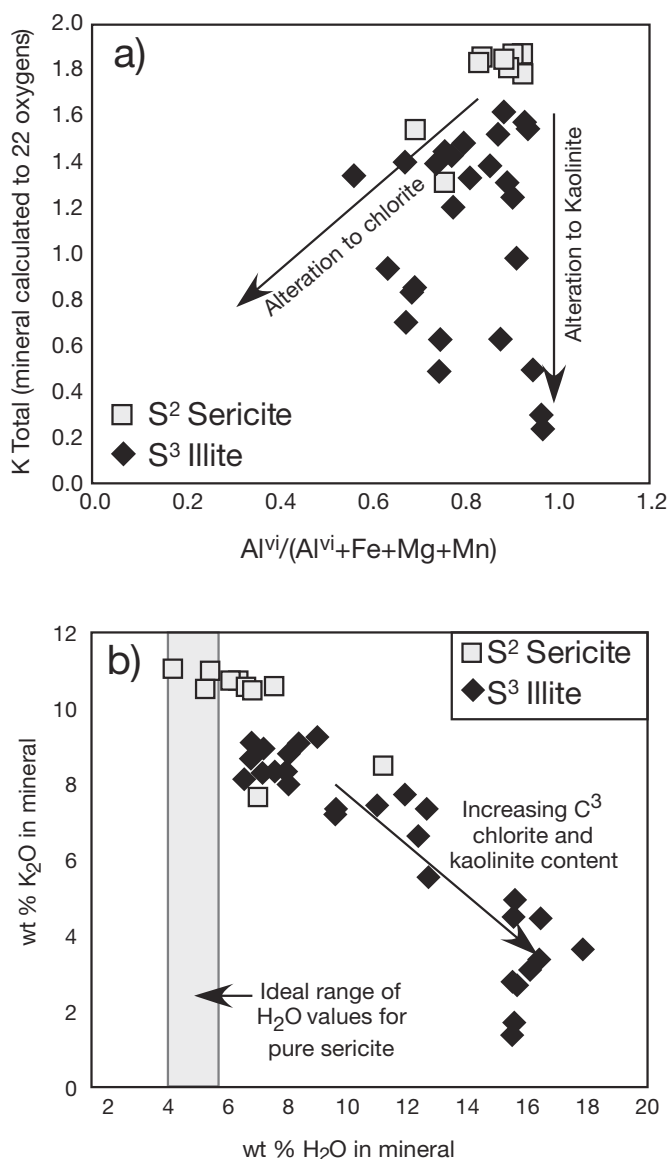


FIG. 9. a. Relationship between total K and octahedral cation ratios in S^2 sericite and S^3 illite. Effects of alteration to late, postmineralization C^3 chlorite and recent kaolinite are indicated. Elements expressed in units per weight formula calculated to 22 oxygens. b. Relationship between wt % K_2O and wt % H_2O in S^2 sericite and S^3 illite. Few samples are unaltered as indicated by the loss of K^+ and increase in H_2O .

between 2,780 and ca. 5,000 m, these veins formed at ca. $230^\circ \pm 30^\circ$ to $260^\circ \pm 35^\circ C$. In contrast, the $CaCl_2$ - $MgCl_2$ - KCl - $NaCl$ fluid inclusions exhibit a range of homogenization temperatures between ca. 95° and $170^\circ C$ (Fig. 11). These fluid inclusions form a trend that is consistent with mixing between two fluids (Shepherd et al., 1985). In such a scenario, it is difficult to calculate an exact formation temperature for vein formation. However, it is most likely that these veins formed between ca. $165^\circ \pm 30^\circ$ and $270^\circ \pm 30^\circ C$ based on the pressure correction mentioned above.

Type D fluid inclusions have eutectic temperatures indicating the presence of a $CaCl_2$ - $MgCl_2$ - KCl - $NaCl$ brine (Table 3). Salinities calculated from the dissolution temperature of the halite daughter minerals equate to ca. 28.7 wt percent $NaCl$ equiv, similar to the younger, high-salinity type C fluid inclusions (Fig. 11). Complete homogenization was achieved by vapor disappearance at 134° and $158^\circ C$ and by halite disappearance at $168^\circ C$ (Table 3). Entrapment temperatures for these fluid inclusions fall within the range calculated above for the younger type C fluid inclusions.

Four type B fluid inclusions were observed at the tips of one Q^{3b} quartz vein (Table 3). Ordinarily, this may be an indication that the type B fluid inclusions occurring in the preore Q^1 quartz veins were associated with a late-stage fluid and contemporaneous with the high-salinity, type C and/or D fluid inclusions. However, these four fluid inclusions all formed ice that has eutectic temperatures indicative of $CaCl_2$ - $MgCl_2$ - KCl - $NaCl$ brines, similar to the saline type C and/or D fluid inclusions but distinct from the type B fluid inclusions in the Q^1 quartz veins (Table 3). Homogenization of the vapor phase in the late type B fluid inclusions ranges from 106° to $137^\circ C$, but few of the daughter minerals reduced in size during heating and decrepitation was observed between 262° and $360^\circ C$. In one type B fluid inclusion, dissolution of the daughter minerals occurred at 78° and $137^\circ C$. Salinity estimates indicate that this fluid inclusion may belong to the population identified as the late type C and D fluid inclusions (Fig. 11).

Stable Isotope Compositions of the Alteration Minerals

Q^1 vein quartz has $\delta^{18}O$ values between 8.9 and 12.6 per mil, which are similar to that reported by Ypma and Fuzikawa (1980) for early-stage quartz veins (Table 4). Assuming formation temperatures of ca. $370^\circ C$, the Q^1 veins formed from fluids with $\delta^{18}O_{fluid}$ values of 4.1 to 7.9 per mil and δD_{fluid} values of -48 and -32 per mil (Fig. 12). Two metamorphic muscovite separates, representative of the metamorphic mineral assemblage that reached middle to upper amphibolite grade during the Barramundi-Top End orogeny, record $\delta^{18}O_{fluid}$ values of 7.4 per mil and δD_{fluid} values between -42 and -30 per mil (Table 4; Fig. 12).

Preore $C^{2a/b}$ chlorite has $\delta^{18}O$ values from 4.3 to 7.5 per mil, δD values from -65 to -42 per mil, and water contents of 10.5 to 13.5 wt percent H_2O (Table 4). The values indicate $\delta^{18}O_{fluid}$ values of 2.0 to 5.2 per mil and δD_{fluid} values of -15 to -36 per mil based on a temperature of $200^\circ C$ from chlorite geothermometry (Fig. 12).

Two S^2 sericite samples have $\delta^{18}O$ values of 6.3 and 8.1 per mil, δD values of -55 and -54 per mil, and water contents of 4.9 and 8.9 wt percent H_2O (Table 4). Water contents greater than 5 wt percent H_2O generally indicate incorporation of retrograde molecular water in the sericite, which can change the δD and $\delta^{18}O$ value of the mineral depending on the degree of alteration (Graham, 1981; Wilson et al., 1987; Kotzer and Kyser, 1991). Mass-balance calculations that correct for the incorporation of postore retrograde water, which is estimated to be ca. -50 per mil (the δD_{fluid} value obtained from Q^3 quartz veins; see below), indicate that the isotopic composition of the fluid in apparent equilibrium with this sericite was 2.0 per mil in $\delta^{18}O$ and -38 to -25 per mil in δD , given formation at ca. $200^\circ C$ (Fig. 12).

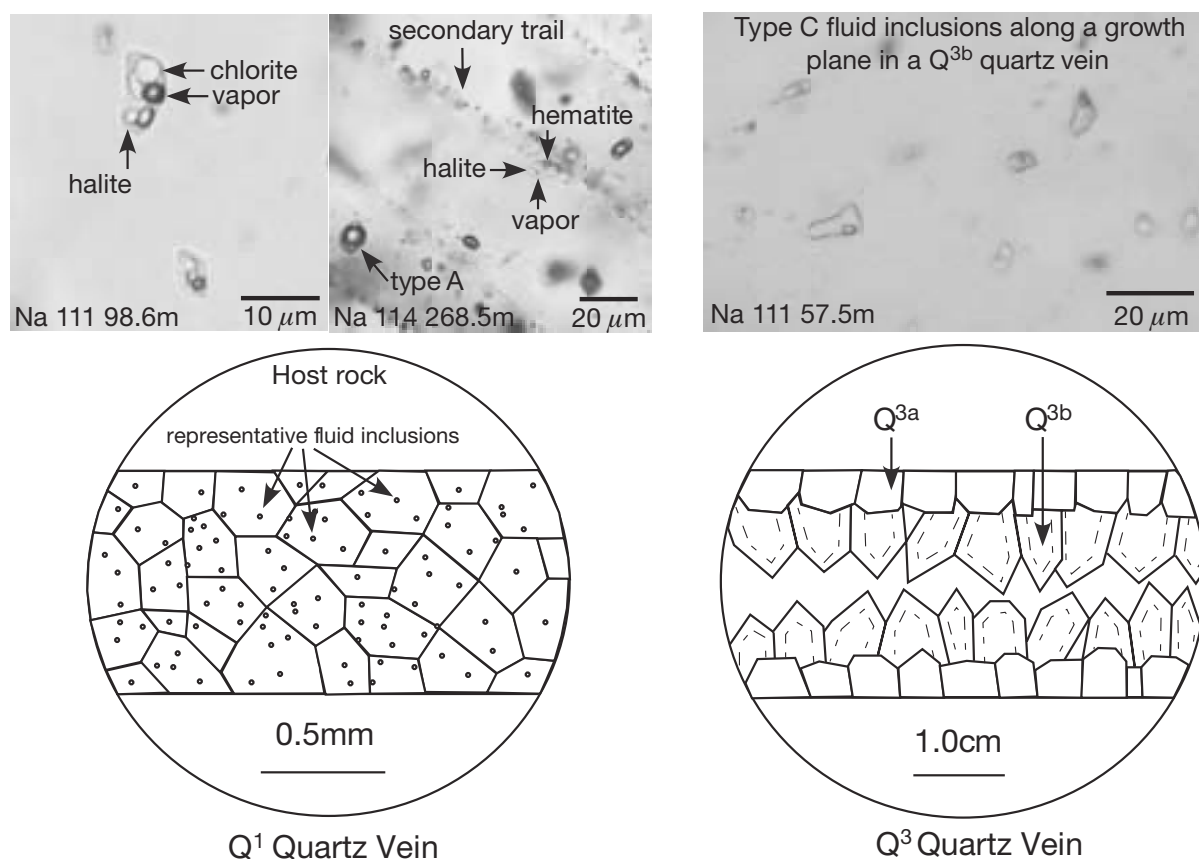


FIG. 10. A schematic representation of the two types of quartz veins used for fluid inclusion microthermometry at Nabarlek and the fluid inclusions that they host. a. Q¹ quartz with anhedral, interlocking quartz grains, host primary, type A and B fluid inclusions and trails of secondary type C and D fluid inclusions. Daughter minerals that occur in the type B fluid inclusions include halite, dolomite, chlorite, white mica, calcite, hematite, and anhydrite (Ypma and Fuzikawa, 1980). b. Q³ quartz veins, divided into massive, anhedral early quartz (Q^{3a}) and sparry euhedral quartz with clearly defined growth zones (Q^{3b}) host liquid-rich type C and D fluid inclusions.

Synore S³ illite is difficult to separate from postore C³ chlorite and kaolinite. However, one pure separate was obtained from a relatively low grade sample (Na 168 29.0m, 0.1% U₃O₈) having a $\delta^{18}\text{O}$ value of 8.5 per mil, a δD value of -59 per mil, and a water content of 7 wt percent H₂O. Using the same mass-balance calculations mentioned above to account for the effects of alteration by retrograde molecular water, S³ illite formed from a brine with a $\delta^{18}\text{O}_{\text{fluid}}$ value of 2.8 per mil and a $\delta\text{D}_{\text{fluid}}$ value of -32 per mil at ca. 200°C (Fig. 12).

Two S³ illite separates from high-grade U ore samples containing ca. 30 and ca. 50 percent late kaolinite, respectively, were analyzed to better constrain the isotopic value of the ore-forming fluid (Table 4). The two samples have water contents and isotopic values that cannot be explained by the presence of kaolinite alone and suggest that a low-temperature, low- to mid-latitude meteoric water exchanged hydrogen and oxygen with the interlamellar water and the octahedral hydroxyl groups of the illite without completely altering the crystallography of the illite (Graham, 1981; Wilson et al., 1987; Kotzer and Kyser, 1991). This process can be expected

during weathering and at high water/rock ratios (Kotzer and Kyser, 1991).

Four $\delta^{18}\text{O}$ values from Nabarlek uraninite were obtained by microdrilling thick sections of uraninite and analyzing the powders by the fluorination technique described by Kotzer and Kyser (1991). These samples have $\delta^{18}\text{O}$ values ranging from -5.3 to +1.1 per mil (Table 4). Ten in situ oxygen isotope analyses by high-resolution ion microprobe (Fayek et al., 2002) reveal that homogeneous uraninite (Fig. 6c) has $\delta^{18}\text{O}$ values between -13.9 and -8.9 per mil, whereas heterogeneous uraninite (Fig. 6d) has $\delta^{18}\text{O}$ values between -13.4 and -1.2 per mil. Using the experimental uraninite-water fractionation factor for temperatures between 100° and 200°C (Fayek and Kyser, 2000), the uranium-bearing brine had $\delta^{18}\text{O}_{\text{fluid}}$ values ranging from -2.9 to +12.1 per mil (Table 4). Few uraninites have the $\delta^{18}\text{O}_{\text{fluid}}$ values of 3.5 ± 2 per mil calculated from the phyllosilicate minerals in the alteration halo or from S³ illite that is intergrown with the uraninite. This is thought to be due to the incorporation of SiO₂ and CaO in the uraninite structure (Table 5) as a result of alteration to coffinite, which produces high $\delta^{18}\text{O}_{\text{fluid}}$ values

TABLE 3. Fluid Inclusion Microthermometry Results from Q¹ and Q³ Quartz Veins

Sample ¹	Description	T _m (CO ₂)	T _h (CO ₂)	T _e (ice)	T _m (ice)	T _m (hyd)	T _h (clath)	NaCl equiv	T _h (Vp)	T _h (H)	T _h (S1)	T _h (S2)	T _h (S3)
Type A													
Na114 268.5m	90% Vp			n.d.	n.d.	n.d.			336(L)				
Na114 268.5m	80% Vp	-56.9	25.2				10.1	0.0	320(L)				
Na114 268.5m	75% Vp	-57.1	29.2				10.6	0.0	325(L)				
Na114 268.5m	80% Vp			-20.2	-1.5			2.6	392(V)				
Na114 268.5m	85% Vp	-57.6	26.8	-20	0.1		n.d.	0.0	375(C)				
Na114 268.5m	80% Vp			-18.6	0.1			0.0	373(V)				
Na114 268.5m	70% Vp			-19.7	0.1			0.0	375(L)				
Na114 268.5m	70% Vp			-18.9	-0.1			0.2	379(C)				
Na111 57.5m	70% Vp	-56.7	n.d.	-16.2			9.8	0.1	379(L)				
Na111 57.5m	80% Vp	-56.7	n.d.	-16			9.2	1.0	354(L)				
Na111 57.5m	65% Vp			-18.3	-0.5			0.9	338(L)				
Na111 57.5m	40% Vp			-21.5	-3.8			6.2	383(L)				
Na111 98.6m	90% Vp	-56.8	n.d.	-40.1			10.1	0.0	324(D)				
Na111 98.6m	80% Vp	-56.8	n.d.	-20.4			10.0	0.0	349(L)				
Na83 372.8m	70% Vp			-26.5	-0.2			0.4	385(D)				
Na83 372.8m	70% Vp			-14	0.1			0.0	382(V)				
Na77 61.7m	60% Vp			-19.9	0.1			0.0	361(L)				
Na77 61.7m	60% Vp			-22.5	0.2			0.0	370(L)				
Na77 61.7m	70% Vp			-31.8	-0.4			0.7	378(C)				
Type B													
Na114 268.5m	10% Vp, + 2 dm			n.d.	n.d.	n.d.		32.2	151(L)	208	334		
Na114 268.5m	10% Vp, + 2 dm			n.d.	n.d.	n.d.		35.9	261(L)	270	326		
Na114 268.5m	15% Vp, + 3 dm			n.d.	n.d.	n.d.		41.0	301(L)	216	335	354	
Na114 268.5m	10% Vp, + 3 dm			n.d.	n.d.	n.d.		28.2	306(L)	108	333	375	
Na111 98.6m	20% Vp, + 4 dm			n.d.	n.d.	n.d.			450(D)	450(D)	397	450(D)	450(D)
Na111 98.6m	8% Vp, + 3 dm			n.d.	n.d.	n.d.			165(L)	409(D)	409(D)	409(D)	
Na111 98.6m	8% Vp, + 3 dm			-22.7		25.9		28.6	133(L)	102	295	375	
Na111 98.6m	10% Vp, + 2 dm			-23.1		23.1		29.5	136(L)	147	305(D)		
Na111 98.6m	10% Vp, + 2 dm			-22.6		9.5		29.1	109(L)	136	360		
Na111 98.6m	10% Vp, + 3 dm			-18.6		16.8		27.6	115(L)	86	130	295(D)	
Na111 98.6m	15% Vp, + 3 dm			n.d.	n.d.	n.d.		30.9	327(L)	179	350	392	
Na111 98.6m	8% Vp, + 3 dm			n.d.	n.d.	n.d.		43.3	126(L)	360	388	Hematite	
Type C													
Na111 57.5m	15% Vp			-57.9	-41.3			33.9	115(L)				
Na111 57.5m	15% Vp			-21.9	-2.7			4.5	169(L)				
Na111 57.5m	15% Vp			-35.5	-1.6			2.7	190(L)				
Na111 57.5m	15% Vp			-66.2	-34.1			31.0	116(L)				
Na111 57.5m	8% Vp			-21.6	-1.8			3.1	160(L)				
Na111 57.5m	8% Vp			-21.7	-1.8			3.1	156(L)				

(>5.5‰; Kotzer and Kyser, 1993; Fayek and Kyser, 1997; Fayek et al., 2002). On the other hand, low $\delta^{18}\text{O}_{\text{fluid}}$ values (<1.5‰) are most likely due to the reequilibration of uraninite with late meteoric water, which in northern Australia ranges from -3 to -7 per mil (IAEA, 2001, GNIP Maps and Animations, International Atomic Energy Agency, Vienna: <http://isohis.iaea.org>). Samples that have calculated $\delta^{18}\text{O}_{\text{fluid}}$ values closest to those indicated by chlorite and sericite from the inner and outer alteration zone exhibit relatively limited alteration by postmineralization fluid (Fig. 6c) and have low SiO₂ and CaO concentrations (Table 5).

Two postore Q³ quartz veins from within the Oenpelli Dolerite have $\delta^{18}\text{O}$ values of 14.8 and 16.6 per mil. These veins suggest that brines with $\delta^{18}\text{O}_{\text{fluid}}$ values between 0.1 and 1.9 per mil and $\delta\text{D}_{\text{fluid}}$ values of -54 and -53 per mil were responsible for vein formation at a temperature of 165°C (Table 4; Fig. 12). The low $\delta^{18}\text{O}_{\text{fluid}}$ and $\delta\text{D}_{\text{fluid}}$ values indicate formation from a moderately evolved meteoric fluid (Longstaffe, 1987, 2000). However, the low- to high-salinity type C fluid inclusions trapped in these veins suggest that a contribution from an evaporite source such as those found in the McKay

Sandstone or the Cottee Formation cannot be discounted (Sweet et al., 1999b).

One postore kaolinite sample extracted from a piece of high-grade U ore (Na 4 40 m) has a $\delta^{18}\text{O}$ value of 14 per mil, a δD value of -63 per mil, and a water content of 16 wt percent H₂O. Assuming formation at <40°C, this kaolinite formed from low-latitude meteoric water with a $\delta^{18}\text{O}_{\text{fluid}}$ value of -7 per mil and a $\delta\text{D}_{\text{fluid}}$ value of -41 per mil (Fig. 12), equivalent to values obtained from modern meteoric water in northern Australia (IAEA, 2001, GNIP Maps and Animations, International Atomic Energy Agency, Vienna: <http://isohis.iaea.org>).

U-Pb and Pb-Pb Isotope Systematics

Uraninite grains analyzed by LA-HR-ICPMS have ²⁰⁷Pb/²⁰⁶Pb ages that range from 718 to 1642 Ma (Table 6). Distinct modes occur at ca. 900, 1100, and 1360 Ma (Fig. 13). The oldest age of 1642 Ma was obtained from highly reflective homogeneous uraninite (Fig. 6c). The younger ages are common in all samples analyzed and are associated with less reflective and often heterogeneous uraninite grains (Fig. 6d). A similar spread of ²⁰⁷Pb/²⁰⁶Pb ages has been reported from

TABLE 3. (Cont.)

Sample ¹	Description	T _{m(CO₂)}	T _{h(CO₂)}	T _{e(ice)}	T _{m(ice)}	T _{m(hyd)}	T _{h(clath)}	NaCl equiv	T _{h(Vp)}	T _{h(H)}	T _{h(S1)}	T _{h(S2)}	T _{h(S3)}
Na111 57.5m	10% Vp			-21.4	-1.7			2.9	163(L)				
Na111 57.5m	8% Vp			-21.6		5.7			144(L)				
Na83 99.2m	8% Vp			-60.2	-25.5			26.0	137(L)				
Na83 99.2m	15% Vp			-65.8	-24.7			25.5	134(L)				
Na83 99.2m	8% Vp			-30.8	-1.3			2.2	250(L)				
Na83 99.2m	8% Vp			-65.3	-26.2			26.9	158(L)				
Na83 99.2m	8% Vp			-62	-22.8			24.9	159(L)				
Na83 372.8m	8% Vp			-68	-20.6			22.8	159(L)				
Na83 372.8m	10% Vp			-65	-15.8			19.3	171(L)				
Na83 372.8m	10% Vp			-23.6	-4.7			7.5	219(L)				
Na83 372.8m	10% Vp			-24	-2.5			4.2	176(L)				
Na77 61.7m	15% Vp			-57.2	-20			22.4	149(L)				
Na77 61.7m	10% Vp			-59.3	-26.2			26.9	139(L)				
Na77 61.7m	10% Vp			-68	-26.2			26.9	123(L)				
Na77 61.7m	10% Vp			-69	-23.9			25.3	134(L)				
Na83 93.3m	10% Vp			-67.5	-37			33.0	94(L)				
Na83 93.3m	15% Vp			-67	-41.6			34.1	109(L)				
Na83 93.3m	10% Vp			-65.5	-35.9			31.9	133(L)				
Na83 93.3m	10% Vp			-62.5	-35.2			31.7	127(L)				
Na83 93.3m	10% Vp			-69.1	-31.4			29.6	148(L)				
Na83 93.3m	15% Vp			-67	-29.8			28.2	137(L)				
Type D													
Na111 98.6m	20% Vp, + 1 dm			n.d.	n.d.	n.d.			265(D)				
Na83 99.2m	15% Vp, + 1 dm			-56.4	-30.2			29.0	134(L)	124			
Na83 99.2m	5% Vp, + 1 dm			-60.2	-22.6			22.8	158(L)	123			
Na83 93.3m	10% Vp + 1 dm			-60.9	-36			31.5	140(L)	169			
Type B "late"													
Na83 93.3m	15% Vp + 2 d.m.			-61	-40.8			33.5	124(L)		234(D)	234(D)	
Na83 93.3m	15% Vp + 2 d.m.			-68.5	-40.9			33.5	117(L)		262(D)	262(D)	
Na83 93.3m	5% Vp + 2 d.m.			-67.6	-32			29.9	137(L)		350(D)	350(D)	
Na83 93.3m	15% Vp + 2 d.m.			-65	-33.5			30.9	106(L)		366(D)	366(D)	
Na114 268.5m	15% Vp, + 3 dm			n.d.	n.d.	n.d.		27.4	149(L)	78	138	Hematite	

¹ Drill hole number and depth

Abbreviations: D = fluid inclusion decrepitated at this temperature; dm = daughter minerals; n.d. = not determined; T_{m(CO₂)} = melt temperature in °C of CO₂ phase; T_{h(CO₂)} = homogenization temperature in °C of CO₂ phase; T_{e(ice)} = initial melt temperature in °C of ice; T_{m(ice)} = final melt temperature in °C of ice; T_{m(hyd)} = melt temperature in °C of hydrate; T_{h(clath)} = melt temperature in °C of CO₂ clathrate; NaCl equiv = wt percent NaCl equiv; T_{h(Vp)} = homogenization temperature in °C of vapor and liquid [(V) = homogenization into the vapor phase, (L) = homogenization into the liquid phase, (C) = critical behavior, vapor displays fading of the meniscus, (D) =]; Th(H) = homogenization temperature in °C of halite; T_{h(S1)} = homogenization temperature in °C of first solid; T_{h(S2)} = homogenization temperature in °C of second solid; T_{h(S3)} = homogenization temperature in °C of third solid

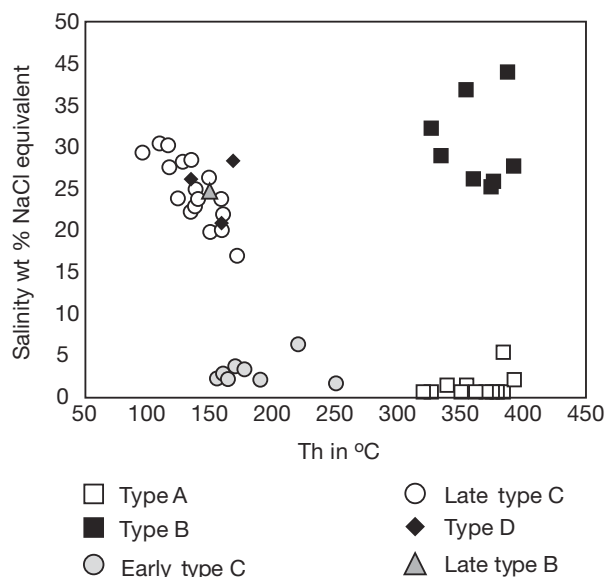


FIG. 11. Homogenization versus salinity plot for fluid inclusions from Nabarlek (Table 3). Four distinct populations are evident. Type A and B fluid inclusions are likely related, having overlapping homogenization temperatures, whereas type C and D fluid inclusions display two distinct fluid types (see text for details). The high salinities of type C and D fluid inclusions indicate that mixing with a third low-temperature, saline population cannot be ruled out.

TABLE 4. Stable Isotope Values and Water Contents of Quartz, Chlorite, Sericite, Phengite, and Kaolinite Varieties from the Nabarlek Deposit

DDH no.	Depth (m)	Mineral analyzed	Paragenesis	$\delta^{18}\text{O}_{\text{min}}$	$\delta\text{D}_{\text{min}}$	H_2O yield	Temp (°C)	$\delta^{18}\text{O}_{\text{fluid}}$	$\delta\text{D}_{\text{fluid}}$
NA26	80.4	Muscovite	Metamorphic	8.7	-60	5.7	500	7.4	-42
NA4	22.8	Muscovite	Metamorphic	8.7	-48	5.9	500	7.4	-30
NA83	372.8	Q ¹ quartz vein	Metamorphic	12.6	n.d.	n.d.	370	7.8	n.d.
NA83	1252	Q ¹ quartz vein	Metamorphic	10.6	n.d.	n.d.	370	5.8	n.d.
NA111	57.5	Q ¹ quartz vein	Metamorphic	8.9		0.2	370	4.1	-46
NA111	98.6	Q ¹ quartz vein	Metamorphic	11.4		0.2	370	6.6	-32
NA114	26.9	Q ¹ quartz vein	Metamorphic	12.7		0.2	370	7.9	-48
NA4	13.6	C ^{2a} chlorite	Preore alteration	8.1	-50	12.9	200	5.8	-21
NA4	26.2	C ^{2a} chlorite	Preore alteration	6.6	-53	12.6	200	4.3	-24
NA83	365.5	C ^{2a} chlorite	Preore alteration	7.7	-54	12.7	200	5.4	-25
NA26	51.4	C ^{2a} chlorite	Preore alteration	5.8	-53	12.9	200	3.5	-24
NA26	87.4	C ^{2a} chlorite	Preore alteration	6	-58	13.5	200	3.7	-29
NA83	316.1	C ^{2a} chlorite	Preore alteration	6.4	-56	12.2	200	4.1	-27
NA83	368.5	C ^{2a} chlorite	Preore alteration	7.7	-65	12.7	200	5.4	-36
NA26	21	C ^{2b} chlorite	Preore alteration	6.6	-53	13.5	200	4.3	-26
NA26	80.4	C ^{2b} chlorite	Preore alteration	4.7	-42	12.7	200	2.4	-15
NA26	91.9	C ^{2b} chlorite	Preore alteration	4.8	-48	13.1	200	2.5	-21
NA26	97.1	C ^{2b} chlorite	Preore alteration	4.3	-54	13.2	200	2.0	-27
NA83	40.0	C ^{2b} chlorite	Preore alteration	7.5	-46	13.0	200	5.2	-19
NA83	299.8	C ^{2b} chlorite	Preore alteration	4.4	-57	12.8	200	2.1	-30
NA83	364.2	S ² sericite	Preore alteration	6.3	-54	8.9	200	2.0	-25
NA83	372.8	S ² sericite	Preore alteration	8.1	-55	4.9	200	2.0	-38
NA168	29	S ³ phengite	Synore alteration	8.5	-59	7.0	200	2.8	-32
NA4	43.1	50% kaolinite/50% S ³ illite	Syn/postore alteration	13.2	-60	9.4	200	n.d.	n.d.
NA88	29	30% kaolinite/70% S ³ illite	Syn/postore alteration	14	-63	8.8	200	n.d.	n.d.
NA83	93.3	Q ³ quartz veins	Postore alteration	14.8		0.2	165	-0.3	-54
NA83	99.2	Q ³ quartz veins	Postore alteration	16.6		0.2	165	1.5	-53
NA88	29.0	Uraninite	Synore mineral	1.1			200	12.1	
NA4	40.0	Uraninite	Synore mineral	-5.3			200	5.7	
NA4	40.0	Uraninite	Synore mineral	-2.0			200	9.0	
NA4	40.0	Uraninite	Synore mineral	-1.5			200	9.5	
NA4	40.0	Uraninite*	Synore mineral	-13.9			200	-2.9	
NA4	40.0	Uraninite*	Synore mineral	-8.9			200	2.1	
NA4	40.0	Uraninite*	Synore mineral	-11.6			200	-0.6	
NA4	40.0	Uraninite*	Synore mineral	-12.1			200	-1.1	
NA4	40.0	Uraninite*	Synore mineral	-11.5			200	-0.5	
NA1	n.d.	Uraninite*	Synore mineral	-5.8			200	5.2	
NA1	n.d.	Uraninite*	Synore mineral	-1.2			200	9.8	
NA1	n.d.	Uraninite*	Synore mineral	-6.8			200	4.2	
NA1	n.d.	Uraninite*	Synore mineral	-11.4			200	-0.4	
NA1	n.d.	Uraninite*	Synore mineral	-13.4			200	-2.4	
NA4	40	Kaolinite	Late weathering	14	-63	16.0	40	-7.0	-41

Notes: Stable isotope values of the fluid from which these minerals precipitated are calculated using temperatures estimated in this paper (see text for details); oxygen isotope fractionation factors used are those suggested by Eslinger and Savin (1973) for water-illite, Land and Dutton (1978) for water-kaolinite, Yeh and Savin (1976) for water-chlorite, O'Neil and Taylor (1969) for water-muscovite, Clayton et al. (1972) for water-quartz, and Fayek and Kyser (2000) for water-uraninite; hydrogen isotope fractionation factors used are those suggested by Suzuoki and Epstein (1976) for water-muscovite, Yeh (1980) for water-illite, Lambert and Epstein (1980) for water-kaolinite, and Graham et al. (1984, 1987) for water-chlorite.

Abbreviations: $\delta^{18}\text{O}_{\text{min}}$ = $\delta^{18}\text{O}$ value of the mineral analyzed; $\delta\text{D}_{\text{min}}$ = δD value of mineral analyzed; uraninite* = analyzed using a CAMECA IMS 1270 ion microprobe; H_2O yield = wt percent of H_2O released from mineral during heating; n.d. = not determined

unconformity-related U deposits in the Paleoproterozoic Athabasca basin (Kotzer and Kyser, 1993; Fayek and Kyser, 1997; Fayek et al., 2002). The two oldest $^{207}\text{Pb}/^{206}\text{Pb}$ ages (Table 6) are comparable to the 1616 ± 50 Ma Sm-Nd age obtained for Nabarlek ore deposition by Maas (1989) and are the minimum and most accurate ages for original uraninite

precipitation. The apparent episodic resetting of uraninite recorded by $^{207}\text{Pb}/^{206}\text{Pb}$ ages younger than 1620 Ma indicates that the U-Pb isotope systems have been partially to completely disturbed by subsequent events.

U-Pb discordia (Fig. 14) indicate ages similar to the $^{207}\text{Pb}/^{206}\text{Pb}$ ages at 948 ± 47 , 1178 ± 56 , 1393 ± 76 , and 1648

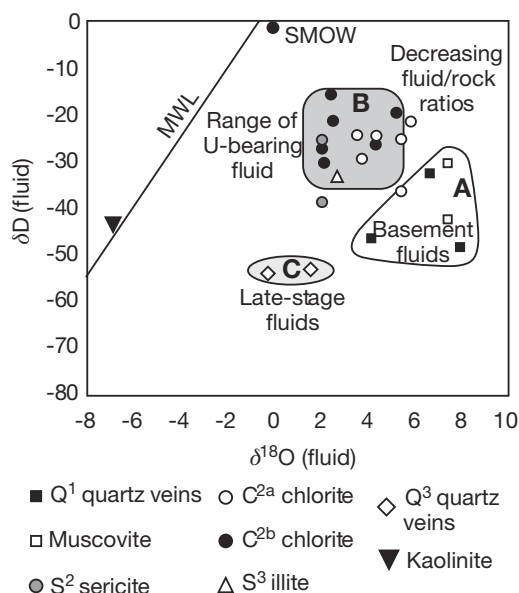


FIG. 12. Calculated $\delta^{18}\text{O}$ and δD values for various fluids associated with alteration at Nabarlek (Table 4). Area A represents the isotopic value of minerals in equilibrium with metamorphic (basement-derived) water (350°–500°C). Shaded area B represents the calculated isotopic value of the fluid that transported and deposited uranium in the Nabarlek fault (200°C). $\delta^{18}\text{O}$ values outside of area B indicate alteration at low fluid/rock ratios (high $\delta^{18}\text{O}$ values) or subsequent alteration by late-stage basinal fluids (light δD values). Area C corresponds to the fluid values calculated for late-stage fluids represented by Q^3 quartz veins. One kaolinite sample plots adjacent to the meteoric water line indicating recent formation at <40°C.

TABLE 5. Chemical Composition and Calculated Chemical U-Pb Ages from Electron Microprobe Analyses of Variably Altered Uraninite at Nabarelek

Sample no.	Spot no.	Mineral	$\delta^{18}\text{O}$ per mil ¹	SiO_2	PbO	UO_2	FeO	CaO	TiO_2	Total	Chem. age ² (Ma)	²⁰⁷ Pb/ ²⁰⁶ Pb age ³ (Ma)
NA4 40m	7.2	Uraninite		0.52	12.12	82.11	0.41	1.30	0.58	97.03	916	1642
NA4 40m	7.3	Uraninite		0.59	12.00	81.70	0.31	1.42	0.68	96.71	911	1525
NA4 40m	7.4	Uraninite		0.55	10.32	82.59	0.20	1.53	0.73	95.91	775	
NA4 40m	7.6	Uraninite	-11.5	0.57	11.97	81.41	0.38	1.46	0.68	96.47	912	1494
NA4 40m	7.7	Uraninite		0.54	11.80	81.63	0.42	1.52	0.72	96.63	896	1459
NA4 40m	7.8	Uraninite		0.52	11.69	81.43	0.38	1.41	0.71	96.14	891	1439
NA4 40m	7.10	Uraninite		0.53	9.96	83.81	0.38	1.55	0.84	97.06	737	1115
NA4 40m	8.1	Uraninite	-12.1	0.52	11.40	81.84	0.37	1.48	0.55	96.17	864	1002
NA4 40m	8.2	Uraninite		0.43	11.37	81.55	0.28	1.34	0.63	95.60	865	886
NA4 40m	9.6	Uraninite	-8.9	0.88	7.40	85.10	0.17	1.63	0.79	95.99	539	898
NA4 40m	9.7	Uraninite		0.30	6.30	87.07	0.18	1.66	0.54	96.03	449	
NA4 40m	9.8	Uraninite		1.90	3.09	84.26	0.01	1.06	2.37	92.68	227	784
NA4 40m	10.5	Uraninite	-13.9	0.32	6.74	85.75	0.11	1.73	0.66	95.30	488	1340
NA4 40m	10.6	Uraninite	-11.6	0.80	4.34	85.18	0.11	1.56	1.03	93.02	316	1198
NA4 40.5m	2.1	Uraninite		0.49	11.71	81.69	0.26	1.25	0.52	95.92	889	
NA4 40.5m	2.2	Uraninite		0.50	12.96	81.08	0.29	1.29	0.44	96.56	992	1188
NA4 40.5m	2.3	Uraninite		0.54	12.96	81.13	0.33	1.36	0.56	96.89	991	
NA4 40.5m	2.4	Uraninite		0.74	8.87	82.69	0.24	1.49	0.82	94.86	666	1049
NA4 40.5m	2.5	Uraninite		0.74	7.74	84.16	0.20	1.59	0.71	95.15	571	1124
NA4 40.5m	2.6	Uraninite		0.74	8.47	83.44	0.27	1.41	0.84	95.17	630	
NA4 40.5m	3.1	Uraninite		0.49	10.83	82.42	0.25	1.44	0.50	95.93	815	1157
NA4 40.5m	3.2	Uraninite		0.50	10.70	82.48	0.27	1.53	0.53	96.00	805	
NA4 40.5m	3.3	Uraninite		0.40	10.81	82.61	0.28	1.43	0.56	96.08	812	
NA4 40.5m	3.4	Uraninite		0.43	10.25	82.72	0.28	1.56	0.56	95.79	768	
NA4 40.5m	4.1	Uraninite		0.69	10.79	82.13	0.29	1.30	0.58	95.79	815	1091
NA4 40.5m	4.2	Uraninite		0.91	8.29	83.43	0.17	1.49	1.06	95.34	616	
NA4 40.5m	4.3	Uraninite		1.13	4.49	87.01	0.08	2.17	0.34	95.22	320	1119
NA4 40.5m	4.4	Uraninite		0.60	10.66	82.65	0.33	1.49	0.63	96.36	800	
NA4 40m	9.9	Brannerite		4.00	2.47	51.88	0.02	0.39	30.45	89.22	296	
NA4 40m	10.1	Brannerite		4.06	3.96	49.71	0.23	0.77	34.30	93.03	494	
NA4 40m	10.2	Brannerite		4.16	3.11	48.67	0.13	0.56	35.02	91.66	397	
NA4 40m	10.3	Brannerite		4.11	5.33	50.21	0.14	0.39	33.27	93.45	659	
NA4 40m	10.4	Brannerite		3.66	4.32	49.27	0.24	0.45	34.62	92.57	544	

¹ Oxygen isotope data as presented in Table 4

² Chemical age calculated using the equation: $t = \text{Pb} \times 10^4 / (1.612\text{U})$ (Bowles, 1990)

³ ²⁰⁷Pb/²⁰⁶Pb ages calculated from ratios obtained by LA-HR-ICPMS presented in Table 6

TABLE 6. LA-HR-ICPMS Acquired U-Pb and $^{207}\text{Pb}/^{206}\text{Pb}$ Ratio Data from Uraninite Samples from Nabarlek with Calculated $^{235}\text{U}/^{207}\text{Pb}$, $^{238}\text{U}/^{206}\text{Pb}$, and $^{207}\text{Pb}/^{206}\text{Pb}$ Ages

Sample no.	Spot no.	Isotopic ratios			Calculated ages ¹ (Ma)		
		$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{235}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{235}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$
NA4 40.0m	1.1	0.06650	0.69360	0.07253	415	535	1002 ± 20
	1.2	0.08150	0.90343	0.07688	505	654	1118 ± 22
	2.1	0.14971	2.03143	0.09971	899	1126	1620 ± 32
	2.2	0.10930	1.24886	0.08703	669	823	1362 ± 27
	3.1	0.09978	1.11310	0.08703	613	759	1362 ± 27
	3.2	0.15802	1.51560	0.07007	946	937	930 ± 19
	7.2	0.09146	1.31773	0.10099	564	854	1642 ± 33
	7.3	0.12192	1.54143	0.09480	742	947	1525 ± 30
	7.6	0.10853	1.41109	0.09331	664	894	1494 ± 30
	7.7	0.10175	1.29177	0.09159	625	842	1459 ± 29
	7.8	0.09992	1.31701	0.09061	614	853	1439 ± 29
	7.10	0.11176	1.23069	0.07675	683	815	1115 ± 22
	8.1	0.17900	1.79674	0.07257	1061	1044	1002 ± 20
	8.2	0.18348	1.71455	0.06854	1086	1014	886 ± 18
	9.6	0.04571	0.45741	0.06897	288	382	898 ± 18
	9.8	0.04202	0.37487	0.06531	265	323	784 ± 16
	10.5	0.08770	1.03590	0.08603	541	721	1340 ± 27
	10.6	0.09391	1.03732	0.08005	579	723	1198 ± 24
	a	0.08433	1.04851	0.08714	522	728	1364 ± 28
	b	0.09469	1.18484	0.08726	583	794	1366 ± 27
	c	0.08313	0.97343	0.08732	514	690	1368 ± 27
NA4 40.5m	2.2	0.08031	0.90432	0.07963	498	654	1188 ± 24
	2.4	0.08600	0.87851	0.07430	532	640	1049 ± 21
	2.5	0.09912	1.04693	0.07711	609	727	1124 ± 22
	3.1	0.09545	0.93759	0.07840	588	672	1157 ± 23
	4.1	0.08071	0.89983	0.07584	500	652	1091 ± 22
	4.3	0.08608	0.94336	0.07691	532	675	1119 ± 22
NA88 29.0m	1.2	0.06920	0.63682	0.06329	431	500	718 ± 14
	2.1	0.07111	0.68236	0.06716	442	528	842 ± 17
	2.2	0.07038	0.67884	0.06705	438	526	840 ± 17
	2.3	0.07580	0.69426	0.06607	471	535	808 ± 16
	2.4	0.07534	0.76260	0.07272	468	576	1006 ± 20
	3.1	0.07479	0.72786	0.06748	465	555	852 ± 17
	3.2	0.07411	0.69553	0.06442	461	536	756 ± 15
	3.3	0.08061	0.79117	0.06883	500	592	894 ± 18
	4.4	0.08273	0.73893	0.06825	512	562	876 ± 17
	4.5	0.07274	0.71481	0.06865	453	548	888 ± 18
Na1 36.1m	1.1	0.07033	0.75624	0.07577	438	572	1089 ± 22
	1.2	0.08344	0.94246	0.08003	517	674	1198 ± 24
	1.3	0.09515	1.08268	0.08309	586	745	1271 ± 25
	1.4	0.08328	0.97613	0.08181	516	691	1241 ± 25

¹ $^{206}\text{Pb}/^{238}\text{U}$, $^{207}\text{Pb}/^{235}\text{U}$, and $^{207}\text{Pb}/^{206}\text{Pb}$ ages calculated using equations reported by Ludwig (1993)

± 120 Ma (Fig. 14). High error margins and MSWD values for the U-Pb age calculations are most likely due to uraninite mottling (Fig. 6d) and the LA-HR-ICPMS spot area being no less than 25 μm . Indeed, 2-mm-diam uraninite grains can produce $^{207}\text{Pb}/^{206}\text{Pb}$ ages that vary on the order of 400 m.y. (sample Na 4 40m, spots 3.1 and 3.2; Table 6). The high MSWD for our U-Pb discordia indicates that the scatter of the sample points is considerably greater than the analytical errors for each grain analyzed. Further, the majority of our U-Pb isotope ratios plot well below concordia, a phenomenon that is common in unconformity-related uranium deposits in Australia and Canada (Hills and Richards, 1976; Ludwig et al., 1987; Kotzer and Kyser, 1993; Fayek and Kyser, 1997; Fayek et al., 2000) and is indicative of radiogenic Pb loss.

Chemical composition of uraninite and U-Pb chemical ages

Electron microprobe data and backscattered electron images from uraninite and brannerite reveal that multiple generations of uraninite are preserved at the micron scale within the inner alteration halo (Fig. 6d). Uraninite at Nabarlek has highly variable chemical compositions in terms of U, Pb, Si, Ca, and Fe contents, which affect the brightness of the uranium-bearing phase under backscattered electron imaging. Few of the uraninite grains analyzed are pure UO_2 and most contain elevated concentrations of SiO_2 , CaO , FeO , and H_2O (Fig. 13a; Table 5). Older generations of uraninite have the highest PbO concentrations and the lowest SiO_2 , CaO , FeO , and H_2O concentrations (Fig. 15b; Table 5). In relative terms, this shows that more chemically heterogeneous

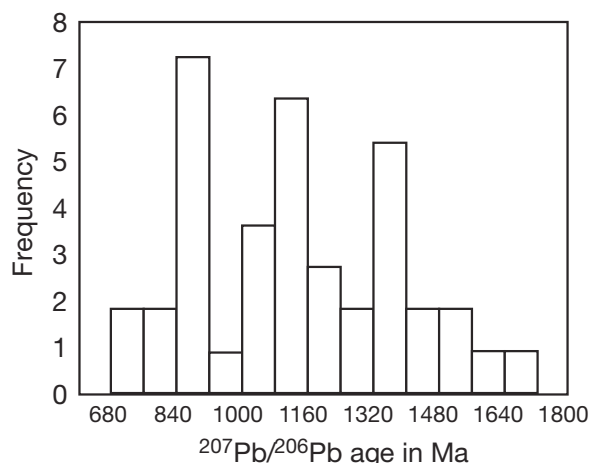


FIG. 13. Histogram of $^{207}\text{Pb}/^{206}\text{Pb}$ ages obtained from uraninite grains by LA-HR-ICPMS (Table 6). The peaks suggest that discrete events may have occurred at ca. 900, 1100, 1360, and 1640 Ma.

uraninite formed later than the more homogeneous uraninite, probably during the incursion of late fluids along the Nabarlek fault. This postformation alteration of uraninite has also been reported from unconformity-related uranium deposits in the Athabasca basin, Canada (Kotzer and Kyser, 1993; Fayek and Kyser, 1997; Fayek et al., 2002).

Electron microprobe data from uraninite and brannerite were used to calculate chemical ages for these minerals (Table 5). Calculation of chemical ages assumes that the total lead present in the sample is of radiogenic origin. The accuracy of the calculated age relies on there being no Pb loss, no U gain, and no addition of radiogenic Pb since the time of crystallization (Kotzer and Kyser, 1993; Fayek and Kyser, 1997). Chemical ages calculated from the uranium-bearing minerals range from 227 to 992 Ma (Table 5). In all cases, these ages are much younger than the 1642 Ma $^{207}\text{Pb}/^{206}\text{Pb}$ formation age obtained by LA-ICPMS. However, the majority of chemical U-Pb ages overlap the youngest $^{207}\text{Pb}/^{206}\text{Pb}$ ages obtained by LA-ICPMS and coincide with the $^{207}\text{Pb}/^{206}\text{Pb}$ and U-Pb concordia ages obtained by Hills and Richards (1976). These results suggest that the majority of the uraninite at Nabarlek experienced significant lead loss at ca. 900 Ma, coincident with fluid incursions during reactivation of the Nabarlek fault.

$^{40}\text{Ar}/^{39}\text{Ar}$ Dating of Phyllosilicate Minerals

Samples of metamorphic muscovite from relatively unaltered Hanging-Wall schist, S² sericite from the outer alteration halo, and S³ illite from the inner alteration halo were analyzed using the $^{40}\text{Ar}/^{39}\text{Ar}$ dating technique. Most of the samples analyzed gave disturbed age spectra, probably due to ^{40}Ar loss, ^{39}Ar recoil, and phase mixing caused in part by

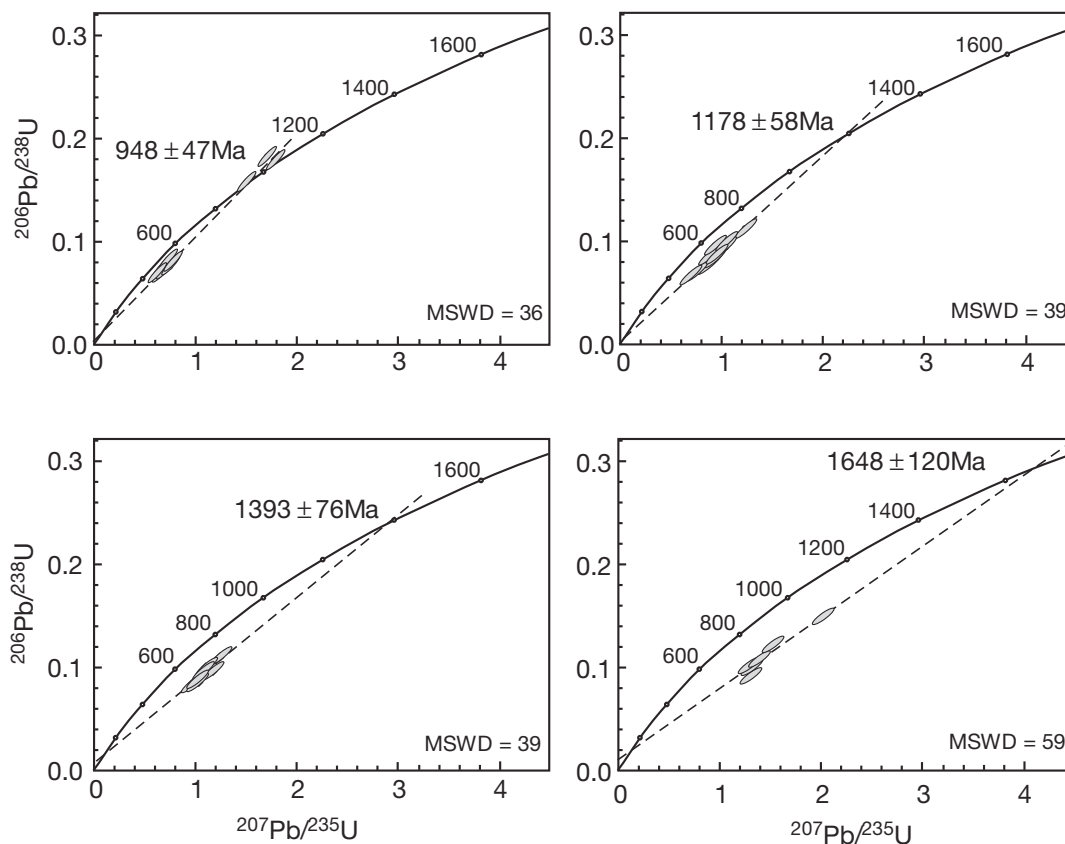


FIG. 14. U-Pb concordia diagrams from in situ isotopic analysis by LA-HR-ICPMS of uraninite from Nabarlek. Each plot represents a distinct population of uraninite alteration or reprecipitation. The four discordia ages coincide with distinct $^{207}\text{Pb}/^{206}\text{Pb}$ ages. Plots were constructed using ISOPLOT (Ludwig, 1993) from the isotopic ratios presented in Table 6.

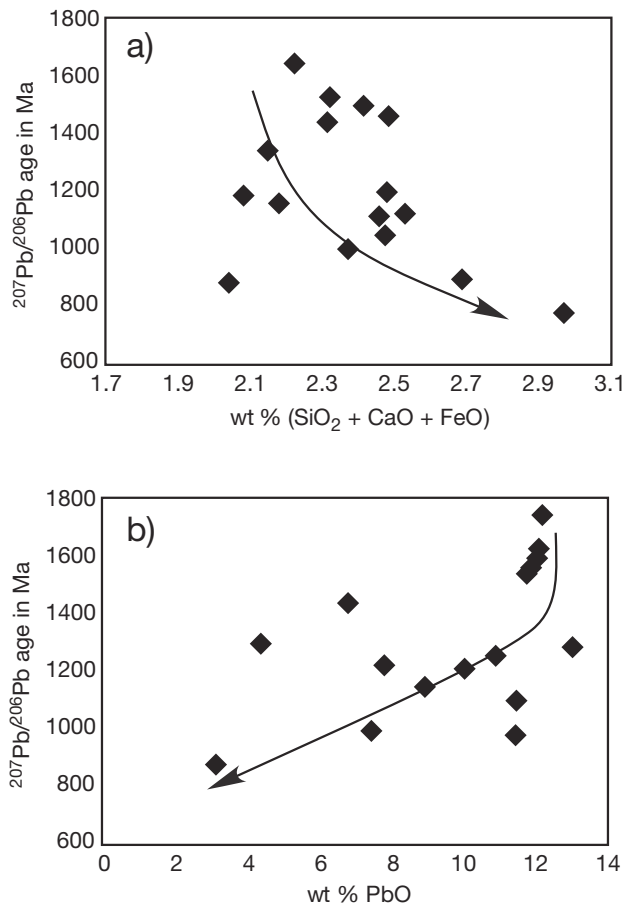


FIG. 15. a. Relationship between $^{207}\text{Pb}/^{206}\text{Pb}$ ages and SiO_2 , CaO , FeO contents in uraninite (Table 5). Increases in the contents of these components correlate with decreasing ages, indicative of postformation alteration by retrograde fluids as shown by arrow. b. Relationship between $^{207}\text{Pb}/^{206}\text{Pb}$ ages and PbO highlighting decreasing PbO values with decreasing age (Table 5). This relationship indicates that radiogenic Pb loss from the system has occurred throughout time.

multiple thermal and fluid overprints. For these reasons, $^{40}\text{Ar}/^{39}\text{Ar}$ ages are reported as integrated ages, which are equivalent to K-Ar ages.

One 500- μm grain of metamorphic muscovite separated from sample Na 83 12m gives an integrated age of 1737 ± 11 Ma (Fig. 16; Table 7). The other 500- μm grain of metamorphic muscovite separated from sample Na 26 37.5m gives an integrated age of 1748 ± 11 Ma (Table 7). Neither sample shows evidence of alteration. These ages are younger than most K/Ar ages obtained from metamorphic muscovite outside of the mineralized zones in the Alligator Rivers uranium field but concur with K/Ar ages obtained from metamorphic muscovite previously separated from Nabarlek schist (Page et al., 1980). The young age could be considered to correspond to the time of cooling below the closure temperature for muscovite (330°C; McDougall and Harrison, 1999), since peak metamorphism occurred prior to 1800 Ma. However, it is most likely that these ages reflect resetting of the isotopic system as a result of high temperatures associated with the

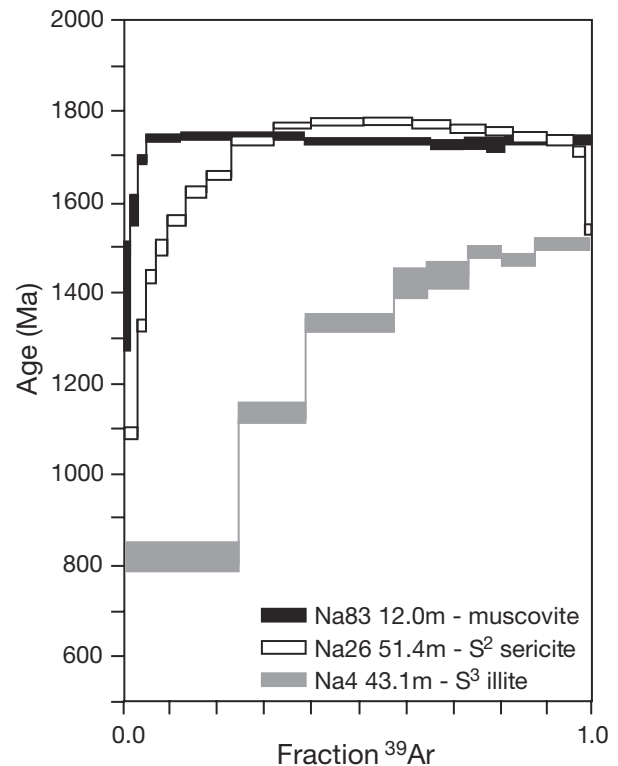


FIG. 16. $^{40}\text{Ar}/^{39}\text{Ar}$ age spectra for three representative samples of metamorphic and alteration minerals at Nabarlek. Sample Na83 12.0m represents metamorphic muscovite from the Hanging-Wall schist. Sample Na26 51.4m represents S^2 sericite from the outer alteration zone. Sample Na4 43.1m represents S^3 illite from the ore zone. Complete step-heat data and interpreted ages are provided in Table 7.

intrusion of the 1723 Ma Oenpelli Dolerite (Page et al., 1980; Kyser et al., 2000; Edgecombe et al., 2002).

Three, 2- to 5- μm fractions of S^2 sericite were analyzed to constrain the age of alteration in the outer alteration zone around the Nabarlek fault. Petrographically, this sericite formed after peak metamorphism but prior to S^3 illite alteration associated with uraninite precipitation. All three samples contain minor amounts (<10%) of C^{2a} chlorite that could not be removed from the sample. The samples produce bell-shaped spectra indicative of superficial ^{40}Ar loss (McDougall and Harrison, 1999) with narrow, pseudo-plateaus that give a range of old ages (Fig. 15; Table 7). The bell-shaped spectra could be indicating that the S^2 sericite analyzed in this study replaced metamorphic muscovite, which was able to retain some of its initial $^{40}\text{Ar}/^{39}\text{Ar}$ ratio. However, this is unlikely given that metamorphic muscovite does not retain any semblance of an age indicative of peak metamorphism and because S^2 sericite primarily replaces metamorphic minerals other than muscovite in the outer alteration zone. Instead, the spectra may be caused by disturbances to the $^{40}\text{Ar}/^{39}\text{Ar}$ ratio due to retrograde alteration of the sericite as indicated by low K_2O , high H_2O , and high FeO contents (Table 2; Fig. 9b). Indeed, the integrated ages for these samples (1696 ± 7 , 1701 ± 7 , and 1715 ± 7 Ma; Table 7) are geologically reasonable, agree with paragenetic

TABLE 7. Individual Spot Fusion Ages, Argon Isotope Ratios, % ^{40}Ar Atmospheric Contents and % ^{39}Ar Released from Muscovite, S² Sericite and S³ Illite from Nabarlek

Na26 37.5 muscovite, integrated age = 1748 ± 11 Ma							Na26 51.4 S ² sericite, integrated age = 1696 ± 7 Ma						
$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)	$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)
0.001	0.0178	0.003	35.10	0.10	36.43	1387 ± 630	0.000	0.029	9.699	17.99	2.84	28.16	1092 ± 20
0.000	0.0185	0.000	11.20	0.23	47.98	1671 ± 239	0.000	0.026	11.780	6.28	2.05	36.64	1324 ± 12
0.000	0.0197	0.011	2.49	1.54	49.43	1703 ± 32	0.000	0.024	9.629	3.41	2.16	41.05	1433 ± 10
0.000	0.0190	0.001	1.28	1.57	51.75	1754 ± 36	0.000	0.022	5.720	2.24	2.96	43.52	1492 ± 7
0.000	0.0186	0.008	0.27	8.94	53.53	1792 ± 14	0.000	0.021	5.036	1.75	4.62	46.24	1554 ± 7
0.000	0.0196	0.008	0.26	29.49	50.71	1731 ± 15	0.000	0.020	3.384	0.95	3.83	48.94	1614 ± 7
0.000	0.0186	0.000	0.34	4.63	53.49	1791 ± 23	0.000	0.019	2.000	0.68	5.47	51.15	1662 ± 6
0.000	0.0191	0.013	0.47	31.60	52.00	1759 ± 13	0.000	0.018	2.119	0.15	9.00	54.39	1729 ± 6
0.000	0.0196	0.001	0.07	6.69	50.93	1736 ± 16	0.000	0.018	0.566	0.26	7.66	56.07	1763 ± 6
0.000	0.0195	0.006	0.00	2.96	51.16	1741 ± 20	0.000	0.018	1.350	0.20	10.14	56.47	1771 ± 6
0.000	0.0197	0.021	0.30	12.25	50.52	1728 ± 14	0.000	0.018	1.077	0.00	9.95	56.51	1772 ± 6
							0.000	0.018	1.341	0.25	9.25	56.17	1765 ± 6
							0.000	0.018	0.353	0.07	7.80	55.73	1756 ± 6
							0.000	0.018	0.889	0.08	5.80	55.32	1748 ± 6
							0.000	0.018	0.486	0.20	6.56	54.86	1739 ± 6
							0.000	0.018	0.763	0.06	6.25	54.69	1735 ± 6
							0.000	0.018	1.113	0.52	3.16	53.93	1720 ± 6
							0.000	0.022	5.928	1.31	0.50	45.61	1540 ± 13
Na83 12.0 muscovite, integrated age = 1737 ± 11 Ma							Na88 29.0 S ³ illite, integrated age = 1319 ± 7 Ma						
$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)	$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)
0.002	0.013	0.135	81.94	0.04	13.21	633 ± 1730	0.000	0.033	0.019	26.88	5.49	21.97	901 ± 23
0.001	0.019	0.031	37.80	0.07	31.88	1263 ± 635	0.000	0.028	0.021	22.60	15.02	27.87	1083 ± 10
0.000	0.025	0.012	6.07	0.36	36.47	1389 ± 123	0.000	0.026	0.016	9.15	13.12	34.56	1268 ± 10
0.000	0.022	0.004	1.67	1.16	44.15	1582 ± 35	0.000	0.024	0.014	4.81	12.35	38.97	1381 ± 10
0.000	0.019	0.051	4.08	1.55	49.03	1695 ± 7	0.000	0.023	0.011	2.89	10.98	42.55	1467 ± 10
0.000	0.019	0.009	0.41	9.40	51.34	1746 ± 13	0.000	0.023	0.009	3.03	4.25	42.53	1467 ± 21
0.000	0.019	0.010	0.15	26.54	51.72	1754 ± 14	0.000	0.022	0.004	2.98	3.22	44.20	1506 ± 26
0.000	0.019	0.011	0.30	26.24	50.82	1734 ± 13	0.000	0.024	0.017	4.93	35.57	38.91	1379 ± 8
0.000	0.019	0.007	0.28	6.41	50.62	1730 ± 13							
0.000	0.019	0.006	0.13	5.17	50.70	1732 ± 15							
0.000	0.019	0.008	0.31	4.21	50.55	1728 ± 19							
0.000	0.019	0.011	0.00	18.85	51.10	1740 ± 12							
Na83 40.0 S ² sericite, integrated age = 1715 ± 7 Ma							Na26 43.1 S ³ illite, integrated age = 1252 ± 9 Ma						
$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)	$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)
0.000	0.0219	0.037	2.14	11.43	44.55	1515 ± 6	0.000	0.030	0.049	45.14	25.26	18.41	816 ± 18
0.000	0.0231	0.550	0.37	8.57	43.12	1482 ± 5	0.000	0.029	0.091	18.82	14.70	28.11	1132 ± 18
0.000	0.0193	0.448	0.28	4.84	51.67	1672 ± 6	0.000	0.025	0.077	11.06	18.03	35.29	1335 ± 16
0.000	0.0172	0.454	0.03	11.42	57.85	1797 ± 6	0.000	0.024	0.036	8.36	7.50	38.54	1419 ± 28
0.000	0.0172	0.267	0.00	15.40	58.11	1802 ± 6	0.000	0.023	0.049	8.68	7.86	39.46	1443 ± 28
0.000	0.0172	0.196	0.19	2.40	57.83	1797 ± 7	0.000	0.022	0.028	8.99	7.12	41.55	1495 ± 19
0.000	0.0174	0.410	0.24	1.75	57.32	1787 ± 6	0.000	0.022	0.027	9.79	7.10	40.47	1468 ± 19
0.000	0.0175	0.832	0.09	9.60	56.78	1777 ± 11	0.000	0.022	0.026	7.76	12.42	42.52	1518 ± 19
0.000	0.0177	3.808	0.86	8.14	55.92	1759 ± 14							
0.000	0.018	0.179	0.09	10.96	55.24	1745 ± 5							
0.000	0.0182	0.697	0.04	8.24	54.91	1739 ± 6							
0.000	0.0183	0.796	0.11	7.25	54.44	1729 ± 6							
Na26 80.4 S ² sericite, integrated age = 1701 ± 7 Ma							Na4 40.0 S ³ illite, integrated age = 860 ± 12 Ma						
$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)	$^{36}\text{Ar}/^{40}\text{Ar}$	$^{39}\text{Ar}/^{40}\text{Ar}$	Ca/K	^{40}Ar atm %	^{39}Ar %	$^{40}\text{Ar}^{\circ}/^{39}\text{Ar}_K$	Age (Ma)
0.000	0.0295	7.133	9.92	10.05	30.49	1158 ± 11	0.002	0.012	2.521	71.84	29.48	22.68	924 ± 28
0.000	0.0217	8.059	2.12	6.42	44.91	1523 ± 9	0.001	0.019	0.71	40.19	7.67	31.29	1179 ± 16
0.000	0.0199	6.693	1.10	5.62	49.65	1629 ± 8	0.001	0.022	1.822	31.65	6.58	30.98	1171 ± 15
0.000	0.0186	3.888	0.85	12.00	53.18	1703 ± 7	0.001	0.023	10.446	38.58	6.00	26.42	1039 ± 21
0.000	0.0178	1.998	0.43	7.51	55.92	1759 ± 6	0.001	0.021	2.589	48.85	7.62	24.58	983 ± 17
0.000	0.0171	2.059	0.19	7.56	58.14	1803 ± 7	0.002	0.017	1.337	68.62	13.59	18.85	799 ± 23
0.000	0.0168	1.284	0.26	6.41	59.05	1821 ± 7	0.002	0.011	2.781	87.01	12.63	12.32	559 ± 47
0.000	0.0167	1.834	0.26	7.74	59.51	1830 ± 6	0.003	0.007	1.101	91.65	7.19	11.33	520 ± 70
0.000	0.0168	0.101	0.08	8.59	59.38	1828 ± 7	0.003	0.005	1.415	93.74	9.23	12.78	577 ± 111
0.000	0.0171	0.792	0.09	9.79	58.37	1808 ± 7							
0.000	0.0177	0.713	0.30	9.58	56.24	1766 ± 6							
0.000	0.0181	0.593	0.02	6.41	55.00	1741 ± 6							
0.000	0.0187	2.115	0.43	2.31	53.04	1701 ± 7							

Abbreviations: $^{40}\text{Ar}^{\circ}$ = radiogenic argon, $^{39}\text{Ar}_K$ = argon from potassium during sample irradiation

observations, and suggest that alteration around the Nabarlek fault occurred at ca. 1700 Ma.

Three, 2- to 5- μ m fractions of variably altered S³ illite were dated to determine the age of alteration associated with uranium deposition. Of the three samples analyzed, one contained up to 50 percent kaolinite that could not be separated, one contained up to 30 percent kaolinite, and one contained ca. 90 percent kaolinite. The Ar release patterns are highly disturbed (Fig. 16), but the integrated age for each sample is 1319 ± 7 , 1252 ± 9 , and 860 ± 16 Ma, respectively. A plot of these total fusion ages against the wt percent H₂O yield from stable isotope analyses (Fig. 17) suggests that retrograde alteration of S³ illite by incorporation or exchange of interlamellar water with the octahedral hydroxyl groups must displace K⁺ ions from the interlamellar site and facilitate the loss of Al and Fe and the gain of Si to form kaolinite (Kotzer and Kyser, 1991). This relationship has previously been observed in the Athabasca basin, where it was shown that illites

with the highest water contents also have the youngest K-Ar ages (Kotzer and Kyser, 1991).

Discussion

Detailed mineral paragenesis, electron microprobe and stable and radiogenic isotope analyses confirm that the formation of the Nabarlek deposit involved the incursion of basinal fluids at specific times during the formation of the Alligator Rivers uranium field (Fig. 18, Table 8). It has been shown that the basement lithologies in the Nabarlek area, specifically the Myra Falls Metamorphics, reached mid to upper amphibolite facies (550°–630°C and 5–8 kbars) during the Barramundi and Top End orogenies (1890 and 1800 Ma; Ferguson, 1980; Page et al., 1980; Needham, 1988). K-Ar ages from metamorphic muscovite and biotite, which have closure temperatures for radiogenic argon of ca. 330° and 350°C, respectively (McDougall and Harrison, 1999), cluster around 1800 Ma (Page et al., 1980). These K/Ar ages represent the end of the Top End orogeny and reflect uplift, cooling, and erosion of basement rocks at that time, bringing the Myra Falls Metamorphics to the surface.

Q¹ quartz veins most likely formed toward the end of the Top End orogeny and represent the earliest recorded fluid event (Table 8). Fluid inclusion and stable isotope data reveal that a basement-derived NaCl-dominated fluid formed the quartz veins at temperatures in excess of 370°C. These results concur with Ypma and Fuzikawa (1980) who reported that some quartz veins formed during the waning stages of peak metamorphism. The occurrence of type A and B fluid inclusions in the same veins that homogenize between 320° and 392°C suggests that a circulating fluid, heated by a cooling Nabarlek Granite at ca. 1830 Ma, most likely led to the formation of these veins (Fig. 18a). Regular faulting associated with rapid uplift would have caused pressure variations and phase separation that might explain the critical behavior of some fluid inclusions.

Movement along the Nabarlek fault and the Upper and Lower Footwall shears (Fig. 3) most likely began during uplift and erosion of the Myra Falls Metamorphics. Erosion of the Myra Falls Metamorphics was followed by the deposition of the Kombolgie Subgroup sometime between 1800 and

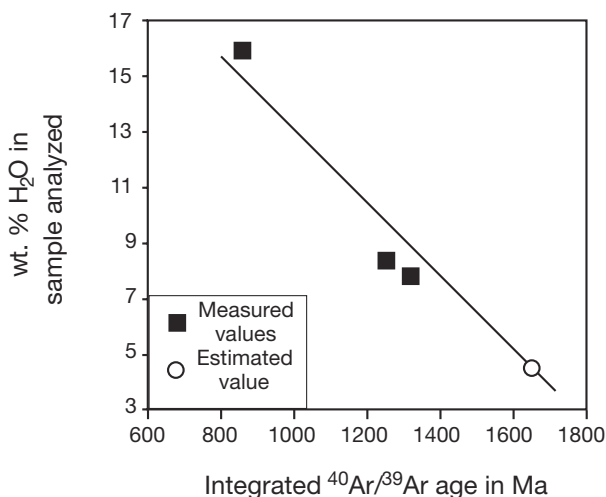


FIG. 17. Integrated $^{40}\text{Ar}/^{39}\text{Ar}$ ages vs. wt % H₂O yield from stable isotope analyses (filled squares), showing that the degree of S³ illite alteration to kaolinite, as indicated by increasing water contents, lowers the age of the illite. Open circle represents theoretically ideal, unaltered illite with ca. 4.5 wt % H₂O coprecipitated with the original uraninite at ca. 1640 Ma.

TABLE 8. A Summary of the Major Results Presented in this Paper Showing the Different Geologic Events, Mineral Assemblages, Formation Temperatures, and Isotopic Compositions of Fluids Recorded at Nabarlek

Event	Peak-postpeak metamorphism	Pre-Oenpelli Dolerite	Preore alteration	Synore alteration	Postore alteration
Major minerals formed and analyzed	Q ¹ quartz veins	S ¹ sericite, C ¹ chlorite	S ² sericite, C ² chlorite	S ³ illite, uraninite	Q ³ quartz veins, C ³ chlorite
Age	~1820 Ma	~1800–1720 Ma	~1700 Ma	~1640 Ma	~1360–750 Ma
Formation temperature	320°–400°C		200° ± 25°C	200° ± 25°C	165°–310°C
$\delta^{18}\text{O}_{\text{fluid}}$ range	4.1–7.9 per mil		2.0–5.2 per mil	1.5–5.5 per mil	0.1–1.9 per mil
$\delta\text{D}_{\text{fluid}}$ range	–48 to –32 per mil		–38 to –15 per mil	–38 to –25 per mil	–54 to –53 per mil
Proposed fluid source	Metamorphic fluids		Basinal brines, minor meteoric component	Basinal brines, minor meteoric component	Meteoric fluids mixed with an evaporite source

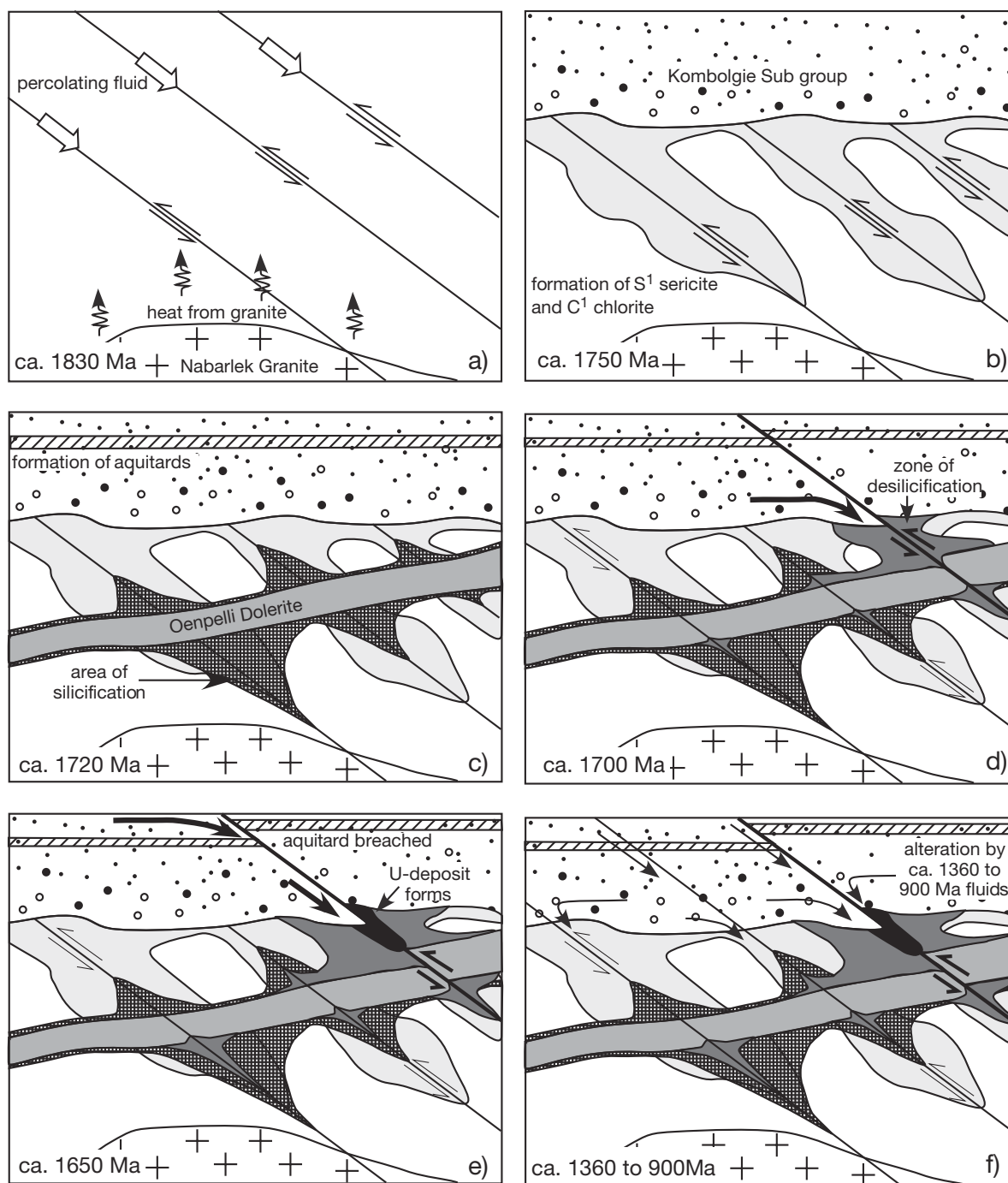


FIG. 18. A simplistic, schematic representation of the formation of alteration styles surrounding the Nabarlek uranium deposit. a. Faults and shear zones, most likely to have been active during rapid uplift of the Myra Falls Metamorphics, allow basement-derived fluids (large arrows) to circulate and be heated by the cooling Nabarlek Granite (small rising arrows). Precipitation of Q¹ quartz veins occurred at this time. b. The Komolgie Subgroup was deposited and fluids possibly derived from overlying sources percolated into the now relatively inactive faults, resulting in alteration of the metamorphic assemblage to fine-grained S¹ sericite and C¹ chlorite. c. Intrusion of the flat-lying Oenpelli Dolerite at ca. 1720 Ma was accompanied by intense quartz cementation around the faults, which effectively sealed them. In the Komolgie Subgroup, unrelated shallow burial diagenesis formed effective aquitard lithologies. d. The Nabarlek fault was reactivated at ca. 1700 Ma and allowed significant volumes of basinal fluid to alter the metamorphic assemblage, resulting in the formation of an extensive alteration halo (not depicted due to its ca. 1-km radius around the deposit), which included the removal of quartz. e. Movement along the Nabarlek fault at 1640 Ma created significant porosity and may have breached aquitard units in the Komolgie Subgroup that allowed uraniferous brines to travel down to the unconformity where they were focussed into the Nabarlek fault forming the deposit. f. Reactivation of the Nabarlek fault at discrete periods of time between 1380 and 750 Ma related to major tectonic events. Precipitation of C³ chlorite and Q³ quartz veins, alteration of uraninite, and replacement of pre- and synore alteration minerals (S² sericite, C² chlorite, and S³ illite) occurred at this time.

1720 Ma (Sweet et al., 1999a). Fluids, probably derived from overlying sources, percolated into the faults and caused minor alteration of metamorphic biotite, amphibole, plagioclase, and muscovite to fine-grained C¹ chlorite and S¹ sericite (Fig. 18b, Table 8).

Drill hole logs indicate that Q² quartz enveloped the upper and lower side of the Oenpelli Dolerite (Fig. 18c) prior to the formation of the inner and outer alteration halo. This suggests that the intrusion of the Oenpelli Dolerite at ca. 1723 Ma (Kyser et al., 2000; Edgecombe et al., 2002) was the most likely trigger for silica mobilization that resulted in the precipitation of the Q² quartz cement. The Q² quartz cement effectively sealed the Upper and Lower Footwall shears.

Petrographic evidence from elsewhere in the Kombolgie Subgroup indicates that burial diagenesis within the sandstones was in progress by 1723 Ma (Kyser et al., 2000; Hiatt and Kyser, 2002). Indeed, early diagenesis at shallow depths in the well-sorted, aeolian and marine sandstones resulted in the formation of aquitard units prior to the intrusion of the Oenpelli Dolerite (Carson et al., 1999; Kyser et al., 2000; Hiatt et al., 2001; Hiatt and Kyser, 2002; Fig. 18c). These aquitards, the Nungbargari Volcanics and the Oenpelli Dolerite, served to compartmentalize the Kombolgie Subgroup and isolated the poorly sorted, fluvial clastic sediments where diagenetic alteration in the presence of basinal fluids was ongoing well after 1723 Ma (Kyser et al., 2000). These poorly sorted sandstones are thought to have been the metal-bearing aquifer lithologies and source rocks for Nabarlek mineralization (Kyser et al., 2000).

Integrated ⁴⁰Ar/³⁹Ar ages indicate that by 1700 Ma the metamorphic assemblage had begun altering to a S² sericite-C² chlorite-dominated assemblage (Fig. 18d, Table 8). This suggests that the Nabarlek fault may have been reactivated by a tectonic event, possibly coincident with the introduction of the 1705 ± 11 Ma West Branch Volcanics (Sweet et al., 1999b) or stresses associated with a change in the direction of plate movement, which is recorded as bend 1 (B1) on the apparent polar wander path for Australia (Loutit et al., 1994; Idnurm and Giddings, 1995; Idnurm, 2000). Such intraplate stresses are a major influence on faulting, uplift, and fluid migration in basinal settings (Loutit et al., 1994; Southgate et al., 2000). Metamorphic and Q² quartz was largely removed from this time onward from above the Oenpelli Dolerite. However, its preservation below the Oenpelli Dolerite (Fig. 3) and in the Hanging-Wall and Footwall shears suggests that significant volumes of fluid failed to penetrate below the Oenpelli Dolerite or into these shear zones after 1700 Ma.

The formation of S² sericite and C² chlorite is considered to have been the result of basinal fluids flowing into the Nabarlek fault. Wilde and Wall (1987) first suggested that desilicification around the Nabarlek fault was caused by a fluid derived from the Kombolgie Subgroup sandstone and dismissed a basement-sourced fluid that would have deposited quartz instead. Komninou and Sverjensky (1996) supported this suggestion by demonstrating that a silica-saturated fluid derived from the Kombolgie Subgroup would have become undersaturated with quartz when it reacted with an amphibolite-dominated basement. The isotopic compositions of S² sericite and C^{2a} and C^{2b} chlorite indicate that a fluid with δ¹⁸O and δD values of 3.5 ± 2 and -25 ± 10 per mil, respectively (Fig. 12,

Table 8), altered the metamorphic assemblage and simultaneously removed quartz. These δ¹⁸O_{fluid} values are too low to be derived from a metamorphic-derived fluid, whereas the high δD_{fluid} values rule out normal magmatic sources (Taylor, 1997). Instead, a basinal brine that evolved from typically marine values with minor contributions from a meteoric source (Longstaffe, 1987, 2000) are invoked, consistent with the models proposed by Wilde and Wall (1987) and Komninou and Sverjensky (1996). Stable isotope variations outside of this range are most likely due to low fluid/rock ratios or incorporation of additional low-temperature, low- to midlatitude meteoric water into the interlamellar and octahedral sites. Integrated ⁴⁰Ar/³⁹Ar ages indicate that this alteration developed up to 60 m.y. before the oldest known uraninite first precipitated.

By 1642 ± 33 Ma, uraninite was precipitating together with S³ illite and H² hematite (Table 8). Importantly, chlorite did not precipitate with primary uraninite. The timing of original uraninite precipitation appears to coincide with two bends on the Australian apparent polar wander path at 1650 and 1640 Ma (Idnurm, 2000), either of which could indicate changes in plate motion and tectonism that would have reactivated the Nabarlek fault. The mineralizing brine contained uranium that was most likely leached from detrital zircon, monazite, and apatite (Fayek and Kyser, 1997) and potassium from altered feldspar and lithic clasts that comprised much of the Kombolgie Subgroup prior to burial diagenesis (Kyser et al., 2000). Previous studies have shown that Proterozoic sediments derived from uranium-rich provinces similar to the Kombolgie sub-basin contain ca. 70 ppm U prior to diagenetic alteration (Macleod, 1992; Fayek and Kyser, 1997) and similar sediments are most likely to have been the source of U at Nabarlek (Kyser et al., 2000). Brine flow to the unconformity appears to have been induced by gravity and was likely focussed by aquitard units (Hiatt et al., 2001; Hiatt and Kyser, 2002). Topographic relief may have been present at the unconformity, similar to that described at some Canadian uranium deposits hosted by reverse faults (Fayek and Kyser, 1997; Harvey et al., 2002) and would have acted as a local barrier that further focussed the uranium-bearing brine into the Nabarlek fault (Fig. 18e). Brittle fracturing associated with movement along the Nabarlek fault at certain times would have created additional permeability for fluids, particularly at Nabarlek where the preexisting foliation is at a high angle to the Nabarlek fault (Fig. 3; Johnson, 1984; Wilde and Wall, 1987). Interaction of the oxidized fluid with the reduced basement lithologies is thought to have resulted in the precipitation of uraninite, illite, and hematite. In this model, there is minimal to no contribution from basement-derived fluids. In fact, we calculate that 96,000,000 m³ of brine can account for a 200-m-thick chlorite-sericite-illite-hematite assemblage extending up to 1,000 m from the deposit (Wilde and Wall, 1987). If this volume of fluid contained 86 ppm U it also had the potential to form an orebody containing 1.82 Mt of uranium metal.

Stable isotope results show that the synore S³ illite and uraninite formed from basinal fluids with δ¹⁸O and δD values that were indistinguishable from those that formed the S² sericite and the C^{2a} and C^{2b} chlorite (Fig. 12, Table 8). However, the absence of synore chlorite from the inner alteration

zone indicates that the ore-bearing fluid was relatively depleted in Mg^{2+} while being enriched in U and K^+ . The other principal difference between the ca. 1700 Ma basinal brine and the ca. 1640 Ma mineralizing fluid is that the latter likely had a higher salinity and oxidation state and lower pH (Komninou and Sverjensky, 1996). The abundance of red hematite in the inner alteration halo and the absence of graphite support this finding and suggest that the Fe^{2+} in the Fe-rich C^{2b} chlorite may have acted as a primary reductant of U^{6+} . Other workers have proposed that uraninite precipitation may have been caused by the interaction between methane-bearing fluids and oxidizing U-bearing fluids (Hoeve and Sibbald, 1978; Wall et al., 1985; Hoeve and Quirt, 1987; Derome et al., 2003), but evidence for this reaction is lacking given that methane-bearing fluid inclusions have only been reported in quartz veins that are either distal to or post-date mineralization (Ypma and Fuzikawa, 1980; Wilde et al., 1989). The reaction path predicted by the model of Komninou and Sverjensky (1996), which ends with chlorite and hematite replacing white mica, was proposed to explain previously published mineral assemblages that suggested chlorite and uraninite were coeval. We propose that chlorite was removed from the ore zone during uraninite precipitation and such a lengthy reaction path did not occur.

Previous studies have suggested that quartz veins around Nabarlek were part of the ore-forming system (Ypma and Fuzikawa, 1980; Wilde et al., 1989; Derome et al., 2003) and that a distinct mineralizing fluid is preserved in the fluid inclusions in these quartz veins. However, it is now clear that Q^1 quartz veins formed prior to uranium mineralization. The saline, complex, low-temperature brines previously linked to uranium transport only occur in Q^3 quartz veins that postdate mineralization and have $\delta^{18}\text{O}_{\text{fluid}}$ and $\delta\text{D}_{\text{fluid}}$ values, which are distinct from the basinal brines that transported and deposited uranium.

Multiple incursions of fluid into the Nabarlek fault after ca. 1640 Ma are recorded (Fig. 18f). The sulfides, and in particular galena, most likely formed during postmineralization fluid events that mobilized radiogenic Pb out of the uraninite (Kotzer and Kyser, 1993). These events are thought to correspond to regional tectonic events, including (1) the intrusion of the Maningkorriir phonolitic dike swarm at ca. 1316 ± 40 Ma (Page et al., 1980; Needham, 1988) or the regional Derim Derim Dolerite intrusion at 1324 ± 4 Ma (Sweet et al., 1999b); (2) the amalgamation of Australia and Laurentia during the Grenville orogeny at ca. 1140 Ma (Wingate et al., 2002); and (3) the breakup of Rodinia between 1000 Ma (Tack et al., 2001) and 750 Ma (Wingate and Giddings, 2000). The high temperatures recorded by the C^{3a} and C^{3b} chlorite may record the migration of fluids that were heated by, and subsequently migrated from, areas around the Maningkorriir phonolitic dikes or the more regional Derim Derim Dolerite.

Finally, stable isotope evidence from late kaolinite alteration associated with anatase, hematite-goethite, diginite, covellite, native copper, and disseminated secondary uranium minerals shows that this assemblage is most likely to have occurred relatively recently as a result of exposure to, and interaction with, near-surface meteoric water.

Previously published models do not take into account the multiple stages of fluid overprinting that occurred over a

period lasting for at least 800 m.y. We have refined the model for Nabarlek to take into account the incursion of chemically distinct basinal fluids at different times in the history of the Alligator Rivers uranium field.

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