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CHLORITIZATION AND ASSOCIATED ALTERATION AT THE JABILUKA UNCONFORMITY-TYPE URANIUM DEPOSIT, NORTHERN TERRITORY, AUSTRALIA

CONSTANCE J. NUTT
U.S. Geological Survey, M.S. 905, Box 25046, Denver Federal Center, Denver, Colorado 80225, U.S.A.

ABSTRACT

Jabiluka is the largest of four known unconformity-type uranium deposits that are hosted by brecciated and altered metasedimentary rocks in the Pine Creek geosyncline, Northern Territory, Australia. The alteration zone at Jabiluka is dominated by chlorite, but also contains white mica, tourmaline and apatite; hematite is present, but only in minor amounts. Added quartz is mainly restricted to fractures and breccias. Chlorite, which formed during episodic fluid movement, partly to totally replaced all pre-existing minerals. Chloritized rocks are enriched in Mg, and depleted in K, Ca, Na and Si. Five types of chlorite are optically and chemically distinguishable in the rocks at Jabiluka: (1) Mg-rich and coarse-grained, (2) Mg–Fe-rich and coarse-grained, (3) Mg–Fe-rich, typically 10–70 μm in size, (4) clay-size and Mg-rich, and (5) 7 Å amesite. Chloritization is proposed as a mechanism that lowered the pH of the circulating fluid, and also caused significant loss of silica from the altered rocks. The proposed constraints on alteration, and presumably on at least part of the uranium mineralization, neither require nor preclude the existence of the unconformity as necessary for the formation of ore.

Keywords: Jabiluka, unconformity-type uranium deposit, alteration, chlorite, sericite, dravite, apatite, Australia.

SOMMAIRE

Le gisement d'uranium de Jabiluka est le plus important des quatre exemples connus de gisements associés à une non-conformité, situés dans un encaissant de roches métasédimentaires bréchifiées et altérées dans le géosynclinal de Pine Creek (Northern Territory, en Australie). La zone d'altération contient surtout de la chlorite, mais aussi mica blanc, tourmaline et apatite; l'hématite y est accessoire. La présence de quartz est surtout localisée près des fissures et dans les brèches. La chlorite, formée lors de flux épisodiques d'une phase fluide, a remplacé, en partie ou *in toto*, tous les minéraux primaires. Les roches ainsi chloritisées sont enrichies en Mg, et appauvries en K, Ca, Na et Si. On distingue optiquement et chimiquement cinq variétés de chlorite: 1) chlorite magnésienne à granulométrie grossière; 2) chlorite Mg–Fe, à granulométrie grossière; 3) chlorite Mg–Fe, en grains de 10 à 70 μm; 4) chlorite magnésienne dans la fraction argileuse, et 5) amesite en feuillets de 7 Å. La chloritisation serait responsable d'un abaissement du pH de la phase fluide, ainsi que d'une perte importante de silice des roches altérées. Les caractéristiques des phénomènes d'altération, et présumément de la minéralisation en uranium, au moins en partie, n'impliquent pas de façon nécessaire l'existence de la non-conformité dans la formation du minerai.

(Traduit par la Rédaction)

Mots-clés: Jabiluka, gisement d'uranium associé à une non-conformité, altération, chlorite, sericite, dravite, apatite, Australie.

INTRODUCTION

The extensive chlorite-dominated zone of alteration that encloses the ore at the world-class Jabiluka uranium deposit, Northern Territory, Australia, is characteristic of unconformity-type deposits in Australia. These unconformity-type deposits, as well as similar deposits in Canada, were first discovered in the late 1960s and early 1970s; therefore, current geological studies are at the stages of description and preliminary genetic modeling. Because an understanding of the complex alteration that affected Jabiluka is a fundamental part of any model for the deposit, I document in this study the properties of chlorite and associated minerals in the alteration zone, and describe the chemistry of altered rocks. The observations incorporated in this study are based on core logging and core samples collected in 1980 during a three-week stay at the Jabiluka camp, during which Pancontinental Mining Limited kindly allowed R.I. Grauch and me, both of the U.S. Geological Survey, access to drill cores and company data. Nineteen cores were logged and about 1100 samples, mostly metasedimentary rocks and ore, were collected; detailed descriptions of some of the core sampled at Jabiluka are reported in Nutt (1983, 1984a, 1986). Because most of the deposit is overlain by sandstone and is not yet mined, this investigation is based solely on drill-core samples.

GEOLOGICAL SETTING

Jabiluka is the largest of four known unconformity-type deposits located in the Alligator Rivers Uranium Field (ARUF) of the Pine Creek geosyncline, Northern Territory, Australia (Fig. 1). These deposits are called "unconformity-type" because they occur near a regional unconformity that separates Early Proterozoic metasedimentary rocks

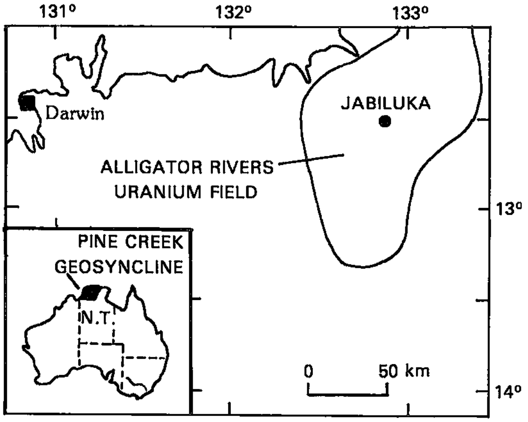

FIG. 1. Location of the Pine Creek geosyncline and the Alligator Rivers Uranium Field (ARUF) in northern Australia.

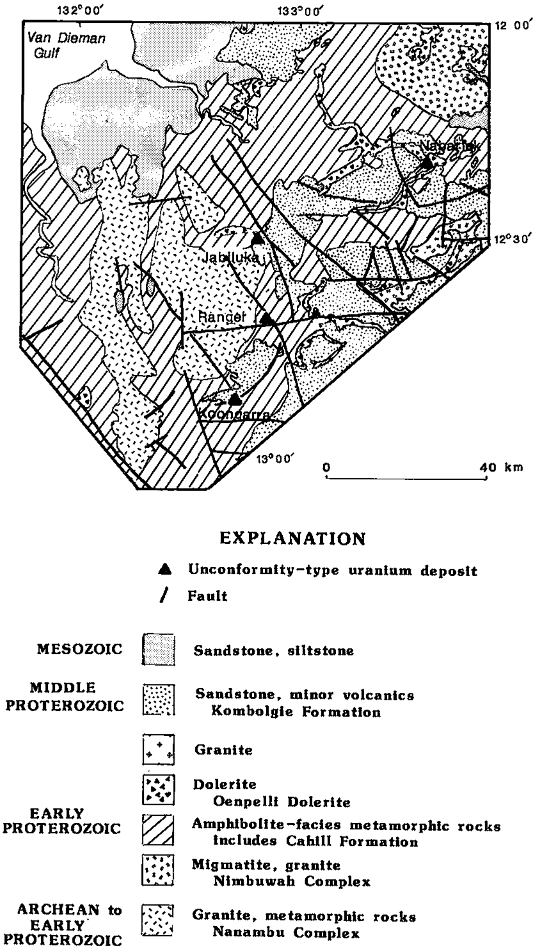

FIG. 2. Geological map of the Alligator Rivers Uranium Field and location of the unconformity-type deposits; modified from Ewers *et al.* (1984).

and Middle Proterozoic sedimentary rocks. The geology of the ARUF is described in Needham (1984), Needham & Stuart-Smith (1980) and Ewers *et al.* (1984), and is only briefly summarized here.

In the ARUF, most uranium deposits and prospects are located in the Cahill Formation or equivalent rocks (Fig. 2). This formation is part of a thick (as much as 14 km) sequence of Early Proterozoic pelitic, carbonate and evaporite sediments, and subordinate volcanic rocks; the sequence was deposited on Archean basement, which is now exposed as the Nanambu Complex (Fig. 2). Zircon from the complex yielded a U–Pb isotopic age of about 2470 Ma (Page *et al.* 1980). The Early Proterozoic sequence was intruded by dolerite and, between 1890 and 1870 Ma (Page *et al.* 1980), by granitic rocks of the Nimbuwah Complex. This igneous activity in the Nimbuwah Complex marked the onset of metamorphism, which continued until about 1800 Ma (Page *et al.* 1980) and which multiply deformed and metamorphosed ARUF rocks to amphibolite-facies conditions. Peak metamorphism was followed by intrusion of granitic magma at 1755 to 1732 Ma, and the Oenpelli Dolerite at 1688 ± 64 Ma (Page *et al.* 1980, reinterpreted by Ludwig *et al.* 1987). Following a period of erosion, the Early Proterozoic sequence was covered by sediments of the Middle Proterozoic Kombolgie Formation. This formation, greater than 650 m thick, consists of quartz-rich sandstone with two volcanic members, one of which has a Rb–Sr isotopic age of 1648 ± 29 Ma (Page *et al.* 1980). Post-Kombolgie dykes, some of which are near deposits, have been isotopically dated by Rb–Sr and K–Ar methods at 1370–1200 Ma (Page *et al.* 1980), which should be considered as minimum ages (Ludwig *et al.* 1987).

The Cahill Formation, about 3000 m thick, consists of a lower member, which hosts the Jabiluka, Ranger and Koongarra unconformity-type deposits (Fig. 2), and an upper member dominated by psammitic rocks. The Nabarlek deposit (Fig. 2) is in stratigraphically equivalent rocks that have been metamorphosed to a higher grade. The lower member of the Cahill Formation consists of carbonate rocks (primarily dolomite) that attain a thickness of 500 m near the base of the unit, graphitic schist, and quartz ± feldspar ± muscovite ± biotite schists. The Cahill Formation has been altered by regional retrograde metamorphism; in particular, the lower member is now mildly chloritized and sericitized.

Isotopic data indicate that ARUF unconformity-type deposits are Early to Middle Proterozoic in age. At Jabiluka, U–Pb whole-rock isotopic data were interpreted by Ludwig *et al.* (1987) to show that the ore is 1437 ± 40 Ma, an age that clearly postdates deposition of sandstones of the Kombolgie Formation. Data from Ranger, however, indicate that ore formed at 1737 ± 20 Ma (Ludwig *et al.* 1987), apparently before erosion and subsequent deposition of the Kombolgie Formation. Sm–Nd isotopic ages of primary ores and uraninite from Jabiluka indicate an older, but still post-Kombolgie age of about 1600 Ma, which is similar to Sm–Nd ages determined from ore at Nabarlek and Koongarra (Maas 1987). Sm–Nd data from Ranger cannot be unambiguously interpreted, but Nd–Sr initial ratios are consistent with the pre-unconformity age determined by the U–Pb study (Maas 1987).

Jabiluka consists of two orebodies (Figs. 3, 4): Jabiluka 1, which contains 3500 tonnes of U_3O_8 at a grade of 0.26%, and Jabiluka 2, which contains more than 206,000 tonnes of U_3O_8 at a grade of 0.39% (Ewers *et al.* 1984). Jabiluka 2 also contains 8100 kg of Au at a grade of 15.3 g/t. Gold-bearing assemblages, which include uraninite and tellurides, apparently formed late in the ore paragenesis (Grauch *et al.* 1985). Uranium ore minerals are confined to metasedimentary rocks, even though the deposit formed after deposition of the Kombolgie Formation and even though the Kombolgie Formation sandstone is altered.

The host rock belongs to a brecciated and altered pelitic sequence that is now composed of quartz + chlorite ± muscovite ± sericite ± graphite schist, which commonly contains chlorite-replaced garnet porphyroblasts, and massive chlorite rock. In places, the schists contain apatite- or tourmaline-rich layers. Unaltered amphibolite-facies schists northwest of Jabiluka are probably compositionally and texturally similar to the pre-alteration schists at Jabiluka (Binns *et al.* 1980a). These relatively unaltered schists are medium to coarse grained and are composed of quartz, feldspar, biotite, muscovite, garnet and sillimanite. Carbonate rocks are found between and beneath Jabiluka orebodies, but are largely absent within the orebodies except where they occur at depth in the easternmost part of Jabiluka 2. Intrusive rocks recognized at Jabiluka include tourmaline pegmatite and dolerite.

The Kombolgie Formation at Jabiluka does not host ore and, therefore, has not been studied in the same detail as sandstones of the Athabasca Group, which are major hosts for unconformity-type ore deposits of the Athabasca Basin, in western Canada. The most detailed study of the Kombolgie Formation at Jabiluka is by Gustafson & Curtis (1983), who reported that the unit consists of well-bedded sandstone and subordinate conglomerate and mudstone. The sandstone is composed of detrital quartz grains cemented by quartz, chlorite, sericite and carbonate. Some of this chlorite, and probably sericite, was also formed by hydrothermal solutions. Post-diagenetic chlorite, quartz and carbonate veins cut the sandstone, and in places massive chlorite replaces sandstone. Hematite occurs along quartz grain boundaries, as a minor constituent in quartz veins, and in veins and disseminations formed during subaerial weathering and oxidation.

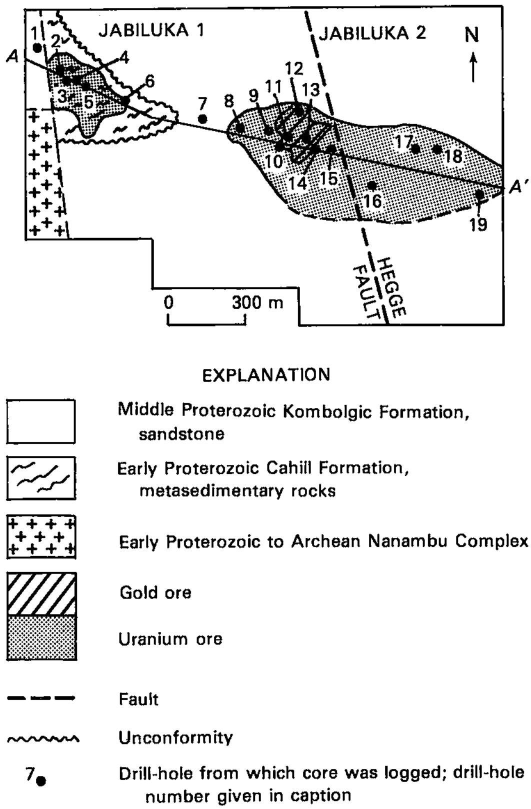

FIG. 3. Bedrock geological map and surface projections of the orebodies at Jabiluka; map modified from Ewers *et al.* (1984). Drillholes from which samples were collected are: 1 GN042V, 2 AN045V, 3 AN048V, 4 BN054V, 5 Y057ND, 6 Y069V, 7 X102V, 8 V111V, 9 V123V, 10 T129V, 11 U132V, 12 V135V, 13 U138V, 14 T141V, 15 T147V, 16 0162ND, 17 S177V, 18 S189V, and 19 N204V.

HOST-ROCK ALTERATION

Alteration at Jabiluka was so pervasive that an alteration halo at least 200 m wide encircles the ore-bearing rocks (Binns *et al.* 1980a) and extends into the overlying Kombolgie sandstone. Chlorite is by far the most abundant mineral in the alteration zone, but white mica, tourmaline and apatite also occur. Added quartz is primarily restricted to fractures and brecciated zones. Hematite, which is prominent at some ARUF deposits, is irregularly distributed and generally is a minor constituent of altered rocks in the cores that were sampled. In the cores sampled from Jabiluka 2, the rocks on the eastern side of the

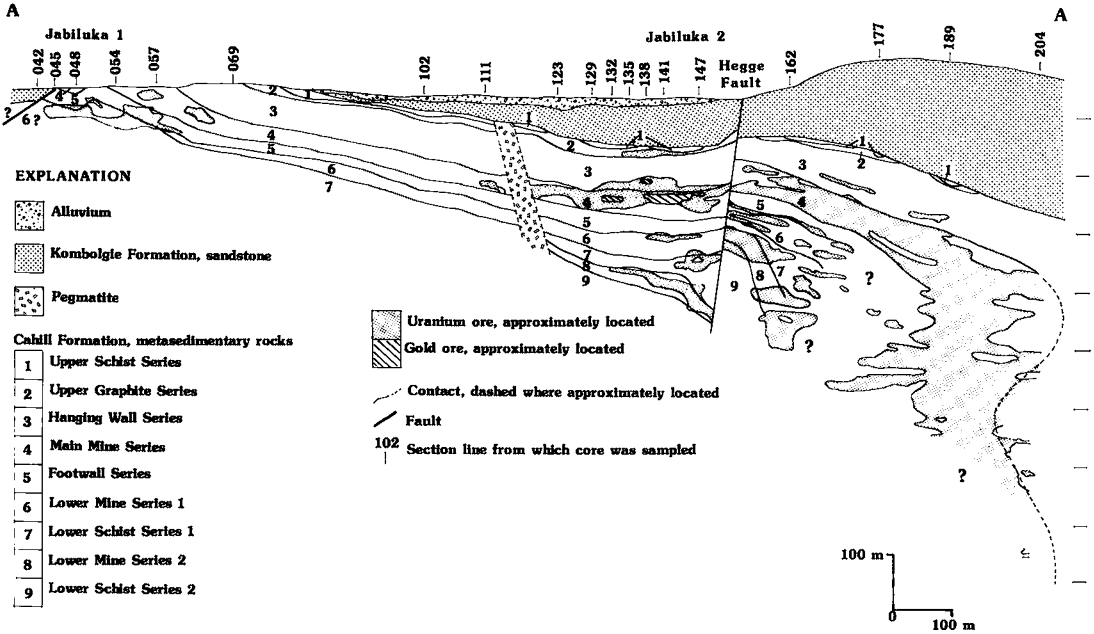

FIG. 4. Long section A–A′ (see Fig. 3) through the Jabiluka 1 and 2 orebodies. Section constructed from cores that were logged for this study, and from data supplied by Pancontinental Mining Limited. Section lines from which a core was sampled are numbered. Contacts are not shown in the east because of poor stratigraphic control.

Hegge fault (Fig. 4) are less intensely altered than those on the western side; compared to the rocks on the western side of the fault, the rocks to the east are less intensely chloritized and contain more white mica. These differences suggest that the fault either separates rocks of different porosity or juxtaposed different parts of the deposit.

The widespread occurrence of chlorite was recognized early during the drilling of Jabiluka (Hegge 1977). Ewers & Ferguson (1980) and Wilde (1988) studied suites of chlorite samples from all the deposits in the ARUF, and Gustafson & Curtis (1983) primarily studied chlorite from altered sandstone of the Kombolgie Formation and massive chlorite zones near the unconformity. These studies contain a limited amount of microprobe data on chlorite. Ewers & Ferguson (1980) noted that in the breccias, the chlorite has high Mg and ^{VI}Al contents and low oxide totals. Because their data are presented mainly on triangular plots, the exact composition of most chlorite samples is unknown. Binns *et al.* (1980b) reported the presence of septechlorite, better known as 7 Å chlorite, and proposed that uranium minerals were deposited with the 7 Å chlorite. Wilde (1988) identified the white mica at Jabiluka as phengite. The presence of phosphorus enrichment and the formation of apatite in altered rocks are reported by Frishman *et al.* (1985), Binns *et al.* (1980a) and Gustafson & Curtis (1983). Binns *et al.* (1980a) noted a slight boron enrichment in the ore zones, whereas Nutt (1984b) and Durak *et al.* (1983) reported the presence of Mg-rich tourmaline in, respectively, the metasedimentary rocks and the sandstone of the Kombolgie Formation.

The zone of pervasive alteration that surrounds the orebodies is characterized by a chlorite and white mica assemblage that is similar to but more intense than the regional retrograde alteration assemblage of the lower member of the Cahill Formation. This white mica is distinguished from muscovite by its fine grain-size and typical association with chlorite. Wilde (1988) identified the white mica associated with fine-grained chlorite as phengite, but in this study examination of samples with the electron microprobe using energy dispersion showed that some of the coarser, but still fine-grained, mica is compositionally similar to the muscovite and is not Mg-rich. Although it is recognized that the white mica may be phengitic, it is herein called sericite because in most samples examined white mica is so intimately mixed with chlorite that it is difficult to determine if the mica is Mg-rich or if sericite and chlorite are mixed or interlayered at a submicroscopic scale. Powder-camera X-ray-diffraction analyses of fine-grained sericite in the matrix show that the mica is a 3*T* polytype.

In the vicinity of Jabiluka, feldspars and ferromagnesian minerals were totally replaced by sericite and chlorite, respectively, so that these rocks now consist of coarse-grained quartz, chlorite and muscovite set in a fine-grained to clay-size chlorite-sericite matrix that makes up 5–95% of the rock. Where in contact with chlorite matrix, quartz grains are embayed. Quartz dissolution apparently is proportional to the intensity of chloritization because the amount of quartz dissolved is greatest in the rocks with the greatest proportion of chlorite matrix. Fine-grained acicular tourmaline, identified as dravite by X-ray diffraction (powder method), is disseminated in the schist matrix and is concentrated along cleavage planes of coarse-grained chlorite. Typically, the dravite occurs in sprays composed of fine grains (up to 0.2 mm but in most cases much shorter) that are clear to light green in transmitted light and that, in many instances, lack detectable birefringence (Fig. 5).

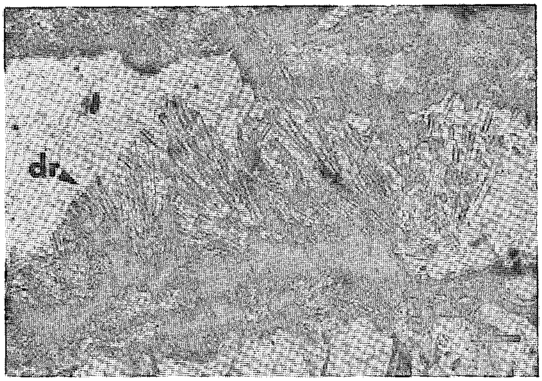

FIG. 5. Photomicrograph (transmitted light) of acicular dravite (dr) in a quartz + dravite vein that cuts across foliation of schist. Bar scale 25 μm.

Ore-bearing brecciated and altered zones, typically a few meters or less in thickness, occur in chloritic schist, graphitic schist, siliceous zones and, where present, carbonate rocks. In these zones uranium occurs in fragments of chloritized schist and broken uraninite and quartz–uraninite veins, as well as in the breccia matrix. Fragments are cemented by quartz, quartz + dravite, chlorite, chlorite + dravite or chlorite + apatite. The quartz is coarse grained and euhedral to subhedral, whereas the chlorite is clay-size in most places. Coarse-grained quartz commonly has growth zones that are easily visible in hand specimen. Apatite occurs both as stubby clear grains and euhedral grains containing hematite inclusions. The presence of chloritized fragments cemented by quartz, which in turn is replaced by chlorite, attests to the complexity of the alteration sequence in these zones.

Veins cut foliation in the schists and bedding in the Kombolgie Formation sandstone, and also occur parallel to metamorphic foliation and along the unconformable contact. Most veins fill fractures, which are no wider than 1 cm, with assemblages of chlorite, quartz ± chlorite, quartz + dravite (Fig. 5), organic matter, or, less commonly, chlorite +

TABLE 1. CHEMICAL COMPOSITION OF METASEDIMENTARY AND SEDIMENTARY ROCKS FROM JABILUKA

	A	B	C	D	E	F	G	H	I
SiO_2 wt.%	63.84	64.14	67.78	67.85	63.86	56.22	43.18	96.80	41.43
Al_2O_3	16.24	14.88	13.15	14.23	16.86	14.69	18.45	1.21	18.14
FeO	3.54	3.20	1.75	1.79	2.25	1.71	4.01	0.12	4.58
Fe_2O_3	1.18	1.07	1.03	0.83	0.86	3.00	1.99	0.42	2.15
MgO	3.64	6.39	7.51	6.48	6.36	8.35	18.42	0.47	17.26
CaO	1.73	0.27	0.17	0.14	0.12	0.18	0.42	0.01	2.55
Na_2O	1.48	0.26	0.04	0.05	0.12	0.06	0.05	0.01	0.02
K_2O	3.58	2.59	1.74	2.33	3.24	2.18	0.44	0.01	0.64
TiO_2	0.54	0.33	0.23	0.26	0.32	0.30	0.24	0.02	0.13
P_2O_5	0.15	0.09	0.11	0.08	0.09	0.10	0.25	0.03	2.03
MnO	0.06	0.05	0.02	0.03	0.02	0.02	0.04	0.01	0.03
LOI	3.57	5.06	5.61	5.15	5.02	12.07	10.94	0.58	9.76
Total	99.55	98.33	99.14	99.22	99.12	98.88	98.43	99.69	98.72
Th (ppm)	19	16	15	19	18	16	18	3	45
U (ppm)	5	64	1,536	810	1,162	2,633	5,583	2	11

A Average composition of lower member Cahill Formation metapelite from Ewers and Higgins (1985); 34 samples
B Slightly altered quartz-chlorite-(biotite)-muscovite-schist; 10 samples
C Quartz-chlorite schist; 11 samples
D Quartz-chlorite-sericite schist; 62 samples
E Quartz-chlorite-muscovite schist; 70 samples
F Quartz-chlorite-graphite schist; 45 samples
G Massive chlorite rock; 14 samples, oxides only from 11 because of interference in rocks with high U
H Least altered sandstone, Kombolgie Formation; 2 samples
I Highly altered sandstone, Kombolgie Formation; 8 samples
For B-I: SiO_2, Al_2O_3, MgO, CaO, K_2O, P_2O_5 determined by XRF; Na_2O, MnO, Th, U determined by INAA; TiO_2 determined by ICP; FeO determined by titration; Fe_2O_3 calculated, Loss on Ignition (LOI) determined at 900°C.

apatite, quartz + apatite, dolomite or sericite. Ore minerals are predominantly associated with the chlorite, quartz ± chlorite, quartz + dravite and organic matter.

Massive chlorite rocks, which are major hosts of disseminated uranium minerals, occur as zones of light to dark green rocks that are composed of up to 95% chlorite. Embayed quartz is present as remnants of dissolved grains. Apatite, florencite (Ce-Al phosphate) and dravite occur disseminated in the chlorite rock, but sericite is rare.

Whole-rock chemistry of altered rocks

The average chemical composition of whole-rock samples from Cahill Formation metasedimentary rocks and Kombolgie Formation sandstones is shown in Table 1. The average composition of metapelites from the lower member of the Cahill Formation (Ewers & Higgins 1985) is shown for comparison. No adjustments could be made for changes in volume because no conclusive evidence exists for any such change. Similarly, a quantitative comparison of altered and unaltered rocks from core material was not made because it is uncertain that similar rock-types are being compared, and because the extent of open space in breccia zones prior to alteration is unknown. Treating Ti as an immobile element actually suggests a volume increase in altered metasedimentary rocks (Table 1), which might be reasonable given all the water that is added to these rocks during chloritization. The major chemical trends described below should be valid because they hold true for up to 30% volume reduction during alteration of schist to massive chlorite rock, and because they correspond to the observed mineralogical changes.

The chemical data for the metasedimentary rocks confirm the observation that the rocks have undergone intense alteration that added Mg, and suggest a relationship between uranium mineralization and chloritization. Binns *et al.* (1980a) found similar relationships in their geochemical study of Jabiluka. All the Jabiluka schists (Table 1, columns B–F), including rocks that show minimal megascopic effects of alteration, are enriched in Mg and depleted in Ca, Na and K (Fig. 6); Si is not noticeably depleted, whereas Al content is variable and apparently related in part to the original composition of the rock. In the massive chlorite rocks (column G), which are assumed to be the end product of chloritization, the chief chemical changes that produced chlorite alteration were a dramatic increase in Mg and a loss of Si, Ca, Na and K (Fig. 6). The Ti, Mn and P con-

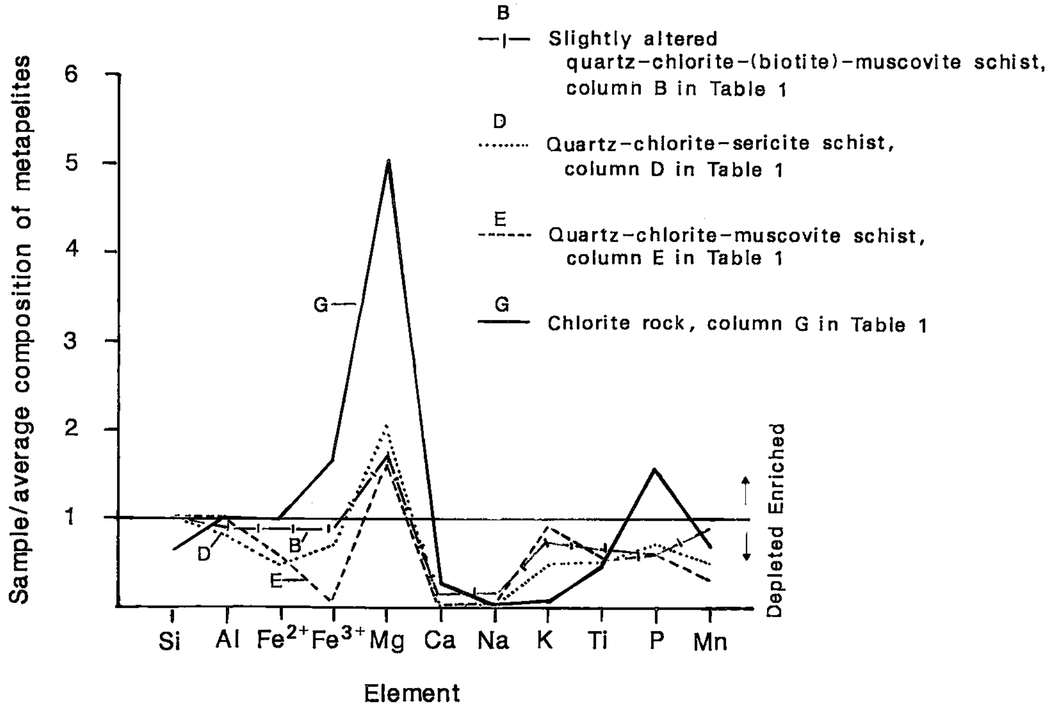

FIG. 6. Variation diagram, normalized to average composition of the metapelite in the lower member of the Cahill Formation, showing changes in composition associated with alteration of some metasedimentary rocks.

tents in all the Jabiluka rocks are about the same as in rocks outside the deposit, although within Jabiluka the P concentrations are higher in the massive chlorite rock. Uranium abundances are, as expected, high in the Jabiluka rocks, but they are not accompanied by an enrichment in Th.

Samples of sandstone from the Kombolgie Formation have been divided into two groups according to the extent of alteration. Compared with the least altered samples (column H), the altered sandstone samples (column I) are enriched in Mg, Al, Fe, Ca, K, Ti, P, U and Th, and strongly depleted in Si.

The chemical compositions of the rocks are consistent with the observed mineralogy. In the altered schists (columns B–F), Ca, Na and K, like the feldspars that originally contained them, are depleted. Introduction of Mg caused chloritization of ferromagnesian minerals and the incipient replacement of all minerals by chlorite matrix. The most altered chlorite rocks are best characterized by their enrichment in Mg and their depletion in Si, Ca, Na and K. The high Mg content is due to the presence of ubiquitous chlorite, and the decrease in Si is a result of the observed dissolution of quartz. Binns *et al.* (1980a) did not report Si depletion, but the rocks in their category of low-grade ore are notably high in Mg and low in Si, and possibly these are highly chloritized rocks from which Si has been lost. Depletion of K is expected inasmuch as sericite is rare in the most chloritized rocks. The enrichment in P and Ca, relative to other altered rocks, is due to the presence of apatite.

This data set does not show the K enrichment reported to be present in the "greisen" of Binns *et al.* (1980a). This disparity, which may be due in part to the fact that different cores were sampled, emphasizes the need for further study of the distribution of white mica. The lack of detectable K-enrichment in this study does suggest that the sericite largely replaced pre-existing K-rich minerals such as K-feldspar and muscovite. Potassium is depleted in the U-rich, highly altered rocks, as expected from the optically observed replacement of all minerals, including sericite and muscovite, by chlorite.

The analytical data on sandstones of the Kombolgie Formation show that alteration there was similar to that which affected the metasedimentary rocks, although the altered sandstones are also enriched in iron. High Mg and Al concentrations in the altered rocks reflect the presence of chlorite, but in these rocks chlorite is not accompanied by significant enrichment in U. As in the metasedimentary rocks, chloritization was accompanied by depletion of Si. High Ca and P concentrations reflect the addition of apatite to the sandstone but, as with chlorite, apatite is not associated with significant enrichment in U. Potassium in altered sandstones resides in either sedimentary clay layers or sericitized rocks. Iron, more abundant in the sandstone than in the metasedimentary rocks, occurs in both chlorite and hematite.

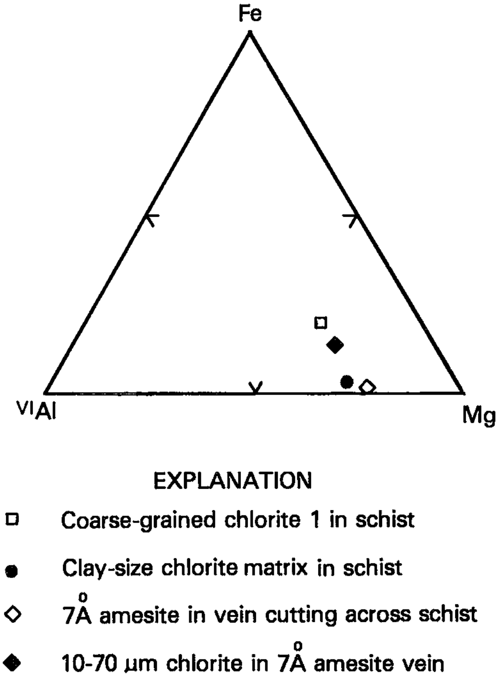

FIG. 7. Triangular plot of Fe–^{VI}Al–Mg showing the variation in chlorite compositions that can occur in a single thin section. Sample Y057ND-160.5.

GENERAL OBSERVATIONS ON CHLORITE

Chlorite is ubiquitous at Jabiluka and invariably is associated with uranium minerals but, because of complex textural relationships and variety in its size, morphology and color, the mineralogy of the chlorite has been poorly described and understood. To better document the mineralogy, chlorite samples

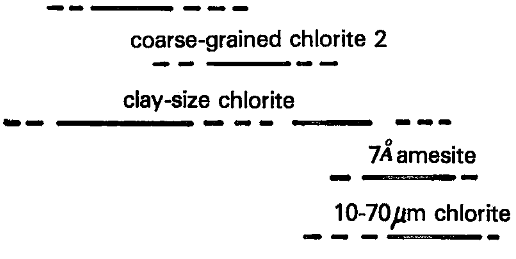

FIG. 8. Paragenetic sequence of the chlorites.

were examined petrographically and grouped according to color, texture, and mode of occurrence. Examples of each type of chlorite were analyzed by electron microprobe to determine if optical and chemical properties are correlative. Microprobe analyses were performed on an ARL-SEMQ apparatus using a wavelength-dispersion system (use of the trade name is for identification purposes and does not imply endorsement by the U.S.G.S.). The following operating conditions were used: 15 kV; 10 nA on brass; 15 μm beam; 40-s counting time for Si, Al, K, and 20-s counting time for Fe, Mg, Ca, Na, Ti, Mn. The

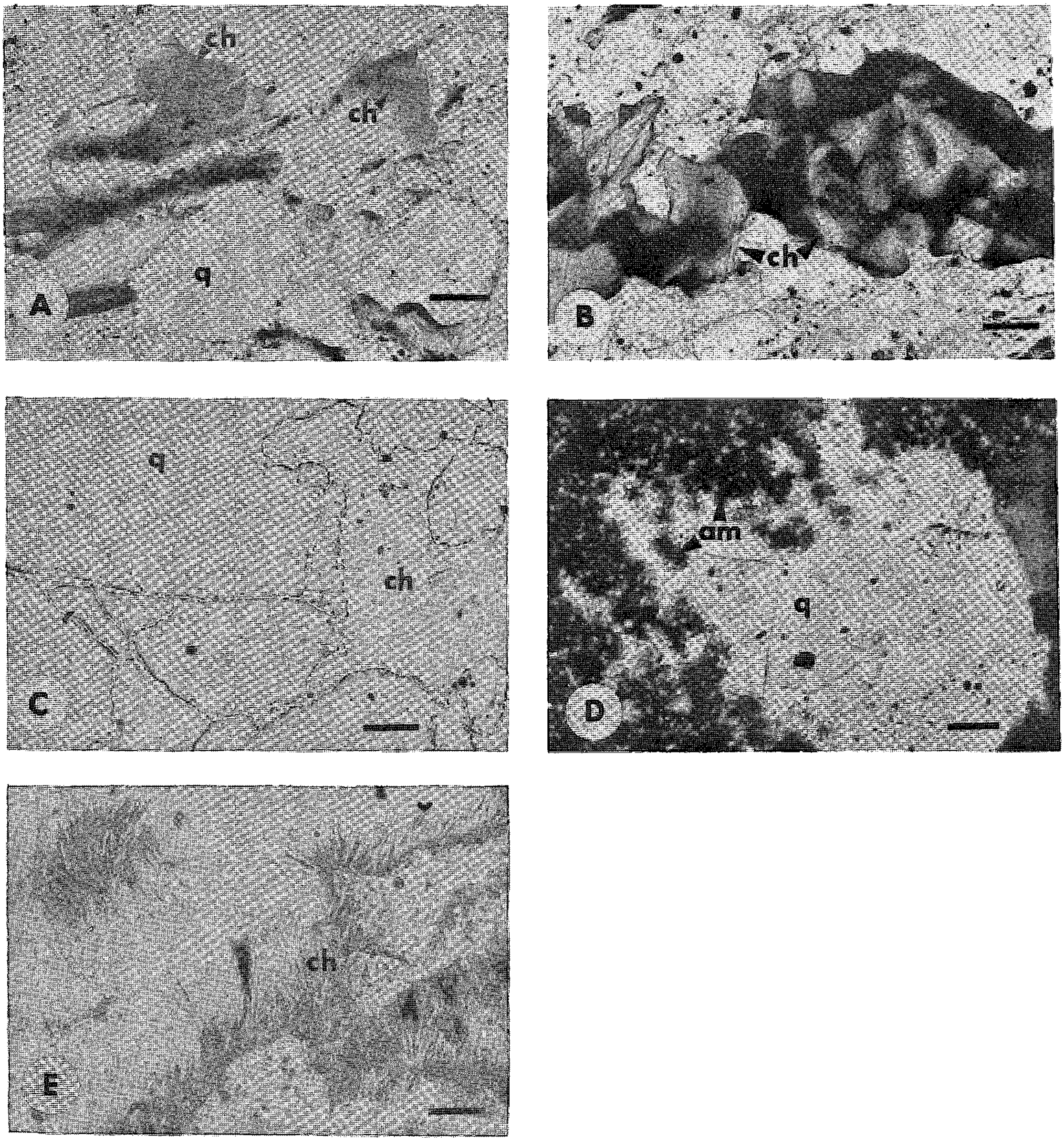

FIG. 9. Photomicrographs (transmitted light) of chlorite from the alteration zone (ch chlorite, q quartz, am amesite). A. Coarse-grained chlorite 1 (ch) partly replaced by the surrounding clay-size chlorite. Bar = 0.1 mm. B. Coarse-grained chlorite 2 (ch) in a vein that cuts quartz–chlorite schist. Bar = 0.1 mm. C. Clay-size chlorite (ch) surrounds embayed quartz grains. Bar = 25 μm. D. 7 Å amesite (dark) has partly replaced quartz grain (crossed polars). Bar = 50 μm. E. 10–70 μm chlorite (ch) cuts clay-size chlorite matrix. Bar = 25 μm.

TABLE 2. ELECTRON MICROPROBE DATA ON THE CHLORITES

	Coarse-grained chlorite 1				Coarse-grained chlorite 2						Clay-size chlorite									
	1 n=10	2 n=7	3 n=8	4 n=13	5a n=9	5b n=10	6 n=9	7 n=10	8 n=8	9 n=5	10 n=10	11 n=20	12 n=12	13 n=7	14 n=10	15 n=9	16 n=20	17 n=15	18 n=10
SiO_2	29.3	30.1	27.6	30.3	24.0	25.6	25.4	24.2	24.6	24.9	34.2	31.7	32.1	32.3	30.2	31.5	33.2	31.9	32.0
Al_2O_3	23.0	21.3	21.8	21.2	22.4	23.9	20.8	23.7	21.8	20.8	23.6	24.9	23.4	21.0	21.7	23.3	21.5	23.8	23.0
FeO	14.7	9.8	19.0	9.6	31.0	27.9	27.3	30.1	30.5	31.2	6.0	5.9	5.1	3.6	3.0	2.1	2.4	5.5	2.8
MgO	18.0	25.6	16.3	22.8	9.2	9.8	13.1	8.5	9.5	10.6	19.0	19.5	22.3	28.7	30.0	28.5	28.8	24.6	29.3
CaO	-	-	-	-	-	-	-	-	0.1	-	0.2	0.2	0.2	0.1	-	-	0.1	-	-
Na_2O	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
K_2O	0.3	-	-	0.2	-	-	-	-	-	-	-	-	-	-	-	0.2	0.4	0.4	-
TiO_2	0.8	0.2	-	1.2	-	-	-	-	-	0.1	-	-	-	-	-	-	-	-	-
MnO	0.2	-	0.2	-	1.1	1.0	0.5	0.9	0.7	0.3	-	-	-	-	-	-	-	-	-
Total	86.3	87.0	84.9	85.3	87.7	88.2	87.1	87.4	87.2	87.9	83.0	82.2	83.1	85.7	84.9	85.6	86.4	86.2	87.1
							Structural formula on the basis of 14 oxygen atoms												
Si	2.93	2.92	2.88	2.99	2.62	2.71	2.73	2.63	2.68	2.71	3.33	3.13	3.14	3.06	2.90	2.97	3.11	3.03	2.97
Al	1.07	1.08	1.12	1.01	1.38	1.29	1.27	1.37	1.32	1.29	0.67	0.87	0.86	0.94	1.10	1.03	0.89	0.97	1.03
Al	1.64	1.35	1.56	1.45	1.50	1.69	1.36	1.66	1.49	1.37	2.04	2.03	1.84	1.41	1.35	1.56	1.48	1.70	1.49
Fe	1.23	0.79	1.66	0.79	2.83	2.47	2.45	2.73	2.79	2.84	0.49	0.49	0.42	0.29	0.24	0.17	0.19	0.44	0.22
Mg	2.68	3.70	2.54	3.35	1.49	1.55	2.10	1.38	1.54	1.72	2.76	2.87	3.25	4.06	4.29	4.00	4.01	3.48	4.06
Ca	-	-	-	-	-	-	-	-	0.01	-	0.02	0.02	0.02	0.02	-	-	0.01	-	-
K	0.04	-	-	0.02	-	-	-	-	-	-	-	-	-	-	-	0.02	0.04	0.05	-
Ti	0.06	0.01	-	0.09	-	-	-	-	-	0.02	-	-	-	-	-	-	-	-	-
Mn	0.02	-	0.02	-	0.10	0.09	0.05	0.08	0.06	0.01	-	-	-	-	-	-	-	-	-
Octahedral total	5.67	5.85	5.78	5.70	5.92	5.80	5.96	5.85	5.89	5.96	5.31	5.41	5.53	5.78	5.88	5.75	5.73	5.67	5.77
Fe/(Fe+Mg)	0.31	0.18	0.40	0.19	0.66	0.61	0.54	0.66	0.64	0.62	0.15	0.15	0.11	0.07	0.05	0.04	0.05	0.11	0.05

	7Å amesite			10-70 μm chlorite				Chlorite in sandstone			Chlorite at the unconformity	Chlorite from sample outside mineralized area	
	19a n=22	19b n=11	20 n=8	21 n=8	22 n=12	23 n=12	24 n=8	25 n=25	26 n=11	27 n=10	28 n=15	29 n=15	30 n=13
SiO_2	28.2	30.0	28.6	29.6	27.9	28.4	27.4	28.5	27.5	33.8	30.6	28.2	34.6
Al_2O_3	24.7	21.8	25.5	24.0	22.6	21.3	22.0	21.9	21.5	17.6	24.1	26.6	26.9
FeO	1.1	0.9	1.2	8.7	15.5	13.1	14.2	13.8	21.0	2.5	7.4	1.9	5.1
MgO	30.0	32.6	29.5	22.2	19.2	22.7	22.2	22.5	17.2	31.8	23.4	27.9	20.3
CaO	-	-	-	-	-	-	-	-	-	0.1	-	-	0.1
Na_2O	-	-	-	-	-	-	-	-		-	-	-	-
K_2O	-	-	-	-	-	-	-	-	-	0.2	-	-	0.4
TiO_2	-	-	-	-	-	-	-	-	-	-	-	-	-
MnO	-	-	-	0.1	-	-	-	-	-	-	-	-	0.1
Total	84.0	85.3	84.8	84.6	85.2	85.5	85.8	86.7	87.2	86.0	85.5	84.6	87.5
	Structural formula on the basis of 7 oxygen atoms						Structural formula on the basis of 14 oxygen atoms						
Si	1.35	1.42	1.36	2.92	2.84	2.86	2.77	2.83	2.83	3.19	2.96	2.69	3.19
Al	0.65	0.58	0.64	1.08	1.16	1.14	1.23	1.17	1.17	0.81	1.04	1.31	0.81
Al	0.75	0.64	0.79	1.71	1.56	1.38	1.39	1.40	1.43	1.15	1.71	1.68	2.11
Fe	0.04	0.04	0.05	0.72	1.32	1.10	1.20	1.15	1.81	0.20	0.60	0.15	0.39
Mg	2.15	2.30	2.09	3.26	2.92	3.40	3.34	3.30	2.63	4.47	3.37	3.97	2.80
Ca	-	-	-	-	-	-	-	-	-	0.01	-	-	0.01
K	-	-	-	-	-	-	-	-	-	0.02	-	-	0.05
Ti	-	-	-	-	-	-	-	-	-	-	-	-	-
Mn	-	-	-	0.01	-	-	-	-	-	-	-	-	0.01
Octahedral total	2.94	2.98	2.93	5.70	5.80	5.88	5.93	5.85	5.87	5.85	5.68	5.80	5.37
Fe/(Fe+Mg)	0.02	0.02	0.02	0.18	0.31	0.25	0.26	0.26	0.41	0.04	0.15	0.04	0.12

n = number of spots analyzed; - = not detected. Total Fe calculated as FeO; TiO_2 value may be due to the presence of small inclusions of rutile. Sample 9 from slightly altered schist from the edge of the deposit. Sample numbers, in order: T147V-300, Y057ND-160.5, T147V-302.6, T147V-204.2, S189V-219C, S189V-219D, Y069V-34.4, T147V-302.6, V123V-156, T147V-233.3A, T129V-205.2, T147V-302.6, V123V-156, AN048-51.0B, Y067V-73.5, T147V-233.3, Y069V-67.0, T147V-300, Y057ND-160.5, Y057ND-160.5A, Y057ND-160.5B, Y067V-73.5, T129V-198.6B, V123V-156, Y069V-73.5, Y057ND-160, V123V-44.1, S177V-147.1, S189V-161.0A, T129V-85.8, CA75-124, CA76-203-6. Description of clay-size chlorite samples; where measured, whole-rock uranium values are shown: no. 10, breccia matrix, 12200 ppm; no. 11, breccia matrix, 6500 ppm; no. 12, alteration zone, 950 ppm; no. 13, chlorite vein; 14 ppm; no. 14, chlorite vein, 359 ppm; no. 15, schist matrix near quartz-uraninite vein, 17100 ppm; no. 16, schist matrix, 18 ppm; no. 17, schist matrix, 1400 ppm; no. 18, schist matrix, 1105 ppm.

following standards were used: albite (Si, Na, Al), orthoclase (K), diopside (Mg, Ca), spessartine garnet (Mn), hematite (Fe) and rutile (Ti). Data reduction was performed using the Magic IV program (Colby 1971). A 15-μm beam was used to minimize volatilization and, in the case of the clay-size to cryptocrystalline chlorite, to obtain an average composition. Analyses of numerous samples confirm that five petrographically identified categories (two types of coarse-grained chlorite, clay-size chlorite, 10-70 μm chlorite and 7 Å amesite) are chemically distinct. Commonly, more than one chlorite occurs at the scale of a thin section (Fig. 7). The proposed paragenetic sequence among the chlorites is shown on Figure 8.

Coarse-grained chlorite

Chlorite grains that are coarse grained (0.1 to 2.0 mm across) relative to the clay-size matrix in schists and breccias predominantly replaced pre-existing biotite and amphibole grains in the altered metasedimentary rocks. The coarse-grained chlorite can be divided into two groups based on optical and chemical characteristics. Both types of chlorite are found throughout the sequence, but chlorite 1 is most prominent in altered schist, whereas chlorite 2 is common in highly altered, ore-bearing chloritic rocks and breccias and occurs in veins as well as disseminations in the schist. In addition, only chlorite 2 contains (rarely) small uraninite grains. Chlorite 1 is pleochroic green to brown, has low birefringence, contains abundant inclusions of dravite and TiO_2 minerals, is corroded, and is typically partly replaced by matrix chlorite (Fig. 9a). In contrast, chlorite 2 is pleochroic green to tan, has purple to bronze birefringence, and in places has radial extinction (Fig. 9b). Compared with chlorite 1, chlorite 2 appears fresher, has fewer TiO_2 inclusions and no inclusions of dravite, and shows only minor embayment along boundaries that are in contact with the clay-size matrix chlorite. These two types of chlorite, which were originally classified on their optical characteristics, have distinctly different compositions (Table 2). Compared to chlorite 2, chlorite 1 is enriched in Mg and Si (Fig. 10), contains less Fe and Mn, and gives slightly lower oxide totals; in addition, the range of Fe and Mg contents in chlorite 1 is much larger than

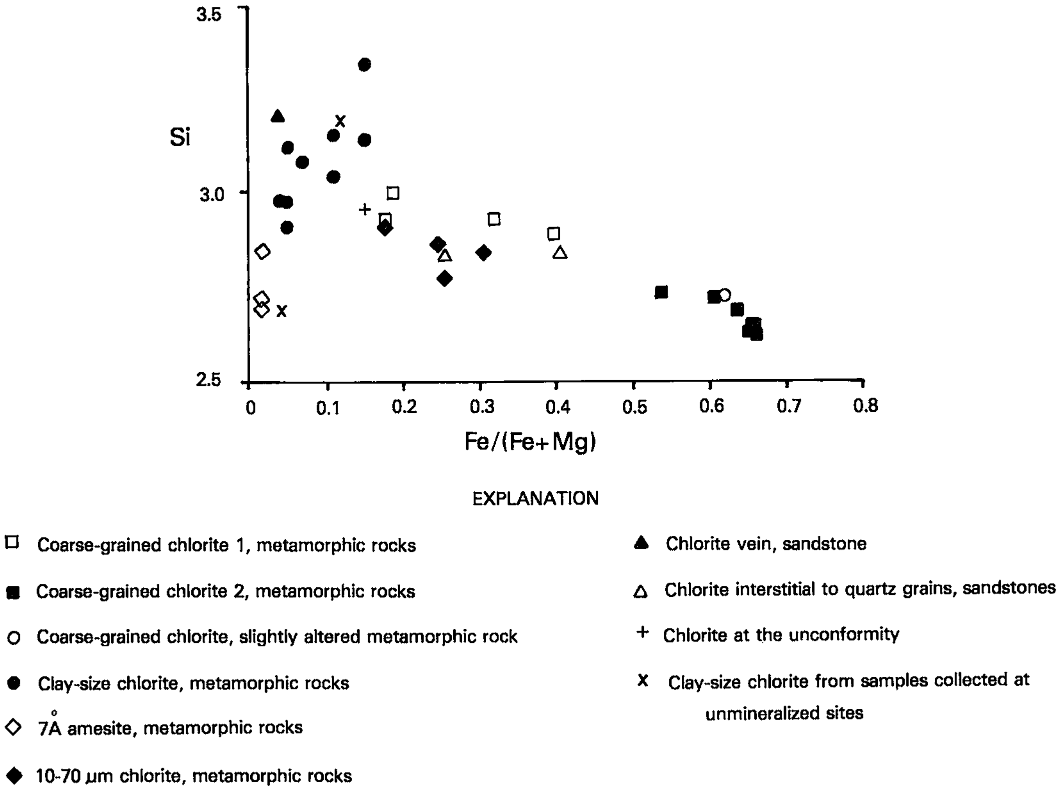

FIG. 10. Variation of Fe/(Fe+Mg) and Si (both expressed as atomic fractions) in the different varieties of chlorite.

that in chlorite 2. The oxide totals of chlorite 2 are the highest of all the analyzed chlorites and are the only ones that fall in the range expected for "normal" chlorite. Chlorite 1 ranges from clinochlore, $(Mg_{3.70}Al_{1.35}Fe_{0.79}Ti_{0.01})(Si_{2.92}Al_{1.08})O_{10}(OH)_8$, to iron clinochlore, $(Mg_{2.54}Fe_{1.66}Al_{1.56}Mn_{0.02})(Si_{2.88}Al_{1.12})O_{10}(OH)_8$; the nomenclature follows Bailey (1980) and Bayliss (1975). Chlorite 2 is classified as magnesian aluminian chamosite; a structural formula for a typical chlorite 2 is $(Fe_{2.47}Al_{1.69}Mg_{1.55}Mn_{0.09})(Si_{2.71}Al_{1.29})O_{10}(OH)_8$.

A chlorite from a slightly altered schist from the eastern edge of Jabiluka 1 was also analyzed (Table 2, no. 9; Fig. 10). This chlorite is fresh, coarse grained, and is similar in composition to chlorite 2 but does not occur with radial extinction, or in veins.

Clay-size chlorite

Clay-size chlorite, which is the main constituent of schist and breccia matrix as well as massive chlorite rock and veins, is the chlorite that is most conspicuously associated with ore minerals. This chlorite, which ranges from 50 μm in length to so fine grained as to appear cryptocrystalline, is called clay-size to emphasize that it is most commonly less than 5 μm across. Optically, clay-size chlorite varies from white to green, and has low to negligible birefringence. At Jabiluka, to some extent this chlorite has replaced all of the primary minerals in the metasedimentary rocks. Textures are massive to pseudomorphic after the primary minerals. Quartz is invariably embayed along contacts (Fig. 9c) and even in rocks having little matrix, chlorite partly rims some quartz grains, whereas grains of coarse-grained chlorite 1 are surrounded and partly to totally replaced by clay-size chlorite. Muscovite grain boundaries are feathery where in contact with chlorite, and small grains are in places replaced by a mixture of chlorite and sericite. Even sericite, which is mixed with chlorite in the schist matrix, is less abundant in the more intensely chloritized schist.

The clay-size chlorite is characterized by high Mg and, compared to other chlorites at Jabiluka, high Si contents (Table 2, Fig. 10). The ratio of ^{VI}Al to ^{IV}Al is highly variable but, as found by Ewers & Ferguson (1980), is consistently high (Fig. 11). The observed range in compositions of the clay-size chlorite may reflect both the compositions of the replaced minerals and variations in the intensity of chloritization. Samples were picked in part for their low content of sericite, but the analyses do show that the K content, and presumably the amount of intergrown or partly replaced sericite, is highest in schistose rocks and decreases in highly altered rocks. The small amounts of Ca that were detected, particularly in the breccia matrix, veins and alteration zones, may be due to the presence of submicroscopic grains of apatite. Oxide totals range from 83.0 to 87.1% but are generally low. Causes for these low totals may be the presence of interlayered or intergranular water, the presence of an interlayered mineral, or unrecognized problems in the analysis of such fine-grained material.

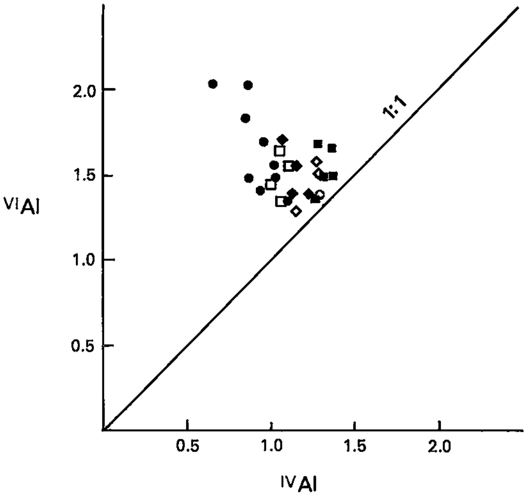

FIG. 11. Values of the ratio of ^{VI}Al to ^{IV}Al in the chlorites in metasedimentary rocks at Jabiluka. Clay-size chlorite shows by far the largest range in values. Symbols as in Figure 10.

X-ray-diffraction studies give *d*(060) values between 1.53 and 1.54, indicating that the clay-size chlorite is trioctahedral (Bailey 1975). The clay-size chlorite is mainly clinochlore, although the chlorite with the highest Si content would more properly be classified as silician clinochlore. Structural formulas range from $(Mg_{4.29}Al_{1.35}Fe_{0.24})(Si_{2.90}Al_{1.1})O_{10}(OH)_8$ to $(Mg_{2.76}Al_{2.04}Fe_{0.49}Ca_{0.02})(Si_{3.33}Al_{0.67})O_{10}(OH)_8$. Cation totals, which range from 5.3 to 5.9 and typically are about 5.7, likely reflect the presence of vacant sites that compensate for the substitution of Al^{3+} for divalent cations in the octahedral sheet.

Microprobe data and X-ray-diffraction patterns both suggest that the clay-size chlorite is mixed with an unidentified mineral, such as 7 Å amesite, that is compositionally similar to chlorite. The X-ray-diffraction pattern of clay-size chlorite shows weak (001) and (003) reflections (Fig. 12), which is more typical of chlorite containing heavy cations, such as Fe-rich chlorite (Bailey 1975); examination of the samples with the electron microprobe using energy dispersion, however, revealed no elements except those shown in Table 2. X-ray-diffraction patterns of two glycolated samples did not show any expansion that would be associated with the presence of expandable interlayered clays. Three of the samples,

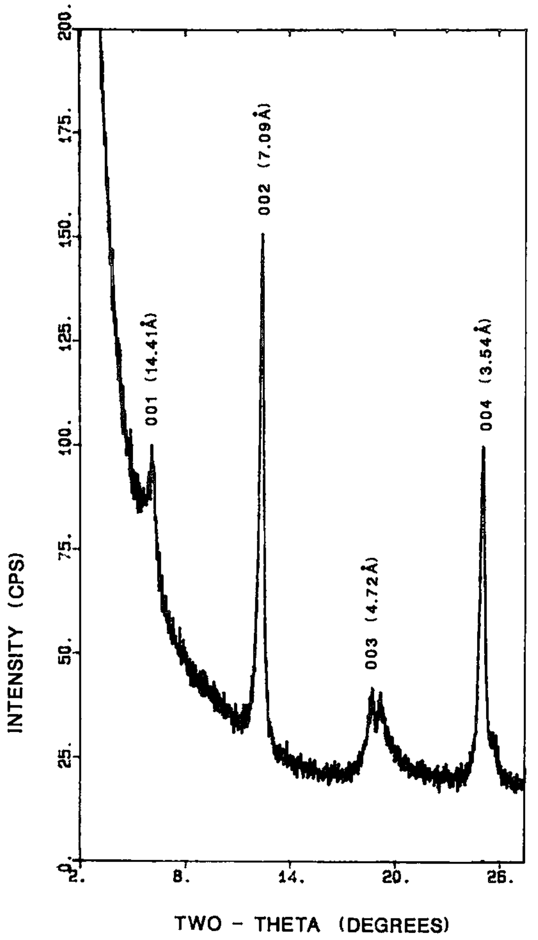

FIG. 12. Part of X-ray-diffraction pattern typical of clay-size chlorite at Jabiluka. Sample V123V-156.

which occur in breccia matrix (Table 2, nos. 10, 11) and in a highly altered zone (no. 12), have noticeably low oxide totals and high ^{VI}Al. In these samples, the ^{IV}Al content expected as a function of $d(001)$ is higher than that calculated from microprobe data, a disparity that suggests a lower Si content in the chlorites than indicated by the microprobe analyses (Bailey 1975). High Si contents in these samples may reflect the presence of cryptocrystalline intergrowths of quartz or another intergrown silica-rich phase.

7 Å amesite

The 7 Å amesite occurs in veins and pods that cut the clay-size schist matrix, and in breccia matrix both of barren lithologies and ore zones. This mineral has a composition similar to that of chlorite, but is characterized by serpentine-like layers with a $d(001)$ of 7 Å. Amesite was identified by X-ray-diffraction data obtained using a Gandolfi camera (Cu radiation, 40 kV, 30 mA, 18-hour exposure); d values are similar to those reported for amesite in Brindley *et al.* (1951). The 7 Å amesite, which is typically less than or equal to 50 µm in size, is distinguishable from the clay-size chlorite in thin section by its birefringence (1st order, but slightly higher than the chlorite), its equidimensional shape, and its white to brown color. The 7 Å amesite is invariably intergrown with, and in places replaced, clay-size chlorite in the matrix and, to a lesser extent, in veins. Quartz in contact with the 7 Å amesite is highly embayed (Fig. 9d). Where the paragenetic relationship can be established, the 7 Å amesite typically is later than much of the clay-size chlorite. Uranium minerals are commonly associated with 7 Å amesite; however, the relationship is incidental, in part, because the 7 Å amesite has replaced the minerals in pre-existing uranium-bearing rocks.

The compositions in Table 2 were obtained from veins of optically white 7 Å amesite. The 7 Å amesite is Mg-rich. Silica content and Fe/(Fe+Mg) and $^{VI}Al/^{IV}Al$ values are lower than those of the clay-size chlorite (Figs. 10, 11). The average structural formula for the 7 Å amesite is $(Mg_{2.18}Al_{0.73}Fe_{0.04})(Si_{1.38}Al_{0.62})O_5(OH)_4$.

10–70 µm chlorite

The 10–70 µm chlorite, so called because most of this variant occurs in this size range, is not distinguishable in hand specimen, but optically it is identifiable by (1) size, which is slightly coarser than the clay-size chlorite, (2) a flaky appearance, and (3) occurrence in veins that cut across the clay-size matrix chlorite (Fig. 9e). The veins are narrow, 0.1 to 0.2 mm in width, and discontinuous. In addition to veins, this chlorite also occurs in clumps scattered through the clay-size chlorite and 7 Å amesite. Although this chlorite occurs throughout the ore zone, only rarely are uranium minerals directly associated with it.

The 10–70 µm chlorite ranges in composition from clinochlore $(Mg_{3.26}Al_{1.71}Fe_{0.72}Mn_{0.01})(Si_{2.92}Al_{1.08})O_{10}(OH)_8$, to iron clinochlore $(Mg_{2.92}Al_{1.56}Fe_{1.32})(Si_{2.84}Al_{1.16})O_{10}(OH)_8$. The composition of the 10–70 µm chlorite, including a trace amount of Mn in one sample, is similar to that of the coarse-grained chlorite 1 (Table 2, Fig. 10). The 10–70 µm chlorite appears to have two origins: (1) its existence in veins suggests that it formed from relatively Fe-rich fluids, and (2) its occurrence elsewhere in clumps, as well as its composition, suggest that the chlorite is relict coarse-grained chlorite 1 grains that were replaced by clay-size chlorite.

Chlorite in Kombolgie Formation sandstone

Chlorite from three samples of Kombolgie sandstone were analyzed, as was a sample of massive chlorite collected from along the unconformity (Table 2). Two sandstone samples (nos. 25, 26) have chlorite that is interstitial to quartz grains, whereas one sample (no. 27) contains chlorite in a cross-cutting vein and associated alteration halo. In the two samples containing interstitial chlorite, the chlorite compositions are similar to those of the 10–70 μm chlorite and coarse-grained chlorite 1 (Fig. 10). In contrast, the chlorite in the cross-cutting vein and alteration halo and in the massive chlorite near the unconformity have low Fe/(Mg + Fe) values and high Si contents similar to those of the clay-size chlorite. As in the highly chloritized metasedimentary rocks, the quartz in contact with this chlorite is highly embayed.

Chlorite outside the deposit

The data set includes two samples, kindly supplied by Stuart Needham of the Australian Bureau of Mineral Resources, that were collected as part of a suite representative of unmineralized rocks (Table 2). The rocks are a quartz–chlorite–muscovite–sericite schist (no. 29), and a chloritized dolomitic schist (no. 30). Both have a chlorite matrix similar in composition to the clay-size chlorite at Jabiluka (Fig. 10). It is interesting that dravite also occurs in one sample.

PARAGENETIC SEQUENCE OF ALTERATION

Minerals that formed during alteration of the rocks at Jabiluka include chlorite (coarse-grained 1, coarse-grained 2, clay-size, 10–70 μm, and 7 Å amesite), sericite, apatite and dravite; quartz was also deposited but is largely confined to breccias and fractures. A proposed paragenetic sequence for these minerals is shown in Figure 13. The sequence is largely schematic because the rocks were subjected to overlapping periods of alteration that are not easily separated into discrete events. As proposed in the Figure, chlorite, dravite, quartz and apatite formed episodically in response to fluctuations in fluid composition.

In contrast to the other minerals in the alteration zone, most sericite formed during the early stages of alteration. Evidence for early formation of sericite includes geochemical data, which show that K is depleted in the more altered rocks; petrography, which shows that sericite is rare in breccia matrix and massive chlorite rocks; and microprobe data, which show that some types of chlorite contain trace amounts of K that may reside in submicroscopic sericite that was mostly replaced by chlorite. The relationship between sericitization and chloritization is unknown; sericite may have been replaced by chlorite as fluid conditions such as a_{K^+} progressively changed, or sericite alteration may have preceded and been unrelated to chloritization. Although chloritization largely overprinted minerals and textures formed during sericitization, the abundance of sericite in ore-bearing rocks from the eastern part of Jabiluka 2 permits speculation that some mineralization occurred with sericitization.

Dravite and apatite are only sporadically associated with uranium minerals. Dravite, which occurs in quartz + dravite veins and breccia cement, and along cleavage planes in coarse-grained chlorite and disseminated in clay-size chlorite, apparently formed over an extended period. The presence of dravite in a chloritized rock from outside the deposit suggests that boron metasomatism accompanied regional retrograde metamorphism. Apatite, which is not as common as dravite, is mainly associated with clay-size chlorite.

Clay-size Mg-rich chlorite, the most abundant chlorite, can be considered the end product of Mg alteration at Jabiluka. The presence of cross-cutting veins of clay-size chlorite is evidence that episodic chloritization affected the rocks. The 7 Å amesite, which predominantly occurs mixed with clay-size chlorite or in veins that cut clay-size chlorite, formed after most mineralization and largely remobilized uranium. The 7 Å amesite may have formed either during the late stages of hydrothermal alteration or

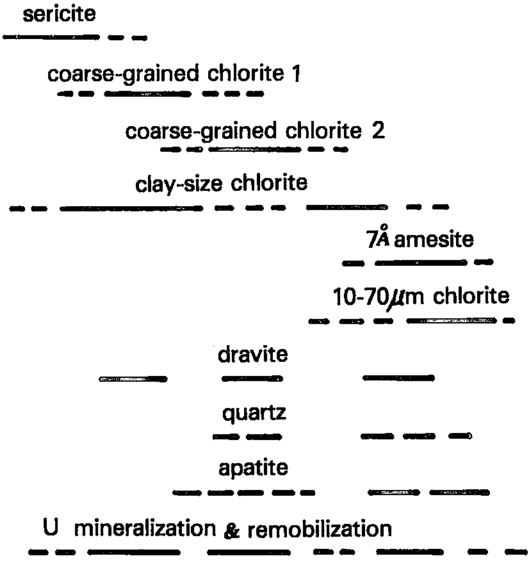

FIG. 13. Proposed paragenetic sequence of minerals in the alteration zone.

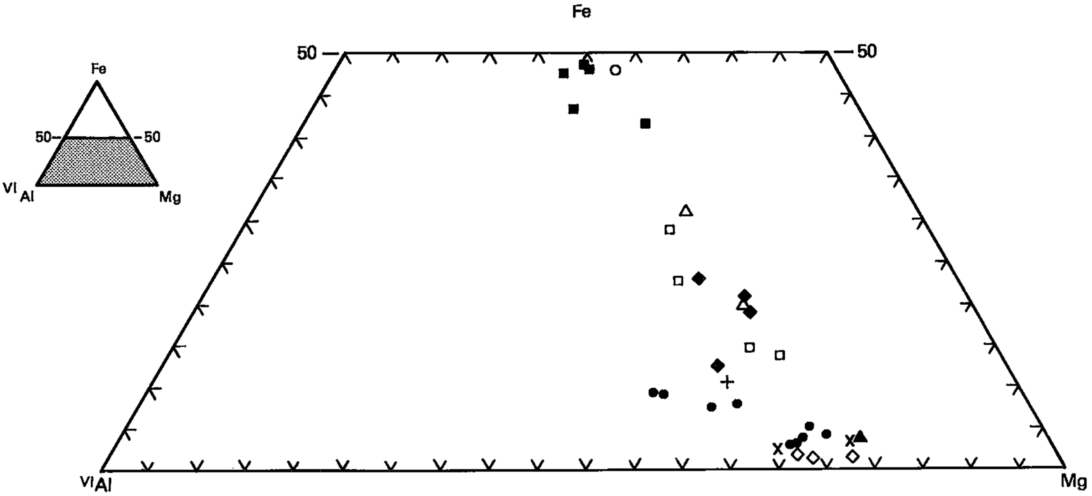

FIG. 14. Part of triangular plot of Fe - ^{VI}Al - Mg showing the compositional variations among the chlorites. Symbols as in Figure 10.

much later, during circulation of cooler, near-surface fluids.

The general trend toward Mg enrichment during alteration is illustrated in Figure 14. The paragenetic sequence, consisting of coarse chlorite 1, clay-size chlorite, and 7 Å amesite, is one of progressively more Mg-rich phases. The intermediate Fe/(Fe + Mg) value of the 10–70 μm chlorite may be inherited from the coarse-grained chlorite 1 that it replaced, or, alternatively, may be due to crystallization from more Fe-rich fluids that circulated long after the major alteration events. Chlorite 2, which appears to have formed contemporaneously with some of the clay-size chlorite and perhaps at the time of mineralization, is the exception to the trend of Mg enrichment during alteration, and is enigmatic in that this Fe-rich chlorite occurs in highly altered rocks. The similarity between the composition of chlorite 2 and coarse-grained chlorite from only slightly altered rocks is unexpected, but suggests that chlorite 2 formed by recrystallization of earlier Fe-rich chlorite.

Chlorite, in particular the clay-size chlorite, is spatially related to ore, but the presence of chlorite-replaced quartz + uraninite vein and breccia cement suggests that some chlorite merely replaced uranium-bearing rocks. Certainly some alteration must have only remobilized and redistributed uranium concentrations at the nearby Ranger deposit. Ranger has a U–Pb isotopic age of ore formation that apparently predates deposition of the Kombolgie sandstone (Ludwig *et al.* 1987), but sandstone as well as ore-bearing schists are chlorite-altered there.

COMPARISON WITH ALTERATION AT OTHER UNCONFORMITY-TYPE DEPOSITS

Alteration zones around other unconformity-type deposits in the ARUF and the Athabasca Basin of Saskatchewan, as well as those associated with uranium deposits in the western part of the Pine Creek geosyncline (Rum Jungle Uranium Field), have mineral assemblages similar to that found at Jabiluka, although it is recognized that each deposit has a unique assemblage. Chlorite, sericite–illite, dravite and, in some cases, hematite and apatite are common constituents of the alteration zones at deposits in all of these districts.

In the ARUF deposits, the clay-size chlorite is typically Mg-rich and has high ^{VI}Al/^{IV}Al and low oxide totals (Ewers & Ferguson 1980, J.T. Nash, pers. comm. 1986, Wilde & Wall 1987). Apatite is reported from Ranger (Nash & Frishman 1985, Frishman *et al.* 1985), Nabarlek (Ewers *et al.* 1983) and White's East deposit in the Rum Jungle (Paterson *et al.* 1984), and dravite has been detected at Ranger (D. Frishman, pers. comm. 1986) and the White's East deposit. Silica depletion is reported at Nabarlek (Wilde & Wall 1987) and in the altered schists at Ranger (Nash & Frishman 1985).

In the Athabasca deposits, altered zones are characterized by the presence of illite–sericite, chlorite and, in some deposits, dravite (Hoeve & Quirt 1984). A halo of quartz dissolution surrounds ore and extends along the unconformity and into sandstone overlying ore. Sandstone is bleached and, in a shell surrounding the high-grade ore, hematite-altered (Hoeve & Quirt 1984). Illite–sericite in altered rocks at unconformity-type deposits and in sandstones of the Athabasca Group has been extensively documented by Hoeve & Quirt (1984). Results from this and previous work have led to the "diagenetic-hydrothermal" model in which fluids associated with diagenesis of the sandstone transport the uranium to a site of mixing with reduced hydrothermal fluids (Hoeve & Sibbald 1978, Hoeve & Quirt 1984, 1987). Chlorite, which is predominantly in altered metasedimentary rocks, is typically Mg- or Mg–Al-rich and is di–trioctahedral or trioctahedral (Hoeve & Quirt 1984). In the deposits of the Cluff Lake area Mg-, Al- and Mg–Fe-rich chlorites that have high $^{VI}Al/^{IV}Al$, low oxide totals, and low cation totals are found in ore zones (Ey *et al.* 1985, Pagel & Svab 1985). Mellinger (1985) proposed that, at Maurice Bay, chloritization overprinted earlier illitization and mineralization.

The Rabbit Lake deposit in Saskatchewan is hosted by a metamorphosed pelitic and carbonate sequence and is probably most similar to Jabiluka in geological setting. As at Jabiluka, altered metasedimentary rocks are enriched in Mg and depleted in Si, Na, K, Ca and Fe; at least three generations of chlorite, one associated with dravite, have been identified (Hoeve & Sibbald 1978). In contrast to the Jabiluka occurrence, hematite is more abundant, and 7 Å amesite is absent in the altered rocks at Rabbit Lake. At Jabiluka, no hematite shell has been identified.

DISCUSSION OF ALTERATION AT JABILUKA

Alteration at Jabiluka consisted of sericitization followed by repeated episodes of chloritization. Uranium minerals are principally in chlorite-altered rocks and in quartz veins and breccia cement. Some uranium mineralization may have accompanied sericitization, but because chlorite overprinting has obscured many textures, no genetic relationship was established by this study. The presence both of chlorite-altered, silica-depleted rocks, and of quartz ± uraninite veins and breccia cement, suggests that the fluid fluctuated between being silica-saturated and silica-undersaturated.

Clay-size Mg-rich chlorite that has high $^{VI}Al/^{IV}Al$ and low oxide totals occurs not only at Jabiluka, but in all the ARUF deposits and also in some Canadian deposits, and it may be typical of unconformity-type deposits. Because chlorite is not in equilibrium with quartz, the chlorite solid-solution model of Walshe (1986) cannot be used to compute the temperature and oxidation state of the solution associated with chloritization. It is not apparent if sparse pyrite, originally sedimentary, is stable or metastable in massive chlorite rock; the equations based on pyrite–chlorite equilibrium that Shikazono & Kawahata (1987) used to characterize the fluid associated with Kuroko massive sulfide mineralization cannot be applied with confidence to Jabiluka rocks. Cathelineau & Nieva (1985) showed that in a suite of chlorites collected from a geothermal field, the temperatures of chlorite formation were inversely related to octahedral vacancy. Using this relationship in the most general way, the data from Jabiluka indicate that the clay-size chlorite formed at a maximum temperature of 200°C. The large number of octahedral vacancies in some of the clay-size chlorite from breccia matrix and massive chlorite zones could be cited as evidence that these chlorite samples formed at lower temperatures than did the chlorite in schist matrix, but the calculated octahedral vacancy may be inordinately low because of contamination by another mineral. Because in this data set the clay-size chlorite samples having the lowest oxide totals and lowest octahedral site occupancy are from permeable breccia and massive chlorite zones, it is possible that these clay-size chlorite samples were subsequently altered by circulating fluids.

The significance of the presence of 7 Å amesite is difficult to assess because 7 Å chlorite is mainly a low-temperature form of chlorite in sedimentary rocks (Velde 1985). Velde (1973) found that instead of chlorite, 7 Å amesite formed and coexisted with chlorite in extremely Mg-rich systems. Although Velde's experimental work was done at 300–700°C, probably hotter than most temperatures of chloritization at Jabiluka, and 7 Å amesite replaces, instead of coexists with, chlorite, it may be that fluids were particularly Mg-rich during the last stages of alteration during which 7 Å amesite formed.

The chemical and textural data in this paper support the suggestion by Gustafson & Curtis (1983) that the widespread chloritization at Jabiluka could have lowered the pH and may have caused destabilization of the U in solution (probably carried as a carbonate or phosphorous complex). In the presence of a reducing agent, which could have been structurally trapped fluid or organic matter similar to that occurring in fractures, U^{6+} in solution could have been reduced and precipitated as a U^{4+} mineral. The Mg-rich alteration mainly involved exchange of Mg for Fe during replacement of earlier-formed chlorite by clay-size chlorite, but conversion of the quartz – feldspar – muscovite assemblage consumed OH and therefore drove down the pH. The reaction quartz + muscovite + K-feldspar + Mg^{2+} + H_2O → Mg-chlorite + K^+ + $H_4SiO_4(aq)$ + H^+ illustrates in a simplified manner how a drop in pH, as well as a loss of Si, should accompany chloritization of a quartz – K-feldspar – muscovite schist. Dissolution of dolomite and magnesite, which are so noticeably absent in the ore zones at Jabiluka and are a likely source of Mg, may have prevented extreme drops in pH. The presence of carbonate in the eastern part of Jabiluka 2, which is also the least chloritized part of the deposit, supports a link between chloritization and carbonate dissolution.

Other studies have documented a drop in solution pH and a loss of Si in the rocks during Mg metasomatism. Experimental studies of water–rock interaction under the conditions of formation of massive sulfides (seawater–basalt and seawater–greywacke at 200°C and higher temperatures) established that Mg is quickly removed from the fluid and incorporated in clays; accompanying the fixation of Mg in the altered rocks are a loss of Ca and K, and a drop in the pH of the fluid (Bischoff & Dickson 1975, Bischoff *et al.* 1981). Reed (1983) calculated seawater–basalt reactions at 300°C and reported that the formation of chlorite, accompanied by loss of Si, occurred at high water–rock ratios. In addition, Goldhaber *et al.* (in press) propose that in a zone of quartz dissolution, chloritization of the rocks in a sandstone-type uranium deposit lowers the pH. The results of these studies are not directly applicable to the Jabiluka deposit; it probably formed at temperatures higher than those relevant to the sandstone-type uranium deposits, but probably near the lower limit of temperatures used in the experiments. The results do suggest, however, that chlorite formation and quartz dissolution, as well as a loss of Ca and K, can be expected from interaction of a Mg-rich fluid with country rock at a high water–rock ratio.

Silica solubility would have been enhanced by any increase in fluid temperature (Kennedy 1950). Wilde & Wall (1987) and Hoeve & Quirt (1984) have attributed the dissolution of quartz at the Nabarlek and Athabasca deposits, respectively, to increases in fluid temperature. Wilde & Wall proposed that the temperature increase was due to heating during downward movement of the fluid through the sandstone, whereas Hoeve & Quirt suggested that heating was caused by the introduction of warm fluids from the basement rocks. This study emphasizes that although a rise in temperature increases quartz solubility, a Mg-rich hydrothermal fluid, in particular a silica-undersaturated brine, interacting at high fluid–rock ratios with the country rock, can produce silica-depleted altered rocks.

The alteration at Jabiluka was more intense than, but similar to, the alteration accompanying the regional retrograde metamorphism that affected the lower member of the Cahill Formation. This similarity suggests that either the alteration at Jabiluka was related to the regional retrograde event or, alternatively, that the fluid involved in both the regional event and the localized alteration at Jabiluka were derived from a common source; such fluids may have moved down large fractures and faults through the metamorphic rocks. The most likely source of the Mg, P and B that were enriched in the alteration zone is the metasedimentary sequence, which contains Mg-rich carbonate and layers that are apatite- or tourmaline-rich. The original source of the fluid is unknown, although Nd–Sr isotope data from Jabiluka permit speculation that some component of the fluid moved through the Kombolgie Formation (Maas 1987). Note, however, that at Ranger, where rocks are altered to an assemblage quite similar to that at Jabiluka, Nd–Sr data from ore is consistent with a fluid that only had contact with Early Proterozoic and Archean rocks (Maas 1987).

The present study suggests that chloritization and quartz dissolution were the products of alteration by a Mg-rich fluid, perhaps a brine, at high values of the fluid–rock ratio. The low Fe/(Fe + Mg) values of the clay-size chlorite attest to the Mg-rich composition of the fluid throughout chloritization. Lowering of pH may have caused dissolution of carbonate rocks and destabilized uranium in solution, although the presence of chlorite-dominated alteration in sandstone indicates that falling pH was not the sole control on mineralization. The presence both of chlorite-altered, silica-depleted rocks, and of quartz–uraninite veins and breccia cement, suggests that during alteration and mineralization the fluid fluctuated between being silica-saturated and silica-undersaturated. These fluctuations could have been caused by periods of heating and cooling, by repeated influxes of fluid that underwent changes in chemistry during alteration and cooling, or by repeated influxes of fluids having different compositions. Fluctuations in temperature or composition could have been caused by fluid mixing at the unconformity, as proposed by Hoeve & Quirt (1987) for Athabasca deposits and by Wall *et al.* (1985) for ARUF deposits, but alternative hypotheses such as repeated influxes of hydrothermal fluid cannot be dismissed. The constraints on alteration as presented in this paper neither require nor preclude the unconformity as necessary for ore concentration.

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