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Determination and refinement of the crystal structure of margarosanite, $PbCa_2Si_3O_9$ *

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Auszug

Margarosanit, $PbCa_2Si_3O_9$, hat die Gitterkonstanten $a = 6{,}768 \pm 0{,}004$ Å, $b = 9{,}575 \pm 0{,}004$ Å, $c = 6{,}718 \pm 0{,}005$ Å, $\alpha = 110{,}36 \pm 0{,}03°$, $\beta = 102{,}98 \pm 0{,}04°$, $\gamma = 83{,}02 \pm 0{,}05°$. Raumgruppe ist $P\bar{1}$. Die Struktur wurde durch Anwendung der Minimumfunktion für $P(xyz)$ und der Schweratom-Methode bestimmt und mittels Ausgleichsrechnung verfeinert. Die Elementarzelle enthält im Idealfall 2 $PbCa_2Si_3O_9$. Die Grundzüge der Struktur sind: 1. ebene tetraedrische Lagen wechseln mit ebenen Calciumlagen zwischen dichtesten Kugelpackungen von Sauerstoffatomen parallel $(\bar{1}01)$ ab; 2. Pb und Ca sind geordnet; 3. die Ca(1)-Polyeder sind durch gemeinsame Kanten zu endlosen Ketten parallel [101] verbunden; 4. die Tetraeder bilden dreigliedrige Ringe, die mit den Ketten der Ca(1)-Polyeder verbunden sind; 5. die beiden Ca-Atome haben Sechser-Koordination, das Pb-Atom ist von sieben O-Atomen umgeben.

Abstract

The crystal structure of margarosanite with $a = 6.768$ Å $\pm .004$, $b = 9.575$ Å $\pm .004$, $c = 6.718$ Å $\pm .005$, $\alpha = 110.36° \pm .03$, $\beta = 102.98° \pm .04$, $\gamma = 83.02° \pm .05$, and with space group $P\bar{1}$ was solved by application of the minimum function to $P(xyz)$, and the use of heavy-atom techniques. This procedure yielded a trial structure which was refined by a least-squares method using the full matrix. The unit cell ideally contains $2(PbCa_2Si_3O_9)$. The margarosanite structure has the following principle features: 1. planes of tetrahedral sites alternate with planes of calcium sites between sheets of close-packed oxygen atoms parallel to $(\bar{1}01)$; 2. Pb and Ca are ordered; 3. Ca(1) polyhedra form an infinite edge-sharing chain parallel to [101] and Pb and Ca(2) sites alternate along the edge of this chain; 4. tetrahedral sites form three-membered rings which are linked to the Ca(1) polyhedral chain; and 5. the two Ca atoms have 6-fold coordination and the Pb atom has 7-fold coordination.

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Introduction

Margarosanite was initially described by FORD and BRADLEY (1916). A chemical analysis and some physical and chemical data were presented for specimens from the Parker Shaft, North Mine Hill, New Jersey.

FLINK (1917) described the occurrence of margarosanite at Långban, Sweden, and provided a chemical analysis. FLINK gave data on a supposed crystal of margarosanite, but ARMSTRONG (1963) was unable to reconcile these data with data from the Franklin material. Margarosanite was included in two studies (PALACHE 1928, 1929), but no other work was attempted prior to 1963. ARMSTRONG (1963) verified that margarosanite is triclinic and gave an unindexed powder pattern listing ninety-nine peaks between 10° and 135° 2θ for copper radiation. ARMSTRONG gives the value of the specific gravity as 4.33 (measured on a Berman Torsion Balance) and 4.30 (calculated). Other physical properties and the optical properties are listed by ARMSTRONG (1963) and WILKERSON (1962).

GLASSER and GLASSER (1964) compared margarosanite to synthetic $Ca_2BaSi_3O_9$ and pseudowollastonite. It was stated that the structures of these compounds are probably very similar. GLASSER and GLASSER (1961) suggested that $Ca_2BaSi_3O_9$ belongs to the group of compounds based on X_3O_9 rings, and this has been recently verified (DENT GLASSER and GLASSER, 1966). GLASSER and GLASSER (1964) also stated that the series of metasilicates containing large cations and having structures based on X_3O_9 rings also exhibit cation ordering. Thus, it is suggested that margarosanite has a structure based on X_3O_9 rings and has a characteristic Pb/Ca ratio.

Experimental

The crystal used in the structure determination was kindly supplied by RICHARD LEE ARMSTRONG. It is from the Yale Brush Collection, number 5938, and was collected in 1898 from the Parker Shaft, North Mine Hill, Franklin, New Jersey. An analysis of this material is given by FORD and BRADLEY (1916, page 161). The x-ray work was performed using this analyzed type material. The unit-cell contents were calculated using a specific gravity of 4.33 g/cm^3 (ARMSTRONG, 1963) and a cell volume of 401.5 Å^3 (this study), and corresponds to the following formula:

$$\begin{array}{l} Ca_{4.06} \\ Pb_{2.04}\ Mn_{0.17}\ Si_{5.87}\ O_{18.01}\,. \\ \overline{} \\ 4.23 \end{array}$$

The final cell parameters for margarosanite are given in Table 1. They were obtained by least-squares refinement (BURNHAM, 1962*a*) of data from *a*-, *b*- and *c*-axis zero-level Weissenberg photographs. The normal representation (all acute angles) is not given in Table 1, but the interaxial angles used in this study (representation $2R$) can be changed to the normal representation simply by changing the sense of the *a* and *b* axes. The cell parameters are compared to those for the isotypic mineral walstromite, $BaCa_2Si_3O_9$ (ALFORS *et al.*, 1965), in Table 1. The space group is $P1$ or $P\bar{1}$. Space group $P\bar{1}$ was chosen because the two formula units present in the unit cell are consistent with equipoint requirements, and because GLASSER and GLASSER (1961) determined, by means of statistical methods applied to 225 $0kl$ and 165 $hk0$ reflections, that the isotypic compound $Ca_2BaSi_3O_9$ has space group $P\bar{1}$.

Table 1. *Crystallographic data for margarosanite and walstromite*

	Walstromite (ALFORS *et al.*, 1965)	Margarosanite (This study)
a	6.734 Å ± .005 Å	6.768 Å ± .004 Å
b	9.607 ± .005	9.575 ± .004
c	6.687 ± .005	6.718 ± .005
α	69°51′	110.36° ± .03°
β	102 14′	102.98 ± .04
γ	97 6½′	83.02 ± .05

The specimen of margarosanite used for intensity measurements was a prismatic cleavage fragment 0.15_7 mm in length parallel to *a*, and 0.09_9 by 0.14_6 mm in cross section. The crystal was mounted with the *a* axis as the rotation axis. Intensities were gathered using a Philips scintillation detector and an equi-inclination counter diffractometer. Copper $K\alpha$ radiation ($\lambda = 1.5418$) was used. All observations were graphically recorded and the area under each peak was measured with a planimeter. Although approximately 1770 nonequivalent reflections were theoretically observable, only 1395 reflections were within the range of the instrument. Reflections in seven layer lines ($h = 0$ to 6) were recorded.

Absorption and Lorentz-polarization corrections were computed in modified versions of IBM 7090 Fortran II programs written by PREWITT (1964) and BURNHAM (1962*b*).

Cell geometry

FLINK (1917) described margarosanite as occurring in masses and in crystal "stalks". However, in a sample from Långban, kindly loaned by the University of Toronto Mineralogical Museum, specimen number M13635, it occurs as intersecting radial aggregates of crystalline needles. A zero-level Weissenberg photograph with the crystal rotating around the fiber axis has axial lattice rows corresponding to [011]* and [101]*. This indicates that the fiber direction in direct space is tentatively [101] for this Långban material.

A pseudo-orthorhombic cell can be defined as follows: $a' = [2\bar{1}\bar{2}]$; $b' = [0\bar{1}\bar{2}]$; and $c' = [101]$. A pseudo-monoclinic cell can be defined as follows: $a' = [2\bar{1}\bar{2}]$; $b' = [111]$; and $c' = [101]$.

Structure determination

Two-dimensional analysis

The two-dimensional Patterson synthesis, $P(vw)$, was computed using a computer program, 3DFPAT5, written by Dr. C. E. NORDMAN of the University of Michigan. Margarosanite contains only one heavy atom, Pb, in the asymmetric unit. A computation of relative peak values within Patterson space (BUERGER, 1959) shows that the single-weight Pb—Pb peak should have the highest value of the peaks represented on a Patterson map. $P(vw)$ showed one large peak per asymmetric unit, and three smaller peaks which had approximately equal magnitudes. A minimum function $M_2(yz)$ was formed assuming the large peak to be at the end of a projected Pb—Pb line image. This $M_2(yz)$ map showed only the large peak corresponding to Pb plus the equivalents of the three smaller peaks of $P(vw)$.

It seemed reasonable to assume that at least one of the three smaller peaks in $M_2(yz)$ could be a Ca atom. Therefore, each of the three smaller peaks in $P(vw)$ were used for forming new $M_2(yz)$ functions as follows: (1) single-weight Ca inversion peaks, and (2) double-weight Ca peaks. However, none of these approaches to finding the Ca positions gave unambiguous solutions. It was concluded that the three smaller peaks in $P(vw)$ were either doubled Pb—Ca, Si, O peaks, or that overlap of atoms in projection caused individual single inversion peaks to be unresolved.

The $P(vw)$ and $M_2(yz)$ functions all showed a striking linear feature parallel with the c axis, with three such linear elements per cell. This well-defined lineation is parallel with {010} and the [101] fiber direction.

Three-dimensional analysis

The three-dimensional Patterson function, $P(uvw)$, was computed. The large peak corresponding to the Pb position in $P(vw)$ was located at (12/60, 19/60, 33/60). A minimum function $M_2(xyz)$ was prepared assuming this peak to be the appropriate image point representing the end of a Pb—Pb line image. The peaks of $M_2(xyz)$ for the asymmetric unit of margarosanite are shown projected along a in Fig. 1. Only those peaks from $x = 0$ to $x = 1/2$ are shown. The linear trends parallel to the c axis are readily seen in this drawing. The largest

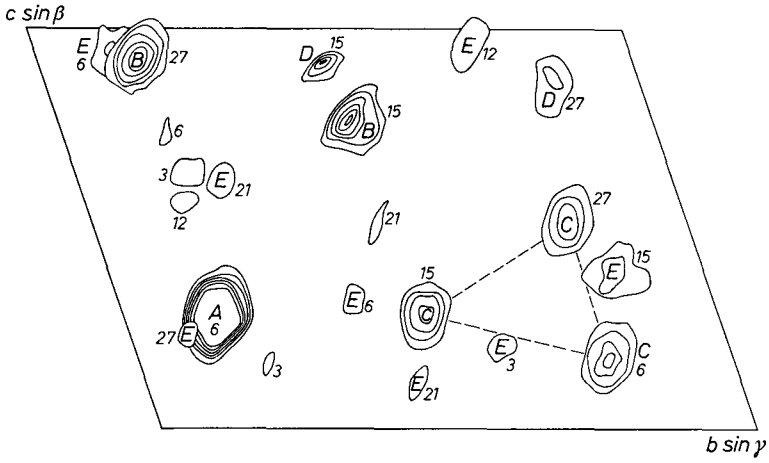

Fig. 1. Peaks of $M_2(xyz)$ projected along a, labelled with the level, in 60ths, on which they occur

peaks occur on $(0k0)$ planes separated from each other by approximately $\frac{1}{3}b$. The Pb-atom position described above is designated by the letter A. Two of the remaining six peaks have magnitudes distinctly larger than the others. These were provisionally assigned to Ca, and are designated in Fig. 1 by the letter B. The placing of the Ca atoms in these sites rules out the possibility of any cation occupying the site marked by letter D. This leaves three relatively large peaks, all of which are designated by the letter C. It seemed reasonable to assume that these are Si atom sites, on the basis of their heights and interpeak distances, and also because these form a three-membered ring. The possibility that the assigned Si positions form a chain was not completely discounted, but it is difficult to arrange geometrically the

Si sites to form a continuous chain. The ring is denoted in Fig. 1 by dashed lines. Eight sites were chosen from $M_2(xyz)$ as oxygen positions E in Fig. 1 plus a ninth site ($x = .45$, $y = .63$, $z = .50$) because they reproduce tetrahedra around Si positions, and acceptable polyhedra around the cations.

Refinement

A modified version of an IBM 7090 Fortran II program written by PREWITT (1962) was used to refine the atom parameters by a least-squares method using the full matrix. The various functions based on Fourier analysis were calculated either by Dr. NORDMAN's computer program or by a computer program written by one of the authors (R.L.F.). Refinement proceeded with all atoms assumed to be half-ionized. The weighting scheme recommended by CRUICKSHANK (1960) was used. Individual isotropic temperature factors were assigned initially as follows: $B_{Pb} = 0.75$; $B_{Ca} = 0.75$; $B_{Si} = 0.25$; $B_O = 0.50$. All atoms were assigned to general positions and the scattering curves were scaled by a factor of 1.0. The F_o's which were to small to measure were not used in the refinement, but they were included in the calculation of the discrepancy factor, R.

Structure factors calculated from the cations alone gave a discrepancy factor $R = 21.0\%$. The positional parameters and isotropic

Table 2. *Atom coordinates and isotropic temperature factors for margarosanite*
Standard deviations are given in parentheses

Atom	x	y	z	B
Pb	.0886 (.0001)	.1582 (.0001)	.2865 (.0001)	.57 (.01)
Ca(1)	.2700 (.0005)	.4951 (.0003)	.7629 (.0005)	.19 (.05)
Ca(2)	.4430 (.0006)	.1677 (.0004)	.9297 (.0005)	.43 (.05)
Si(1)	.0957 (.0007)	.7817 (.0005)	.1590 (.0007)	.28 (.07)
Si(2)	.2328 (.0007)	.5149 (.0005)	.2803 (.0007)	.38 (.07)
Si(3)	.4382 (.0007)	.7982 (.0005)	.5148 (.0007)	.30 (.07)
O(1)	.2274 (.0020)	.7469 (.0013)	.9779 (.0020)	.35 (.19)
O(2)	.1092 (.0019)	.1245 (.0013)	.9071 (.0019)	.24 (.18)
O(3)	.2299 (.0023)	.8790 (.0016)	.3955 (.0024)	1.15 (.23)
O(4)	.0469 (.0019)	.6266 (.0013)	.1967 (.0019)	.34 (.19)
O(5)	.3732 (.0020)	.4380 (.0014)	.1057 (.0020)	.49 (.19)
O(6)	.1238 (.0021)	.4072 (.0014)	.3561 (.0021)	.72 (.21)
O(7)	.3569 (.0020)	.6373 (.0013)	.5073 (.0020)	.37 (.18)
O(8)	.4795 (.0020)	.0977 (.0013)	.2392 (.0020)	.39 (.19)
O(9)	.3885 (.0019)	.2379 (.0013)	.6244 (.0019)	.35 (.19)

Table 3. *Listing of observed and final calculated structure factors for margarosanite*

h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c
0	3	-8	1.47	1.60	0	7	-2	13.10	13.14	1	7	-4	20.75	19.51	1	0	1	17.89	17.03
	4		2.60	2.64		8		14.64	14.01		8		4.77	4.36		1		21.89	24.71
	5		14.21	17.14		9		16.17	14.95		9		17.06	15.90		2		16.97	17.21
0	-2	-7	2.95	2.77		10		10.99	9.68		10		20.45	18.75		3		9.95	10.04
	-1		6.39	6.77		11		12.16	10.38		11		5.94	5.88		4		30.95	30.91
	0		7.88	8.24	0	-10	-1	2.83	3.53	1	-8	-3	10.78	10.02		5		19.58	15.23
	1		4.84	5.34		-9		1.69	1.64		-7		7.87	7.13		6		5.54	4.82
	2		4.58	5.02		-8		18.20	18.62		-6		1.45	0.93		7		29.73	28.25
	3		12.51	12.74		-7		7.63	6.63		-5		19.58	17.31		8		12.10	10.46
	4		3.60	3.68		-6		4.66	4.27		-4		13.00	14.43		9		5.40	5.17
	5		2.52	2.74		-5		18.03	17.21		-3		1.15	0.12		10		18.92	17.69
	6		9.47	9.23		-4		15.45	14.53		-2		20.63	22.67	1	-11	2	8.23	7.49
	7		8.50	8.47		-3		7.09	7.75		-1		12.19	13.47		-10		10.75	8.77
	8		2.44	2.51		-2		16.11	19.14		0		1.65	1.46		-9		1.00	0.50
0	-4	-6	7.07	6.81		-1		13.35	14.47		1		24.34	24.71		-8		6.11	4.04
	-3		11.30	11.59		0		2.25	2.01		2		8.24	9.12		-7		6.11	5.76
	-2		7.63	8.60		1		20.03	14.80		3		4.60	4.30		-6		9.29	9.54
	-1		10.86	12.31		2		18.99	21.52		4		20.42	17.15		-5		6.21	5.53
	0		8.21	8.19		3		7.71	7.93		5		19.32	16.91		-4		18.35	18.32
	1		15.55	16.66		4		8.19	9.27		6		2.43	2.63		-3		12.45	10.31
	2		19.13	20.24		5		21.53	22.36		7		9.36	9.84		-2		4.69	3.66
	3		10.46	10.09		6		10.00	9.76		8		15.04	14.72		-1		28.80	28.74
	4		17.34	17.77		7		11.12	10.51		9		6.07	5.99		0		20.76	19.14
	5		23.09	24.28		8		20.42	20.25		10		7.04	5.62		1		17.89	16.98
	6		9.48	9.11		9		2.86	2.76		11		10.98	9.00		2		24.29	26.88
	7		17.61	16.93		10		6.25	5.79	1	-9	-2	15.55	14.79		3		14.23	14.41
	8		15.73	16.18		11		17.18	14.88		-8		3.37	3.32		4		5.95	5.53
	9		5.07	5.01	0	-11	0	3.85	3.70		-7		16.90	15.35		5		28.23	24.10
	10		8.72	9.60		-10		7.62	7.01		-6		21.35	21.36		6		14.03	11.26
0	-6	-5	14.48	14.66		-9		17.06	15.79		-5		2.19	1.61		7		0.90	1.14
	-5		3.11	0.52		-8		4.53	4.28		-4		16.01	17.50		8		15.71	13.37
	-4		15.08	13.54		-5		6.80	6.73		-3		23.54	29.48		9		11.48	9.81
	-3		17.90	19.60		-4		14.24	13.01		-2		5.28	5.55	1	-11	3	6.42	6.08
	-2		3.83	3.72		-3		37.33	47.66		-1		13.63	14.81		-10		11.76	10.90
	-1		17.93	17.90		-2		15.06	13.14		0		30.49	38.41		-9		19.26	17.43
	0		15.28	18.44		-1		7.02	7.48		1		13.09	10.90		-8		11.33	9.44
	1		2.67	2.63	1	2	-8	10.03	10.49		2		23.64	20.05		-7		11.13	9.95
	2		14.94	16.41		3		10.23	9.87		3		25.20	28.80		-6		31.15	26.04
	3		15.88	16.25		4		10.15	9.82		4		19.79	20.32		-5		13.10	10.20
	4		6.44	6.45		5		13.46	13.97		5		17.92	18.48		-4		13.78	12.73
	5		12.53	11.78		6		8.48	9.01		6		26.08	27.38		-3		21.03	21.26
	6		16.69	16.57	1	-2	-7	1.45	0.99		7		10.04	9.07		-2		17.89	17.60
	7		10.88	10.67		-1		11.82	12.69		8		8.84	8.06		-1		14.84	13.69
	8		9.02	8.48		0		15.70	17.16		9		19.73	18.75		0		21.14	20.51
	9		13.54	13.17		1		1.92	1.57		10		7.01	5.94		1		14.06	13.15
	10		11.50	11.02		2		13.49	13.97		11		4.65	4.17		2		5.16	4.66
	11		6.22	6.69		3		15.52	16.19	1	-10	-1	4.80	4.26		3		14.87	14.59
0	-7	-4	1.76	1.04		4		3.31	3.30		-9		4.02	3.63		4		15.66	15.40
	-6		17.71	15.82		5		14.87	14.88		-8		18.27	18.80		5		3.68	2.98
	-5		6.28	5.18		6		11.86	11.93		-7		9.64	9.63		6		8.46	6.43
	-4		3.83	3.96		7		3.13	3.03		-6		5.89	5.46		7		16.92	14.40
	-3		15.32	16.36		8		8.64	8.37		-5		21.92	21.24		8		0.00	0.26
	-2		7.69	7.55		9		10.32	12.09		-4		10.85	9.73	1	-11	4	8.84	9.96
	-1		9.21	9.04	1	-4	-6	1.69	1.39		-3		1.91	1.51		-10		16.78	16.35
	0		19.33	18.80		-3		15.41	16.22		-2		20.40	27.69		-9		1.82	1.63
	1		11.65	13.42		-2		5.81	6.90		-1		9.37	10.05		-8		15.70	14.45
	2		6.93	7.13		-1		2.21	2.29		0		14.47	14.47		-7		22.38	21.20
	3		16.12	16.29		0		11.66	12.99		1		25.33	22.67		-6		0.75	0.33
	4		11.60	11.54		1		2.89	3.46		2		8.23	8.85		-5		21.37	19.38
	5		12.36	11.55		2		3.49	3.62		3		11.24	11.14		-4		23.03	22.01
	6		19.14	18.12		3		9.49	9.63		4		19.46	22.17		-3		1.25	0.45
	7		3.61	3.35		4		2.99	3.47		5		8.33	8.51		-2		16.89	17.45
	8		12.55	11.78		5		11.41	11.27		6		5.25	4.83		-1		24.04	26.17
	9		10.65	9.73		6		8.34	8.08		7		10.61	9.81		0		1.68	1.13
	10		4.83	4.23		7		9.40	9.19		8		20.72	20.42		1		14.98	13.05
	11		8.32	8.12		8		12.39	12.15		9		9.85	10.07		2		27.67	25.93
0	-8	-3	10.71	14.27		9		6.34	6.22		10		10.48	9.24		3		2.26	1.73
	-7		19.04	17.84		10		10.42	11.65		11		18.38	16.53		4		10.56	9.49
	-6		3.94	3.39	1	-6	-5	1.70	1.41	1	-10	0	14.23	12.82		5		24.09	18.81
	-5		23.59	21.25		-5		18.07	16.46		-9		7.59	6.75		6		3.42	2.74
	-4		15.21	17.67		-4		15.37	13.32		-8		16.11	15.87		7		2.67	2.64
	-3		7.98	8.32		-3		7.09	7.52		-7		18.19	18.14	1	-10	5	10.43	10.45
	-2		22.36	25.57		-2		16.68	17.10		-6		5.94	5.45		-9		11.62	10.89
	-1		25.09	27.30		-1		17.16	18.80		-5		7.75	8.58		-8		1.73	1.32
	0		9.70	8.79		0		8.40	8.52		-4		23.22	24.10		-7		12.44	11.03
	1		16.63	14.76		1		11.42	13.94		-3		7.50	7.27		-6		9.53	9.16
	2		29.13	33.85		2		18.85	20.28		-2		6.74	6.93		-5		2.44	2.10
	3		15.89	15.05		3		10.79	10.89		-1		16.85	17.08		-4		9.53	9.07
	4		6.59	5.45		4		13.10	13.23		0		26.48	15.87		-3		9.13	8.76
	5		26.65	21.41		5		14.75	14.85		1		0.85	0.16		-2		3.77	3.72
	6		18.19	15.16		6		12.53	12.14		2		16.47	19.42		-1		10.19	10.79
	7		7.79	7.77		7		5.54	4.97		3		24.03	20.08		0		5.72	6.11
	8		18.32	16.21		8		15.37	13.97		4		3.11	2.73		1		11.81	11.85
	9		19.04	16.29		9		13.40	12.53		5		16.38	15.11		2		4.35	3.41
	10		6.77	5.98		10		1.59	1.39		6		19.05	17.48		3		13.42	11.80
	11		15.15	13.25		11		9.60	10.31		7		5.71	4.99		4		16.81	13.02
0	-7	-2	1.32	0.92	1	-7	-4	8.05	7.64		8		11.48	10.03		5		2.49	1.94
	-6		17.01	14.82		-6		13.08	12.11		9		12.10	10.88	1	-9	6	12.74	12.97
	-5		7.23	6.48		-5		4.89	4.44		10		7.17	6.33		-8		10.94	10.17
	-4		4.21	4.47		-4		2.08	1.95	1	-11	1	13.71	12.49		-7		8.48	7.70
	-3		20.67	24.04		-3		13.59	15.03		-10		3.89	3.60		-6		9.51	8.87
	-2		5.51	6.12		-2		4.08	4.77		-9		16.33	15.99		-5		13.85	13.83
	-1		12.74	12.55		-1		11.69	11.45		-8		21.04	21.13		-4		13.73	13.31
	0		23.21	25.54		0		13.99	14.23		-7		3.80	3.44		-3		12.59	11.82
	1		19.93	17.20		1		17.90	18.56		-6		19.55	19.69		-2		12.67	12.66
	2		20.82	19.21		2		4.31	4.54		-5		24.59	20.85		-1		14.84	14.68
	3		21.86	24.67		3		19.20	19.34		-4		2.18	1.67		0		10.11	10.16
	4		19.02	19.28		4		19.19	19.35		-3		21.80	23.79		1		10.09	10.67
	5		20.54	20.81		5		6.93	6.09		-2		20.85	22.65		2		11.89	12.21
	6		21.37	23.36		6		26.56	24.09		-1		7.43	5.62		3		5.95	5.84

Table 3. *(Continued)*

h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c
1	-7	7	10.87	11.47	2	-6	-2	7.99	7.17	2	-7	3	13.86	12.57	3	1	-5	12.60	14.94
	-6		9.02	8.26		-5		3.94	5.70		-6		22.40	19.44		2		9.97	11.09
	-5		0.97	0.46		-4		24.70	23.97		-5		3.91	2.42		3		4.60	4.94
	-4		9.23	9.11		-3		1.17	1.32		-4		15.15	14.27		4		16.38	16.82
	-3		13.00	11.66		-2		1.07	1.40		-3		16.38	16.04		5		19.77	20.19
	-2		6.91	6.46		-1		14.78	16.54		-2		2.06	2.14		6		4.94	4.92
	-1		10.71	10.73		0		14.98	14.81		-1		5.21	5.71		7		18.28	17.56
	0		16.94	16.03		1		14.81	12.18		0		14.82	13.56		8		19.16	19.22
	1		10.15	9.41		4		15.78	15.07		1		9.75	8.89		9		3.32	3.04
2	2	-8	2.69	2.11		5		14.93	15.57		2		1.69	1.82		10		15.64	14.07
	3		5.33	5.77		6		7.40	6.89		3		16.53	15.83	3	-6	-4	2.70	2.50
	4		8.88	9.81		7		12.27	12.19		4		12.91	12.47		-5		5.49	4.99
	5		1.06	0.91		8		17.04	16.35		5		1.82	0.54		-4		14.50	14.09
	6		2.00	2.07		9		7.85	6.96		6		15.52	12.86		-3		4.79	5.30
2	-2	-7	10.82	10.40		10		10.90	10.31		7		17.57	15.61		-2		6.07	6.29
	-1		10.92	11.94		11		17.08	14.80		8		2.58	1.89		-1		12.19	12.39
	0		2.20	2.30	2	-9	-1	15.26	15.33	2	-10	4	1.42	0.44		0		4.86	5.23
	1		8.81	9.08		-8		5.12	5.13		-9		22.24	19.45		1		1.49	1.34
	2		14.33	15.26		-7		2.45	2.21		-8		6.01	5.33		2		13.91	15.45
	3		5.16	5.87		-6		21.03	20.66		-7		6.08	5.45		3		3.58	3.74
	4		6.27	5.74		-5		17.49	16.07		-6		12.84	12.46		4		6.18	5.70
	5		16.63	16.54		-4		2.88	1.67		-5		9.11	8.58		5		13.81	12.72
	6		9.31	9.06		-3		29.63	29.55		-4		16.50	16.08		6		12.99	12.18
	7		6.89	6.14		-2		17.97	22.33		-3		10.81	10.56		7		6.54	5.50
	8		15.53	15.19		-1		9.37	9.47		-2		8.44	8.85		8		12.59	11.17
	9		6.80	7.61		0		28.01	33.54		-1		18.18	19.92		9		14.63	13.33
2	-4	-6	7.08	7.23		1		26.19	30.75		0		8.15	8.45		10		5.40	4.85
	-3		10.39	11.28		2		4.68	4.19		1		13.88	12.90		11		9.10	9.42
	-2		8.29	10.02		3		16.87	18.36		2		15.27	14.65	3	-8	-3	1.16	1.19
	-1		7.48	8.70		4		29.93	31.86		3		10.27	9.74		-7		5.54	5.13
	0		8.89	10.62		5		3.64	3.59		4		11.94	11.01		-6		14.69	13.83
	1		17.29	19.24		6		16.49	15.86		5		16.84	13.46		-5		6.38	5.72
	2		9.91	10.37		7		21.73	20.21		6		11.16	9.99		-4		3.68	3.37
	3		10.94	10.97		8		7.76	7.10	2	-9	5	10.48	10.06		-3		14.43	15.14
	4		20.14	20.65		9		11.02	10.27		-8		5.49	5.19		-2		12.37	13.07
	5		2.90	2.62		10		18.52	16.49		-7		15.16	14.18		-1		6.64	6.35
	6		14.26	13.55		11		8.37	7.04		-6		19.30	18.90		0		28.17	30.05
	7		20.79	21.09	2	-10	0	12.15	12.01		-5		8.53	8.25		1		26.10	26.27
	8		2.06	1.73		-9		2.07	2.23		-4		16.29	16.09		2		1.45	1.17
	9		10.62	10.01		-8		6.97	6.46		-3		21.11	20.16		3		31.57	30.49
	10		20.96	21.25		-7		17.19	16.82		-2		2.59	2.67		4		32.42	30.75
2	-6	-5	6.44	5.90		-6		0.98	0.41		-1		20.18	20.62		5		2.27	1.90
	-5		9.71	8.44		-5		7.28	6.93		0		15.38	15.72		6		23.01	26.45
	-4		10.83	8.38		-4		25.60	26.95		1		1.17	0.20		7		16.52	16.26
	-3		1.89	1.96		-3		1.51	0.96		2		12.91	14.44		8		6.33	6.02
	-2		10.10	10.44		-2		0.60	1.15		3		15.60	15.60		9		14.13	13.55
	-1		18.02	20.19		-1		29.30	33.08		4		4.68	3.72		10		15.20	12.68
	0		2.04	1.37		0		13.53	10.79		5		12.43	11.47		11		3.77	3.19
	1		13.51	16.31		1		5.75	5.08	2	-8	6	6.42	6.15	3	-8	-2	8.50	8.44
	2		19.58	22.07		3		15.80	15.42		-7		6.20	5.47		-7		18.10	18.58
	3		1.66	1.56		4		8.33	6.67		-6		14.07	13.99		-6		6.12	5.62
	4		14.72	15.16		5		37.56	36.42		-5		8.93	8.53		-5		12.23	11.75
	5		17.97	17.60		6		11.35	10.05		-4		8.78	8.09		-4		25.06	24.11
	6		5.42	4.87		7		1.02	0.43		-3		10.96	11.40		-3		4.10	4.50
	7		8.55	7.99		8		25.93	23.98		-2		11.54	11.60		-2		13.57	14.80
	8		7.08	6.66		9		8.35	6.77		-1		8.60	8.16		-1		22.76	25.66
	9		2.97	2.51		10		1.81	1.63		0		11.01	10.89		0		9.37	3.34
	10		7.54	6.72	2	-10	1	7.19	6.57		1		10.65	10.77		1		12.72	12.14
	11		2.93	2.98		-9		18.00	17.22		2		7.07	7.45		2		23.53	23.28
2	-7	-4	19.54	19.06		-8		2.76	1.33		3		12.83	11.47		3		13.55	13.44
	-6		13.56	12.61		-7		6.58	6.21	2	-6	7	2.22	2.04		4		10.60	10.19
	-5		1.71	1.52		-6		25.46	26.64		-5		13.64	13.90		5		19.15	19.73
	-3		14.74	16.23		-5		10.85	10.67		-4		11.02	10.36		6		13.26	15.24
	-2		1.43	0.68		-4		9.44	9.21		-3		2.94	2.80		7		3.03	2.63
	-1		27.46	30.89		-3		26.13	27.93		-2		10.51	10.24		8		17.53	16.77
	0		12.33	12.11		-2		23.37	24.78		-1		8.53	8.39		9		14.47	13.51
	1		6.34	5.72		-1		14.41	13.15	3	3	-8	13.23	14.05		10		1.16	0.82
	2		20.54	23.86		0		29.48	30.13		4		10.51	11.19		11		13.61	11.39
	3		11.33	11.24		1		4.09	4.15		5		7.62	7.67	3	-9	-1	12.48	12.22
	4		13.23	12.54		2		5.41	5.55	3	-1	-7	10.71	11.74		-8		1.14	1.22
	5		18.66	15.89		3		16.75	17.45		0		3.92	3.80		-7		8.43	8.41
	6		17.02	12.45		4		2.42	2.50		1		5.53	6.25		-6		15.41	15.24
	7		15.19	13.79		5		9.84	7.86		2		11.88	12.19		-5		5.23	5.01
	8		14.71	13.23		7		9.05	7.35		3		1.08	1.08		-4		10.83	10.03
	9		13.49	11.64		8		1.37	0.67		4		8.02	8.38		-3		20.96	19.80
	10		13.39	12.16		9		7.21	5.99		5		12.73	13.48		-2		2.97	3.24
	11		11.70	11.28		10		10.74	9.35		6		1.73	1.71		-1		8.38	9.09
2	-8	-3	9.24	8.91	2	-10	2	14.30	13.19		7		8.10	7.68		0		21.45	24.80
	-7		6.65	6.32		-9		12.01	10.79		8		10.41	11.22		1		1.29	1.51
	-6		13.41	11.74		-8		18.41	18.39	3	-4	-6	14.26	14.87		2		3.44	4.22
	-5		13.04	11.66		-7		15.05	15.17		-3		11.63	11.82		3		19.57	21.39
	-4		12.05	11.82		-6		14.43	14.54		-2		3.57	3.77		4		10.33	9.84
	-3		13.19	14.85		-5		17.11	16.30		-1		16.76	20.45		5		7.91	7.73
	-2		16.32	18.64		-4		25.99	26.20		0		8.29	10.11		6		13.05	12.28
	-1		15.59	16.32		-3		17.58	14.10		1		2.10	2.00		7		9.85	9.08
	0		15.77	16.05		-2		16.12	15.09		2		20.44	22.92		8		2.52	2.09
	1		20.94	20.99		-1		28.09	32.75		3		10.80	12.04		9		9.64	8.88
	2		16.10	17.66		0		9.34	7.97		4		2.51	2.65		10		5.42	4.43
	3		6.32	6.06		1		11.96	11.55		5		17.63	18.17	3	-9	0	10.60	10.02
	4		28.45	22.63		2		31.25	33.79		6		8.85	8.63		-8		21.15	21.64
	5		14.79	14.30		3		4.83	4.46		7		5.97	5.26		-7		7.54	7.31
	6		1.22	1.01		4		10.64	10.53		8		10.81	10.29		-6		15.63	15.31
	7		15.15	15.71		5		27.47	24.05		9		8.04	7.29		-5		20.48	20.51
	8		16.46	16.33		6		3.74	3.04		10		7.97	9.60		-4		12.34	12.71
	9		3.36	3.09		7		9.70	8.47	3	-5	-5	8.24	8.88		-3		15.39	16.36
	10		14.32	11.74		8		21.57	19.93		-4		8.83	7.41		-2		28.24	29.09
	11		14.54	12.87		9		4.18	3.43		-3		14.16	14.49		-1		14.21	14.40
2	-9	-2	0.86	0.74	2	-10	3	7.83	7.19		-2		9.47	10.12		0		6.93	6.39
	-8		12.44	12.23		-9		15.87	15.19		-1		7.57	7.45		1		27.27	31.83
	-7		21.50	21.48		-8		2.85	2.90		0		8.68	8.53		2		19.72	21.21

Table 3. *(Continued)*

h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c
3	3	0	1.72	1.61	3	1	6	20.19	21.04	4	1	-1	10.99	11.87	4	-1	5	15.72	15.81
	4		21.88	19.02	4	0	-7	9.61	4.20		2		13.94	14.40		0		3.44	3.28
	5		21.77	18.52		1		13.52	14.74		3		21.58	20.99		1		11.21	11.30
	6		14.61	13.19		2		4.89	4.76		4		12.59	11.91		2		13.81	14.15
	8		19.86	18.27		3		3.33	3.43		5		18.66	17.83	4	-3	6	9.78	9.64
	9		9.56	8.77		4		11.99	12.58		6		22.45	21.70		-2		1.36	1.19
	10		8.47	7.99		5		10.75	10.97		7		8.69	7.84	5	1	-7	9.44	10.87
3	-10	1	10.17	9.51		6		4.63	4.64		8		10.87	10.32		2		12.61	12.96
	-9		9.86	9.65		7		14.86	14.73		9		21.72	19.79		3		9.96	10.50
	-8		3.30	3.17		8		7.98	9.19		10		8.83	8.47		4		8.98	9.69
	-7		5.17	5.55	4	-3	-6	8.03	8.01	4	-9	0	5.68	5.33		5		6.74	6.50
	-6		14.52	14.58		-2		4.94	5.69		-8		13.55	13.22		6		7.63	8.35
	-5		3.61	3.64		-1		8.54	9.93		-7		2.82	2.93	5	-2	-6	12.34	13.68
	-4		9.66	9.73		0		11.26	12.09		-6		8.50	9.07		-1		8.90	10.79
	-3		15.10	15.95		1		6.00	6.61		-5		20.66	21.33		0		2.47	2.11
	-2		9.01	9.39		2		8.47	8.89		-4		2.61	2.34		1		13.77	15.29
	-1		9.74	9.54		3		14.28	14.28		-3		7.22	7.79		2		13.48	15.01
	0		27.16	26.80		4		5.90	5.78		-2		24.08	26.26		3		3.41	3.23
	1		8.30	7.91		5		7.77	8.11		-1		0.60	0.43		4		16.26	16.02
	2		9.97	10.03		7		6.48	6.34		0		10.28	10.31		5		14.98	15.29
	3		20.48	21.71		8		6.25	6.22		1		19.16	20.63		6		5.55	5.43
	4		12.64	12.38		9		14.41	14.75		2		1.47	1.38		7		11.99	12.19
	5		8.84	7.31	4	-4	-5	12.27	11.44		3		4.22	4.30		8		12.31	14.04
	6		18.59	16.27		-3		14.80	14.99		4		23.11	22.59	5	-3	-5	6.97	7.28
	7		12.73	10.58		-2		10.98	11.67		5		3.22	2.95		-2		1.25	1.04
	8		1.18	0.71		-1		6.75	6.90		6		3.67	3.24		-1		10.87	12.16
	9		15.74	14.37		0		15.94	16.76		7		19.37	18.26		0		6.70	8.14
3	-10	2	1.41	1.18		1		15.10	16.62		8		4.41	3.67		1		7.46	7.71
	-9		13.27	12.18		2		5.25	4.85		9		3.34	3.30		2		9.84	11.01
	-8		12.91	12.11		3		12.11	12.39	4	-9	1	1.05	0.52		3		6.63	7.31
	-7		2.10	1.91		4		19.53	20.71		-8		7.04	7.00		4		9.82	9.53
	-6		12.04	12.40		5		1.92	1.24		-7		22.65	23.07		5		2.82	1.58
	-5		17.16	15.89		6		15.70	14.46		-6		5.63	5.47		6		9.17	8.38
	-4		6.85	5.72		7		17.00	16.60		-5		6.67	6.81		7		9.70	10.21
	-3		9.98	9.04		8		1.04	0.86		-4		27.42	28.73		8		3.22	2.99
	-2		16.17	15.59		9		14.97	13.44		-3		8.68	8.80		9		6.05	5.36
	-1		1.41	1.79		10		14.27	13.63		-2		5.12	4.78	5	-5	-4	10.71	12.22
	0		3.93	3.99	4	-6	-4	3.98	3.55		-1		26.89	28.56		-4		2.40	2.29
	1		17.72	17.85		-5		12.36	12.40		0		14.87	13.58		-3		9.48	9.18
	2		12.89	12.33		-4		15.59	14.47		1		5.64	5.41		-2		12.90	14.05
	3		2.44	1.94		-3		11.57	12.01		2		23.31	24.17		-1		7.59	7.98
	4		20.42	21.18		-2		17.40	18.47		3		17.48	18.14		0		8.29	8.22
	5		14.84	12.47		-1		16.01	17.59		4		8.05	7.58		1		19.52	19.30
	6		4.55	3.69		0		12.96	12.93		5		23.96	22.16		2		9.11	9.57
	7		20.29	18.53		1		22.14	21.67		6		14.66	11.84		3		1.78	2.48
	8		6.51	5.66		2		14.23	16.08		7		5.22	4.70		4		21.81	21.19
3	-10	3	11.11	10.63		3		11.05	10.00		8		15.94	15.03		5		12.66	12.06
	-9		6.21	6.01		4		24.77	24.42	4	-9	2	11.13	10.59		6		3.41	2.72
	-8		5.64	5.15		5		9.18	8.72		-8		16.47	15.33		7		16.59	15.95
	-7		15.82	14.54		6		7.88	6.57		-7		6.69	6.16		8		11.81	10.39
	-6		16.81	16.09		7		14.77	12.62		-6		12.78	11.97		9		1.94	1.35
	-5		9.03	8.17		8		10.59	9.05		-5		19.19	16.68	5	-6	-3	4.86	5.63
	-4		24.86	24.83		9		11.81	10.89		-4		6.29	5.57		-5		1.58	1.09
	-3		21.58	20.42		10		7.13	6.22		-3		11.37	11.40		-4		16.79	17.20
	-2		5.57	5.59	4	-7	-3	17.17	16.29		-2		17.64	17.82		-3		7.66	7.92
	-1		31.92	36.49		-6		8.46	8.27		-1		10.64	11.32		-2		1.17	0.78
	0		21.39	19.52		-5		3.69	3.26		0		5.34	5.40		-1		14.79	15.66
	1		8.31	7.58		-4		19.17	17.79		1		17.52	16.76		0		14.66	14.70
	2		22.95	23.09		-3		11.06	11.63		2		6.87	6.24		1		2.02	1.53
	3		19.99	20.16		-2		2.17	2.15		3		5.34	5.14		2		12.78	13.53
	5		15.01	12.37		-1		11.71	11.93		4		16.19	16.00		3		16.51	18.62
	6		15.93	13.16		0		7.80	8.48		5		2.00	1.72		4		11.54	11.99
	7		1.29	0.70		1		10.51	9.99		6		1.74	1.10		5		15.02	15.71
3	-9	4	9.41	9.23		2		9.24	9.47		7		22.14	21.15		6		19.64	21.06
	-8		13.17	12.81		3		6.19	6.16	4	-9	3	12.87	12.90		7		11.32	11.63
	-7		5.25	4.99		4		14.42	14.16		-8		11.18	10.38		8		13.67	14.01
	-6		9.17	8.67		5		4.85	4.61		-7		10.88	10.16		9		20.38	18.90
	-5		13.29	12.91		6		10.88	11.31		-6		5.59	4.73	5	-7	-2	1.26	1.50
	-4		1.18	1.28		7		12.04	12.17		-5		10.82	10.65		-6		7.86	8.97
	-3		9.55	9.23		8		1.21	1.14		-4		15.59	14.66		-5		8.15	8.83
	-2		17.31	17.15		9		12.49	11.63		-3		1.45	1.28		-4		3.39	3.57
	-1		2.12	1.98		10		10.71	9.67		-2		8.31	8.30		-3		8.69	8.85
	0		5.93	6.41	4	-8	-2	17.60	18.23		-1		16.51	17.46		-2		18.07	20.63
	1		18.09	16.99		-7		2.59	2.63		0		1.74	1.76		-1		8.20	8.41
	2		3.24	3.02		-6		8.73	8.34		1		10.81	10.26		0		13.59	14.82
	3		6.53	6.01		-5		26.82	26.46		2		15.27	14.67		1		21.12	22.60
	4		15.04	13.28		-4		6.84	6.60		3		1.37	1.52		2		7.34	7.79
	5		5.31	4.43		-3		9.22	9.12		4		5.69	5.22		3		11.81	12.48
	6		1.33	0.96		-2		26.13	31.14		5		14.00	12.03		4		17.87	19.10
3	-8	5	13.26	13.07		-1		5.08	5.53		6		5.48	4.70		5		1.60	1.52
	-7		15.18	14.59		0		3.46	3.53	4	-8	4	7.24	6.52		6		5.31	5.19
	-6		3.55	3.37		1		25.92	27.36		-7		15.80	15.48		7		8.08	7.38
	-5		16.98	16.80		2		6.55	6.46		-6		14.69	14.36		8		5.43	4.95
	-4		14.64	13.94		3		3.52	3.22		-5		7.84	7.17		9		4.85	4.32
	-3		3.02	2.95		4		21.69	20.98		-4		14.58	14.68	5	-7	-1	20.08	17.48
	-2		14.52	14.58		5		9.30	9.65		-3		8.46	8.48		-6		0.81	0.87
	-1		11.59	11.92		6		8.67	8.39		-2		13.00	12.68		-5		13.71	13.94
	0		1.63	1.93		7		19.97	19.31		-1		10.35	10.42		-4		19.22	21.65
	1		8.71	9.37		8		11.94	10.85		0		7.59	7.70		-3		1.32	1.36
	2		8.80	9.73		9		8.23	7.84		1		16.88	17.64		-2		16.02	16.73
	3		5.40	5.63		10		14.79	14.89		2		5.49	5.46		-1		19.69	20.89
	4		5.88	5.78	4	-8	-1	3.78	3.98		3		13.01	12.65		0		6.12	6.26
3	-7	6	11.06	11.37		-7		12.51	13.11		4		20.16	18.08		2		20.35	22.84
	-6		9.99	9.56		-6		3.69	4.08		5		3.69	3.91		3		9.31	8.21
	-5		9.39	9.01		-5		7.37	6.89	4	-7	5	10.30	10.64		4		6.37	5.88
	-4		7.92	7.64		-4		14.03	13.96		-6		1.14	1.04		5		19.11	18.02
	-3		12.01	11.54		-3		4.71	5.32		-5		6.62	6.86		6		10.96	10.19
	-2		14.97	14.68		-2		19.48	18.09		-4		12.63	12.44		7		3.35	2.93
	-1		2.89	3.08		-1		10.62	10.70		-3		1.26	0.91		8		16.08	14.71
	0		10.49	10.37		0		18.07	20.33		-2		9.80	9.92		9		9.61	8.48

Table 3. *(Continued)*

h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c	h	k	l	F_o	F_c
5	-8	0	6.06	7.19	5	-6	3	4.61	5.17	6	3	-4	11.55	11.83	6	-5	0	3.55	3.03
	-7		2.33	2.67		-5		11.74	11.85		4		8.35	8.91		-4		5.31	5.76
	-6		13.72	15.27		-4		9.20	9.28		5		9.01	8.79		-3		16.82	18.44
	-5		11.75	12.47		-3		1.31	1.40		6		13.02	12.53		-2		4.05	4.05
	-4		7.32	7.80		-2		13.38	13.72		7		7.53	7.56		-1		10.99	11.34
	-3		17.21	18.77		-1		13.75	14.34		8		10.31	9.78		0		24.25	27.10
	-2		12.99	13.52		0		3.28	3.15	6	-5	-3	15.64	15.99		1		1.65	1.79
	-1		6.53	6.41		1		14.32	14.50		-4		11.96	13.02		2		10.44	10.76
	0		17.13	18.40		2		17.81	17.90		-3		2.77	2.98		3		26.24	29.87
	1		12.60	12.93		3		2.61	2.49		-2		18.27	19.95		4		2.25	2.33
	2		2.87	2.81		4		16.67	14.74		-1		10.80	12.28		5		11.71	10.56
	3		14.54	15.51		5		16.07	14.33		0		3.89	4.53		6		22.23	20.57
	4		13.37	13.33	5	-6	4	11.41	13.54		1		19.31	20.70		7		4.42	3.61
	5		8.92	7.44		-5		7.22	7.64		2		13.02	14.20	6	-6	1	1.73	1.74
	6		10.98	10.35		-4		12.54	12.53		3		3.48	3.32		-5		18.53	19.01
	7		15.06	13.60		-3		16.28	16.45		4		15.04	15.78		-4		5.65	5.75
	8		8.61	8.19		-2		8.45	8.05		5		9.63	9.64		-3		1.37	1.10
5	-8	1	12.32	14.59		-1		10.81	11.21		6		9.01	9.21		-2		20.60	21.00
	-7		2.81	3.13		0		9.56	9.99		7		7.34	7.22		-1		4.95	5.38
	-6		7.23	7.05		1		3.59	3.80		8		9.25	8.43		0		1.22	0.54
	-5		12.25	13.04		2		6.10	6.25	6	-5	-2	5.78	6.20		1		18.86	18.73
	-4		7.39	7.97		3		8.43	8.56		-4		3.69	4.02		2		10.02	9.89
	-3		3.97	4.03	5	-3	5	9.33	10.88		-3		14.02	14.78		3		2.50	2.15
	-2		6.15	6.12		-2		14.19	16.44		-2		6.51	6.60		4		13.33	13.93
	-1		13.62	14.29		-1		4.41	4.28		-1		4.89	4.79		5		11.64	10.69
	0		5.36	4.87	6	0	-6	6.05	7.10		0		18.72	21.11		6		3.25	3.37
	1		5.72	5.87		1		7.98	8.58		1		7.25	7.75	6	-6	2	11.40	11.66
	2		12.76	13.04		2		7.52	8.09		2		6.02	5.86		-5		8.38	8.20
	3		6.88	8.39		3		5.67	5.81		3		18.93	20.27		-4		10.46	10.43
	4		3.57	3.49		4		12.80	13.09		4		12.14	13.07		-3		10.65	10.55
	5		14.71	13.47		5		12.79	13.38		5		4.12	4.61		-2		5.65	8.31
	6		7.69	7.10		6		11.19	11.28		6		17.99	18.29		-1		7.31	10.72
	7		6.88	5.89	6	-2	-5	5.89	5.78		7		11.51	11.15		0		9.07	10.49
5	-8	2	0.97	0.79		-1		11.78	13.25		8		7.99	7.94		1		8.10	7.55
	-7		8.39	8.85		0		16.79	17.12	6	-6	-1	0.85	0.63		2		8.33	8.14
	-6		15.38	16.24		1		5.24	5.02		-5		15.00	16.23		3		12.74	12.54
	-5		2.32	2.21		2		12.74	14.27		-4		3.70	4.12		4		7.58	6.79
	-4		11.54	11.08		3		15.65	16.47		-3		1.55	0.91		5		3.84	3.72
	-3		20.22	20.57		4		3.53	3.43		-2		12.83	13.56	6	-5	3	12.95	14.60
	-2		8.61	8.49		5		13.25	13.15		-1		1.74	2.43		-4		8.68	8.66
	-1		12.82	13.84		6		15.74	15.93		0		2.91	2.70		-3		11.24	11.18
	0		23.06	24.81		7		1.14	0.44		2		2.33	3.25		-2		13.29	13.51
	1		5.39	4.73		8		9.90	10.13		3		9.64	9.67		-1		2.80	2.73
	2		6.82	6.65	6	-3	-4	10.57	10.60		4		8.85	8.67		0		8.83	8.65
	3		21.60	23.12		-2		8.02	8.85		5		10.81	10.39		1		15.82	15.99
	4		5.50	4.97		-1		5.54	5.75		6		10.62	10.19		2		3.66	3.69
	5		4.42	4.06		0		10.93	11.51		7		7.27	8.20		3		5.86	5.87
	6		17.03	15.22		1		7.02	7.75		8		13.17	13.03					
5	-7	3	5.19	6.01		2		7.46	8.44	6	-6	0	11.53	12.46					

temperature factors for all atoms were refined to values which gave $R = 13.2\%$. Refinement of form factor, scale factors, and the calculated interatomic distances gave reasonable values. Structure factors calculated by using only Pb were compared to structure factors computed at this stage of refinement. Approximately 2/3 of the total number of intensity values were found to correlate satisfactorily. These were used to calculate the three-dimensional difference Fourier syntheses $\Delta\varrho(xyz)$. No major discontinuities were discovered in the resulting map.

Corrections for average real and imaginary components of anomalous scattering (DAUBEN and TEMPLETON, 1955; TEMPLETON, 1955) were made at this time and positional parameters were refined to values which gave $R = 11.8\%$.

The electron-density function $\varrho(xyz)$ was computed using all structure factors at this point in refinement. The locations and relative weights of the peaks verified that the atom parameters were correctly chosen. Several errors in the definition of the crystal shape were discovered; after correcting for these errors the refinement proceeded

to completion. The final parameters and their standard errors are listed in Table 2. The observed and calculated structure factors are listed in Table 3. Selected interatomic distances and angles are listed in Table 4. These were computed with an IBM 7090 Fortran II program written by BUSING, MARTIN and LEVY (1964). The final conventional discrepancy factor was 7.4%.

Table 4. *Selected interatomic distances and angles of margarosanite*
Standard deviations are in parentheses

Si(1)—O(1)	1.587Å	(.013)	Si(2)—O(4)	1.679Å	(.013)
O(2)	1.621	(.013)	O(5)	1.596	(.013)
O(3)	1.656	(.015)	O(6)	1.608	(.014)
O(4)	1.672	(.013)	O(7)	1.680	(.013)
average	1.634		average	1.641	
Si(3)—O(7)	1.678	(.013)	Ca(1)—O(1)	2.360	(.012)
O(8)	1.617	(.012)	O(5)	2.481	(.013)
O(9)	1.595	(.013)	O(5)′	2.458	(.014)
O(3)	1.698	(.015)	O(6)	2.555	(.013)
average	1.647		O(6)′	2.751	(.014)
			O(9)	2.413	(.012)
Si(1)—Si(2)	2.929	(.007)	average	2.503	
Si(1)—Si(3)	2.907	(.007)			
Si(2)—Si(3)	2.954	(.007)	O(1)—O(2)	2.600	(.017)
			O(1)—O(3)	2.643	(.018)
Ca(2)—O(1)	2.344	(.014)	O(1)—O(4)	2.705	(.017)
O(2)	2.308	(.014)	O(2)—O(3)	2.698	(.019)
O(5)	2.482	(.013)	O(2)—O(4)	2.740	(.017)
O(8)	2.440	(.012)	O(3)—O(4)	2.623	(.019)
O(8)′	2.351	(.013)	O(4)—O(5)	2.717	(.018)
O(9)	2.313	(.012)	O(4)—O(6)	2.623	(.018)
average	2.373		O(4)—O(7)	2.589	(.017)
			O(5)—O(6)	2.741	(.019)
Pb—O(1)	2.721	(.012)	O(5)—O(7)	2.724	(.017)
O(2)	2.460	(.012)	O(6)—O(7)	2.633	(.018)
O(2)′	2.908	(.012)	O(7)—O(8)	2.719	(.017)
O(3)	3.020	(.015)	O(7)—O(9)	2.665	(.018)
O(6)	2.293	(.013)	O(7)—O(3)	2.661	(.019)
O(8)	2.697	(.013)	O(8)—O(9)	2.645	(.017)
O(9)	2.657	(.012)	O(8)—O(3)	2.740	(.019)
average	2.679		O(9)—O(3)	2.699	(.020)
			average	2.676	

Table 4. *(Continued)*

Angles					
Si(1)—Si(2)—Si(3)	59.23°	(.15)	O(4)—Si(2)—O(6)	105.84°	(.70)
Si(2)—Si(1)—Si(3)	60.82	(.16)	O(4)—Si(2)—O(7)	100.82	(.62)
Si(2)—Si(3)—Si(1)	59.96	(.16)	O(5)—Si(2)—O(6)	117.63	(.71)
Ca(1)—Ca(1)′—Ca(1)″	177.27	(.17)	O(5)—Si(2)—O(7)	112.50	(.70)
O(1)—Si(1)—O(2)	108.26	(.64)	O(6)—Si(2)—O(7)	106.40	(.68)
O(1)—Si(1)—O(3)	109.14	(.74)	O(7)—Si(3)—O(8)	111.17	(.66)
O(1)—Si(1)—O(4)	112.16	(.66)	O(7)—Si(3)—O(9)	108.97	(.65)
O(2)—Si(1)—O(3)	110.81	(.70)	O(7)—Si(3)—O(3)	104.01	(.73)
O(2)—Si(1)—O(4)	112.41	(.67)	O(8)—Si(3)—O(9)	110.89	(.69)
O(3)—Si(1)—O(4)	104.01	(.71)	O(8)—Si(3)—O(3)	111.47	(.71)
O(4)—Si(2)—O(5)	112.09	(.65)	O(9)—Si(3)—O(3)	110.09	(.71)

Results

Margarosanite is similar to other metasilicates in that its structure is composed of planes of tetrahedral sites alternating with planes of calcium sites between sheets of approximately close-packed oxygen atoms. The plane of close-packing is $(\bar{1}01)$. This plane is the (100) plane in both the pseudo-orthorhombic and pseudo-monoclinic cells discussed above. The tetrahedral sites are arranged in groups of three-membered rings which are shown in Figs. 2 and 3. Figure 2 shows the structure in the same orientation as Fig. 1. Figure 3 is a composite view of the tetrahedral and octahedral layers projected onto a plane parallel to the close-packed layers. The major feature of the octahedral layer is a continuous chain of edge-sharing Ca(1) polyhedra. Pb and Ca(2) polyhedra alternate along the edges of the chain.

Two tetrahedral layers are shown in Fig. 3. The rings are in groups of two with vertices pointing towards each other. The octahedral layer is situated between the two planes of rings, and the vertex oxygen atoms join the rings to the Ca(1) chain. One Ca(1) chain ties together a continuous series of ring groups, each group composed of one ring above and one ring below. The angles and distances given in Table 4 show that the silicon atoms form a nearly perfect equilateral triangle.

The Ca(1) chain is the major feature of the octahedral cation sheet. All other structural features of margarosanite can be described and discussed in terms of this chain. The chain direction is [101] which is the fiber direction of the Långban margarosanite. Individual Ca(1)

polyhedra have only approximate octahedral coordination. A Ca(1) polyhedron shares two edges with other Ca(1) polyhedra, two edges with Pb polyhedra, and two edges with Ca(2) polyhedra.

Ca(2) atoms have a 6-fold coordination polyhedron which is essentially a regular octahedron. A Ca(2) octahedron shares two edges

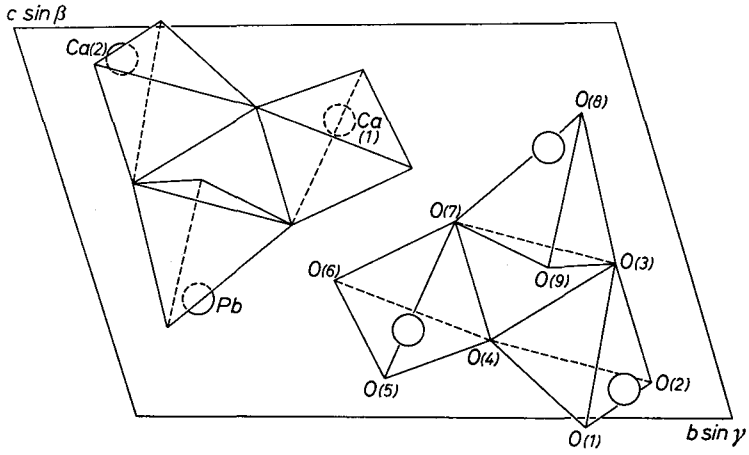

Fig. 2. Projection along a of the structure of margarosanite

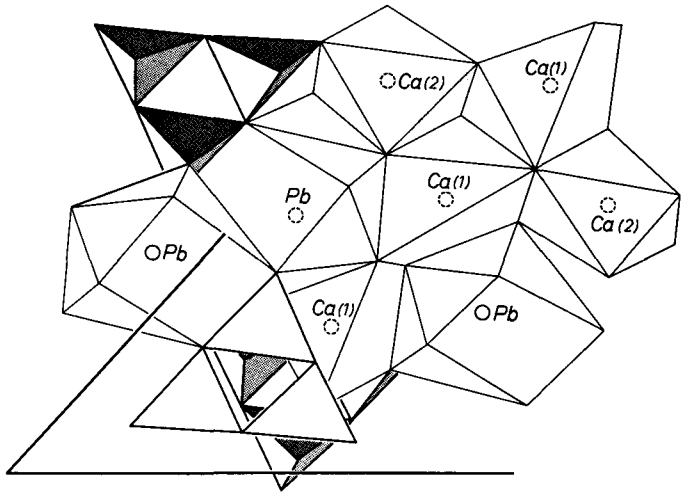

Fig. 3. The structure of margarosanite projected onto a plane parallel with the approximately close-packed layers. Two tetrahedral layers and one octahedral layer are shown. The horizontal line is the b' axis ($b' = [111]$), the other is the c' axis ($c' = [101]$), and the a' axis ($a' = [2\bar{1}\bar{2}]$) is normal to the diagram. This cell is the pseudo-monoclinic cell described in the text.

with Ca(1) polyhedra and shares one edge with another Ca(2) octahedron producing a two-membered unit of Ca(2) octahedra. These units are separated from similar units by the Ca(1) chain. Thus, they link Ca(1) chains forming a two-dimensional network of calcium polyhedra.

The Pb atoms are situated in part of the oxygen close-packed layers, and, in particular, are in the layers containing those oxygen atoms which join the tetrahedral rings to the Ca(1) chain. The Pb atom has 7-fold coordination. The Pb-coordination polyhedron resembles a six-sided half-shell, with the Pb atom occupying the center of the opening. The Pb polyhedron on the right in Fig. 3 has the opening towards the viewer. The other Pb polyhedron has the surface of the half-shell towards the viewer. A single Pb polyhedron shares two edges with Ca(1) polyhedra, two edges with Si(1) tetrahedra, and one edge with a Si(3) tetrahedron. Two Pb polyhedra share edges forming a two-membered unit. These units are transverse to the close-packed plane and are separated from similar units by Ca(1) chains. The Pb polyhedral units link Ca(1) chains from two different two-dimensional networks of calcium polyhedra. Thus, the octahedral cations form a continuous three-dimensional network.

Valency sums may be calculated for each oxygen atom by assuming a bond strength of 2/7 for each Pb—O bond, 2/6 for each Ca—O bond, and 1 for each Si—O bond. Four groups of oxygen atoms are present: (1) three oxygen atoms, O(3), O(4), O(7), are all coordinated to two Si and one Pb for a total bond strength of $2\frac{2}{7}$ each: (2) four oxygen atoms, O(1), O(6), O(8), O(9), are all coordinated to one Si, two Ca, and one Pb for a total bond strength of $1\frac{2}{21}$ each; (3) O(2) is coordinated to one Si, one Ca, and two Pb for a total bond strength of $1\frac{19}{21}$; (4) O(5) is coordinated to one Si and three Ca for a total bond strength of 2. These deviations from the ideal are compensated, in general, by distinct lengthening of bonds associated with group (1), and normal lengths of bonds associated with the remaining oxygen atoms.

Margarosanite is related to benitoite ($BaTiSi_3O_9$; ZACHARIASEN, 1930) and wadite ($Zr_2K_4Si_6O_{18}$; HENSHAW, 1956) by the fact that all three have X_3O_9 rings. However, the cation distribution for each is different. DORNBERGER-SCHIFF (1962) has proposed that pseudowollastonite ($CaSiO_3$) has X_3O_9 rings. The unit-cell parameters for the pseudo-orthorhombic cell of margarosanite ($a' = 20.40$ Å, $b' = 11.54$ Å, $c' = 9.50$ Å, $\alpha' = 82°$, $\beta' = 85°$, $\gamma' = 94°$) and those of pseudowollastonite ($a = 6.90$ Å, $b = 11.78$ Å, $c = 19.65$ Å, $\alpha = \beta = 90°$,

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