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RELATION BETWEEN CHEMICAL COMPOSITION AND LATTICE CONSTANTS OF EPIDOTE

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Abstract

Chemical, optical and *x*-ray data on five members of the clinozoisite-pistacite series are presented. The unit cell becomes larger with increasing ferric iron content. It is shown that the members of the clinozoisite-pistacite series can be readily distinguished from zoisite by the *x*-ray powder method.

Introduction

The *x*-ray study of epidote was attempted by Gossner and Mussgnug (1930), Bujor (1931), Strunz (1935), Ito (1950), Ito, Morimoto and Sadanaga (1954) and Lapham (1957). The most elaborate analysis of the crystal structure was given by Ito, Morimoto and Sadanaga (1954).

The writer was greatly interested in the epidote minerals (clinozoisite, pistacite, piemontite and zoisite) during his study of regional metamorphism in the Kanto Mountains, central Japan (Seki, 1958). The behavior of epidote in this progressive metamorphic terrain has been described and discussed by Miyashiro and Seki (1958).

In the present paper, some relations between the chemical composition and lattice constants for the members of the clino-zoisite-pistacite series and the distinction of the members of this series from zoisite by the *x*-ray powder method will be described in some detail. The nomenclature of the members of the epidote group adopted in this paper are the same as that adopted by Miyashiro and Seki (1958).

Chemical Compositions and Optical Properties of Clinozoisites and Pistacites

The localities and modes of occurrence of the five epidotes used in this paper are shown in Table 1.

The samples were purified by means of the isodynamic separator and Clerici solution.

Among them, the chemical composition and optical properties of the Hawleyville pistacite were already given by Lapham (1957). The chemical analysis and optical measurements on the other four minerals were newly carried out by the present writer and Miss Chigusa Kato of the Saitama University. These data are summarized in Table 2.

In the table, the analyses are arranged in the order of increasing $Fe^{+3}/(Al+Fe^{+3})$ ratio. The atomic ratios were calculated on the anhydrous basis of $O=25$, because the water contents shown in Table 2 are somewhat unreliable.

The indices of refraction, double refraction and the intensities of

TABLE 1. LOCALITIES AND MODES OF OCCURRENCE OF THE CLINOZOISITES AND PISTACITES USED

No.	Name	Locality	Mode of occurrence
1	Sasaguri clinozoisite	Nakakoti, Sasaguri, Hukuoka Prefecture, Japan (YS-C1-3)	Occurs in leucocratic veins and nodules in serpentinites. Minerals associated with the clinozoisite are sericite, quartz and albite. The serpentinites are intruded into low grade crystalline schists including crossite-bearing green schists
2	Kanto clinozoisite	Huppu, Yorii-mati, Saitama Prefecture (Kanto Mts.), Japan (TT57032206)	Occurs as fine grained crystals in chlorite-clinozoisite-albite-titanite schist derived from mafic pyroclastic materials. This metamorphic rock with conspicuous albite porphyroblasts represents the highest grade of metamorphism in the Kanto Mountains (Miyashiro and Seki, 1958; Seki, 1958)
3	Hawleyville pistacite	East of Danbury, Connecticut, U.S.A.	Occurs in single crystals or parallel groups in the pink feldspar-epidote-chlorite pegmatites intrusive into medium to coarse grained diorite. Minerals associated with this pistacite are apatite, quartz, prochlorite, muscovite and orthoclase (Lapham, 1957)
4	Kanto pistacite	Northwestern side of Mt. Hoto, Minano-mati, Saitama Prefecture (Kanto Mts.), Japan (YS57062609)	This pistacite is associated with chlorite, actinolite, quartz, albite and titanite in a low-grade green schist derived from mafic pyroclastic rock. This green schist without any conspicuous porphyroblast of albite belongs to the writer's non-spotted schist formation (Miyashiro and Seki, 1958; Seki, 1958)
5	Dauphine pistacite	Dauphine, Bourg d'Oisans, Swiss	Unknown to the writer. It is composed of an aggregate of dark green crystals in parallel growth, up to several cm. in length

pleochroism of these minerals generally increase with the ferric iron content.

Optically, clinozoisites (with 0–10 per cent Fe^{+3} end-member) are positive, while pistacites (with more than 10 per cent Fe^{+3}) are negative.

X-RAY POWDER DATA AND LATTICE CONSTANTS OF CLINOZOISITES AND PISTACITES

X-ray powder diffraction data of the above five epidotes were obtained by means of the Philips Geiger counter *x*-ray diffractometer using CuKα-radiation. Silicon powder or Brazilian quartz was used as the internal standard. The data are listed in Table 3, where about fifty peaks between 10 and 60 degrees in angle 2θ were indexed.* The results

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* Lapham (1957) stated that the Hawleyville pistacite is characterized by the presence of the 7.02 Å and 1.590 Å lines. However, no reflections with these *d* values were found in the writer's *x*-ray powder data of epidotes including the Hawleyville pistacite. It is probable that these reflections in the Lapham's data are due to admixed chlorite and/or other minerals. Lapham also said that the 6.60 Å and 3.325 Å lines are characteristic reflections of his Timmins clinozoisite. However, these lines were not found in any of the clinozoisites treated here.

There are minor differences between this indexing and that of Fisher (*Am. Mineral.*, **43**, 588–589) obtained on single crystal precession photographs.

TABLE 2. CHEMICAL COMPOSITIONS AND OPTICAL PROPERTIES* OF CLINOZOISITES AND PISTACITES

	1† Clinozoisite (Sasaguri)	2† Clinozoisite (Kanto)	3 Pistacite (Hawleyville)	4† Pistacite (Kanto)	5† Pistacite (Dauphine)
$\mathrm{SiO_2}$	37.35	37.41	37.96	37.69	36.88
$\mathrm{Al_2O_3}$	29.53	29.88	27.34	20.73	18.61
$\mathrm{Fe_2O_3}$	4.03	4.09	8.88	14.03	17.93
FeO	0.29	0.58	0.97	1.17	0.37
MnO	0.60	0.14	0.16	0.27	0.48
MgO	0.02	tr.	0.34	0.22	tr.
CaO	23.65	23.92	22.07	22.72	23.06
$\mathrm{Na_2O}$	—	0.11	—	0.09	—
$\mathrm{K_2O}$	—	—	—	—	—
$\mathrm{H_2O+}$	3.21	3.12	2.14	2.77	2.30
$\mathrm{H_2O-}$	0.62	0.34	0.08	0.35	0.02
Total	99.30	99.59	99.94	100.04	99.65
Si	6.01	5.90	5.96	6.12	6.06
		0.10 } 6.00	0.04 } 6.00		
Al	5.46	5.45	5.02	3.97	3.60
$\mathrm{Fe'''}$	0.47	0.48	1.05	1.72	2.21
$\mathrm{Fe''}$	0.04 } 6.05	0.08 } 6.03	0.13 } 6.28	0.16 } 5.94	0.05 } 5.93
Mn	0.08	0.02	0.02	0.04	0.07
Mg	0.00	—	0.08	0.05	—
Ca	3.97	4.03	3.71	3.96	4.06
Na	— } 3.97	0.03 } 4.06	— } 3.71	0.03 } 3.99	— } 4.06
K	—	—	—	—	—
O	25.00	25.00	25.00	25.00	25.00
$\mathrm{H_2O+}$	1.67	1.66	1.11	1.50	1.26
$\frac{\mathrm{Fe'''}}{\mathrm{Al+Fe'''}}$	0.08	0.08	0.20	0.30	0.38
α	1.710	1.710	1.726	1.729	1.740
β	1.713	1.714	1.735	1.754	1.768
γ	1.719	1.719	1.741	1.776	1.787
$\gamma-\alpha$	0.009	0.009	0.015	0.047	0.047
2V	(+)65°	(+)74°–78°	(−)>50°	(−)73°–74°	(−)74°
X	Colorless	Colorless	Colorless	Pale yellow	Pale yellow
Y	Colorless	Pale yellow	Pale pink	Pale greenish yellow	Greenish yellow
Z	Colorless	Colorless	Colorless	Pale greenish yellow	Pale yellow
$c \wedge X$	<2°	2°	7°	8°	13°
Color to the unaided eye	Pale pink	Pale grey	Straw brown	Yellowish green	Dark green

* The maximum possible error for α, β and γ is ±0.003. 2V was determined by means of the universal stage, using Na-light.

† Analyzed by Y. Seki and Chigusa Kato. No. 3 was quoted from Lapham (1957).

TABLE 3. *X*-RAY POWDER DATA AND LATTICE CONSTANTS OF CLINOZOISITES AND PISTACITES

hkl	1 Clinozoisite (Sasaguri)		2 Clinozoisite (Kanto)		3 Pistacite (Hawleyville)		4 Pistacite (Kanto)		5 Pistacite (Dauphine)	
	d(Å)	I	*d*(Å)	I	*d*(Å)	I	*d*(Å)	I	*d*(Å)	I
100	8.04	6	8.04	10	8.04	10	8.04	10	8.04	10
101 $10\bar{2}$	5.01	30	5.04	30	5.01	25	5.04	35	5.05	25
011	4.72	25	—	—	—	—	—	—	4.79	10
002	—	—	—	—	4.59	5	—	—	4.59	15
200	4.003	25	4.006	45	4.010	25	4.011	45	4.019	50
$20\bar{2}$	—	—	3.974	10	3.980	25	3.990	20	3.992	10
111	3.748	10	3.749	15	3.754	10	—	—	3.767	20
$21\bar{1}$	3.477	15	3.478	40	3.483	35	3.490	35	3.490	30
102	3.396	15	3.398	20	3.398	20	3.407	35	3.401	40
201	3.197	20	3.201	50	3.206	25	3.209	30	3.209	20
003	3.052	10	3.058	15	3.061	10	3.059	30	3.062	20
$30\bar{1}$	2.913	30	2.914	10	2.917	15	2.925	20	2.930	10
112	2.901	30	2.901	35	—	—	2.920	20	2.920	25
$11\bar{3}$	2.889	100	2.891	100	2.895	100	2.902	100	2.900	100
020	2.796	50	2.794	30	2.803	45	2.815	30	2.817	40
211	2.778	15	2.778	25	2.783	10	—	—	2.786	15
021	2.680	50	2.679	60	2.682	50	2.686	50	2.688	70
300	2.671	50	2.673	30	2.677	50	2.678	40	2.679	100
120	2.642	20	2.640	30	2.650	25	2.656	30	2.656	30
$31\bar{1}$	2.590	40	2.593	50	2.596	45	2.597	40	2.599	50
103 202	2.525	30	2.526	40	2.529	25	2.530	30	2.531	40
121	2.451	15	2.453	20	2.452	25	2.460	20	2.460	50
$31\bar{3}$	2.399	20	2.396	40	2.399	30	2.406	30	2.409	40
022	2.389	30	2.389	30	2.394	40	2.400	30	2.401	40
220 004	2.290	30	2.290	10	2.296	15	2.301	10	2.301	10
$22\bar{2}$	2.287	30	2.287	20	2.289	15	2.295	20	2.294	30
122	2.161	30	2.161	20	2.163	25	2.165	10	2.166	30
$12\bar{3}$	2.151	15	2.154	20	2.157	20	2.160	30	2.163	30
014	2.121	10	2.121	20	2.125	10	2.131	20	2.131	10
221	2.106	30	2.107	30	2.115	25	2.116	30	2.117	25
$22\bar{3}$	2.099	30	2.098	30	2.104	25	2.108	40	2.109	25
$41\bar{2}$ 023	2.062	10	2.062	30	2.068	25	2.072	40	2.072	15
203	2.040	10	2.040	15	2.047	10	2.047	20	2.048	20
$41\bar{3}$	2.021	10	—	—	—	—	—	—	2.026	10
400	2.001	25	2.000	10	2.006	10	2.010	10	2.010	15
$30\bar{5}$	1.951	10	1.952	10	—	—	—	—	—	—
213	1.919	10	1.919	30	1.922	5	1.928	30	—	—

(continued on next page)

TABLE 3.—(*continued*)

hkl	1 Clinozoisite (Sasaguri)		2 Clinozoisite (Kanto)		3 Pistacite (Hawleyville)		4 Pistacite (Kanto)		5 Pistacite (Dauphine)	
	d(Å)	I	*d*(Å)	I	*d*(Å)	I	*d*(Å)	I	*d*(Å)	I
123 114 222	1.873	15	1.873	30	1.879	15	1.884	30	—	—
$11\bar{5}$ $12\bar{4}$ $22\bar{4}$	1.866	20	1.867	30	1.870	30	1.876	20	1.876	20
312	1.856	5	1.857	5	1.860	10	1.864	5	1.866	5
$32\bar{4}$	1.772	5	—	—	—	—	—	—	—	—
$50\bar{2}$	1.765	10	—	—	—	—	—	—	—	—
015	1.759	10	—	—	—	—	—	—	1.748	10
132	1.637	30	1.637	30	1.637	35	1.642	20	1.642	30
420	1.628	20	1.633	20	1.634	20	1.634	20	1.639	30
124	1.621	10	1.621	10	1.625	10	1.627	10	1.628	10
$12\bar{5}$	—	—	1.616	10	1.618	5	—	—	—	—
115	1.574	10	1.574	10	1.578	5	1.578	10	1.579	10
$33\bar{2}$ $11\bar{6}$	1.569	10	1.571	5	1.573	10	—	—	1.574	10
a_0	8.87 Å		8.87 Å		8.88 Å		8.89 Å		8.90 Å	
b_0	5.59 Å		5.59 Å		5.61 Å		5.63 Å		5.63 Å	
c_0	10.15 Å		10.15 Å		10.17 Å		10.19 Å		10.20 Å	
β	115°27′		115°27′		115°25′		115°24′		115°24′	

Note: The *d* values are for CuKα_1, so far as the peak for CuKα_1 can be distinguished from that of CuKα_2.

are in harmony with the symmetry of $C_{2h}{}^2 - P_{2_1/m}$. The unit-cell dimensions obtained are in good accord with those given by Gossner and Mussgnug (1930), Bujor (1931), Strunz (1935), and Ito, Morimoto and Sadanaga (1954).

From the data in Table 2 and 3, it is clear that a_0, b_0, and c_0 generally increase with the $Fe^{+3}/(Al+Fe^{+3})$ ratio of the mineral.

Table 4 shows the variations of the unit cell volume and the "packing index" (Fairbairn, 1943) of the clinozoisite-pistacite series calculated from the data given in Tables 2 and 3. The unit cell volume gradually increases with increasing iron content. On the other hand, the packing index generally decreases with increasing iron content.

These data suggest that the variation of the lattice constants may

TABLE 4. UNIT-CELL VOLUMES AND PACKING INDICES OF CLINOZOISITES AND PISTACITES. THE PACKING INDICES WERE CALCULATED ON THE ASSUMPTION THAT ONE MOLECULE OF WATER IS IN A UNIT-CELL

	Clinozoisite (Sasaguri)	Clinozoisite (Kanto)	Pistacite (Hawleyville)	Pistacite (Kanto)	Pistacite (Dauphine)
Unit-cell volume	454.5 Å^3	454.5 Å^3	457.6 Å^3	460.7 Å^3	461.7 Å^3
Packing index	6.14	6.16	6.08	6.06	6.06
	6.15 (average)				

Note: Packing index = ionic volume × 10/unit-cell volume.

show a break at the composition of about 30 per cent Fe^{+3} end-member.* The possible break may be related to that the epidote structure is probably most "stable" or unstrained when the mineral has compositions near 33 per cent Fe^{+3} end-member, as was discussed by Miyashiro and Seki (1958).

DISCRIMINATION OF CLINOZOISITE FROM ZOISITE BY *X*-RAY POWDER DATA

As is well known, clinozoisite and zoisite are very similar to each other in their optical properties, crystal form and other appearances. Especially, so-called β-zoisite, the optical plane of which is parallel to 001 (normal to the cleavage plane), can hardly be distinguished from clinozoisite by optical examination in thin sections. However, zoisite and clinozoisite show distinctive *x*-ray powder patterns.

The present writer obtained, by means of the Philips Geiger counter *x*-ray diffractometer using CuKα-radiation, the *x*-ray powder data of two zoisites. One of them is α-zoisite (optical plane is parallel to 010) from Cummington, Mass., U.S.A., and the other is β-zoisite from Nagatoro, Saitama Prefecture, central Japan. The data thus obtained are shown in Table 5.† About eighty peaks between 10° and 70° in angle 2θ were indexed on the basis of orthorhombic unit cell with the dimensions

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* Such a break was noticed in the unit cell dimensions of nepheline (Smith and Sahama, 1954).

† No reflections with d-spacings of 4.245 Å, 2.459 Å, 2.238 Å, 1.666 Å, 1.544 Å and 1.453 Å were found in these zoisites, though Lapahm (1957) presented these reflections for his Ducktown zoisite. These reflections are probably due to quartz impurities included in the Ducktown zoisite.

TABLE 5. *X*-RAY POWDER DIFFRACTION DATA OF α- AND β-ZOISITES

Indices	α-zoisite from Cummington, Mass., U.S.A.		β-zoisite from Nagatoro, Saitama Pref., Japan	
	d(Å)	Intensity	*d*(Å)	Intensity
200	8.12	20	8.12	25
002	5.018	17	5.023	20
111	4.662	7	4.657	5
400	4.070	36	4.070	58
401	3.757	5	3.755	19
302	3.678	10	3.681	12
311	3.619	15	3.616	18
402	3.151	17	3.155	9
411	3.108	9	3.112	7
501	3.083	23	3.087	24
013	2.864	51	2.868	40
113	2.824	8	2.824	10
020	2.779	23	2.777	22
502	2.722	30	2.722	40
600	2.699	100	2.698	100
220	2.628	10	2.626	13
313	2.533	12	2.531	13
104	2.481	5	2.481	7
122	2.404	12	2.401	14
204	2.398	8	2.397	7
611	2.358	5	2.354	3
413	2.341	7	2.340	8
222	2.331	11	2.327	15
420	2.289	4	2.289	4
304	2.276	7	2.276	6
701	2.255	7	2.255	7
421	2.236	12	2.235	10
404	2.135	4	2.134	7
702 603	2.102	16	2.103	18
521	2.066	20	2.064	18
800	2.026	26	2.025	46
323	1.990	5	1.990	5
504	1.982	17	1.982	18
205	1.975	6	1.975	8
613	1.967	4	1.967	5
522	1.947	10	1.944	8
703	1.905	4	1.903	5
810 621	1.900	3	1.899	3
801	1.878	6	1.879	7
514	1.870	4	1.870	3
124	1.851	10	1.850	11
604	1.838	5	1.839	4
424	1.693	4	1.695	7

TABLE 5.—(*continued*)

Indices	α-zoisite from Cummington, Mass., U.S.A.		β-zoisite from Nagatoro, Saitama Pref., Japan	
	d(Å)	Intensity	*d*(Å)	Intensity
911 430	1.687	7	1.689	6
722 623	1.676	6	1.677	7
006	1.674	9	1.674	8
812	1.654	4	1.654	3
820	1.637	4	—	—
515	1.633	6	1.633	5
714	1.627	4	1.628	5
033 912	1.622	10	1.622	7
10.0.0	1.620	15	1.621	17
524	1.615	16	1.615	18
10.0.1 306 432	1.599	8	1.599	11
116	1.595	5	—	—
531	1.588	6	1.587	7
821 10.1.0.	1.556	10	1.556	10
333	1.552	8	1.552	5
615	1.548	7	1.548	7
10.1.1.	1.537	12	1.537	10
624	1.534	4	1.533	6
715	1.464	6	1.464	7
10.0.3.	1.459	6	1.459	6
107 126	1.429	4	1.429	4
040 732 633	1.390	10	1.390	5
10.2.1.	1.387	8	1.386	7
240	1.371	6	—	—
10.2.2.	1.349	10	1.349	10
141 317	1.345	7	1.345	5
a_0	16.20 Å		16.20 Å	
b_0	5.56 Å		5.56 Å	
c_0	10.04 Å		10.04 Å	

Note: The values of *d* are for $CuK\alpha_1$ so far as the peak for $CuK\alpha_1$ can be distinguished from that for $CuK\alpha_2$. The reflections due to hematite inclusions in the Cummington zoisite were excluded.

shown in the same table. Brazilian quartz was used as the internal standard. These *x*-ray diffraction data of α- and β-zoisites are very similar to each other and are in harmony with the symmetry of V_h^{16}-*Pnma*. These unit cell dimensions are close to those given by Gossner and Mussgnug (1930), Gossner and Reichel (1932), Waldbauer and McCann (1935) and Ito (1950).

The chemical compositions of these zoisites are shown in Table 6. The writer attempted to purify these zoisites by means of the isodynamic separator and Clerici solution, but a small amount of hematite impurity included in the Cummington zoisite could not be removed. Accordingly, the chemical analysis of the Cummington zoisite shows an unusually large content of ferric iron.

TABLE 6. CHEMICAL COMPOSITION AND OPTICAL PROPERTIES† OF α- AND β-ZOISITE

α-zoisite from Cummington, Mass.					β-zoisite from Nagatoro, Saitama Pref., Japan*				
Analysis		Atomic ratio			Analysis		Atomic ratio		
SiO_2	38.97	Si	6.04		SiO_2	37.47	Si	5.78	6.00
Al_2O_3	29.13				Al_2O_3	33.44	Al	0.22	
Fe_2O_3	5.11	Al	5.32	5.98	Fe_2O_3	1.33		5.86	6.02
FeO	0.37	Fe'''	0.60		FeO	0.00	Fe'''	0.15	
MnO	0.10	Fe''	0.05		MnO	0.04	Fe''	—	
MgO	tr.	Mn	0.01		MgO	0.00	Mn	0.01	
CaO	24.01	Mg	—		CaO	24.52	Mg	—	
Na_2O	0.00	Ca	3.97	3.97	Na_2O	0.00	Ca	4.04	4.04
K_2O	n.d.	Na	—		K_2O	n.d.	Na	—	
H_2O+	2.26	K	—		H_2O+	2.73	K	—	
H_2O-	0.12	O	25.00		H_2O-	0.05	O	25.00	
Total	100.07	H_2O+	1.14		Total	99.58	H_2O+	1.40	
α		1.702			1.694				
β		1.707			1.698				
γ		1.714			1.707				
$\gamma-\alpha$		0.012			0.013				
(+)2V		72°–87°			39°–49°				
Color in thin section		Colorless			Colorless				
Color to the unaided eye		Pale grey			Colorless				

† The maximum possible error for α, β and γ is ±0.003; 2V was determined by means of the universal stage, using Na-light.

* Analyzed by Y. Seki and Chigusa Kato. The high Fe_2O_3 content of the Cummington zoisite is due to hematite inclusions.

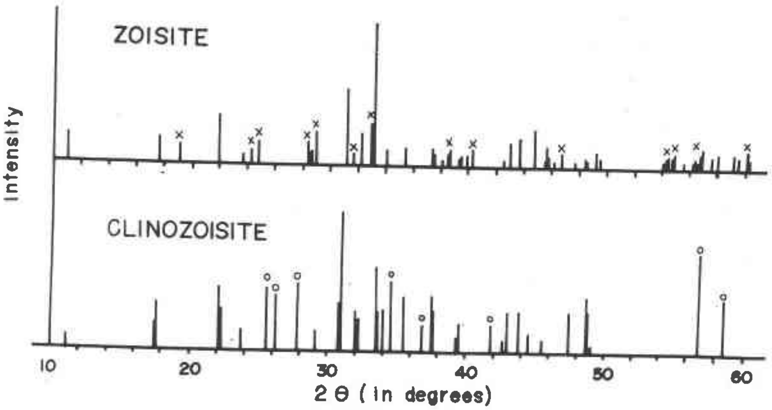

FIG. 1. Comparison of the x-ray powder patterns of zoisite and clinozoisite (see Tables 3 and 4).
x = Peaks characteristic of zoisite.
o = Peaks characteristic of clinozoisite.

The x-ray powder patterns of clinozoisite and zoisite are diagrammatically compared in Fig. 1. The difference between them is very clear.

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