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STRUCTURES, COMPOSITIONS, AND X-RAY DIFFRACTION IDENTIFICATION OF DIOCTAHEDRAL CHLORITES

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Abstract—Al-rich di,trioctahedral chlorite exists as the species cookeite and sudoite. Di,dioctahedral chlorite exists as the species donbassite. Cookeite has essential Li in its structure, sudoite has essential Mg, and donbassite has only small amounts of either element. To date, sudoite has been reported to have only II*b* structural units and donbassite to have only I*a* structural units. Cookeite is based primarily on I*a* structural units, but II*b* units are present in specimens from two localities. Most Al-rich chlorite species have regular-stacking "*r*" or "*s*" 2-layer stacking sequences, but 1-layer I*a*-2 and I*a*-6 polytypes also are known. The structural units (I*a* or II*b*) and the specific stacking sequences can be explained by a combination of local charge balance and minimization of cation-cation repulsion involving the interlayer and tetrahedral cations. X-ray powder diffraction data are adequate to differentiate Al-rich chlorite from trioctahedral chlorite and to identify the type of structural unit present, but single crystal study is necessary to identify the 2-layer and 1-layer sequences with certainty.

Key Words—Chemical composition, Chlorite, Cookeite, Crystal structure, Donbassite, Sudoite, X-ray powder diffraction.

INTRODUCTION

Al-rich chlorite is being reported in the literature with increasing frequency. Three well-crystallized species have been defined to date—cookeite, sudoite, and donbassite. These species can be differentiated from trioctahedral chlorites by their compositions, smaller d(060) values (1.49 to 1.51 Å compared with 1.52 to 1.56 Å for trioctahedral species), slightly smaller d(001) values, and a distinctly more intense 003 reflection near 4.70 Å. The three species are more difficult to distinguish from one another by X-ray diffraction data alone. Cookeite and sudoite are di,trioctahedral chlorites, i.e., the 2:1 layer is dioctahedral, but the interlayer is trioctahedral. Donbassite is di,dioctahedral with two dioctahedral sheets. No examples are known as yet having a trioctahedral 2:1 layer and a dioctahedral interlayer. The present paper summarizes the compositions and several polytypic forms known to occur for these Al-rich species and presents indexed X-ray powder diffraction data to facilitate their identification.

COOKEITE

Cookeite is a di,trioctahedral chlorite commonly found as an alteration product of Li-bearing minerals in pegmatites and hydrothermal veins. The ideal composition is $Al_2(Si_3Al)O_{10}(OH)_2 \cdot (Al_2Li)(OH)_6$. Its essential Li content (~3–4 wt. % Li_2O) distinguishes it from sudoite and donbassite, which contain only minor amounts of Li. Černý (1970), in a study of the most reliable analyses of cookeite, found tetrahedral Si to be nearly constant at 3.0 Si per 4.0 positions, but with tetrahedral Al of some specimens substituted by small amounts of B or Be. Li ranges between 0.8 and 1.4 atoms per formula unit and is concentrated mainly in the interlayer sheet. Fluorine sometimes substitutes for OH in small amounts. Small amounts of Ca + Na + K are often reported in chemical analyses of dioctahedral chlorites as cations residing between the 2:1 layers and the interlayer sheet, but they are more likely to be impurities according to Peacor *et al.* (1988). Formula units of 13 cookeite specimens, modified from Černý (1970), are presented in Table 1. The total octahedral occupancy ranges between 4.8 and 5.3 atoms per 6.0 sites in these formulas, but if the Ca + Na + K totals are excluded as impurities, both the total occupancy and the individual atom values require small revisions.

By far the great majority of cookeite specimens are based on I*a* structural units in the terminology of Bailey and Brown (1962). The symbol I indicates that the interlayer octahedral sheet has the same orientation or slant as the octahedral sheet within the 2:1 layer, whereas the symbol II indicates opposite orientations. The symbol *a* indicates that the interlayer sheet is positioned by long hydrogen bonds so that the interlayer cations project normal to the layers onto tetrahedral cations and the centers of 6-fold rings of the 2:1 layers above and below. The symbol *b* indicates lack of such superposition. A following integer (1 to 6) then indicates the precise location of the 6-fold rings of the upper 2:1 layer relative to the symmetry plane of the layer plus interlayer below for regular-stacking polytypes. Several polytypic modifications are known. Vrublevskaja *et al.* (1975) described an unusual occurrence of cookeite in metamorphosed bauxite from Djalair in Middle Asia, where it formed as a result of interaction of Li-rich solutions with pyrophyllite-2*M*. The structure is unique also in that it is a regular-stacking 1-layer triclinic struc-

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Table 1. Structural formulae of cookeite specimens.

Sample	Locality	Composition
1	Hebron, Maine	$(Na_{0.03}K_{0.015})(Al_{3.795}Fe^{3+}_{0.03}Li_{1.44})_{5.265}(Si_{3.045}Al_{0.955})O_{10}(OH)_{7.94}F_{0.06}$
2	Ward's Natural Science Establishment, unknown locality	$(Al_{3.825}Li_{1.365})_{5.19}(Si_{3.165}Al_{0.835})O_{10}(OH)_8$
3	Buckfield, Maine	$(Na + K)_{0.075}(Al_{3.825}Fe^{3+}_{0.045}Li_{1.265})_{5.135}(Si_{3.06}Al_{0.94})O_{10}(OH)_{7.985}F_{0.015}$
4	Northwest U.S.S.R.	$(Na_{0.12}K_{0.075})(Al_{3.63}Fe^{3+}_{0.115}Fe^{2+}_{0.14}Mg_{0.05}Li_{1.22})_{5.155}(Si_{2.98}Al_{1.02})O_{10}(OH)_8$
5	Dobrá Voda, Czechoslovakia	$(Ca_{0.055}Na_{0.01})(Al_{3.80}Fe^{3+}_{0.015}Mg_{0.14}Li_{1.13})_{5.085}(Si_{3.04}Al_{0.96})O_{10}(OH)_8$
6	Muiane, Mozambique	$(Ca_{0.01}Na_{0.01})(Al_{4.025}Li_{1.12})_{5.145}(Si_{2.98}Al_{0.74}Be_{0.22}B_{0.06})O_{10}(OH)_{7.955}F_{0.045}$
7	Kalbinski Range, U.S.S.R.	$(Ca_{0.04}Na_{0.01}K_{0.01})(Al_{3.89}Fe^{2+}_{0.05}Mg_{0.025}Li_{1.11})_{5.075}(Si_{2.95}Al_{1.05})O_{10}(OH)_8$
8	Lipovka, Urals, U.S.S.R.	$(Ca_{0.025}Na_{0.19}K_{0.015})(Al_{3.75}Fe^{3+}_{0.005}Fe^{2+}_{0.015}Mg_{0.175}Li_{0.97})_{4.915}(Si_{3.135}Al_{0.865})O_{10}(OH)_8$
9	Manono, Katanga	$(Ca_{0.025}Na_{0.02})(Al_{3.94}Fe^{3+}_{0.005}Mg_{0.025}Li_{0.945})_{4.915}(Si_{3.09}Al_{0.905}B_{0.005})O_{10}(OH)_8$
10	North Little Rock, Arkansas	$(Ca_{0.01})(Al_{4.02}Fe^{3+}_{0.005}Mg_{0.005}Li_{0.86})_{4.89}(Si_{3.035}Al_{0.965})O_{10}(OH)_8$
11	Ogofau, Carmarthenshire, England	$(Na_{0.01})(Al_{4.00}Fe^{3+}_{0.02}Fe^{2+}_{0.065}Li_{0.855})_{4.94}(Si_{3.035}Al_{0.965})O_{10}(OH)_8$
12	Radkovice, Czechoslovakia	$(Ca_{0.055})(Al_{3.90}Fe^{3+}_{0.02}Fe^{2+}_{0.01}Mg_{0.11}Li_{0.815})_{4.855}(Si_{3.085}Al_{0.66}B_{0.255})O_{10}(OH)_8$
13	Wait-a-bit Creek, British Columbia	$(Ca_{0.16}Na_{0.12}K_{0.01})(Al_{3.88}Mg_{0.11}Li_{0.77})_{4.76}(Si_{2.93}Al_{1.07})O_{10}(OH)_8$

Modified from Černý (1970). See that author for references to samples.

ture of the I*a*-6 type. The space group is $C\bar{1}$, and the crystallographic angle α is distorted to 90°33′. Most cookeite specimens are less well crystallized and are based on monoclinic 2-layer structures. Lister (1966) pointed out that most of the cookeite specimens observed in her survey of the globular rosettes in Li-bearing pegmatites appeared to be imperfect 2-layer structures similar to the monoclinic "*s*" structure derived by Mathieson and Walker (1954) in a study of vermiculite. The symbol of Lister and Bailey (1967) for this 2-layer structure is $\bar{X}_1$-I*a*-4:$\bar{X}_1$-I*a*-6. This symbol indicates that the $a/3$ shift within each 2:1 layer is directed along $-X_1$ and successive 2:1 layers are positioned so that the centers of the 6-fold rings are located alternately at points 4 and 6 on opposite sides of the symmetry plane below (related to one another by a $b/3$ shift). The 2-layer nature is only apparent by observation of weak diffuse reflections of index $k \neq 3n$ on single crystal patterns. These reflections do not show up well on Debye-Scherrer patterns (Table 2a).

Lister (1966) also identified a better crystallized 2-layer cookeite from Wait-a-bit Creek, British Columbia, and at several localities in the Magnet Cove district and around North Little Rock, Arkansas. The latter includes cookeite from the Jeffrey quarry, which is part of the Source Clays Repository of The Clay Minerals Society. The structure can be correlated with the monoclinic "*r*" and "*q*" structures of Mathieson and Walker (1954), which have polytypic symbols "*r*" = $\bar{X}_1$-I*a*-4:$\bar{X}_3$-I*a*-4 and "*q*" = $\bar{X}_1$-I*a*-6:$\bar{X}_3$-I*a*-2. Here the $a/3$ shift is directed along $-X_1$ of the first 2:1 layer and along $-X_3$ of the second layer. Six-fold rings of successive 2:1 layers are positioned at points 4 and 4 for the "*r*" structure and at points 6 and 2 for the "*q*" structure. The two structures are enantiomorphic in space group *Cc*. Bailey (1975) summarized results of an incomplete structural refinement of one of the North Little Rock crystals. In this structure Li was found to be ordered in one interlayer site, where it apparently achieves the best local charge balance. Interlayer Li^+ lies on a line directly between a tetrahedral cation in the 2:1 layer below and a tetrahedral cation in the 2:1 layer above. The more highly charged interlayer Al^{3+} cations, however, only have a tetrahedral cation on one side and the center of a 6-fold ring on the other side. Although different T–O bond lengths were determined during structural refinement, the poor quality of the film data does not permit a firm conclusion as to ordering of tetrahedral Si and Al, nor has it been possible to obtain a better crystal to date. The powder pattern in Table 2b shows clearly $k \neq 3n$ reflections that identify the 2-layer nature of the specimen, but the more intense $k = 3n$ lines are very similar to those of the more common but less well crystallized cookeite specimens.

Černý *et al.* (1971) described a cookeite from a pegmatite at Dobrá Voda, Czechoslovakia, as having II*b* structural units. The present writers have confirmed a second occurrence of cookeite-II*b* as pink rosettes on elbaite from a pegmatite at Norway, Maine (University of Wisconsin Museum #6001/1). The powder pattern (Table 2c) is distinctly different from those of cookeite specimens based on I*a* units. Single crystals were not available, but a comparison of observed and calculated intensities of $k \neq 3n$ reflections suggests 2-layer "*r*" or "*q*" structures.

Frank-Kamenetskii (1960) and Černý (1970) mentioned manandonite from Madagascar as a possible di,trioctahedral Al-Li chlorite having substantial tetrahedral boron ($B^{IV} \geq Al^{IV}$). Guggenheim *et al.* (1983) confirmed the high B^{IV} content of this material by ion probe analysis and stated that its X-ray powder pattern was that of a I*a* type chlorite. The type manandonite defined by Lacroix (1912, 1922), however, has since been shown to be a trioctahedral 1:1 layer silicate of approximate composition $(Al_2Li)(Si_{1.0}Al_{0.5}B_{0.5})O_5(OH)_4$

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(A.-M. Fransolet, University Liége, Liége, Belgium, personal communication, 1988; Ranorosoa, 1986). This 1:1 type material is intermixed with a B-rich Al-Li "cookeite", which Strunz (1957) and later workers showed to be a chlorite and mistakenly called manandonite. This chloritic material will probably qualify as a new species when the details of its composition and structure are better known.

SUDOITE

Sudoite is also a di,trioctahedral chlorite, and has been identified in hydrothermal alteration zones, sediments, soils, fissure veins, and low-grade metamorphic rocks from many localities. The ideal composition is $Al_2(Si_3Al)O_{10}(OH)_2 \cdot (Mg_2Al)(OH)_6$. Tetrahedral Al is known to range from about 0.4 to 1.1 atoms per formula unit, octahedral Al from 2.5 to 3.4, and Mg from 1.2 to 2.5. Smaller amounts of Fe, Mn, and Li are also present. Total octahedral occupancy ranges from 4.5 to 5.1 atoms per 6.0 sites. Sudoite contains only a minor amount of Li and less Al and considerably more Mg than cookeite. Representative structural formulae for sudoite are listed in Table 3.

Stanton (1984) reported two groups of iron-rich "sudoites" at the Geco mine, Manitouwadge, Ontario, with $Fe^{2+} > Mg$ for one group. The materials are impure, however, and the average formulae of the two groups differ from those in Table 3 in several respects. Tetrahedral Al is low (0.1 to 0.3 atoms as recalculated by the present authors on the basis of 28 positive charges), the Ca + Na + K totals are moderately high (0.27 to 0.29 atoms), total octahedral occupancy is low (4.3 to 4.4 atoms per 6.0 sites, excluding Ca + Na + K), and the total octahedral charge is negative (rather than positive) relative to an ideal complement of anions. Purer material is needed to verify the nature of this mineral.

Eggleton and Bailey (1967) and Shirozu and Higashi (1976) attempted partial refinements of the structures of different fine-grained sudoite specimens on the basis of X-ray powder diffraction data. Their results confirmed that these specimens are based on II*b* structural units in which the 2:1 layer is dioctahedral and the interlayer is trioctahedral. Drits and Lazarenko (1967) and Lin and Bailey (1985) used single crystals to show that specimens from the Ural Mountains and Belgium, respectively, have 2-layer structures of the "*s*" type. The powder pattern in Table 4 is indexed best by incorporation of a slight distortion to triclinic geometry. Di,dioctahedral and di,trioctahedral chlorites have d(001) values that are systematically smaller than those for trioctahedral specimens of similar tetrahedral compositions, thus invalidating the usage of existing d(001) graphs and equations for estimating tetrahedral Si,Al contents of such specimens. Lin and Bailey (1985) concluded from 1-dimensional electron density projections of sudoite that the smaller d(001) value is due primarily to a thinner dioctahedral sheet within each 2:1 layer.

DONBASSITE

Donbassite is defined as having two dioctahedral sheets and an ideal composition near $Al_2(Si_3Al)O_{10}(OH)_2 \cdot Al_{2.33}(OH)_6$. For a variable amount (x) of tetrahedral substitution the formula can be written as $Al_2(Si_{4-x}Al_x)O_{10}(OH)_2 \cdot Al_{2+\frac{x}{3}}(OH)_6$. It has been reported from soils, sediments, hydrothermal alteration zones, and fissure veins. Substitution of Al for Si in the tetrahedral sheets of the best crystallized specimens ranges from 0.6 to 1.3 atoms per 4.0 positions. The octahedral cation total is always greater than 4.0 (excluding Ca + Na + K), ranging from about 4.2 to 4.5 atoms and indicating that the positive charge on the interlayer sheet arises primarily from the presence of Al + Mg + Fe cations in excess of 2.0 in that sheet, i.e., partly filling the "vacant" octahedron. Small amounts of octahedral Li are commonly present (Rozinova and Dubik, 1983). Representative structural formulae are listed in Table 5.

Whereas sudoite has only been identified to date as having II*b* structural units, donbassite has only been identified as having I*a* structural units. This statement takes into account the evolution of the definition of these names over time, i.e., some sudoite specimens as presently defined have been called donbassite in the past and vice-versa. Drits and Lazarenko (1967) identified a donbassite specimen from the Donbass region, U.S.S.R., as having a 2-layer structure like those of the equivalent "*r*" and "*q*" structures and consisting of I*a* structural units. Aleksandrova *et al.* (1972) refined the crystal structure of a low-Li donbassite from Novaya Zemlya, U.S.S.R. They reported a regular-stacking 1-layer structure of space group *C*2, which is a distorted version of the I*a*-2 polytype of ideal symmetry *C*2/*m*. The vacant octahedral site in the 2:1 layer lies on the symmetry plane of the layer, but the partly vacant octahedral site in the interlayer sheet lies off the projection of the mirror plane (thereby eliminating the mirror for the crystal as a whole). Adjacent layers adopt the mica configuration. Different mean T–O bond lengths observed in this refinement using film-collected data suggest ordering of tetrahedral Si and Al, but a more accurate refinement using counter-collected data is desired for proof of this point.

No accurate powder data for purified donbassite specimens are available for comparison with those of sudoite and cookeite. Observed d(060) values are between 1.49 and 1.50 Å for donbassite and between 1.49 and 1.51 Å for sudoite and cookeite. Table 6 lists a calculated XRD pattern for donbassite based on the atomic coordinates reported by Aleksandrova *et al.* (1972) for the I*a*-2 structure. The strong $k = 3n$ reflections are similar in positions and intensities to those

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Table 2. X-ray powder diffraction patterns of cookeite specimens.

hkl	I		d(obs.) (Å)	d(calc.) (Å)
(a) Cookeite-I*a*(*s*)				
002	75		14.10	14.056
004	60		7.05	7.028
006	100		4.68	4.685
020				4.468
110	30		4.44	4.444
021				4.412
11$\bar{3}$				4.197
112	5		4.12	4.110
008	60		3.513	3.514
00,10	20		2.812	2.811
200,13$\bar{2}$	30		2.560	2.561
20$\bar{4}$	50		2.505	2.510
132				2.505
13$\bar{4}$	3		2.47	2.469
202				2.465
20$\bar{6}$	1		2.37	2.377
134				2.370
13$\bar{6}$	90		2.317	2.319
204				2.314
20$\bar{8}$	5		2.202	2.206
136				2.198
13$\bar{8}$	2		2.140	2.143
206				2.137
20,1$\bar{0}$	5		2.020	2.024
138				2.017
00,14	10		2.010	2.008
13,1$\bar{0}$	25		1.962	1.962
208	12		1.953	1.956
00,16	5		1.760	1.759
20,1$\bar{4}$	3		1.689	1.687
13,12	7		1.680	1.681
13,1$\bar{4}$	20		1.636	1.634
20,12	10		1.627	1.629
20,1$\bar{6}$	5		1.545	1.542
13,14	10		1.538	1.537
33$\bar{2}$	50		1.490	1.490
060,33$\bar{1}$				1.489
33$\bar{6}$	8		1.458	1.459
064,332				1.457
33$\bar{8}$	8		1.421	1.422
066				1.419
334				1.418
20,1$\bar{8}$				1.415
13,16	8		1.409	1.410
13,1$\bar{8}$;33,1$\bar{0}$	6		1.375	1.373 Å
068				1.371
20,16;336			1.372	1.370
33,1$\bar{2}$				1.319
06,10	5		1.314	1.316
338				1.314
20,2$\bar{0}$	25		1.300	1.303
13,18				1.299

Specimen from Waisanen-Tamminen pit, Greenwood, Maine, has an imperfect "*s*" structure. Cell dimensions: $a = 5.163$, $b = 8.935$, $c = 28.34$ Å, $\beta = 97.2°$. Pattern of crystal mounted in a Gandolfi camera, 114.6-mm diameter, graphite-monochromatized FeKα radiation. Indexing by comparison with single crystal data, intensities estimated visually.

hkl	I		d(obs.) (Å)	d(calc.) (Å)
(b) Cookeite-I*a*(*r*)				
002	90		14.06	14.077
004	35		7.03	7.039
006	60		4.70	4.692
11$\bar{1}$	20		4.47	4.459
110				4.443
11$\bar{2}$	10		4.35	4.366
111				4.321
11$\bar{3}$			4.17	4.184
112	10	B		4.118
023			4.03	4.031
113	5	B	3.87	3.866
024			3.77	3.770
11$\bar{5}$	1		3.66	3.674
008	60		3.518	3.519
11$\bar{6}$	3		3.410	3.404
116	3		3.060	3.073
00,10	45		2.815	2.815
200	60		2.560	2.561
13$\bar{2}$				2.558
132	80		2.505	2.505
20$\bar{4}$				2.503
202	3		2.465	2.469
13$\bar{4}$				2.464
134	2		2.370	2.372
20$\bar{6}$				2.368
204	100		2.316	2.321
13$\bar{6}$				2.314
02,11	1		2.220	2.220
136		B		2.202
20$\bar{8}$	5		1.200	2.197
206	3		2.140	2.144
13$\bar{8}$				2.138
02,12	1		2.070	2.077
138	20		2.109	2.021
20,1$\bar{0}$				2.016
208	45	B	1.960	1.964
13,1$\bar{0}$				1.958
20,10	1		1.790	1.792
13,1$\bar{2}$				1.787
00,16	5		1.760	1.760
13,12	10	B	1.686	1.685
20,1$\bar{4}$			1.680	1.681
20,12	35	B	1.638	1.637
13,1$\bar{4}$			1.630	1.632
13,14	20		1.542	1.541
20,1$\bar{6}$	15		1.538	1.537
33$\bar{2}$	75		1.487	1.489
060				1.488
33$\bar{1}$				1.487
332				1.457
064	15		1.456	1.456
33$\bar{6}$				1.455
334				1.420
33$\bar{8}$	15		1.419	1.418
066				1.418
13,16	10		1.412	1.414
20,1$\bar{8}$				1.411
20,16	7		1.374	1.377
13,1$\bar{8}$				1.373
336		B		1.373
068	7		1.371	1.370
33,1$\bar{0}$				1.369

Specimen from North Little Rock, Arkansas, has a 2-layer "*r*" structure with I*a* layers. Cell dimensions: $a = 5.158$, $b = 8.927$, $c = 28.351$ Å, $\beta = 96.8°$. Conditions as for cookeite-I*a*(*s*).

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Table 2. Continued.

hkl	I		d(obs.) (Å)	d(calc.) (Å)	*hkl*	I		d(obs.) (Å)	d(calc.) (Å)
(c) Cookeite-II*b*									
002	80		14.10	14.100	204,13$\bar{6}$	20		2.319	2.319
004	60		7.05	7.050	20$\bar{8}$,136	20		2.203	2.203
006	80		4.70	4.700	206,13;$\bar{8}$	1		2.140	2.143
11$\bar{1}$	65		4.45	4.463	20,$\bar{1}$0;138	15		2.021	2.022
110				4.445	208;13,$\bar{1}$0	75		1.962	1.962
11$\bar{2}$	5		4.35	4.373	208;$\bar{1}$2;13,10	12		1.848	1.847
111				4.321	20,10;13,$\bar{1}$2	40		1.792	1.791
112	5		4.13	4.117	00,16	5		1.762	1.762
024	5	B	3.73	3.775	20,$\bar{1}$4;13,12	15		1.686	1.686
11$\bar{5}$				3.684	20,12;13,$\bar{1}$4	3		1.638	1.636
008	40		3.52	3.525	20,$\bar{1}$6;13,14	55		1.542	1.542
026	1		3.22	3.239	060,33$\bar{2}$	90		1.490	1.490
11$\bar{8}$	1		2.92	2.915	064,332,33$\bar{6}$	15		1.458	1.458
00,10	30		2.820	2.820	066,334,33$\bar{8}$	20		1.420	1.420
11$\bar{9}$	5		2.705	2.700	20,16;13,$\bar{1}$8	40	B	1.375	1.376
118	1		2.62	2.630	068;336;33,$\bar{1}$0				1.372
200,13$\bar{2}$	25	B	2.560	2.561	06,10;338;33,$\bar{1}$2	8		1.319	1.317
204,13$\bar{2}$	15		2.505	2.507	Specimen from Norway, Maine. Cell dimensions: a = 5.161, b = 8.938, c = 28.41 Å, β = 97°. Conditions as for cookeite-I*a*(*s*).				
202,13$\bar{4}$	100		2.468	2.468					
206,13$\bar{4}$	80		2.373	2.373					

Table 3. Structural formulae of sudoite specimens.

Specimen	Locality	Composition	Reference
1	Tracy mine, Michigan	$(Al_{2.7}Mg_{2.3})_{5.0}(Si_{3.3}Al_{0.7})O_{10}(OH)_8$	Bailey and Tyler (1960)
2	Colorado Plateau	$(R_{5.1})(Si_{3.4}Al_{0.6})O_{10}(OH)_8$	Schultz (1963)
3	Honko deposit, Kamikita mine, Japan	$(Ca_{0.11})(Al_{3.017}Fe^{3+}_{0.345}Fe^{2+}_{0.003}Mn_{0.004}Mg_{1.175})_{4.544}(Si_{3.261}Al_{0.739})$-$O_{10}(OH)_8$	Hayashi and Oinuma (1964); Sudo and Sato (1966)
4	Furutobe mine, Japan	$(Ca_{0.060}Na_{0.17}K_{0.159})(Al_{3.042}Fe^{3+}_{0.037}Mg_{1.594})_{4.673}(Si_{3.242}Al_{0.758})O_{10}(OH)_8$	Tsukahara (1964)
5	Kaiserbach Valley, Germany	$(Ca_{0.09})(Al_{2.70}Fe^{3+}_{0.09}Fe^{2+}_{0.04}Mg_{1.97})_{4.80}(Si_{3.43}Al_{0.57})O_{10}(OH)_8$	Frenzel and Schembra (1965)
6	Beriozovsk, Urals, U.S.S.R.	$(Ca_{0.03})(Al_{2.9}Fe_{0.22}Mg_{1.96})_{5.08}(Si_{2.86}Al_{1.14})O_{10}(OH)_8$	Drits and Lazarenko (1967)
7	Kazan Mineral Museum, unknown location	$(Ca_{0.14})(Al_{3.04}Fe^{2+}_{0.18}Mg_{1.61})_{4.83}(Si_{3.02}Al_{0.98})O_{10}(OH)_8$	Vlasov and Drits (1967)
8	Niida mine, Japan	$(Na_{0.03}K_{0.03})(Al_{3.03}Fe^{3+}_{0.06}Mg_{1.78})_{4.87}(Si_{3.01}Al_{0.99})O_{10}(OH)_8$	Kimbara and Nagata (1974)
9	Uchinotai deposit, Kosaka mine, Japan	$(Ca_{0.06}Na_{0.03}K_{0.05})$ $(Al_{3.13}Fe^{3+}_{0.01}Fe^{2+}_{0.01}Mg_{1.60})_{4.75}(Si_{3.19}Al_{0.81})O_{10}(OH)_8$	Tsuzuki and Honda (1977)
10	Okunosawa deposit, Kamikita mine, Japan	$(Ca_{0.01}Na_{0.11}K_{0.01})(Al_{3.19}Fe_{0.03}Mg_{1.37})_{4.59}(Si_{3.08}Al_{0.92})O_{10}(OH)_8$	Honda (1975)
11	Hanaoka mine, Japan	$(Al_{3.2}Mg_{1.6})_{4.8}(Si_{3.2}Al_{0.8})O_{10}(OH)_8$	Shirozu and Higashi (1976)
12	Rydnyy Altai, U.S.S.R.	$(Ca_{0.17}Na_{0.02}K_{0.06}(Al_{2.45}Fe^{3+}_{0.03}Fe^{2+}_{0.01}Mg_{2.54})_{5.03}(Si_{3.06}Al_{0.94})O_{10}(OH)_8$	Alysheva *et al.* (1977)
13	Venn-Stavelot Massif, Ardennes, Belgium	(1) $(Al_{2.84}Fe^{3+}_{0.19}Fe^{2+}_{0.02}Mn_{0.02}Mg_{1.91})_{4.98}(Si_{3.01}Al_{0.99})O_{10}(OH)_8$ (2) $(Al_{2.86}Fe^{3+}_{0.17}Mn_{0.02}Mg_{1.94})_{4.99}(Si_{2.99}Al_{1.01})O_{10}(OH)_8$	Fransolet and Bourguignon (1978)
14	Harz Mountains, German Democratic Republic	(1) $(Al_{2.864}Fe^{3+}_{0.136}Fe^{2+}_{0.136}Mn_{0.032}Mg_{1.886})_{5.054}(Si_{3.028}Al_{0.972})O_{10}(OH)_8$ (2) $(Al_{2.871}Fe^{3+}_{0.129}Fe^{2+}_{0.136}Mn_{0.035}Mg_{1.875})_{5.046}(Si_{3.037}Al_{0.963})O_{10}(OH)_8$	Kramm (1980)

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Table 4. Powder pattern of sudoite-II*b*, Ottre, Belgium.

hkl	I	d(obs.) (Å)		d(calc.) (Å)	*hkl*	I	d(obs.) (Å)		d(calc.) (Å)
002	40	14.2		14.16	$1\bar{3},10$				1.871
004	80	7.1		7.08	$2,0,\bar{12}$	5	1.870	B	1.871
006	100	4.72		4.721	1,3,10				1.858
020				4.547	$1,3\bar{12}$	12	1.820		1.820
$0\bar{2}1$	4	4.50		4.496	20,10	8	1.803		1.805
021				4.483	$1,\bar{3},\bar{12}$				1.804
$1\bar{1}1$				4.387	0,0,16	1	1.770		1.770
111				4.384	$3\bar{1}2$				1.717
$0\bar{2}2$	10	4.35		4.340	$3\bar{1}1$		1.711		1.713
022				4.318	0,4,11	10		B	1.712
$11\bar{4}$	10	4.01		4.024	$1,\bar{3},12$				1.706
$1\bar{1}4$				4.004	$15\bar{3}$		1.705		1.705
$1\bar{1}3$	5	3.87		3.915	$31\bar{4}$				1.705
113				3.905	153	5	1.673		1.675
$0\bar{2}5$				3.561	245				1.674
008	40	3.535		3.540	154			B	1.650
025				3.531	$24\bar{6}$	1	1.650		1.650
$1\bar{1}5$	1	3.36		3.361	2,0,12				1.647
115				3.350	$1\bar{3},\bar{14}$				1.646
$0\bar{2}6$	2	3.28		3.289	245	1	1.600		1.600
026				3.260	$15\bar{7}$				1.600
$1\bar{1}6$	2	3.10		3.102	$1,\bar{3},14$	15	1.560		1.559
116				3.092	$2,0,\bar{16}$				1.559
0,0,10	25	2.835		2.932	1,3,14	25	1.549		1.549
$20\bar{2}$				2.619	$33\bar{2}$				1.516
130	10	2.615		2.618	060	50	1.515		1.516
$13\bar{2}$				2.610	$33\bar{2}$				1.514
$20\bar{4}$				2.550	$33\bar{2}$				1.480
132	20	2.545		2.549	$33\bar{6}$	20	1.481		1.480
$13\bar{2}$				2.544	332				1.480
202	90	2.502		2.503	064				1.479
$13\bar{4}$				2.502	$33\bar{8}$				1.447
$20\bar{6}$				2.412	066				1.447
$13\bar{4}$	35	2.408		2.412	$33\bar{4}$	10	1.422	B	1.441
134				2.402	$33\bar{8}$				1.441
$13\bar{6}$				2.364	334				1.440
0,0,12	15	2.358		2.360	066				1.439
1,1,$\bar{11}$			B	2.357	0,0,20	7	1.419		1.416
204				2.348	$0\bar{6}8$	20	1.398		1.398
$13\bar{6}$	10	2.345		2.347	$3,3\bar{10}$				1.398
1,1,11				2.346	336				1.389
$1\bar{3}6$	20	2.237		2.270	068	15	1.384		1.389
$20\bar{8}$				2.269	$1,5,\bar{13}$				1.383
206	5	1.165		2.165					
$13\bar{8}$				2.164					
$1\bar{3}8$	1	2.050		2.051					
$2,0,\bar{10}$				2.050					
0,0,14	10	2.020		2.023					
$1\bar{3},\bar{10}$	20	1.995		1.996					
208	18	1.978		1.980					
$1,3,\bar{10}$				1.979					

Specimen from Ottre, Belgium. $a = 5.247$, $b = 9.094$, $c = 28.557$ Å, $\alpha = 90.5°$, $\beta = 97.3°$, and $\gamma = 89.9°$. Structure is of the *s*-type with II*b* layers. Conditions as listed in Table 2. B = broad.

of cookeite-I*a*. The strong 021 reflection at 4.269 Å is diagnostic of the I*a*-2 polytype.

EVALUATION OF STRUCTURES

The dominance of the I*a* structure in cookeite and donbassite and of the II*b* structure in sudoite and trioctahedral chlorites is linked directly to the magnitude and location of the positive electrostatic charge in the interlayer sheet. Bailey and Brown (1962) and Shirozu and Bailey (1965) showed that the II*b* structure is by far the most stable structural form of trioctahedral chlorite as a result of the complete absence of any cation-cation repulsion between sheets of the layered assemblage. The I*a* and II*a* structures, on the contrary, have exact vertical superposition of interlayer and adjacent tetrahedral cations with large resultant cation-cation repulsion values. Nevertheless, the I*a* structure would have a very high structural stability rating if the

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Table 5. Structural formulae for donbassite specimens.

Specimen	Locality	Composition	Reference
1	Donbass region, U.S.S.R.	(1) $(Ca_{0.17}Na_{0.19})(Al_{3.92}Fe^{3+}_{0.05}Mg_{0.21}Li_{0.05})_{4.23}(Si_{3.09}Al_{0.91})O_{10}(OH)_8$ (2) $(Ca_{0.19}Na_{0.22})(Al_{3.88}Fe^{3+}_{0.08}Mg_{0.23})_{4.18}(Si_{3.06}Al_{0.94})O_{10}(OH)_8$ (3) $(Al_{4.16}Fe^{3+}_{0.07}Mg_{0.11})_{4.34}(Si_{3.10}Al_{0.90})O_{10}(OH)_8$	Lazarenko (1940)
2	Alberni soil, British Columbia	$(Ca_{0.02}K_{0.23}X_{0.24})$ $(Al_{3.22}Fe^{3+}_{0.38}Ti_{0.09}Mg_{0.76})_{4.45}(Si_{2.80}Al_{1.20})O_{10}(OH)_8$	Brydon *et al.* (1961)
3	Saint-Paul-de-Fenouillet, East Pyrenees	$(Al_{4.3}Fe^{3+}_{0.03}Fe^{2+}_{0.02})_{4.35}(Si_{2.70}Al_{1.30})O_{10}(OH)_8$	Caillère *et al.* (1962)
4	Kesselberg, Federal Republic of Germany	(1) $(Al_{4.17}Mg_{0.10})_{4.27}(Si_{3.30}Al_{0.70})O_{10}(OH)_8$ (2) $(Al_{4.27})(Si_{3.2}Al_{0.8})O_{10}(OH)_8$	Müller (1961, 1963)
5	Nagol Tarasovki, Donbass, U.S.S.R.	$(Ca_{0.2})(Al_{4.0}Mg_{0.24})_{4.24}(Si_{3.12}Al_{0.88})O_{10}(OH)_8$	Drits and Lazarenko (1967)
6	Namivu, Alto Ligonho, Mozambique	$(Ca_{0.04})(Al_{3.96}Fe^{3+}_{0.02}Fe^{2+}_{0.016}Mn_{0.008}Mg_{0.04}Li_{0.27})_{4.314}$ $(Si_{3.45}Al_{0.55})O_{10}(OH)_8$	Gomes (1967)
7	Novaya Zemlya, U.S.S.R.	$(Al_{4.10}Fe^{3+}_{0.04}Fe^{2+}_{0.01}Mg_{0.08}Li_{0.26})_{4.49}(Si_{3.14}Al_{0.86})O_{10}(OH)_8$	Aleksandrova *et al.* (1972)
8	Donbass area, U.S.S.R.	$(Al_{3.96}Fe^{2+}_{0.08}Mg_{0.14}Li_{0.20})_{4.38}(Si_{3.36}Al_{0.64})O_{10}(OH)_8$	Rozinova and Dubik (1983)
9	Szabo Bluff, Scott Glacier, Antarctica	$(Al_{4.32}Fe^{3+}_{0.07})_{4.39}(Si_{2.83}Al_{1.17})O_{10}(OH)_8$	Ahn and Buseck (1988)

cation-cation repulsion factor could be minimized. This is exactly what happens in vermiculite, which contains only a few exchangeable interlayer cations and for which the structural type has been shown to be I*a* without exception. This favorable rating of the I*a* structure carries over to dioctahedral chlorite species in which one of the three interlayer cation sites is partly vacant (donbassite) or has a low-charge Li^+ cation present (cookeite).

In the regular-stacking 1-layer chlorite polytypes involving I*a* and II*a* layers, two different dispositions of the 2:1 layers exist on opposing sides of the interlayer sheet. The first, termed type A here, is found only in the monoclinic structures I*a*-2 and II*a*-1, in which successive 2:1 layers adopt the mica configuration with their 6-fold rings opposed. One interlayer cation site thus lies between the centers of these opposed fold rings, and the other two interlayer sites lie on vertical straight lines between tetrahedral cations of the two sets of rings. One form of donbassite adopts the I*a*-2 structure. The structural refinement by Aleksandrova *et al.* (1972) places the partly vacant interlayer site M between one set of superimposed tetrahedral cations and an Al^{3+} cation between the other set of two tetrahedra. The second Al^{3+} interlayer cation lies between the centers of the 6-fold rings in vertical projection. This distribution of interlayer cations gives the minimum amount of cation-cation repulsion possible for the I*a*-2 structure. The authors stated that repulsion is minimized further by tetrahedral cation ordering and local charge balance such that the source of positive interlayer charge is located between the sources of the tetrahedral negative charges, i.e., between Al-rich tetrahedra above and below. In this arrangement, the source of positive interlayer charge is the partly vacant M octahedron. Thus, that cation-cation repulsion would be minimized even further by the opposite pattern of tetrahedral cation ordering in which the partly filled and low-charge M site would be between Si^{4+}-rich tetrahedra and the high-charge interlayer Al^{3+} would lie between Al-rich tetrahedra. Verification of the ordering pattern is desirable.

The other type A structure having a mica configuration of layers is II*a*-1, which is not known in true chlorites, but is present in franklinfurnaceite, a hybrid chlorite-mica in which the chlorite is of the tri,dioctahedral type. In this mineral there is excellent local charge balance, with interlayer R^{3+} cations positioned midway between tetrahedral R^{2+}, interlayer R^{2+} between tetrahedral R^{4+}, and interlayer vacancies between the extra Ca that lie in the 6-fold ring openings (Peacor *et al.*, 1988).

A second disposition of 2:1 layers, termed here type B, exists in the 1-layer triclinic polytypes I*a*-4 and II*a*-3. Here, the unique interlayer site lies on a center of symmetry between tetrahedral sites above and below, and the other two interlayer sites are asymmetric, with a tetrahedral cation on one side and the center of a 6-fold ring on the other side. Cookeite in metamorphosed bauxite from Djalair, Middle Asia, adopts the I*a*-6 = I*a*-4 1-layer structure (Vrublevskaja *et al.*, 1975). Although the cation ordering scheme is not known for that specimen, logically the low-charge Li^+ should exist on the center of symmetry between tetrahedral sites above and below and the two high-charge Al^{3+} cations should be in the asymmetric locations with a tetrahedral cation on only one side. Cookeite and donbassite also adopt either or both of the 2-layer I*a* "*s*" and

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Table 6. Calculated powder pattern of I*a*-2 donbassite, Novaya Zemlya, U.S.S.R.

hkl	I	d(calc.) (Å)	*hkl*	I	d(calc.) (Å)
001	60	14.127	$20\bar{7}$	21	1.702
002	65	7.063	$31\bar{1}$		1.693
003	100	4.709	$15\bar{1}$		1.688
020	54	4.478	136		1.683
$11\bar{1}$	35	4.395	151		1.670
021	49	4.269	$15\bar{2}$	11	1.661
111	14	4.106	241		1.657
$11\bar{2}$	93	3.983	$13\bar{7}$		1.645
022	16	3.782	$24\bar{3}$	42	1.635
112	25	3.578	206		1.627
004	60	3.532			
$11\bar{3}$	11	3.444	$20\bar{8}$	16	1.557
113	28	3.058	137		1.539
$11\bar{4}$	4	2.941	$33\bar{1}$	48	1.493
005	28	2.825	060		1.493
024	5	2.773	062	6	1.460
114	4	2.619	331		1.457
$13\bar{1}$	47	2.569	063	21	1.423
200		2.563	332		1.418
$20\bar{2}$	52	2.522	227		1.414
131		2.508	138		1.413
$13\bar{2}$	4	2.479	00,10		1.413
201		2.463	$33\bar{5}$	7	1.382
$20\bar{3}$		2.392	064		1.375
025	7	2.390	$20,\bar{1}0$	31	1.315
132		2.371	139		1.302
$13\bar{3}$	97	2.331			
202		2.310			
$20\bar{4}$	10	2.223			
133		2.200			
$13\bar{4}$		2.155			
042	4	2.134			
203		2.132			
$20\bar{5}$		2.042			
134	20	2.018			
007		2.018			
$13\bar{5}$	49	1.974			
204		1.952			

Specimen from Novaya Zemlya, U.S.S.R., is a I*a*-2 polytype. Pattern calculated from atomic coordinates of Aleksandrova *et al.* (1972). Cell dimensions: $a = 5.174$, $b = 8.956$, $c = 14.26$ Å, $\beta = 97.83°$. Space group = $C2$.

"*r*" structures (terminology of Mathieson and Walker, 1954) that have the same interlayer-layer type B configurations. The "*r*" structure has been reported for donbassite by Drits and Lazarenko (1967), although the cation-ordering pattern was not investigated. Both structures are known for cookeite. Our refinement of the cookeite "*r*" structure places the low-charge interlayer Li^+ between tetrahedral cations on both sides and the two high-charge interlayer Al^{3+} cations in the asymmetric locations with a tetrahedral cation on only one side. The "*s*" structure has not been refined, but a similar local charge balance arrangement can be anticipated. The I*a* "*s*" structure is also the most stable form of vermiculite (Shirozu and Bailey, 1966), in which the exchangeable cation lies vertically between tetrahedral cations above and below, with vacancies in the two asymmetric locations. At one time, the tetrahedral cation ordering was thought to place Al-rich tetrahedra above and below the exchangeable cation to give optimum local charge balance (Shirozu and Bailey, 1966), but later work (de la Calle *et al.*, 1976) ruled out the existence of long-range tetrahedral cation ordering in vermiculite. Some evidence exists for short-range ordering in local domains (Thompson, 1984).

In contrast to the preceding species, sudoite has a trioctahedral interlayer sheet of composition (Mg_2Al), in which all of the cations have medium to high charges. And, just as in trioctahedral chlorites, the II*b* structure is favored to avoid cation-cation repulsion present in an *a* position of the interlayer sheet. The presence of a vacancy in the dioctahedral 2:1 layer has little or no effect on the position of the interlayer.

It is not prudent to assume that all sudoite is of the II*b* structural type and that all donbassite is of the I*a* structural type, because study of more samples may show structural variations in each, just as it has in cookeite. Most cookeite and most donbassite specimens, however, are based on I*a* structural units and most sudoite specimens are based on II*b* units. These structural units can be identified by X-ray powder diffraction patterns. The I*a* structural unit in a 2-layer dioctahedral chlorite is characterized by a strong d(204) XRD line near 2.32 Å. The II*b* structural unit for a 2-layer dioctahedral chlorite is characterized by a strong d(202) line near 2.47–2.50 Å and a strong d($20\bar{6}$) line near 2.37–2.41 Å. Most dioctahedral chlorite specimens appear to be based on regular-stacking 2-layer structures of the "*s*" or "*r*" and "*q*" types, but regular 1-layer forms are known as well. Single crystal study is usually necessary to identify the specific stacking arrangement present.

Environment of occurrence also can be helpful in identification. The presence of essential Li in cookeite usually restricts its occurrence to Li-rich environments, rather than in sediments and soils. Essential Mg in sudoite is not very restrictive and leads to its ocurrence in many different environments. An analysis for MgO should be definitive. Donbassite lacks both substantial Mg and Li, and although this restricts its occurrence to very Al-rich environments, it does not necessarily rule out occurrences in soils, sediments, hydrothermal veins, metamorphosed aluminous sediments, etc. Donbassite is less abundant in nature than sudoite and cookeite, presumably because of its unique composition. X-ray diffraction study should be supplemented by chemical analysis for suspected occurrences of all Al-rich chlorites.

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