

COMPILATION OF CHEMICAL ANALYSES AND PHYSICAL CONSTANTS OF NATURAL CORDIERITES

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ABSTRACT

A brief review of previous attempts to correlate the composition and optical properties of cordierite is given, followed by a compilation, including four new analyses, of 62 chemical analyses of natural cordierites together with their refractive indices, optic angles and densities.

INTRODUCTION

The relationship between the chemical composition of cordierite and its optical properties and density has been considered by a number of workers, particularly Oppenheimer (1914, p. 257), Pehrman (1932, p. 1), Shibata (1936, p. 205), Winchell (1937, p. 1175), Thiele (1940, p. 64), Folinsbee (1941, p. 485) and Iiyama (1956, p. 372).

The first notable plot relating α , β and γ , 2V, D and

$$\frac{\text{FeO} + 2\text{Fe}_2\text{O}_3}{\text{FeO} + 2\text{Fe}_2\text{O}_3 + \text{MgO}}$$

was drawn by Shibata (1936, p. 223).^{*} Many subsequent diagrams have essentially been modifications of Shibata's diagram.

Early attempts at correlation were frustrated by two main difficulties, one of which is still a considerable drawback. The first problem was the quite unpredictable variation of 2V, which provoked much comment. Thus Brammall & Rao (1936, p. 257) showed that cordierite from the Dartmoor Granite varied in 2V α from 56°–72° while Tsuboi (1936, p. 337) found that the cordierite in a single rock had a 2V variation from positive near 0° to negative near 0° with most grains having a 2V near 90°. Although most cordierites are negative, a number of well established optically positive cordierites are known (Stewart, 1942, p. 262; Heald, 1950, p. 66; Tilley, 1940, p. 340; Pehrman 1932, p. 5; Mahadevan and Sastry, 1948, p. 363). The second difficulty is the relatively limited range of Fe/Mg variation in common cordierites. Until Shibata (1936, p. 209 and 217) had two mixtures of quartz and iron rich cordierite analyzed there was no information about cordierite richer than 50% of the Fe end member.[†] Both these analyzed mixtures contained more than 50%

^{*} Shibata actually used the

$$\frac{\text{FeO} + 2\text{Fe}_2\text{O}_3 + \text{Na}_2\text{O}}{\text{FeO} + 2\text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{MgO}}$$

ratio but the Na₂O is usually negligible.

[†] Read (1929, p. 550) calculated the composition of an Fe rich cordierite from an

quartz and the density and chemical composition had to be calculated from planimetric estimations of the volume of quartz in the cordierite-quartz mixture. Even now, over 20 years later, there is only one more analysis available of an Fe rich cordierite, that by Sambonsugi, quoted by Iiyama (1956, p. 374) and Miyashiro (1957, p. 47). Thus with the exception of these three cordierites, all from Japanese pegmatites, chemical and optical determinations of cordierite cover only the range of 10–54% Fe cordierite, at present.

Winchell (1937, p. 1176) replotted Shibata's data after replacing Na_2O by MnO in the chemical ratio, which then became the same as Niggli's *mg* ratio, $\text{MgO}/\text{MgO}+\text{FeO}+\text{MnO}+2\text{Fe}_2\text{O}_3$ and will be referred to as such below. Winchell's diagram was of uncertain value because "the lack of correlation between chemical composition and optic properties is illustrated by the fact that there are almost as many (seven) samples of cordierite which have been analysed and measured optically that cannot be used in the diagram as the number (eight) used in preparing it" (Winchell, 1937, p. 1178). Neither Winchell nor Shibata considered Pehrman's (1932) data but both showed that despite a poor correlation of refractive indices with Fe/Mg the density increased as the Fe/Mg ratio increased and that when the density was plotted against the refringence a very good correlation was obtained.

In 1940 Thiele (p. 80) made a notable contribution to the data by supplying seven new analyses and he considered that the effect of increase in water content and of substitution of Si and Mg for Al was to decrease the refractive indices and increase the optic angle. Folinsbee (1941, p. 498) rejected these suggestions and proposed that the 2V was related to the alkali content on the grounds that the structures of beryl and cordierite are very similar and it is known that the alkali content has an appreciable effect on the optical properties of beryl. Folinsbee plotted 2V against "the effective alkali content" ($\text{Na}_2\text{O}+\text{K}_2\text{O}-\text{CaO}$ wt.%) and obtained a smooth curve on the basis of six points. As Heald (1950, p. 67) and Subramaniam (1956, p. 346) have pointed out, many of the analyses published since 1941 do not plot anywhere near to this curve, nor do all the pre-1941 data.

In 1942 Winchell (p. 346) slightly modified his earlier plot of β and D against Fe/Mg while the current edition of Winchell (1951, p. 471) contains a re-modified plot of density, refractive indices and Fe/Mg. Tröger (1952, p. 52) has reproduced a graph nearly identical with the

analysis of a cordierite-plagioclase mixture but he did not give the refractive indices or density. Because of its unusually Fe rich composition a re-examination of this cordierite is needed.

original Winchell-Shibata diagram but he does not give the evidence upon which his plot is based.

Since Folinsbee's work (*ibid*) little further discussion occurred until lately. Miyashiro in a series of very important papers (principally 1955 and 1957) has concluded (1957, p. 60) that there are at least six polymorphs of cordierite compositions. The different structural states are dominantly controlled by different thermal histories and they show gradations from one to the other, having different order \rightleftharpoons disorder states (as do the alkali feldspars). Most of these different structural states are poorly understood. However, "most cordierites, if not all, in ordinary metamorphic rocks, pegmatites and quartz veins belong to the low sub-distortional class, pseudohexagonal and unstable at all temperatures" (Miyashiro, 1957, p. 60.) However, this cordierite, known as low cordierite, has a considerable degree of possible structural order \rightleftharpoons disorder variation and clearly this affects the refractive indices slightly and the optic angle very considerably. Thus it is not surprising that the correlation of composition with refringence and $2V$ has not been very successful and Iiyama (1956, p. 380) and Miyashiro (1957, p. 48) have shown that $2V$ is largely but not entirely controlled by the "distortion index" ($=\Delta$) which is obtained from an x -ray diffractogram while β is lower for a given Fe/Mg ratio the higher the distortion index. This index, which is zero for hexagonal indialite and roughly indicates a measure of deviation from hexagonal symmetry, decreases with increase of disorder and of temperature of crystallization but Iiyama (1956, p. 372) and Miyashiro (1957, p. 53) have suggested that cordierites from volcanic rocks have lower refractive indices than cordierites from other natural environments because they form a separate group of cordierites known as high (temperature) cordierite. At present, however, there are only two chemical and optical analyses of volcanic cordierites available and the accuracy of one of these (No. 57, Table 1) is uncertain.*

Heating low temperature cordierite lowers its refractive indices and Iiyama (1956, p. 387) has recommended that the most accurate determination of Fe/Mg without chemical or x -ray analysis is to be obtained by heating a given cordierite for 10 mins. at 1000°C . cooling and then determining the refractive indices and obtaining Fe/Mg from a graph which has been drawn from similarly treated and chemically analyzed natural cordierites. The heating is intended to ensure that the cordierite sample is in a similar structural state (*i.e.* having a similar distortion

* Shand, 1943, *Am. Mineral.*, **28**, p. 393 gives an analysis of an iron-free cordierite (which has 1.24% CaO) from the wall of a glass furnace tank and this high temperature form has very low refractive indices, $\alpha=1.5223$, $\gamma=1.5270$, $2V\alpha=82\frac{1}{2}^{\circ}$, $D=2.520$ at 20°C .

index) to the cordierites used in preparing the graph. At present this plot is based on the data of 8 cordierites.

Iiyama (1956, p. 387) has pointed out that the approximate composition of a cordierite can also be obtained by measurement of $d\theta_{004}$. He suggests that if $2\theta_{004}$ can be measured to $\pm 0.02^\circ$ then $\text{Fe}^{2+}\text{Mn}/\text{Fe}^{2+} + \text{Mn} + \text{Mg}$ can be estimated to $\pm 10\%$ and if the refractive indices can be measured to ± 0.003 and the sample has previously been given the standard heat treatment then the ratio can be estimated to $\pm 5\text{--}8\%$, whereas the accuracy when the refractive indices of unheated cordierite are determined "depends greatly on that of the estimated Δ value." There is, however, no published plot, or list, of all the available data on natural cordierite and so it is difficult to know with what accuracy Fe/Mg can be estimated from unheated samples. In the light of current developments in our understanding of cordierite it is now certain that all really precise cordierite studies must include x-ray analysis. Nevertheless much could be achieved if all the available information on the optics and density of cordierites was fully utilized and so this paper gives a collection of data on natural cordierite (Table 1 and Figs. 1 to 6) including four unpublished analyses (Nos. 32, 50, 55 and 59).

No compilation of the published data is or has been available. Oppenheimer (1914, p. 258) gave a comprehensive list of references to the early literature dealing with cordierite optics and chemistry and he tabulated most of the refractive indices then determined, but only about half a dozen of the chemical analyses are given. Winchell's (1938, p. 1179) list of references is the most comprehensive available at present and Folinsbee (1941, p. 493) supplements this but Winchell did not reproduce the original data in his paper, which is in any case, 20 years old now.

DISCUSSION

The enigmatic role of water in cordierite has been recognized for some time (Gossner and Reindl, 1932, p. 330; Winchell, 1937, p. 1179; Thiele 1940, p. 84; Tilley, 1940, p. 341). Recently Iiyama (1958, p. 796), on the basis of thermobalance experiments on the Laramie Range cordierite No. 2 has claimed that the decrease in refractive indices and increase in $2V$ which occurs when low temperature cordierite is heated and transformed to high temperature cordierite is related to loss of water, the transformation taking place at about 525°C . at atmospheric pressure but at 750°C . when the water vapor pressure is about 600 bars.* An approximate estimate of the correlation between water content and refractive index in natural cordierites was obtained from the data in Table 1 thus. If

* Cf. Thiele, 1940, p. 84 who concluded that increase in water content decreased refraction.

TABLE 1. CHEMICAL ANALYSES, SPECIFIC GRAVITIES AND REFRACTIVE INDICES OF 62 CORDIERITES

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
SiO ₂	49.46	47.09	50.15	49.54	49.18	48.52	48.05	49.17	50.65	49.12	47.96	46.08	49.05	49.60	48.11	48.04	49.47	47.43	48.33	49.50	46.75
Al ₂ O ₃	33.58	34.86	33.07	33.42	32.28	35.25	33.37	33.10	30.26	32.25	31.52	32.85	33.08	31.19	33.02	33.41	33.18	33.34	32.31	33.01	33.64
TiO ₂	0.01	0.03	0.38	0.03	n.d.	tr.	n.d.	n.d.	n.d.	n.d.	0.00	0.05	n.d.	n.d.	tr.	0.06	n.d.	0.03	n.d.	n.d.	0.00
Fe ₂ O ₃	0.14	0.11	1.52	0.14	0.08	0.41	0.09	n.d.	n.d.	n.d.	1.03	0.12	0.83	1.19	0.81	0.01	1.15	0.07	2.22	0.38	0.72
FeO	2.12	2.59	2.22	3.75	4.05	3.43	4.01	4.34	4.10	5.09	3.24	4.08	4.38	4.10	4.49	5.04	4.53	4.91	3.74	5.12	4.89
MgO	12.06	11.92	11.01	11.34	11.25	10.40	11.11	11.45	11.09	11.96	12.16	9.56	11.04	11.13	10.95	10.32	10.80	9.70	10.66	10.42	10.43
CaO	0.03	0.04	0.29	0.10	n.d.	tr.	0.05	n.d.	n.d.	tr.	0.00	1.32	n.d.	0.23	tr.	0.04	n.d.	0.04	0.84	n.d.	0.04
Na ₂ O	0.14	0.78	0.14	0.32	0.21	0.13	0.26	n.d.	n.d.	n.d.	0.33	0.62	n.d.	0.21	0.12	1.12	0.17	0.61	n.d.	n.d.	0.24
K ₂ O	0.30	tr.	0.08	0.01	0.44	0.16	0.39	n.d.	n.d.	n.d.	tr.	0.49	n.d.	0.06	tr.	0.30	0.04	tr.	n.d.	n.d.	0.56
MnO	0.08	0.05	0.12	0.09	n.d.	0.08	tr.	0.04	0.60	tr.	1.09	0.21	n.d.	n.d.	0.07	0.20	n.d.	0.12	n.d.	0.29	0.31
H ₂ O ⁺	1.71	2.21	1.37	1.32	2.04	2.06	2.75	1.20	2.66	0.32	2.80	4.88	1.64	2.33	2.72	1.81	1.16	2.74	1.44	1.62	2.63
H ₂ O ⁻	0.10	0.65	0.09	0.03	0.27	0.12								0.08	0.11	0.23	0.04	1.01		0.10	
	99.73	100.33	100.44	100.10	99.80	100.57	100.08	99.30	100.00	98.74	100.13	100.26	100.02	⁹⁹ 100.22 ¹⁰⁰ 100.16 ¹⁰⁰	100.58	100.54	100.54	100.00	99.54	100.34	100.33
Si	4.99	4.80	5.02	4.98	5.02	4.91	4.93	4.98	5.21	4.99	4.93	4.87	4.97	5.07	4.93	4.91	4.97	4.92	4.94	5.02	4.81
Al	1.01	1.20	0.98	1.02	0.98	1.09	1.07	1.02	0.79	1.01	1.07	1.13	1.03	0.93	1.07	1.09	1.03	1.08	1.06	0.98	1.19
Al	2.98	2.99	2.92	2.94	2.91	3.07	2.96	2.94	2.87	2.85	2.75	2.96	2.92	2.83	2.91	2.93	2.90	3.00	2.83	2.96	2.93
Ti	0.00	0.00	0.03	0.00	0.00	0.00	0.00	0.00	0.05	0.00	0.00	0.01	n.d.	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe ³	0.01	0.01	0.11	0.01	0.01	0.03	0.01	0.03	0.35	0.43	0.08	0.01	0.06	0.09	0.06	0.00	0.08	0.01	0.17	0.03	0.05
Fe ²	0.18	0.22	0.19	0.32	0.34	0.29	0.34	0.37	0.05	0.00	0.28	0.35	0.37	0.35	0.38	0.43	0.38	0.43	0.32	0.43	0.42
Mn	0.01	0.00	0.01	0.01	0.01	0.01	0.00	0.00	0.05	0.00	0.10	0.02	n.d.	0.01	0.01	0.02	0.01	0.01	0.02	0.03	0.03
Mg	1.81	1.80	1.64	1.70	1.71	1.57	1.67	1.73	1.70	1.81	1.86	1.51	1.67	1.69	1.68	1.57	1.62	1.50	1.62	1.57	1.60
Ca	0.00	0.01	0.03	0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.15	0.15	0.02	0.00	0.00	0.04	0.00	0.09	0.00	0.00
Na	0.02	0.16	0.03	0.06	0.04	0.01	0.05				0.07	0.13		0.04	0.02	0.22	0.04	0.12		0.05	0.05
K	0.04	0.00	0.01	0.00	0.06	0.01	0.05				0.00	0.06		0.01	0.00	0.04	0.01	0.00		0.07	0.07
Fe ³ +Mn×100	9.1	11.0	10.9	15.9	16.7	15.8	16.9	17.5	19.1	19.3	17.0	20.2	18.2	17.2	18.8	22.2	19.0	22.6	16.5	22.3	21.9
Fe ³ +Mn+Mg	2.583-		2.588	2.591	2.581		2.581	2.60	2.60	2.59			2.593	2.598	2.589		2.581		2.71	2.607	
D	2.803																				
mg	0.906	0.889	0.840	0.838	0.830	0.830	0.829	0.825	0.809	0.807	0.805	0.795	0.794	0.793	0.788	0.778	0.777	0.771	0.767	0.765	0.761
α	1.550	1.534	1.527	1.539	1.536	1.534	1.536				1.534	1.541	1.539	1.541	1.542	1.539	1.533	1.538	1.540	1.542	1.542
β	1.535	1.539	1.532	1.544	1.541	1.541	1.541				1.543	1.550	1.547	1.545	1.548	1.547	1.541	1.542	1.546	1.546	1.546
γ	1.538	1.543	1.538	1.546	1.543	1.545	1.543				1.543	1.550	1.547	1.547	1.548	1.547	1.541	1.554	1.548	1.548	1.551
2Vα	84½	77	92	66	69½	89	65½				104	71½	70	78½	74	68	80	72	106	65	68

	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42
SiO ₂	49.14	48.58	48.65	48.44	48.32	49.51	50.13	54.63	49.15	48.61	49.58	48.17	48.77	46.99	48.85	48.37	48.15	47.33	48.43	47.69	48.19
Al ₂ O ₃	32.84	31.47	32.58	31.74	35.02	32.46	32.11	29.32	34.60	31.97	32.71	32.53	31.01	32.72	32.52	29.22	30.90	32.59	32.36	32.52	33.45
TiO ₂	n.d.	n.d.	n.d.	tr.	n.d.	0.00	0.00	0.00	n.d.	n.d.	0.00	0.00	0.07	0.10	n.d.	n.d.	tr.	0.15	n.d.	tr.	0.01
Fe ₂ O ₃	0.63	1.85	1.75	1.29	n.d.	0.29	2.24	1.13	1.40	1.45	0.00	1.21	2.83	0.16	2.94	2.20	0.75	0.15	n.d.	0.63	0.55
FeO	5.04	4.90	4.79	4.63	5.76	6.51	6.37	5.15	4.25	7.10	7.26	6.63	5.19	7.12	5.28	7.07	7.75	8.49	8.55	8.04	8.40
MgO	10.40	10.68	10.02	9.39	8.83	9.60	8.47	7.82	7.68	9.95	8.65	8.86	8.91	8.38	8.50	9.54	8.50	8.07	7.81	7.56	7.95
CaO	n.d.	tr.	n.d.	0.70	0.08	0.00	n.d.	0.00	0.55	0.44	0.00	tr.	0.43	0.06	0.43	1.92	0.35	0.09	0.46	0.52	0.17
Na ₂ O	n.d.	0.35	0.15	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.24	0.24	1.39	0.16	0.85	n.d.	0.40	0.53	n.d.	n.d.	0.53	0.22
K ₂ O	n.d.	n.d.	0.04	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.09	0.09	0.50	0.06	0.30	n.d.	n.d.	tr.	0.48	n.d.	0.42	0.02
MnO	0.19	n.d.	n.d.	0.27	0.12	0.10	0.32	0.02	0.94	tr.	0.00	n.d.	0.31	0.42	0.28	0.42	0.06	0.06	n.d.	0.18	0.18
H ₂ O ⁺	1.84	1.82	2.39	3.32	1.73	1.07	0.99	0.82	2.15	1.12	1.31	1.12	1.99	3.07	1.82	1.84	3.35	1.88	1.55	1.85	0.67
H ₂ O ⁻		0.14	0.10	0.20	0.30							0.10	0.29	0.05				0.20	1.32	0.55	0.01
	100.08	99.79	100.26	99.98	100.16	99.54	100.63	99.89	100.72	100.64	99.84	100.51	100.02	100.22	100.62	100.58	100.21	99.94	100.48	100.35	99.82
Si	5.00	4.99	4.97	5.03	4.92	5.04	5.08	5.52	4.99	4.95	5.07	4.94	5.04	4.92	5.00	5.02	5.04	4.94	5.03	4.99	4.95
Al	1.00	1.01	1.03	0.97	1.08	0.96	0.92	0.48	1.01	1.05	0.93	1.06	0.96	1.08	1.00	0.98	0.96	1.06	0.97	1.01	1.05
Al	2.93	2.80	2.89	2.82	3.13	2.93	2.91	3.00	3.13	2.79	3.01	2.87	2.81	2.95	2.91	2.60	2.86	2.95	2.99	3.00	3.00
Ti				0.00		0.00		0.00			0.00		0.01	0.01		0.00	0.00	0.00	0.00	0.00	0.00
Fe ³	0.05	0.14	0.13	0.10		0.02	0.17	0.08	0.10	0.11	0.00	0.09	0.22	0.01	0.22	0.17	0.06	0.01		0.05	0.04
Fe ²	0.43	0.42	0.41	0.40	0.49	0.55	0.54	0.44	0.36	0.61	0.62	0.57	0.44	0.62	0.45	0.61	0.68	0.74	0.74	0.70	0.72
Mn	0.02			0.02	0.01	0.01	0.02	0.00	0.08	0.00	0.00	0.00	0.02	0.04	0.02	0.04	0.01	0.01	0.00	0.00	0.02
Mg	1.58	1.64	1.52	1.46	1.33	1.46	1.28	1.18	1.16	1.51	1.31	1.35	1.37	1.31	1.30	1.48	1.33	1.25	1.21	1.18	1.21
Ca		0.00		0.08	0.01	0.00		0.00	0.06	0.05	0.00	0.00	0.05	0.01	0.04	0.21	0.04	0.01	0.06	0.06	0.02
Na		0.07	0.03								0.05	0.28	0.03	0.17			0.08	0.11		0.05	0.04
K			0.01								0.01	0.06	0.01	0.04			0.00	0.06		0.03	0.00
Fe ² +Mn×100	22.0	20.4	21.3	22.7	27.3	27.8	30.7	27.0	27.5	28.7	31.9	29.5	25.6	33.6	27.0	30.4	34.0	37.5	38.0	37.2	37.7
Fe ² +Mn+Mg																					
D	2.610	2.603	2.614	2.632	2.603	2.588	2.686		2.615	2.60		2.660	2.629		2.620	2.598		2.63	2.642	2.64	2.631
mg	0.760	0.744	0.736	0.735	0.727	0.713	0.698	0.694	0.680	0.680	0.680	0.674	0.666	0.660	0.650	0.644	0.641	0.623	0.620	0.609	0.607
α		1.540	1.541	1.536		1.537	1.540	1.540	1.530	1.538	1.530	1.538	1.546	1.546	1.551		1.537	1.543	1.548	1.538	1.544
β		1.546	1.547	1.543		1.543	1.545	1.545	1.534	1.534	1.544	1.560	1.560	1.560	1.560		1.549	1.555	1.548	1.550	1.550
γ		1.549	1.550	1.546		1.548	1.552	1.552	1.537	1.537	1.547	1.561	1.558	1.562		1.546	1.546	1.555	1.559	1.547	1.556
2Va		65‡	66‡	68‡		81‡		77		81	75-81	43‡			42	85-99		100	53	96	85-95
											av. 78										

(Continued on next page)

TABLE 1 (continued)

	43	44	45	46	47	48	49	50	51	52	53	54	55	56	57	58	59	60	61	62
SiO ₂	40.92	48.88	48.43	48.6	48.09	48.40	48.18	47.18	49.14	49.50	50.09	47.88	47.32	49.62	48.58	49.74	46.69	44.64	43.55	43.27
Al ₂ O ₃	31.06	33.07	32.36	30.5	30.87	32.15	33.86	32.10	30.99	32.45	31.78	32.40	33.36	28.72	32.44	35.21	32.00	29.96	30.68	30.25
TiO ₂	n.d.	n.d.	n.d.	n.d.	n.d.	0.05	n.d.	0.16	tr.	0.06	0.00	0.00	0.00	n.d.	n.d.	n.d.	0.34	n.d.	tr.	tr.
Fe ₂ O ₃	n.d.	0.53	n.d.	n.d.	2.63	0.00	0.34	0.28	2.49	0.40	0.78	0.64	0.96	n.d.	3.15	5.65	0.39	2.72	1.50	1.08
FeO	7.99	8.05	8.55	10.7	7.08	9.80	9.34	9.26	7.03	8.90	8.71	9.65	9.23	11.58	9.17	3.00	12.04	11.02	13.17	13.31
MgO	6.71	7.04	7.81	8.2	7.82	7.25	7.11	6.99	7.38	6.73	6.69	6.94	6.89	8.64	6.63	4.30	5.91	3.08	2.70	1.48
CaO	tr.	0.04	0.46	n.d.	0.80	tr.	n.d.	0.16	0.28	0.57	0.00	0.08	0.04	0.23	n.d.	1.05	0.18	0.02	0.16	0.10
Na ₂ O	0.72	0.09	n.d.	n.d.	n.d.	0.07	n.d.	0.65	0.10	0.46	0.00	0.23	0.43	n.d.	n.d.	n.d.	0.28	2.05	1.82	3.53
K ₂ O	8.60	n.d.	n.d.	n.d.	n.d.	0.39	n.d.	0.59	n.d.	0.39	0.07	0.07	0.19	n.d.	n.d.	n.d.	0.16	0.75	0.32	0.90
MnO	n.d.	0.06	1.32	0.1	1.22	0.23	0.11	0.19	1.01	0.11	0.00	0.10	0.26	1.51	tr.	n.d.	0.09	1.86	2.88	1.20
H ₂ CO ⁺	3.22	2.21	1.55	1.5	2.31	1.58	1.30	1.97	1.69	0.62	1.43	1.92	1.42	n.d.	n.d.	1.74	1.95	3.85	3.72	2.88
H ₂ O ⁻		0.20			0.16				n.d.	0.00		0.24	tr.							
	99.22	100.17	100.48	99.8	100.62	100.11	100.24	99.53	100.11	100.19	99.55	100.15	100.40	100.30	99.97	100.69	100.03	99.95	100.00	100.00
Si	4.62	5.05	5.03	5.08	5.00	5.03	4.95	4.96	5.09	5.07	5.17	5.00	4.90	5.14	4.98	5.03	4.93	4.93	4.83	4.83
Al	1.38	0.95	0.97	0.92	1.00	0.97	1.05	1.04	0.91	0.93	0.83	1.00	1.10	0.86	1.02	0.97	1.07	1.07	1.17	1.17
Al	2.75	3.07	2.99	2.83	2.78	2.97	3.05	2.94	2.87	2.99	3.03	2.98	2.97	2.64	2.90	3.23	2.92	2.82	2.81	2.81
Ti					0.00			0.01	0.00	0.00	0.00	0.00	0.00				0.03	0.00	0.04	0.00
Fe ³⁺	0.75	0.04			0.21	0.00	0.03	0.02	0.19	0.03	0.06	0.05	0.07		0.24	0.43	0.03	0.23	0.13	0.27
Fe ²⁺		0.70	0.74	0.93	0.62	0.85	0.80	0.81	0.61	0.76	0.75	0.84	0.80	1.00	0.79	0.25	1.06	1.02	1.22	1.43
Mn		0.01		0.01	0.11	0.02	0.01	0.01	0.09	0.01	0.00	0.01	0.03	0.13	0.00	0.01	0.01	0.17	0.27	0.11
Mg	1.13	1.08	1.21	1.27	1.21	1.12	1.09	1.10	1.14	1.03	1.03	1.08	1.07	1.33	1.01	0.65	0.93	0.50	0.45	0.25
Ca	0.00	0.01	0.05		0.07	0.00		0.04	0.03	0.06	0.00	0.01	0.01	0.02		0.19	0.02	0.00	0.02	0.01
Na	0.16	0.02			0.08	0.08		0.13	0.02	0.09	0.00	0.05	0.09				0.05	0.44	0.28	0.76
K	1.23				0.01			0.08		0.05	0.01	0.01	0.02				0.02	0.11	0.05	0.13
Fe ³⁺ +Mn×100	40.1	39.4	41.4	42.5	37.4	43.6	42.9	43.1	38.0	42.6	42.1	44.1	43.6	46.0	43.9	27.8	53.5	70.3	76.7	86.1
Fe ²⁺ +Mn+Mg																				
D	2.77	2.647	2.642	2.64	2.636	2.66	2.023		2.651	2.622	2.650			2.65	2.627	2.64			2.72	2.75
mg	0.599	0.594	0.586	0.575	0.566	0.564	0.564	0.562	0.561	0.560	0.559	0.546	0.543	0.540	0.494	0.488	0.458	0.263	0.216	0.128
α		1.549				1.543	1.536	1.543	1.550	1.543	1.543	1.547	1.544				1.548	1.560	1.555	1.568
β						1.533	1.539	1.549		1.550	1.548	1.552	1.550		1.544		1.554	1.574	1.566	1.568
γ		1.560				1.555	1.543	1.552	1.563	1.557	1.553	1.557	1.555				1.558	1.578	1.571	1.573
2Vα		74½				71½	100	74-81		81	91½	77-86	76		86		72-78	55	66	68½
								av. 79									av. 76			

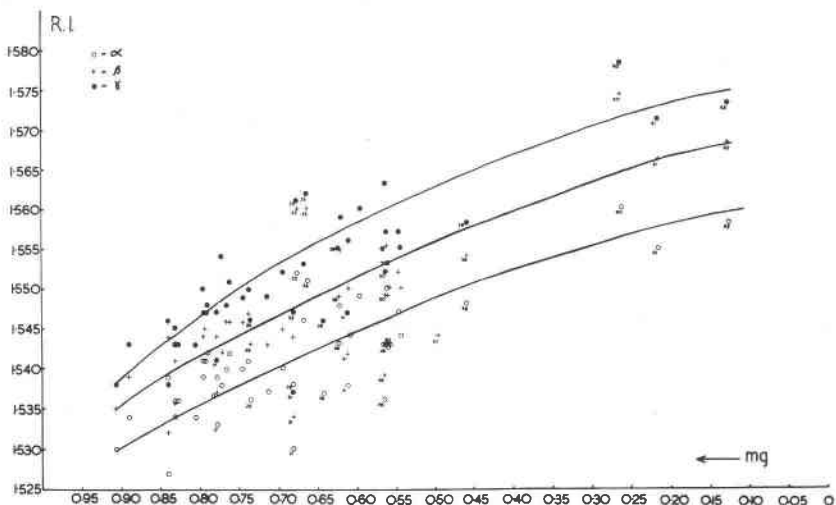


FIG. 1. Plot of refractive indices of natural cordierites against Niggli mg ratio.

cordierites rich in water have higher refringences than those with lower water contents but similar mg ratios then the hydrous cordierites ought to lie above the curves in Fig. 1 and the lower water-bearing cordierites ought to lie below. The average water content (1.94%) of 36 analyzed cordierites, with mg between 0.9 and 0.5, accompanied by refractive indices was obtained (No. 31 was excluded). The differences, positive or negative, from this average, of each of the water contents of the 36 cordierites was calculated. The mean refractive index

$$= \left(\frac{\alpha + \beta + \gamma}{3} \quad \text{or} \quad \frac{\alpha + \gamma}{2} \right)$$

of each of the cordierites was obtained and the difference between this value and the mean value

$$\left(\frac{\alpha + \beta + \gamma}{3} \right)$$

obtained from the curves for α , β and γ in Fig. 1, using the known mg value was calculated. The resulting correlation coefficient between the water differences and the refractive index differences was equal to +0.37, which is not a good correlation coefficient but since Δ has not been considered, may be significant, suggesting that increase in water content tends to increase refractive index. Chambers (1952, p. 62) gives $\gamma = 0.325$ for 37 observations when $P = 0.05$.

In view of the possibility that volcanic cordierites have lower refrac-

tive indices than other natural cordierites, in both Figs. 1 and 2* the numbers of samples with particularly low refractive indices have been inserted. It is notable that in addition to the volcanic cordierites No. 31 from Kasyo-to, Formosa and No. 57 from Cabo de Gata, Spain, Nos. 3, 41 and 49 also have particularly low refractive indices for their *mg* values although No. 3 from an anthophyllite gneiss, Attu, Finland, No. 41 from a hornfels at Belhelvie, Scotland and No. 49 from a cordierite-biotite-microcline-andesine pegmatite at Degero, Finland, are unlikely to be

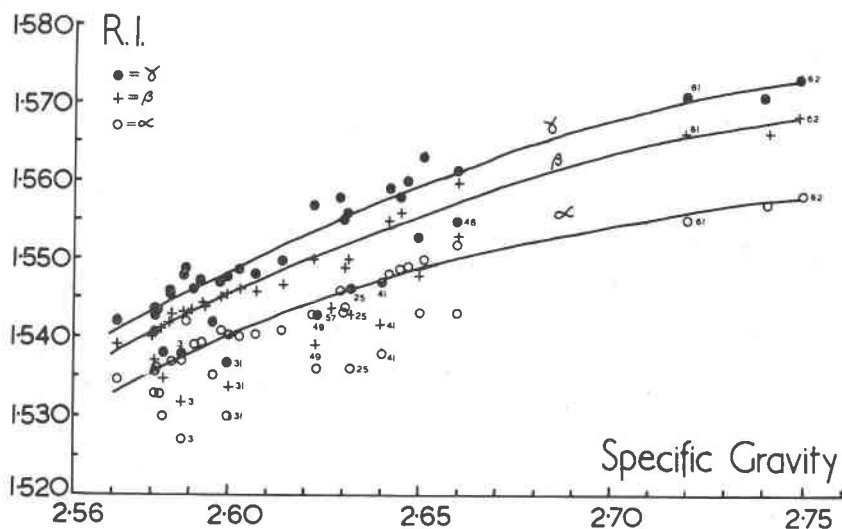


FIG. 2. Plot of specific gravities of natural cordierites against refractive indices.

particularly high temperature cordierites. Although these last three cordierites, with 1.37, 1.85 and 1.30% H_2O respectively, have less than the average water content their deficiency in water is not extreme; many other cordierites have much less water and so water deficiency cannot explain their extremely low refringence. It is possible that these three cordierites are exceptionally ordered and have very high Δ values. It is also of interest that Oppenheimer (1914, p. 299) gives for an unanalyzed cordierite from Cabo de Gata, Spain, $\alpha = 1.5346$, $\beta = 1.5391$, $\gamma = 1.5432$, $2V = 85\frac{1}{2}^\circ$ (presumably negative), $D = 2.571$ which, although β is lower than Osann's (1888, p. 704) determination ($\beta = 1.5438$) on Cabo de Gata cordierite, plots a little above the curves in Fig. 2. Oppenheimer's den-

* For economy of space the data used in compiling Figs. 2 and 6 which do not appear in Table 1 are not reproduced. Most of the information is contained in Shibata, 1936, p. 420.

sity is suspiciously low for cordierite. It is important that a reexamination of the Cabo de Gata cordierite be made for Osann's analysis is over 70 years old, its calcium and water contents were never determined and yet it is one of only two volcanic cordierite analyses available.

Figs. 3 and 4 show great scatter and there is only a very poor correlation of $2V_\alpha$ with either mg or $\text{Na}_2\text{O} + \text{K}_2\text{O} - \text{CaO}$. With the exception of Nos. 19, 28 and 43 which undoubtedly have erroneous density determinations, a good correlation of mg and density is shown by Fig. 5,

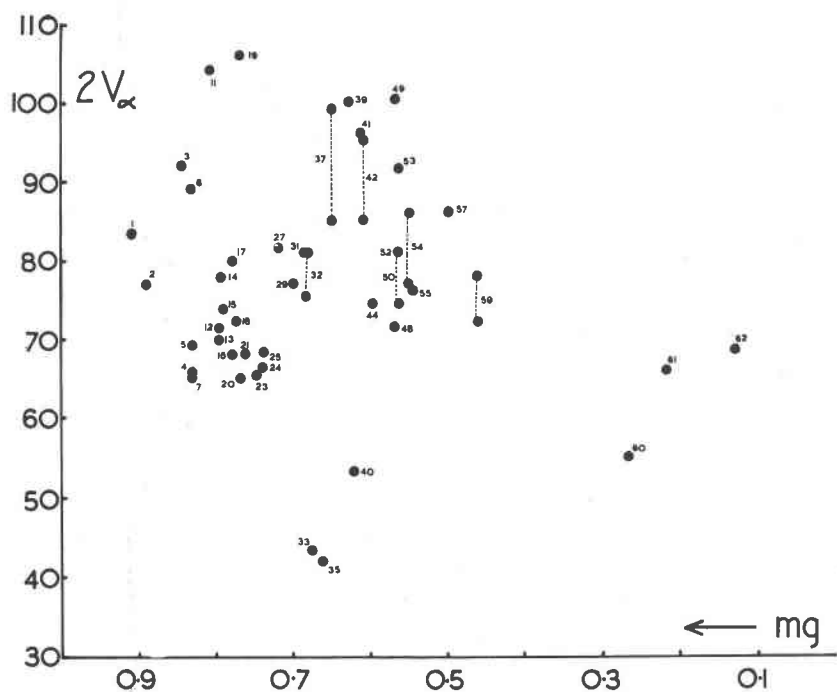


FIG. 3. Plot of $2V_\alpha$ of natural cordierites against Niggli mg ratio.

whereas the plots of mg versus refringence (Fig. 1) show that little reliability can be attached to the drawn curves. Figure 2 (density versus refringence), however, shows rather less scatter of the points from the drawn curves.

This apparent better correlation between density and refringence (Fig. 2) compared with mg and refringence (Fig. 1) is puzzling. There are several possible explanations, the most obvious one being that density has, on the whole, been determined more accurately than Fe/Mg , though proof of this is difficult. Winchell (1937, p. 1178) suggested that

the better correlation of density and refringence resulted from the fact that density could be determined on the same crystal as was used for the refractive index determinations whereas the chemical analysis was an average of all the cordierites in a rock. However, it seems very doubtful whether more than an insignificant proportion of the actual density and refractive index results have been determined on the same grains. Iiyama (1956, p. 389) has indicated that with increase in disorder in cordierite density increases and so does refringence so that some of the better correlation might be the result of this relationship, particularly over the

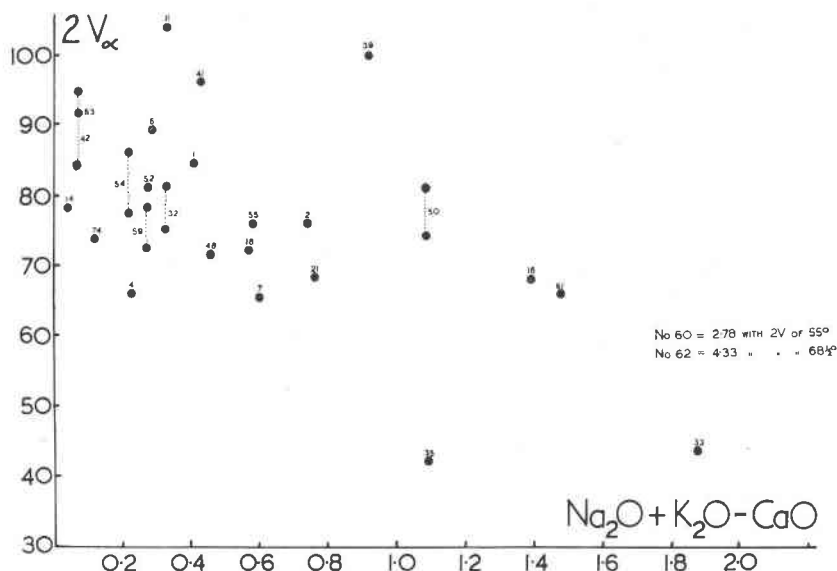
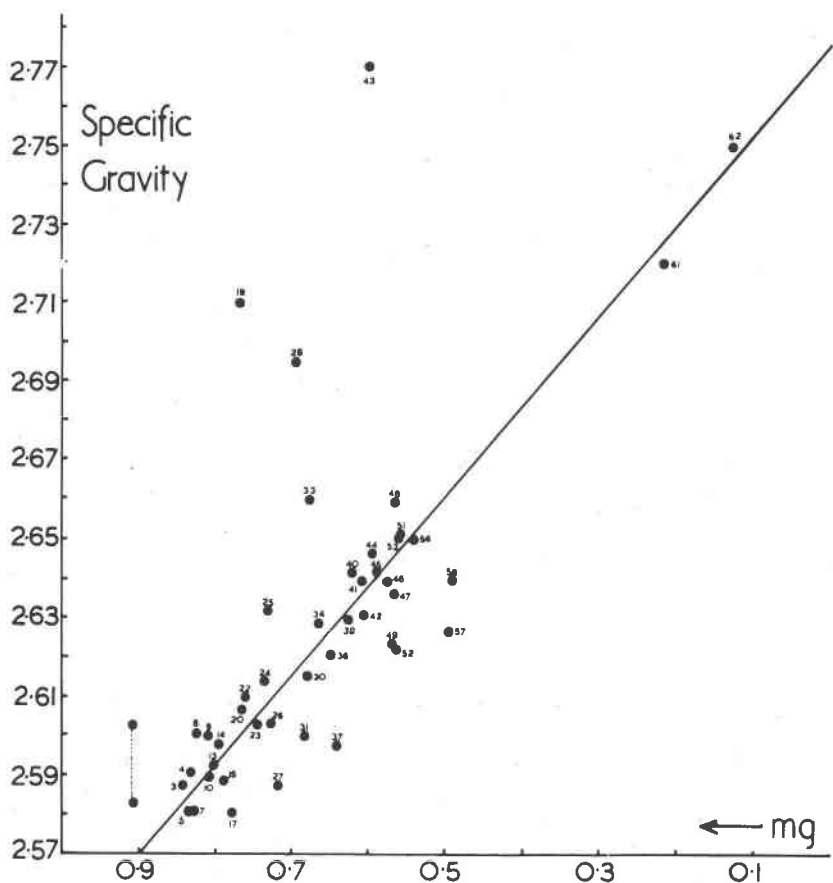
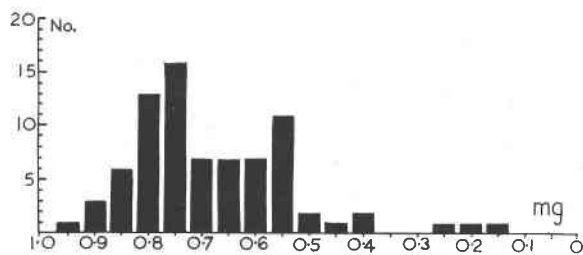


FIG. 4. Plot of $2V\alpha$ of natural cordierites against "effective alkali content" = $\text{Na}_2\text{O} + \text{K}_2\text{O} - \text{CaO}$ wt. %.

density range 2.58–2.61 (Fig. 2). If density does decrease with increase in Δ then volcanic cordierites, which have high Δ values, ought to plot below the line in Fig. 5 and in fact both No. 30 and No. 53 fall well below the line.

Following a suggestion kindly made by Dr. M. H. Hey a plot of the calculated volume of the unit cell (in cubic Ångströms) against mg was made for 34 cordierites. For constant composition this volume ought to depend only on the degree of order-disorder in each individual cordierite. But the two volcanic cordierites, 30 and 53 do not support the view that there is any difference between the cell volumes of differently

FIG. 5. Plot of specific gravities of natural cordierites against Niggli *mg* ratio.FIG. 6. Histogram to show frequency of occurrence of *mg* values of 79 natural cordierites. Class interval 0.05 *mg*.

ordered cordierites as they plot in the middle of a frequency distribution (whose maximum is about 3.9×10^{-22} cubic Å) and the scatter of which seems to be largely the result of scattered density determinations. Such a plot relies unduly on the accuracy of the density determination and cordierites which plot away from the frequency distribution also show excessive scatter in Fig. 5. Dr. M. H. Hey also suggested a correlation of specific refractivity $(n-1)/d$ with *mg* but a plot of 30 cordierites showed that outstanding points did not belong to cordierites with unusual chemistry e.g. H₂O, Na, Ca, Fe or Mg.

Fig. 6 shows that 85% of the analyzed natural cordierites have *mg* values between 0.55 and 0.85, an observation of some value in the calculation of Niggli Katavariants.

Recent discoveries of the various structural states of cordierite suggest that an *x*-ray examination of cordierite from granites, such as the Dartmoor Granite S. W. England, may throw valuable light on the thermal history of the rocks and on the granite problem generally, in a similar manner to that shown by feldspars and quartz.

ACKNOWLEDGMENTS

I wish to thank Drs. Bernard Evans and G. Chinner for allowing me to include unpublished analyses; Dr. A. Miyashiro of the University of Tokyo, for his kind assistance over certain Japanese references; Dr. F. C. Phillips and Mr. R. Bradshaw for revising the manuscript and Drs. M. H. Hey and W. Schreyer for their helpful interest in the problem. The writer would be grateful to know of omissions and additions to Table 1.

LOCALITIES, SOURCES AND NOTES TO TABLE 1

All the cell contents given have been calculated to 18 oxygens excluding water. Most of the pre-1900 analyses (whose value is dubious) do not give the state of oxidation of the iron and the writer has assumed, or calculated it to be FeO. There are many discrepancies and misquotations in the early literature, for example compare Dana 3rd ed, 1850, p. 344, George Putnam, London, and Dana, 6th ed., 1892, p. 420, Chapman & Hill, London, but these have not all been catalogued for economy of space. All, or nearly all of the water contents determined before 1900 are lost on ignition, while later determinations are mostly by the Penfield (1894) method which tends to give consistently low results (Riley, 1958).

An empty space indicates that no information is available; n.d. denotes "not determined."

1. Smith Ridge, Boehls Butte Quadrangle, Idaho, U.S.A. Kyanite-Andalusite-Sillimanite-biotite schist. A. Hietanen, 1956, *Am. Mineral.* 41, p. 7.

2. Laramie Range, Albany County, Wyoming, U.S.A. (Laramie Range No. 2). T. Iiyama 1956, p. 374. A. Miyashiro, 1957, p. 47 determined $\alpha=1.536$, $\beta=1.541$, $\gamma=1.546$, $2V=77^\circ$ for same material.
3. Attu, S.W. Finland. Anthophyllite gneiss. G. Pehrman, 1932, p. 5.
4. W. of Karungalpatti, Salem district, Madras, India. Corundum-sillimanite-anthophyllite-bytownite rock. A. P. Subramaniam, 1956, p. 344. Analysis total includes Cl 0.01 and F 0.01.
5. Twedestrand, Norway, Gneiss. L. Oppenheimer, 1915, p. 176. (also L. Oppenheimer, 1914).
6. Orijarvi, Finland. Gneiss. T. Iiyama, 1956, p. 374. Analysis total includes 0.01 P_2O_5 .
7. Twedestrand, Norway. E. Thiele, 1940, p. 80.
8. Simiutak, Greenland. U. Stromeyer, Untersuchungen über die Mischung der Mineralkörper. etc. Göttingen, 1821, p. 329 quoted by T. H. Holland 1891. *Quart. Jour. Geol. Soc.* **47**, 191. Original specific gravity = 2.5969.
9. Mursinka, Urals, Russia. R. Hermann in N. V. Kokscharow, Min. Russl. Vol. 3, 1858, p. 257 quoted by Dana, 6th Ed., 1892, 420. Chapman & Hall, London.
10. Yötiri, Kéikidö, Korea. Augen gneiss. T. H. Holland, 1891. *Quart. Jour. Geol. Soc.* **47**, 190.
11. Cape Denison, Antarctica. Gneiss. C. E. Tilley, 1940, p. 340. Heald, 1950, p. 66 and Subramaniam, 1956, p. 345, misquote original and give $Al_2O_3=31.56$ total = 100.17.
12. Hitati mine, Hitati, Ibaragi Pref. Japan. Iron ore deposits in a schist. K. Sugiura, 1951, *Jour. Jap. Ceram. Assoc.* **59**, p. 326.
13. Mont Iby, Madagascar. Pegmatite. L. Duparc, M. Wunder & R. Sabot, 1911 *Neues Jahrb., Ref.* II, p. 365. H_2O^+ is loss on ignition. L. Oppenheimer, 1915, p. 176 quotes $\alpha=1.5394$, $\beta=1.5446$, $\gamma=1.5479$, $2V=72\frac{1}{2}$ and gives $\alpha=1.5400$, $\beta=1.5458$, $\gamma=1.5479$, $2V=67^\circ$, $D=2.600$ for another sample from the same locality.
14. Orijarvi, Finland. E. Thiele, 1940, p. 80. Thiele gives the analysis total as 100.22 but the summation of his figures gives 100.12.
15. Iby, Madagascar. E. Thiele, 1940, p. 80. R. E. Folinsbee, 1941, p. 493 gives this analysis and optics as "Twedestrand." Thiele gives analysis total as 100.16, but the summation of his figures gives 100.40.
16. Mont Bity, Madagascar. Quartz vein. T. Iiyama, 1956, p. 374.
17. Orijarvi, Finland. L. Oppenheimer, 1915, p. 176 (also L. Oppenheimer, 1914).
18. New Haven, Connecticut, U.S.A. Pegmatite. T. Iiyama, 1956, p. 374.
19. Pachipenta, Vizagapatam, India. C. Mahadevan & G. Sastry, 1948, p. 361. The determined specific gravity appears to be erroneous (fig. 5), while Fe_2O_3/FeO is unusually large.
20. Guilford, Connecticut, U.S.A. Probably a pegmatitic vein cutting cordierite-quartz-feldspar-biotite-gneiss. Analysis by O. C. Farrington, 1892, *Amer. Jour. Sci.*, **143**, p. 14. Optics and redetermined specific gravity (= 2.605) by L. Oppenheimer, 1915, p. 176.
21. Hitachi (=Hitati) mine, Ibaragi Pref., Japan. Quartz vein. T. Iiyama, 1956, p. 374. Analysis total includes 0.02 P_2O_5 .
22. Haddam, Connecticut, U.S.A. O. C. Farrington, 1892, *Amer. Jour. Sci.*, **143**, p. 14.
23. Bodenmais, Bavaria. L. Oppenheimer, 1915, p. 176.
24. Bodenmais, Bavaria. E. Thiele, 1940, p. 80. Thiele gives the analysis total as 100.25 but the summation of his figures gives 100.47. $2V$ calculated?
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26. Ankaditany, South Madagascar. Pegmatite with cordierite and garnet. A. Lacroix, 1939, *Bull. Soc. Franç. Min.*, **62**, p. 289.
27. Traskhole, Finland, Anthophyllite rock. P. Eskola, 1914, *Comm. Geol. Finlande, Bull.* **40**, p. 195.
28. Rücken, N.E. of Humpoletz, Bohemia. F. Katzer, 1894, *Tsch. Min. Pet. Mitt.*, **14**, p. 515. The specific gravity appears to be erroneous.
29. Sotajoki mouth, Lapland. Quartz-garnet-plagioclase-biotite-orthoclase-sillimanite granulite. P. Eskola, 1952, *Amer. Jour. Sci. Bowen Vol.*, p. 152.
30. Schmelz, near Lam, Bavaria. B. Gossner & E. Ilg, 1932, *Centr. Min. Abt. A*, p. 2. Average of two analyses.
31. Kasyo-to, Formosa. A volcanic rock-biotite-hornblende-andesite. T. Ichimura, 1933, *Taiwan Tiyaiku Kizi*, Vol. 4, p. 32. (Also T. Ichimura, 1936, *Geology of Kasyo-to, Taiwan, Kazan*, Vol. 3, pp. 1-27.)
32. Glen Clova, Angus, Scotland. Hypersthene-quartz-biotite-orthoclase plagioclase hornfels. G. A. Chinner, personal communication, Distortion Index=0.25.
33. Haddam, Connecticut, U.S.A. Pegmatite cutting biotite-sillimanite-garnet-quartz-microcline albite gneiss. L. Oppenheimer, 1915, p. 176. Specific gravity is rather high (fig. 5).
34. Langdorf, (?near Bodenmais, Bavaria). E. Thiele, 1940, p. 80.
35. Haddam, Connecticut, U.S.A. Pegmatite cutting biotite-sillimanite-garnet-quartz-albite gneiss. A. Miyashiro, 1957, p. 47.
36. Langdorf, near Bodenmais. B. Gossner & E. Reindl, 1932, p. 330. Average of two analyses.
37. Madura, Madras, India. Garnet-sillimanite-orthoclase-oligoclase gneiss. M. S. Krishnan, 1924, *Min. Mag.*, **20**, p. 249. Specific gravity is very low.
38. Aldan complex, Russia. Biotite-garnet-plagioclase gneiss. E. I. Lavreno. 1957, *Mem. Soc. russe Min.*, **16**, p. 74.
39. Lovewell Mt. Quadrangle, New Hampshire, U.S.A. quartz-monzonite? M. T. Heald, 1950, p. 66. 4% sericite calculated out of the analysis, D includes 4% sericite? Analysis total includes 0.05 P₂O₅.
40. Watarase, Japan. Hornfels. Analysis by Y. Kukuchi, 1890, *Journ. Coll. Sci. Imp. Univ. Japan*, **3**, p. 313. Optics by H. Shibata, 1936, p. 222 on the same specimen as analysed. On p. 225 Shibata gives $\gamma=1.558$; on p. 222 $\gamma=1.559$. E. Thiele, 1940, p. 81, misquotes $\alpha=1.540$.
41. Sparcraigs, Belhelvie, Aberdeenshire, Scotland. Biotite-plagioclase-garnet spinel hornfels. F. H. Stewart, 1942, p. 262.
42. Great Slave Lake, Canada. Garnet-sillimanite-spinel-biotite-K feldspar-oligoclase-quartz rock. R. E. Folinsbee, 1941, p. 494.
43. Sakuratenzin, Hiedanomura, Kyoto Pref. Japan. Mica-hornfels. Y. Kukuchi, 1889, *Jour. Coll. Sci. Imp. Univ. Tokyo*, Vol. 3, p. 329. The specific gravity appears to be erroneous and the K₂O content is impossibly high for cordierite.
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46. Ostgothland, Sweden. C. Schutz, 1841, p. 565. Quoted by Dana, 3rd Ed. 1850, p. 344 (ibid.). Original includes 0.2 undecomposed. Dana gives total as 100.8.
47. "Goaini Kondes, S.W. Africa." B. Gossner & E. Reindl, 1932, p. 332. Average of two analyses.
48. Upington, Cape Province, South Africa. Quartz-biotite-alkali-feldspar rock. M. Mathias, 1952, *Min. Mag.*, **29**, p. 939. Analysis total includes 0.03 P₂O₅.

49. Degero, Helsingfors, Finland. Biotite-microcline-andesine-pegmatite. G. Pehrman, 1932, p. 5.
50. Glen Muick, Angus, Scotland, Corundum-spinel-biotite-orthoclase-plagioclase hornfels, G. A. Chinner, personal communication. Distortion index=0.25.
51. Kondes (S.W. Africa?). E. Thiele, 1940, p. 80.
52. Laramie Range, Albany County, Wyoming, U.S.A. ("Laramie Range No. 1"). W. H. Newhouse & A. F. Hagner, 1949, p. 13, who give $2V=81^\circ$ ("only one determination") and ? sign = \pm .
53. Santavuori, Ilmajoki, Finland. Vein gneiss. G. Pehrman, 1932, p. 5.
54. Raumanlinna, Rauma, S.W. Finland. Garnet gneiss. A. Hietanen, 1943, *Ann. Acad. Scient. Fennicae, A*, **III**, p. 13.
55. Derryadd West, Connemara, Co. Galway, Eire. Biotite-sillimanite-garnet-andesine hornfels. B. Evans, personal communication. Distortion index=0.28.
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