

# Chromian muscovites and epidotes from Outokumpu, Finland

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## Abstract

The Outokumpu region of Finland is notable for the large number of Cr-rich silicate and oxide phases associated with the massive sulphide deposits. In this paper analyses of Cr-rich phases in the epidote-clinozoisite and muscovite groups are presented. The muscovite-type phase shows a maximum 80% substitution of Cr for Al on octahedral sites with a maximum 24.7 wt.% Cr<sub>2</sub>O<sub>3</sub>. Apparent complete solid solution between this extreme composition and the fuchsitic muscovites is indicated. The clinozoisite type phase has a maximum 15.4 wt.% Cr<sub>2</sub>O<sub>3</sub>, with a composition close to Ca<sub>2</sub>CrAl<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>(OH). As with the micas, complete solid solution of Cr for Al in the clinozoisite structure is indicated.

KEYWORDS: chromium, muscovite, epidote, clinozoisite, Outokumpu, Finland

## Introduction

MANY of the major rock-forming silicate minerals appear to be capable of accommodating substantial substitution of Cr into their crystal structures. While Burns and Strens (1967), Seward (1971) and Burns and Burns (1975) imply that such substitution is often at the expense of octahedrally coordinated Al and Fe<sup>3+</sup>, the degree to which it may extend has rarely been demonstrated. Although complete substitution of Cr for Al may be possible in some phases, such as uvarovite garnet, there may be a physical limit in others. Grapes (1981), for instance, suggested that for epidote the maximum attainable ratio of Cr:Al may be 1:2, although whether this would reflect the strong octahedral M1 site preference of Cr is unclear. As Cr-rich silicate phases are unusual, reported natural examples are few, with the result that data pertaining to the degree of possible Cr for Al substitution in many of them are not available. Analytical data are presented here for two phases, chromian muscovite and chromian epidote, which imply that for both minerals, maximum levels of Cr substitution may be far greater, especially at low pressure, than hitherto suspected.

The Outokumpu district of Finnish Karelia (Fig. 1) is characterized by a number of Cu-Co-Zn-bearing stratiform massive sulphide bodies. These bodies are unique amongst massive sul-

phides in that both they and their host rocks contain anomalously high Cr contents. These high levels of Cr stabilize a wide range of unusual Cr-rich phases, including uvarovite, staurolite, diopside, tremolite, epidote, biotite, muscovite and chlorite together with spinel and eskolaite (Eskola, 1933; Kuovo and Vuorelainen, 1958; Weiser, 1967; Peltola *et al.*, 1968; Vuorelainen *et al.*, 1968; Treloar *et al.*, 1981). Some of these phases, such as diopside and tremolite, are generally confined to calc-silicate horizons. Others, such as staurolite, are restricted to metamorphosed stockwork horizons in the footwall of the massive sulphide bodies (Treloar *et al.*, 1981), while others, in particular epidote, mica and tourmaline, are extensively developed within the quartzite horizons within which the main ore bodies are hosted (Koistinen, 1981). The Cr minerals reflect the metamorphism of variously Cr-enriched rocks during the Svecokarelian orogeny. Metamorphic conditions of about 3.5 ± 1 kbar and 600 ± 50 °C have been estimated for this event (Treloar *et al.*, 1981).

## Petrography

Cr-bearing muscovites have been analysed from two samples, numbers 146093 and 146094 (Harker Collection, Dept. of Earth Sciences, Cambridge University). The former is from underground at the Keretti Mine 4 km SW of Outokumpu, while the latter is from the Vuonos open pit some 4 km NE of Outokumpu. Both sam-

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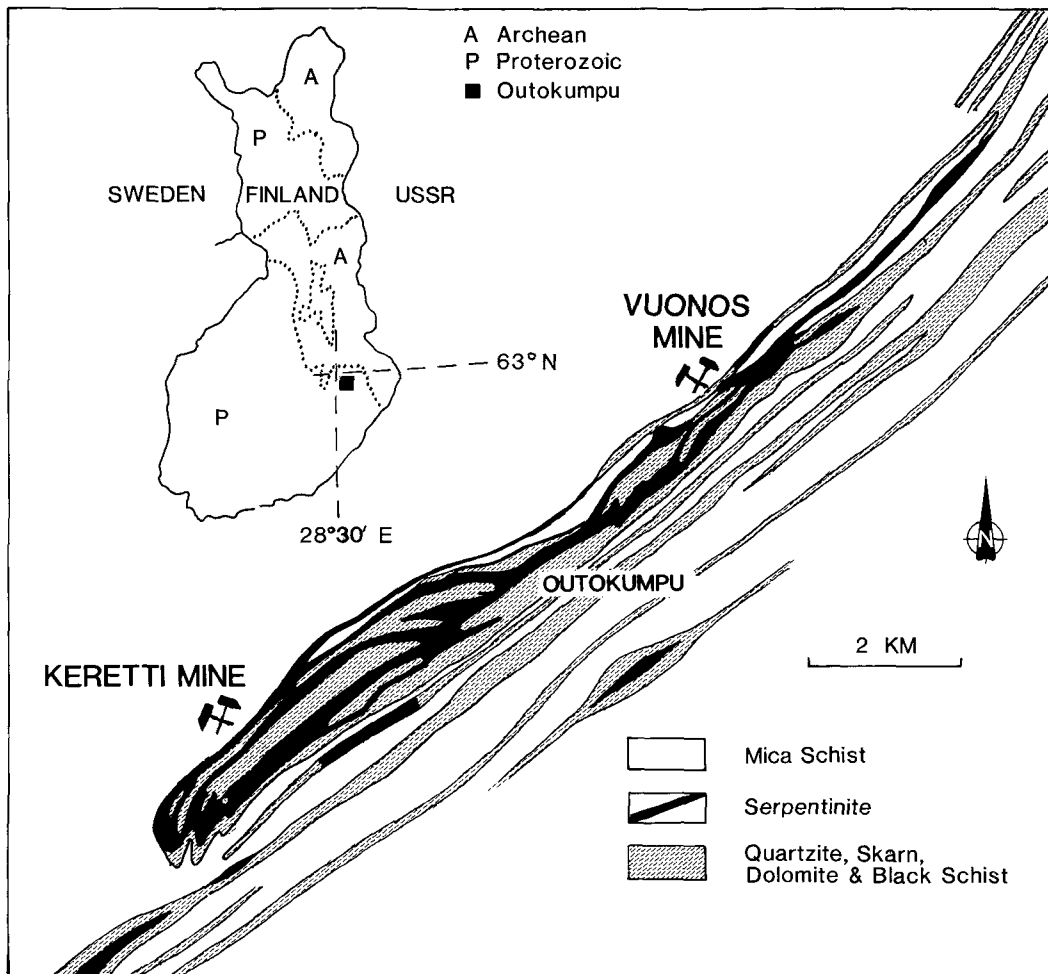


FIG. 1. Geological map of Outokumpu showing the location of the Vuonos and Keretti Mines. The location of Outokumpu is indicated on the inset map of Finland.

ples are quartzites composed predominantly of fine-grained quartz and contain isolated crystals of uvarovite and chlorite and up to 5% pyrrhotite. Mica comprises about 5% by volume normally occurring as small flakes up to 0.25 mm in length. The micas are bright green and pleochroic.

Cr-bearing epidotes have been analysed from three samples, 146090 from Suuri Kokkalampi, 23 km N of Outokumpu, and 146091 and 146092 from the Vuonos open pit. All samples are quartzites and contain minor quantities of epidote and uvarovite, with tremolite, chlorite and chromite in sample 146090, chromite and pyrrhotite in sample 146091 and diopside and tremolite in sample 146092. Epidote constitutes less than 3% of each sample occurring either as individual grains or in

clusters. Grain size is normally less than 0.5 mm although a few crystals may be as large as 2 mm in diameter. The epidotes normally form euhedral crystals (Fig. 2), the cores of which are often extensively altered to a complex hydrated Cr-Mg-Al-Fe-Ca-bearing silicate, the composition of which approaches a hydrogarnet type stoichiometry. The degree of hydration varies from slight to extreme. They are generally pleochroic pale yellow to pale green.

#### Chromian muscovite

Analyses of chromian muscovite are listed in Table 1. Values of  $\text{Cr}_2\text{O}_3$  range between 7.6% and 24.7% corresponding to 0.406 to 1.451 Cr

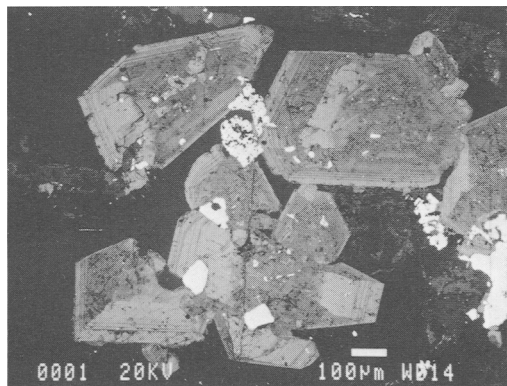


FIG. 2. Back-scattered electron image of epidote crystals, sample 146091.

atoms per formula unit (11 oxygens). Assuming that all Cr is accommodated in octahedral sites (Burns and Burns, 1975; Faye, 1968), this implies a maximum of 75% occupancy of octahedral sites by Cr. The maximum  $\text{Cr}/(\text{Cr} + \text{Al} + \text{Fe}^{3+})$  value recorded is 0.84. Variations in octahedral substitutions of Al, Cr and  $\text{Fe}^{3+}$  are shown in Fig. 3. This plot shows evidence for extensive solid solution between octahedral Al and Cr between Al:Cr ratios of 20:80 through to ratios of 80:20. The gap in the plot at Al:Cr of 30:70 probably arises from the sampling process rather than reflecting a miscibility gap. Other operative substitutions indicated by the analyses are of V for octahedral Al and the presence of Al in the tetrahedral site. The substitution of Si by Al (phengite) may reach 22% but shows no correlation with the amount of Cr. Up to 1.7%  $\text{V}_2\text{O}_3$  (0.081 atoms per formula unit) is present in micas from sample 146094.

### Chromian epidote

Analyses of chromian epidote-type phases are listed in Table 2. Values of  $\text{Cr}_2\text{O}_3$  range between 5.7 and 15.4 wt.% corresponding to the range 0.710 to 1.901 Cr atoms per formula unit (25 oxygens). The upper values are considerably in excess of the 6.79%  $\text{Cr}_2\text{O}_3$  reported from one analysed epidote from the same area by Eskola (1933). Octahedral sites in these analysed epidotes are largely occupied by Al and Cr with only a limited amount of  $\text{Fe}^{3+}$  present. Values of  $\text{Fe}^{3+}/(\text{Fe}^{3+} + \text{Cr} + \text{Al} + \text{V})$  never exceed 0.03. Cr:Al ratios range from 13:87 to 34:66 and, as with the mica analyses, there is no evidence of any miscibility gap (Fig. 4). Some of the large epidote crystals are chemically zoned with an

oscillatory face parallel zonation imposed on a sector zonation (Fig. 5). Within these zones Al and Ca are negatively correlated with Cr, Mg and Fe. As in the micas vanadium is also present in the epidotes at the further expense of aluminium. Up to 2.4 wt.%  $\text{V}_2\text{O}_3$ , corresponding to 0.239 atoms per formula unit, is present in epidote from sample 146092. This represents a significant trend towards the solid solution end-member mukhinite (Shepel and Karpenko, 1970) in the analysed phases.

### Discussion

The degree of Cr substitution reported here for two common silicate phases raises problems of nomenclature. Cr-bearing muscovites have been widely reported in the literature though never yet with Cr values in excess of 5%  $\text{Cr}_2\text{O}_3$  (4.7%, Whitmore *et al.*, 1946; 3.6%, Schreyer *et al.*, 1981; 3.3%, Fabries and Latouche, 1973; 2.5%, Chen and Lee, 1974). Deer *et al.* (1962) recommend that the term 'fuchsite' be used only for muscovites with greater than 1%  $\text{Cr}_2\text{O}_3$  and, as most 'fuchsitic' muscovites seem to have  $\text{Cr}_2\text{O}_3$  contents of between 1 and 5 wt.%, this is not a severe restriction. It may be debatable, however, as to whether it is wise to use the term fuchsite to cover a range in  $\text{Cr}_2\text{O}_3$  content that extends from 1 to 24 wt.%.

Cr-bearing epidotes have been occasionally recorded in the literature (Eskola, 1933; Grapes, 1981; Ward, 1984). Of these, the most chromian epidote reported is from Burma with 11.16%  $\text{Cr}_2\text{O}_3$  (Eskola, 1933) while the most chromian epidote previously recorded at Outokumpu contains 6.8%  $\text{Cr}_2\text{O}_3$  (Eskola, 1933). Neither of these two analyses (Table 3) are rigorously acceptable, the excess of Si in both and of Ca in that from Burma possibly reflecting difficulties in obtaining pure mineral separates. That from Outokumpu also contains 4.41%  $\text{H}_2\text{O}$  and this may well be a function of the common if variable hydration of epidote phases mentioned earlier.

Solid solution chemical variations in the clinzoisite-epidote series are normally restricted to substitutions for Al in two of the three crystallographically different octahedral sites in the clinzoisite structure. Mössbauer spectral analysis and site occupancy refinement (Dollase, 1971) and polarized absorption spectroscopy (Burns and Strens, 1967) show strong site preferences for substitutions of Cr, Fe and Mn for Al. The  $M_2$  site contains only Al atoms with the other octahedrally co-ordinated atoms substituting entirely onto the  $M_3$  and  $M_1$  sites (Dollase, 1971). Cr has a strong preference for the  $M_1$  site (Burns and

Table 1. Microprobe analyses of chromian muscovites. All iron as Fe<sup>3+</sup>. Formulae calculated on the basis of 11 Oxygens. All analyses carried out on the energy dispersive electron microprobe in the Department of Earth Sciences University of Cambridge using operating conditions as described in Treloar *et al* (1981)

Sample	146093	-	-	-	-	-	-	-	-	-	-	-	-	-
SiO <sub>2</sub>	43.17	42.22	43.63	42.73	44.02	45.45	45.03	43.62	42.20	42.60	45.66	44.99	44.96	44.26
TiO <sub>2</sub>		0.12			0.24	0.13			0.23	0.21	0.28	0.41	0.27	
Al <sub>2</sub> O <sub>3</sub>	12.55	11.79	13.38	14.12	14.50	21.34	20.85	19.48	24.46	22.48	24.07	22.32	22.28	24.23
Cr <sub>2</sub> O <sub>3</sub>	24.63	23.62	23.86	23.43	23.12	18.81	17.76	15.15	14.99	13.28	12.13	11.08	11.00	10.15
Fe <sub>2</sub> O <sub>3</sub>	0.37	1.04	2.32	2.26	2.30	0.30	0.18	0.24	0.21	0.84	0.41	0.29	0.37	0.46
V <sub>2</sub> O <sub>3</sub>	0.17	0.24	0.24	0.19	0.13							0.13		
MgO	1.83	2.15	0.53	0.44	0.77	0.31	0.72	5.88		1.27	1.91	1.99	3.69	2.68
MnO				0.17						0.13	0.12		0.12	
CaO	0.10	0.11		0.11	0.09	0.09	0.11	0.15	0.10	0.10				0.14
K <sub>2</sub> O	10.02	8.82	9.10	9.07	9.39	9.79	10.18	10.12	10.11	9.42	10.17	9.99	10.06	10.01
Total	92.84	90.11	93.06	92.52	94.56	96.22	94.83	94.64	92.30	90.33	94.75	91.20	92.75	91.93
Si	3.218	3.228	3.227	3.183	3.198	3.157	3.176	3.088	3.040	3.120	3.165	3.234	3.185	3.150
Al	0.782	0.772	0.773	0.817	0.802	0.843	0.824	0.912	0.960	0.880	0.835	0.764	0.815	0.850
Al	0.321	0.291	0.394	0.423	0.440	0.910	0.915	0.718	1.124	1.067	1.138	1.127	1.051	1.189
Cr	1.451	1.428	1.395	1.380	1.328	1.033	0.991	0.848	0.854	0.769	0.665	0.630	0.616	0.571
Fe	0.021	0.060	0.129	0.126	0.125	0.016	0.010	0.013	0.011	0.046	0.021	0.016	0.020	0.025
V	0.010	0.015	0.014	0.011	0.008							0.007		
Ti		0.007			0.013	0.007			0.012	0.012	0.015	0.022	0.014	
Mg	0.203	0.245	0.058	0.049	0.083	0.032	0.076	0.620		0.139	0.197	0.213	0.390	0.284
Mn				0.011						0.008	0.007		0.007	
Ca	0.008	0.009		0.009	0.007	0.007	0.008	0.011	0.008	0.008				0.011
K	0.953	0.860	0.859	0.862	0.870	0.868	0.916	0.914	0.929	0.880	0.899	0.916	0.909	0.909
Tot	6.966	6.913	6.850	6.870	6.873	6.871	6.915	7.125	6.939	6.929	6.942	6.930	7.007	7.989
Sample	146093	-	-	-	-	-	-	-	146094	-	-	-	-	-
SiO <sub>2</sub>	46.08	45.34	46.49	46.05	46.67	47.02	47.76	44.92	44.41	43.87	43.95	44.51	47.70	
TiO <sub>2</sub>	0.15	0.41		0.46				0.31	0.43	0.37	0.42	0.49	0.36	
Al <sub>2</sub> O <sub>3</sub>	24.48	27.07	23.56	26.71	25.57	30.10	27.17	21.15	22.17	20.86	21.11	21.99	21.10	
Cr <sub>2</sub> O <sub>3</sub>	10.12	10.01	9.80	9.50	9.35	8.95	7.67	11.13	10.72	10.60	10.31	10.39	9.67	
Fe <sub>2</sub> O <sub>3</sub>	0.64		0.31		0.17	0.30	0.34	0.37	0.17	2.36	0.39	0.21	1.62	
V <sub>2</sub> O <sub>3</sub>								1.57	1.60	1.32	1.65	1.72	1.46	
MgO	3.61	0.87	3.94	0.79	1.80	0.32	2.79	3.06	2.67	4.91	3.27	3.10	4.94	
MnO	0.18					0.14		0.12						
CaO	0.12	0.10	0.08	0.18	0.16	0.12	0.13	0.10				0.13	0.15	
K <sub>2</sub> O	10.37	10.12	10.21	9.87	10.86	10.89	10.79	9.97	9.81	8.79	9.31	9.79	9.11	
Total	95.75	93.92	94.39	93.56	94.58	97.84	96.65	92.70	91.98	93.08	90.41	92.33	96.10	
Si	3.153	3.136	3.213	3.184	3.214	3.115	3.197	3.202	3.178	3.115	3.195	3.174	3.246	
Al	0.847	0.864	0.787	0.816	0.786	0.885	0.803	0.798	0.822	0.835	0.805	0.826	0.754	
Al	1.133	1.350	1.138	1.368	1.296	1.473	1.347	1.059	1.048	0.861	1.104	1.022	0.939	
Cr	0.548	0.548	0.536	0.520	0.509	0.469	0.406	0.627	0.606	0.595	0.592	0.586	0.520	
Fe	0.033		0.016		0.009	0.015	0.017	0.020	0.009	0.126	0.021	0.011	0.083	
V								0.090	0.092	0.075	0.096	0.098	0.080	
Ti	0.008	0.021		0.024				0.017	0.023	0.020	0.023	0.026	0.018	
Mg	0.368	0.090	0.406	0.081	0.185	0.032	0.278	0.325	0.285	0.520	0.334	0.329	0.501	
Mn	0.010					0.008		0.007						
Ca	0.009	0.007	0.006	0.013	0.012	0.009	0.009	0.008				0.010	0.011	
K	0.905	0.893	0.900	0.871	0.954	0.921	0.921	0.907	0.896	0.796	0.863	0.891	0.791	
Tot	7.014	6.910	7.001	6.877	6.965	6.926	6.979	6.978	6.958	6.992	6.954	6.974	6.943	

CHROMIAN MUSCOVITES AND EPIDOTES

Table 2. Microprobe analyses of chromian epidotes. All iron as Fe<sup>3+</sup>. Formulae calculated on the basis of 25 Oxygens. Analytical conditions as in Table 1.

Sample	146090	-	-	-	-	-	-	-	-	-	-	-	-
SiO <sub>2</sub>	37.23	38.19	36.61	37.95	38.01	37.07	36.86	37.55	36.73	38.03	37.76	37.98	38.02
TiO <sub>2</sub>	0.15			0.17	0.16		0.17			0.15	0.12	0.12	
Al <sub>2</sub> O <sub>3</sub>	20.26	20.41	20.30	21.23	21.39	20.54	20.86	21.32	21.23	21.78	22.26	22.11	24.27
Cr <sub>2</sub> O <sub>3</sub>	15.37	15.12	14.66	13.66	13.74	13.29	13.35	13.25	12.66	13.00	12.73	12.33	10.04
Fe <sub>2</sub> O <sub>3</sub>	0.48	0.50	0.57	0.57	0.76	1.31	0.95	0.88	1.14	0.92	0.66	0.68	0.48
V <sub>2</sub> O <sub>3</sub>	0.32	0.20	0.27	0.30	0.33	0.45		0.31	0.45	0.25	0.21	0.31	0.41
MgO									0.23				
MnO	0.11			0.18	0.16	0.24	0.18		0.21	0.15	0.12	0.12	0.14
CaO	23.16	23.55	23.15	23.49	23.78	22.01	23.28	23.41	22.17	23.38	23.43	23.37	23.63
Total	97.08	97.97	95.56	97.55	98.28	94.91	95.75	96.72	94.82	97.66	97.29	97.02	96.99
Si	6.021	6.103	6.011	6.073	6.043	6.096	6.020	6.058	6.037	6.066	6.037	6.081	6.041
Al	3.863	3.845	3.929	4.005	4.009	3.982	4.028	4.047	4.113	4.096	4.196	4.173	4.546
Cr	1.965	1.910	1.903	1.723	1.727	1.728	1.725	1.690	1.645	1.639	1.609	1.561	1.261
Fe	0.058	0.060	0.068	0.068	0.090	0.161	0.117	0.106	0.140	0.110	0.079	0.082	0.057
V	0.041	0.026	0.036	0.038	0.042	0.059		0.040	0.059	0.032	0.027	0.040	0.052
Ti	0.018			0.020	0.019		0.021			0.018	0.014	0.014	
Mg									0.056				
Mn	0.015			0.024	0.022	0.033	0.025		0.029	0.020	0.016	0.016	0.019
Ca	4.014	4.033	4.073	4.028	4.051	3.878	4.092	4.047	3.904	3.996	4.014	4.009	4.023
Tot	15.996	15.977	16.021	15.986	16.003	15.939	16.028	15.996	15.984	15.977	15.993	15.977	16.000
X <sub>Cr</sub>	.330	.327	.321	.294	.292	.290	.292	.287	.272	.277	.271	.265	.212

Sample	146091	-	-	-	-	-	146092	-	-	-	
SiO <sub>2</sub>	38.77	39.05	38.31	38.72	38.59	37.90	40.12	38.39	38.05	38.84	38.14
TiO <sub>2</sub>	0.13	0.15	0.16	0.19	0.17						
Al <sub>2</sub> O <sub>3</sub>	24.32	24.22	24.76	25.28	25.03	25.53	26.32	24.32	25.36	26.31	26.60
Cr <sub>2</sub> O <sub>3</sub>	10.83	9.60	9.18	8.83	8.67	8.60	8.21	8.78	6.82	6.20	5.72
Fe <sub>2</sub> O <sub>3</sub>	0.65	0.94	0.99	0.85	1.03	1.01	1.16	1.31	0.54	0.44	0.52
V <sub>2</sub> O <sub>3</sub>	0.20	0.23	0.21	0.20	0.18	0.22	0.22	0.46	2.30	1.86	1.65
MgO									0.27	0.23	0.24
MnO		0.18	0.12		0.12		0.12			0.13	0.11
CaO	23.59	23.59	23.86	23.87	24.08	23.91	24.13	24.29	23.56	24.44	23.69
Total	98.49	97.96	97.59	97.94	97.87	97.17	100.28	97.55	96.90	98.45	96.67
Si	6.064	6.128	6.040	6.062	6.060	5.991	6.114	6.065	6.019	6.034	6.016
Al	4.485	4.481	4.602	4.666	4.632	4.757	4.728	4.529	4.729	4.818	4.946
Cr	1.339	1.191	1.144	1.093	1.076	1.075	0.989	1.097	0.853	0.761	0.713
Fe	0.077	0.111	0.117	0.100	0.122	0.120	0.132	0.155	0.064	0.051	0.061
V	0.025	0.029	0.027	0.025	0.023	0.028	0.027	0.058	0.292	0.232	0.209
Ti	0.015	0.024	0.019	0.022	0.020						
Mg									0.063	0.053	0.056
Mn		0.024	0.016		0.016		0.015			0.017	0.015
Ca	3.954	3.967	4.031	4.005	4.052	4.050	3.940	4.112	3.993	4.068	4.004
Tot	15.958	15.948	15.996	15.973	15.996	16.020	15.947	16.016	16.013	16.035	16.020
X <sub>Cr</sub>	.225	.203	.193	.185	.183	.180	.168	.188	.142	.128	.119
X <sub>V</sub>								.010	.049	.039	.035

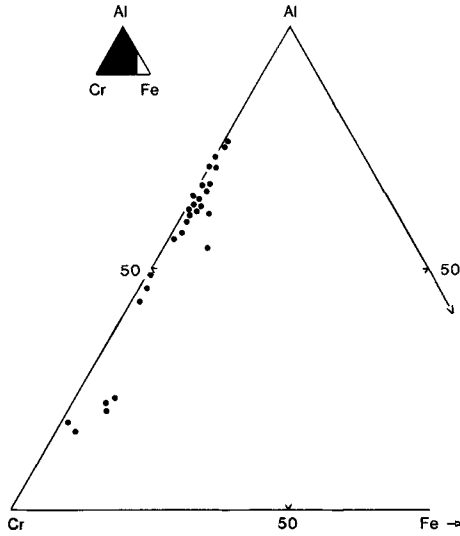


FIG. 3 (left). Plot of octahedral Cr vs. Al vs. Fe in chromian muscovites.

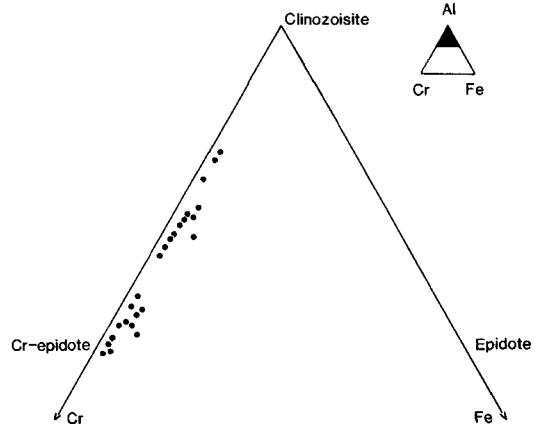


FIG. 4 (right). Plot of Cr vs. Al vs. Fe in chromian epidotes.

Strens, 1967) while Fe, V and Mn show strong preferences for the distorted  $M3$  site. In this context the composition of the 'epidote' end member,  $\text{Ca}_2\text{FeAl}_2\text{Si}_3\text{O}_{12}(\text{OH})$ , of the epidote-clinozoisite solid-solution series with  $X_{\text{Fe}^{3+}}^{\text{oct}} = 0.33$  may be significant in terms of the complete occupancy of  $M3$  sites by  $\text{Fe}^{3+}$ .

In the Cr-epidotes analysed from Outokumpu the observed limit of solid solution is  $X_{\text{Cr}}^{\text{oct}} = 0.33$ . This suggests that Cr may be preferentially partitioned into one of the three octahedral sites, probably  $M1$  (Burns and Strens, 1967). In this case, the component  $\text{Ca}_2\text{CrAl}_2\text{Si}_3\text{O}_{12}(\text{OH})$  may have the same sort of crystal chemical significance within the clinozoisite-Cr-epidote solid solution series as the 'epidote' component appears to have in the clinozoisite-epidote solid solution series.

The term tawmawite has long been applied to the Burmese Cr-rich epidote and the status of this term should be clarified. The analysis of the type tawmawite is a poor one (Table 3) probably reflecting the presence of impurities. As it stands the term applies to Cr-rich epidotes within the solid-solution series between  $X_{\text{Cr}}^{\text{oct}} = 0.00$  and  $0.33$ . In the light of the analyses presented here it may be best for the term tawmawite to be redefined as that epidote family group mineral with  $X_{\text{Cr}}^{\text{oct}} = 0.33$  and  $X_{\text{Al}}^{\text{oct}} = 0.67$ , and a composition of  $\text{Ca}_2\text{CrAl}_2\text{Si}_3\text{O}_{12}(\text{OH})$ .

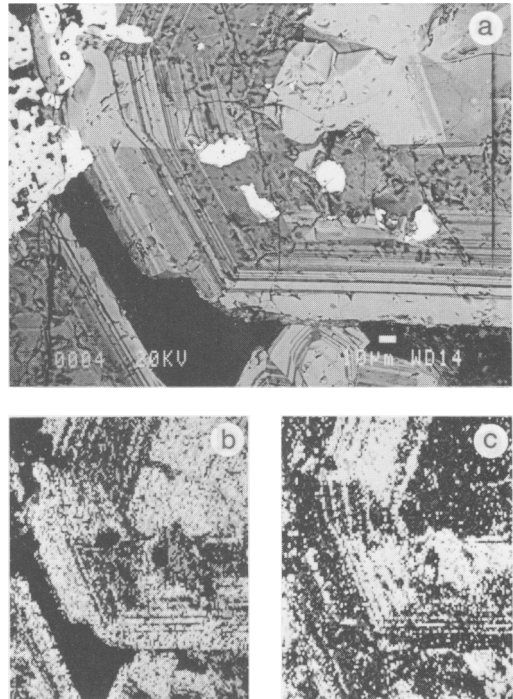


FIG. 5. (a) BSE image showing compositional zonation within a single epidot crystal. Note both a sector zonation and a face parallel oscillatory zonation. (b) and (c) Al and Mg compositional maps of the same crystal as in 5a.

Table 3. Tawmawite analyses from 1) Keretti Mine, Outokumpu, and 2) Tawmoro, Burma (Eskola 1933). Formulae calculated on the anhydrous basis of 25 oxygens per formula unit.

	1	2
SiO <sub>2</sub>	39.26	37.92
Al <sub>2</sub> O <sub>3</sub>	24.38	12.83
Fe <sub>2</sub> O <sub>3</sub>	4.18	9.93
Cr <sub>2</sub> O <sub>3</sub>	6.79	11.16
CaO	20.09	25.35
MgO	0.53	-
H <sub>2</sub> O	4.41	2.32
	99.64	99.57
Si	6.263	6.314
Al	4.587	2.520
Fe	0.502	1.246
Cr	0.857	1.470
Ca	3.433	4.523
Mg	0.126	-
	15.768	16.072

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