

SUDOITE FROM CIGAR LAKE, SASKATCHEWAN

JEANNE B. PERCIVAL

Ottawa-Carleton Geoscience Centre, Carleton University, Ottawa, Ontario K1S 5B6

HIDEOMI KODAMA

Land Resource Research Centre, Agriculture Canada, Ottawa, Ontario K1A 0C6

ABSTRACT

Chlorite in the alteration zone around the Cigar Lake uranium deposit in the Athabasca basin, northern Saskatchewan, has the composition: $(Al_{2.86}Mg_{1.92}Fe_{0.13})(Si_{3.30}Al_{0.70})O_{10}(OH,F)_8$. This is a di-trioctahedral variety, sudoite, with a *I1b* polytype structure, and characterized by an intense 006 reflection at 4.74 Å and a $d(060)$ of 1.515 Å. Abundant sudoite occurs in a well-defined halo around the ore and probably formed at the expense of illite and kaolinite, during hydrothermal alteration associated with uranium mineralization.

Keywords: sudoite, di-trioctahedral chlorite, illite, uranium, Cigar Lake, Athabasca basin, Saskatchewan.

SOMMAIRE

La chlorite de la zone d'altération autour du gisement d'uranium de Cigar Lake, dans le bassin d'Athabasca du nord de la Saskatchewan, a comme composition $(Al_{2.86}Mg_{1.92}Fe_{0.13})(Si_{3.30}Al_{0.70})O_{10}(OH,F)_8$. C'est une sudoite, chlorite di-trioctaédrique, à structure polytypique *I1b*; une réflexion 006 de forte intensité à 4.74 Å et une valeur $d(060)$ de 1.515 Å sont typiques de cette variété de chlorite. La sudoite, abondante, est distribuée dans une auréole bien définie autour de la zone minéralisée, et aurait remplacé l'illite et la kaolinite lors de l'altération hydrothermale associée à la minéralisation en uranium.

(Traduit par la Rédaction)

Mots-clés: sudoite, chlorite di-trioctaédrique, illite, uranium, Cigar Lake, bassin d'Athabasca, Saskatchewan.

INTRODUCTION

Sudoite, a di-trioctahedral chlorite, occurs in a wide variety of environments, for example in hydrothermal deposits such as the iron ores of the Tracy mine, Michigan (Bailey & Tyler 1960) and Kuroko-type deposits in the Kamikita (Hayashi & Oinuma 1964, Sudo & Sato 1966), Itaya (Henmi & Yamamoto 1965) and Shinyo (Fujii *et al.* 1971) deposits, Japan. Sudoite also occurs in sedimentary rocks (Schultz 1963, Müller 1967), in low-grade metamorphic terranes (Fransolet & Bourguignon 1978, Kramm 1980, Franceschelli *et al.* 1989) and in the AB horizon of the Alberni soil series in British

Columbia (Brydon *et al.* 1961). Although Hoeve & Quirt (1984) reported the occurrence of sudoite from hydrothermal, unconformity-type uranium deposits and sedimentary rocks of the Athabasca Group in the Athabasca basin of Saskatchewan, no structural or chemical data were presented. The purpose of this paper is to provide mineralogical details of sudoite from one such setting of the Athabasca basin, the Cigar Lake uranium deposit.

NOMENCLATURE

Diocahedral chlorites have been subdivided into three types, di-trioctahedral, di-dioctahedral and a poorly crystallized type (Eggleton & Bailey 1967). The first, with about 5 octahedral cations per formula unit, $O_{10}(OH)_8$, contains a dioctahedral 2:1 silicate layer and a trioctahedral interlayer hydroxide sheet. Examples of di-trioctahedral chlorites include sudoite and the Li-bearing variety *cookeite*. Theoretically, this first type could contain a trioctahedral 2:1 layer and a dioctahedral interlayer hydroxide sheet, but no examples have been reported. The second type has a dioctahedral 2:1 silicate layer and a dioctahedral interlayer hydroxide sheet and contains slightly more than 4 octahedrally coordinated cations per formula unit. An example of di-dioctahedral chlorite is donbassite. The poorly crystallized variety (type 3) is similar to the di-trioctahedral and di-dioctahedral types, but has an incompletely developed interlayer.

Diocahedral chlorites are identified by their more intense 006 reflection (based on a 2-layer structure) at 4.74 Å with respect to the 004 reflection at 7.1 Å, and a $d(060)$ between 1.49 and 1.51 Å. In contrast, the more common trioctahedral chlorites have a $d(060)$ between 1.53 and 1.56 Å, with the reflection at 7.1 Å stronger than the one at 4.74 Å.

GEOLOGICAL SETTING

The Cigar Lake uranium deposit occurs within the Athabasca basin of northern Saskatchewan (Fig. 1), near the unconformity between Aphebian graphitic metapelites of the Wollaston Group (Domain) and Helikian gritty sandstones of the Manitou Falls For-

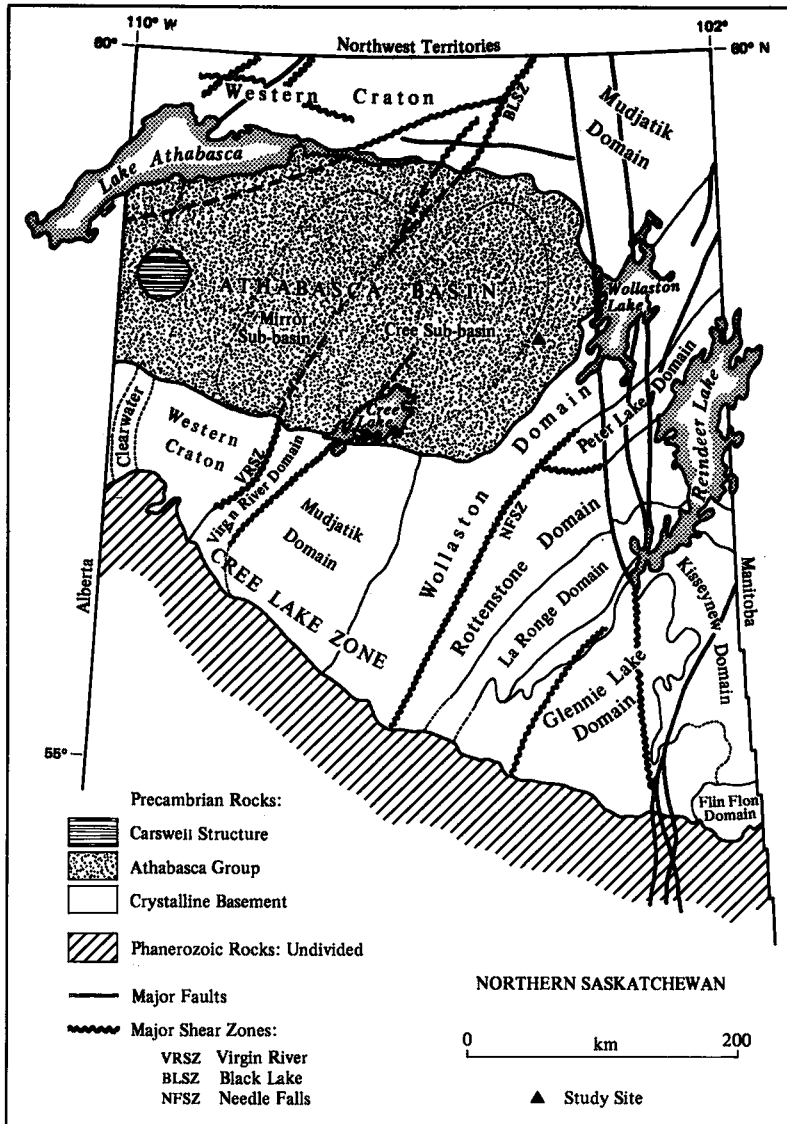


FIG. 1. Regional geology of northern Saskatchewan and location of study area (after Macdonald & Broughton 1980).

mation of the Athabasca Group. The deposit is about 2200 m long, trends in an E-W direction, and varies from 20 to 105 m in width. It is the world's largest and richest known uranium deposit, with estimated reserves of 150,000 tonnes of uranium metal, with an average grade of 8% U_3O_8 (Bruneton 1987).

The predominantly grey to purple sandstones hosting the deposit contain detrital quartz, altered muscovite, relict biotite, zircon, ilmenite and neofomed hematite set in an illite and kaolinite matrix (Bruneton 1987). The ore deposit, at a depth of 410 to 450 m, is surrounded by a zoned, hydrothermal halo

(Fig. 2). From the outside in toward the ore, the halo consists of: (1) a grey alteration zone marked by a minor decrease in quartz content and increase in clay mineral content, (2) a quartz-rich zone (quartz-cemented cap), (3) a grey alteration zone characterized by less quartz than (1) and up to 30% clay (altered sandstone), (4) a massive clay zone (clay-rich halo), and (5) a hematite-rich clay zone that directly encloses the ore (Fouques *et al.* 1986, Bruneton 1987). Hydrothermal alteration of the basement has been observed up to 100 m below the ore body. According to Bruneton (1987), this alteration masks

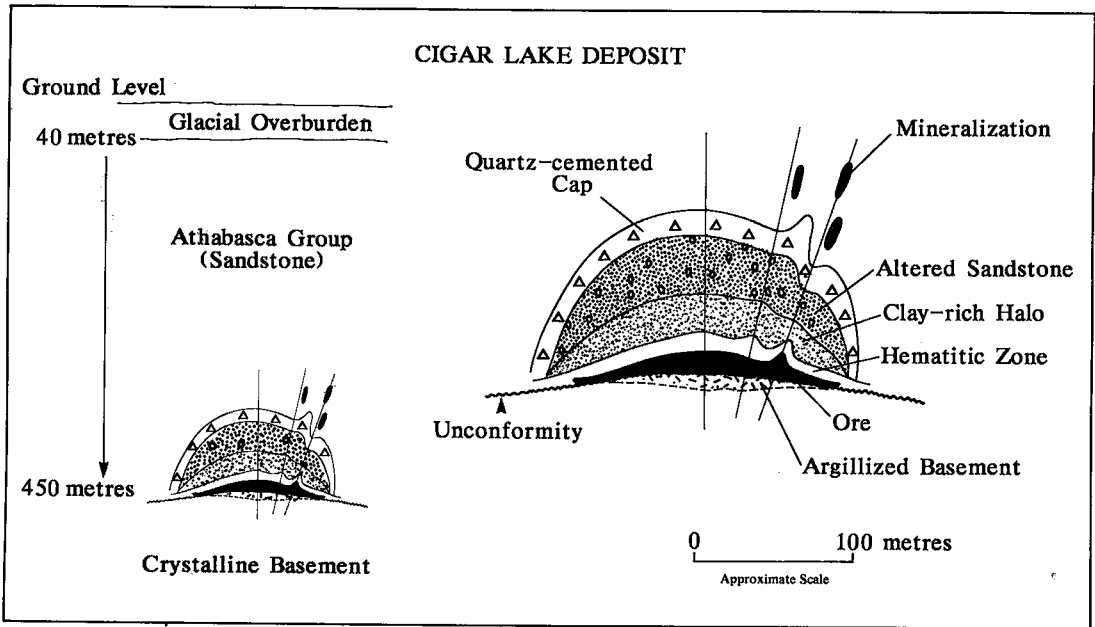


FIG. 2. Schematic cross-section of the alteration halo surrounding the Cigar Lake uranium deposit. The ore deposit is overlain by the Manitou Falls Formation of the Athabasca Group (after Fouques *et al.* 1986, Cramer 1986).

the original, pre-Athabasca paleoweathering, which was described elsewhere in the basin by Hoeve & Sibbald (1978) and Macdonald (1985).

Sudoite occurs in zones above and below the ore deposit and is more abundant near the unconformity. Trioctahedral Fe-chlorite or Mg-Fe-chlorite is present within the ore zone. In contrast, Hoeve *et al.* (1981) reported that unaltered sandstones of the Manitou Falls Formation usually contain equal amounts of kaolinite and illite, but no sudoite. Sudoite, however, is abundant in other units, especially the Upper Wolverine Point Formation, which overlies the Manitou Falls Formation. Macdonald (1980, 1985) reported that a "Mg-rich" chlorite $[(\text{Fe}_{0.05}\text{Mg}_{1.75}\text{Al}_{3.25})(\text{Si}_{3.5}\text{Al}_{0.5})\text{O}_{10}(\text{OH})_8]$ with about 50% dioctahedral character occurs in the green alteration zone of the paleoweathering profile (regolith) developed below the Athabasca unconformity.

MATERIALS AND METHODS

A suite of 152 drill-core samples from twelve boreholes at the Cigar Lake site was selected for a detailed clay-mineral - trace-element study. The suite included samples from a borehole distal to the ore deposit. Fifty of the selected samples are indurated orthoquartzite sandstones, 95 are variably altered sandstone, and 7 are altered basement specimens, which are probably metapelites. The altered sandstone and basement samples contain up to 80% clay-

and silt-sized material, with approximately 23% containing sudoite.

The sudoite-bearing samples were selected from the 440.6-m (53-14) and 444.0-m (53-15) depth of borehole WDG1-53 (Fig. 3) for detailed mineralogical analysis. This borehole intersects the ore zone at a depth of 427.0 - 436.5 m and the unconformity at 436.0 m. Samples 53-14, used for detailed X-ray-diffraction analysis, and 53-15, used for microprobe analysis, are altered basement samples of the Wollaston Group.

The $< 2 \mu\text{m}$ clay-size fraction was separated by a combination of sedimentation and centrifugation. X-ray patterns of nonoriented and oriented samples were recorded on a Scintag PAD V automated powder diffractometer equipped with a graphite monochromator, using Co radiation ($\lambda = 1.7902 \text{ \AA}$). The oriented sample was prepared by drying 30 mg of the sample suspended in 1 mL of water on a 25 mm \times 30 mm glass slide. Glycerolation of a duplicate sample was achieved by substituting 1 mL of 2% glycerol-water solution for water in the above procedure. A Guinier - de Wolff focusing camera (transmission type) was used to record nonbasal reflections, using Co radiation.

A polished thin section of a plastic-impregnated sample (Sheldrick 1984) was used for microprobe analyses on a CAMEBAX wavelength-dispersion microprobe, operating at 15 kV accelerating voltage, with 30 nA regulated beam current. The analytical

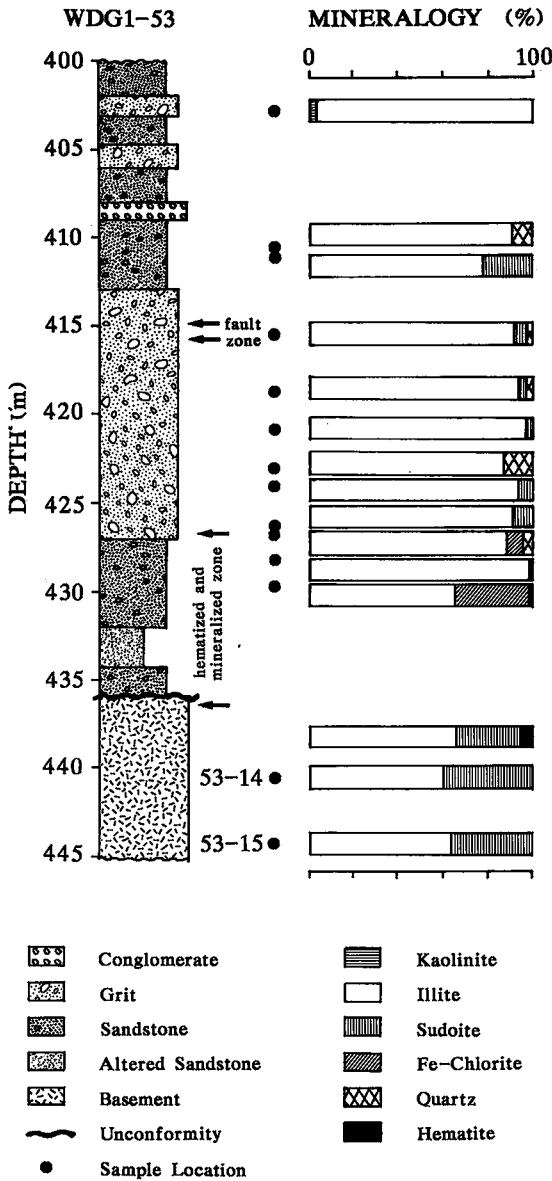


FIG. 3. Lithology and clay composition for samples taken from borehole WDG1-53. Lithology taken from drill core log. Summation of I (illite) + K (kaolinite) + C (chlorite-sudoite) is 100%.

data on sudoite were reduced with a mica routine using a phlogopite standard.

RESULTS

X-ray diffraction

X-ray-diffraction data obtained using a random amount of sample 53-14 are presented in Table 1.

TABLE 1. X-RAY POWDER-DIFFRACTION DATA

hkl	1		2		3		4	
	I	d(obs.)	I	d(obs.)	d(calc.)	I	d(obs.)	
002	19	14.19Å	40	14.2Å	14.16Å	80	14.23Å	
004	92	*10.07	80	7.1	7.08	70	7.13	
006	35	*5.01						
020	100	4.74	100	4.72	4.721	80	4.735	
021					4.747			
111			4	4.50	4.496	85	4.517	
111	8	*4.4 B			4.483			
111			10	4.35	4.387	5	4.314	
112					4.384			
112					4.340			
114					4.318	5	4.180	
113	3	4.08	10	4.01	4.024	5	4.017	
113					4.004			
113	3	3.89	5	3.87	3.913	2	3.840	
025					3.905			
008			54	3.54	3.533	65	3.552	
025					3.561			
115	67	*3.345	1	3.36	3.361			
115					3.350			
026			6	3.126	3.289	2	3.272	
026					3.260			
116	5	*3.072	2	3.10	3.102	4	3.084	
116					3.092	2	2.923	
0,0,10	14	2.851	25	2.835	2.832	20	2.839	
202					2.819			
132	3	*2.694	10	2.615	2.618	10	2.606	
132					2.610			
204					2.590			
132	5	*2.557	20	2.545	2.549	25	2.544	
132					2.544			
202	10	2.512	90	2.502	2.503	100	2.501	
134					2.502			
206					2.412			
134			35	2.408	2.412	30	2.409	
134					2.402			
134					2.384			
0,0,12	3	2.372	15	2.358	2.360	25	2.348	
1,1,11					2.357			
204					2.348			
136			10	2.345	2.347			
1,1,11					2.346			
136	2	2.224	20	2.237	2.270	25	2.230	
208					2.269			
206	2	*2.16 B	5	2.165	2.165	2	2.160	
138					2.164			
138					2.051			
2,0,10			1	2.050	2.050			
0,0,16			10	2.020	2.023	5	2.024	
1,3,10	27	*2.020	20	1.995	1.996			
208			18	1.978	1.980	50	1.984 B	
1,3,10					1.979			
1,3,10					1.871			
2,0,12	2	1.88 B	5	1.870 B	1.871	15	1.868	
1,3,12					1.858			
1,3,12			12	1.820	1.820			
2,0,10	2	1.817	8	1.803	1.805	25	1.812 B	
1,3,12					1.804			
0,0,16	2	1.774	1	1.770	1.770			
312					1.717			
311					1.711			
0,4,11	1	1.70 B	10	1.711	1.712	15	1.710 B	
1,2,12					1.706			
132					1.705			
314					1.705			
123			5	1.673	1.675			
245					1.674			
124	2	1.660			1.650			
240			1	1.650	1.650	5	1.643 B	
2,0,12					1.647			
1,3,14					1.646			
245			1	1.600	1.600			
137					1.600			
1,3,14			15	1.560	1.559	40	1.556 B	
2,0,16	2	1.555			1.559			
1,3,14			25	1.549	1.549			
332					1.516			
060	2	1.512	50	1.515	1.516	60	1.511	
332					1.514			
332					1.480			
336	2	*1.501	20	1.480	1.480	10	1.478	
332					1.480			
066					1.479			
338					1.447			
066					1.447			
334					1.441	5	1.439	
338	3	1.421	10	1.422 B	1.441			
334					1.440			
066					1.439			
0,0,20			7	1.419	1.416	2	1.418	
068			20	1.398	1.398			
3,3,10					1.398			
336	2	1.386			1.389	40	1.389 B	
068					1.389			
1,5,13			15	1.384	1.383			

1. hkl data from Bailey & Lister (1989). 2. Sample 53-14, containing about 60% illite and 40% sudoite. Reflections due to illite or dominantly influenced by illite indicated by *. 3. Data for sudoite from Otré, Belgium, and calculated d-values based on triclinic cell, with cell parameters a 5.247, b 9.094, c 28.357 Å, α 90.5°, β 97.3°, and γ 89.9°, from Bailey (1988) and Lin & Bailey (1985). Frenschel & Bourguignon (1976) first reported X-ray powder-diffraction data for the mineral from Otré, which had been indexed with a monoclinic single-layer structure. 4. Data for sudoite from Tracy mine (Eggleton & Bailey 1967). B denotes broad peak.

Relative intensities of peaks were obtained by direct measurements on the diffractogram using the 4.74-Å reflection as 100% intensity. Illite, identified by its 10.07, 5.01, 3.345 and 2.008 Å basal reflections, makes up about 60% of the sample, as determined by the method outlined in Hoeve *et al.* (1981) and Mellinger (1985). Data for other sudoite samples from the Otrré and Tracy mines, as reported by Bailey & Lister (1989) and Eggleton & Bailey (1967), respectively, are shown in Table 1 for comparison. Eggleton & Bailey (1967) determined that sudoite associated with the Tracy and Kamikita deposits has a chlorite I**b** structure. The sudoite from the latter deposit was originally described by Hayashi & Oinuma (1964). Comparison of the *d*-values of sam-

ple 53-14 with the patterns of these samples, in conjunction with criteria from Bailey (1980), suggests that sample 53-14 contains a I**b** polytype as well. A Guinier - de Wolff photograph of the sample showed the 060 reflection to be a doublet of 1.515 Å and 1.503 Å, the latter due to admixed illite. These values are consistent with a dioctahedral structure.

Figure 4A shows the diffractometer trace of an oriented aggregate of 53-14. Both illite and sudoite exhibit a well-developed series of 00 l reflections. No expansion resulted from glycerol treatment. The 002 reflection increased in intensity by a factor of seven after heating to 550°C for 0.5 hours and decreased in spacing to 13.8 Å. The 008 peak remained, but the 004 and 006 peaks were barely detectable. After

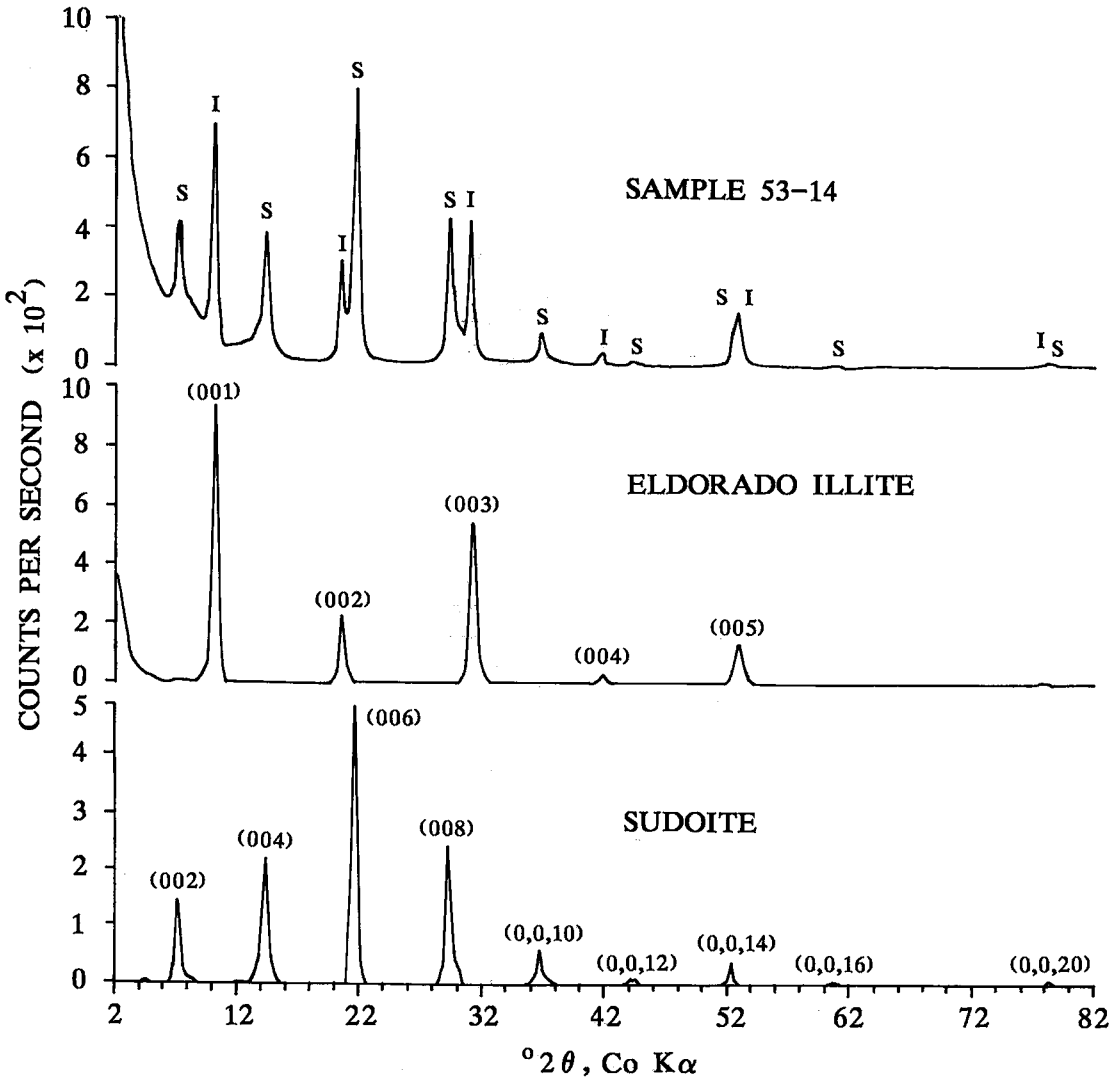


Fig. 4. X-ray-diffraction patterns (CoK α ; air-dried, oriented mounts) for: (A) sample 53-14, (S sudoite, I illite), (B) reference illite from Eldorado (Kodama & Dean 1980), and (C) differential pattern for sudoite.

TABLE 2. BASAL SPACINGS, OBSERVED AND CALCULATED INTENSITIES AND STRUCTURAL FACTORS

00l	d(Å)	d(002)	I _{obs}	I _{corr}	F _{obs}	F _{obs} ¹	F _{calc} ²
2	14.18	14.18	146	146	20.1	31.4	20
4	7.12	14.24	223	232	51.2	60.5	73
6	4.75	14.25	489	555	120.4	118.8	-128
8	3.54	14.16	246	309	122.8	121.0	-190
10	2.84	14.20	60	83	81.3	100.2	-83
12	2.38	14.28	9	14	40.5	39.2	-
14	2.02	14.14	38	64	106.5	85.0	-
16	1.77	14.16	4	7	42.3	45.8	-
18	1.57	14.13	1	2	25.6	24.4	-
20	1.42	14.20	7	15	78.1	75.0	-
	Mean	14.19					
	S.D.	0.05					

¹ Data after Shirozu & Higashi (1976).² Data after Sudo & Sato (1966).

TABLE 3. CHEMICAL COMPOSITIONS OF SUDOITE

	1	2	3	4	5	6	7
SiO ₂	37.93%	35.13%	33.00%	32.46%	35.63%	39.01%	33.58%
TiO ₂	0.02	0.02	-	-	none	0.47	-
Al ₂ O ₃	33.85	31.48	35.69	35.83	34.87	32.15	37.98
Cr ₂ O ₃	0.01	0.01	-	-	-	-	-
Fe ₂ O ₃	-	-	2.74	-	5.01	0.90	-
FeO	1.67	1.67	0.24	2.58	0.43	0.10	-
MnO	0.03	0.03	0.28	0.31	0.05	-	-
ZnO	-	-	0.05	-	-	-	-
NiO	0.04	0.04	0.09	-	-	-	-
MgO	13.70	13.70	14.07	13.64	8.63	10.14	15.02
CaO	0.08	0.08	-	-	1.13	0.54	-
BaO	0.03	0.03	-	-	-	-	-
Na ₂ O	0.02	-	<0.01	-	0.24	0.10	-
K ₂ O	0.71	-	0.00	-	0.46	1.52	-
F	0.14	0.14	-	-	-	-	-
Total	88.17	82.27	86.17	84.82	86.45	84.93	86.58

1. This study; total iron expressed as FeO. "Total" corrected for oxygen equivalent of F content.
2. Composition corrected for illite contamination. "Total" corrected for oxygen equivalent of F content.
3. Sudoite from Otré, Belgium (Fransolet & Bourguignon 1978).
4. Sudoite from Ardennes, Belgium; total iron expressed as FeO (Kramm 1980).
5. Sudoite from Kamikita mine, Japan (Hayashi & Oinuma 1964).
6. Sudoite from Furutobe mine, Japan (Sudo & Sato 1966).
7. Theoretical composition of sudoite: (Al₂Mg₂)(Si₂Al)O₁₀(OH)₂ (Fransolet & Bourguignon 1978).

Note: Li₂O content determined after digestion of whole sample was 0.21%; distribution unknown. Therefore sudoite probably contains <0.5% Li₂O.

heating to 700°C for 0.5 hours, the 002 reflection was reduced in intensity by a factor of 2.5 and decreased in *d*-value to 13.0 Å. There was no observable change in the illite following glycerol and heat treatments.

In order to examine the sudoite structure more closely, the illite peaks were graphically "removed" from the sample diffractogram. The illite in sample 53-14 was compared to illite in the <2 μm separates from the altered sandstone samples above the unconformity and to an illite sample from Eldorado, Saskatchewan (Kodama & Dean 1980). Illite intensities and *d*-values in 53-14 are more similar to the Eldorado reference sample than to the illite from

above the unconformity. Subtraction of the reflections of the Eldorado illite shown in Figure 4B from the reflections shown in Figure 4A resulted in a differential X-ray-diffraction pattern for sudoite (Fig. 4C).

Measurements of the *d*-values of ten basal reflections gave an average *d*(002) value of 14.19 ± 0.05 Å (Table 2). The average *d*(001) value is similar to data reported for sudoite from Kamikita (14.21 Å; Sudo & Sato 1966), Matsumine and Uchinotai (14.18 ± 1 Å and 14.16 ± 1 Å, respectively; Shirozu & Higashi 1976). In order to compare observed intensities with the published data, it was necessary to convert observed intensities to structure factors (F). The

observed intensities were corrected for transparency and Lorentz-polarization effects following the method of Alexander (1969). Observed F values for the ten basal reflections show good agreement with values published by Sudo & Sato (1966) and Shirozu & Higashi (1976), which confirms the dioctahedral nature of this mineral.

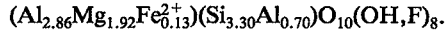
Electron-microprobe analyses

The chemical composition (average of 8 analyses) of sudoite in sample 53-15 is presented in Table 3. Chemical analyses of other samples of sudoite from low-grade metamorphic and hydrothermal environments are shown for comparison. The SiO_2 content tends to be higher than the theoretical composition and that of the other sudoite samples. Al and Mg contents are comparable, but total Fe is generally lower and K higher than in the other samples.

The small amount of K and greater amount of SiO_2 possibly indicate an illite impurity in the sample analyzed. A back-scattered electron image from sample 53-15 (Fig. 5) illustrates the textural relationship between sudoite and illite: sudoite occurs as a fine-grained matrix, and illite occurs both as matrix and coarse clastic grains. Sudoite appears to penetrate cleavage planes at the ragged edges of fine-grained illite. Coarse illite grains contain rutile lenses

along cleavage planes, possibly indicating a biotite precursor (*cf.* Mellinger 1985). Accessory goyazite is present in association with sudoite.

Corrections were made for illite contamination assuming a composition of $(\text{K,Na})\text{Al}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$; the minor elements Ba, Ca, Cr, Mn, Ni and Ti were neglected in the final calculation. If we assume that iron is present as Fe^{2+} , the calculated structural formula is:



The general formula for Al-rich chlorite is $(\text{Al,Fe}^{3+})_{4-y}(\text{Mg,Fe}^{2+})_{x/2+3y/2}(\text{Si}_{4-x}\text{Al}_x)\text{O}_{10}(\text{OH})_8$ (Sudo & Sato 1966). If the chlorite is dioctahedral, then the total number of octahedrally coordinated cations is less than 5, and $0 < (x + y) < 2$. The sum of octahedral cations for sample 53-15 is 4.91, and $(x + y)$ is 1.84. The proportion of $^{\text{IV}}\text{Al}$, x , is 0.70, and the total trivalent cations, y , is 1.14. If iron is present as Fe^{3+} , and Cr, Mn, Ni and Ti are included in the calculations, as these elements can be accommodated in the structure, then the total number of octahedrally coordinated cations would be 4.87, and $(x + y)$ would be 1.75. The difference that results from the assumption of the valency of iron is small; in either case, these data confirm that the mineral is of the di-trioctahedral type.

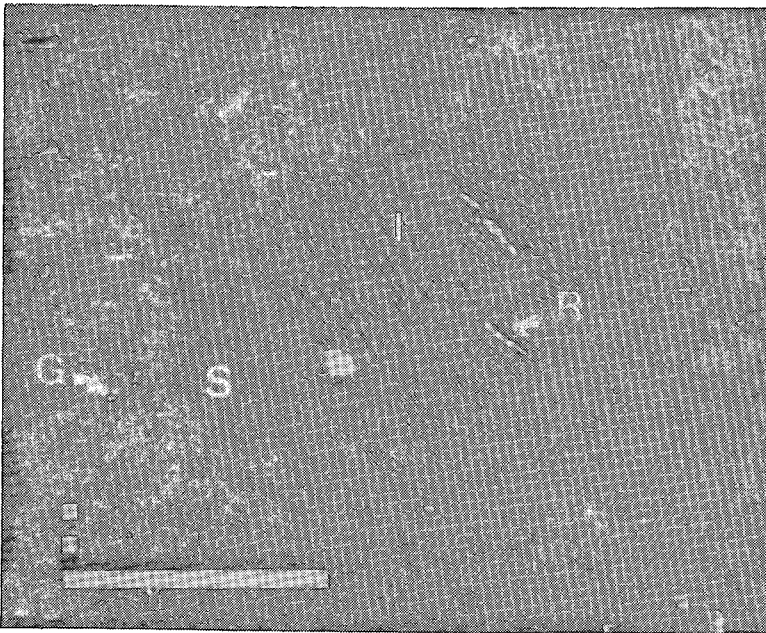


Fig. 5. SEM photomicrograph of illite (I) with sudoite (S). Sudoite appears to penetrate along frayed edges of the illite grain. Rutile (R) occurs along cleavage planes of illite; goyazite (G) is associated with sudoite. Black areas are holes (back-scatter image); long bar scale = 100 μm).

DISCUSSION

Chlorite in borehole WDG1-53 is the Al-rich, dioctahedral variety, sudoite. It is structurally similar to sudoite from hydrothermal, diagenetic and low-grade metamorphic environments.

In the Cigar Lake area, illite crystallinity indices (Kübler 1968), intensity ratios [$I(002)/I(001)$], composition (hydromuscovite) and polytype data (mixtures of $2M_1$ and $3T$) suggest prograde regional diagenesis to anchizonal conditions [*i.e.*, transition between high-grade diagenesis and low-grade metamorphism, as described by Dunoyer de Segonzac (1970)] (Percival *et al.* 1988). According to Hunziker (1986), Al-chlorite is produced by reactions between kaolinite and minor amounts of Fe and Mg near the diagenetic-anchizonal boundary. Illite-smectite mixed-layer minerals, which also can be precursors of chlorite, generally persist to higher temperatures than kaolinite (Hunziker 1986).

Although sudoite can form under high-grade diagenetic conditions, it is more commonly inferred to be of hydrothermal origin, based on its association with ore minerals (Bailey & Tyler 1960, Hayashi & Oinuma 1964) and its high-temperature structural type (IIb polytype; Brown & Bailey 1962, Fransolet & Bourguignon 1978). Abundant sudoite occurs in a well-defined envelope around the ore at Cigar Lake, whereas in the surrounding Manitou Falls Formation, only illite and kaolinite are present. This relationship suggests that sudoite at Cigar Lake formed at the expense of illite and kaolinite; the age relationship is well illustrated by textures showing replacement of illite by sudoite (Fig. 5).

Textures indicating late replacement of illite by chlorite also are observed in altered Athabasca Group rocks at the Maurice Bay uranium deposit (Mellinger 1985). In a similar paragenetic sequence from the Jabiluka unconformity-type uranium deposit, Australia, Nutt (1989) reported complex textures indicating repeated episodes of chloritization following sericitization.

Based on these examples, late chlorite, including sudoite, appears to be a common associate of hydrothermal uranium deposits. Its origin in relation to ore-forming fluids is poorly understood and warrants additional study. Further investigations of sudoite in the Athabasca basin associated with the Upper Wolverine Point Formation (illite-smectite-bearing unit) and other formations (*e.g.*, Lazenby Lake and Fairpoint) in barren areas would help to resolve questions of the regional extent and conditions of chloritization.

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