The optical properties and composition of the acmitic pyroxenes.

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THE occurrence of acmitic pyroxenes in the post-Cambrian intrusions of the north-west of Scotland was first noted by Teall (Horne and Teall, 1892) who recorded aegirine in two dikes discovered by Gunn in the Coigach district of Ross-shire, and aegirine-augite in the borolanite of the Cnoc-na-Sròine mass. Since then, acmitic pyroxenes have been recorded by Teall (1900) from the Assynt felsites, Shand (1910) from dikes of the border zone of Cnoc-na-Sròine, and Phemister (1926) from the dikes and plutonic rocks of the Loch Ailsh laccolite.

Having undertaken a comprehensive examination, in the field and in the laboratory, of the post-Cambrian sills and dikes of the Assynt district, the writer determined some of the optical constants of the acmitic pyroxenes of these intrusions. Arising from this work the considerable data now available on the acmitic pyroxenes has been reviewed. A chemical analysis has also been carried out, and it is hoped that the information assembled and re-interpreted here may be of some general interest.

The name *acmite* (Achmit) was proposed by Berzelius for crystals described by Ström from the parish of Eger in southern Norway (Ström, 1821) and alluded to their pointed shape. *Aegirine* (Aegirin) was the name given by Esmark to a mineral occurring on an island in the sea in the neighbourhood of Skansfjord, and was so called after the Scandinavian sea-god Aegir (Berzelius, 1835).

Aegirine is found as greenish-black crystals having blunt terminations, acmite as pointed brown crystals. Under the microscope aegirine is of green colour and is strongly pleochroic, whilst acmite is usually brown in colour and is only weakly pleochroic. The two species were shown to be essentially the same, $NaFe^{+3}Si_2O_6$, by Tschermak in 1872, but the two names still persist. The term 'acmite' is also used for the theoretical $NaFe^{+3}Si_2O_6$ molecule. The name aegirine-augite was suggested by

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Rosenbusch (Aegirin-Augit) in 1892 for minerals intermediate between aegirine and augite and has been widely used for the green sodic pyroxene of high extinction angle.

Aegirine from the Assynt district, Sutherlandshire.

The aegirine here described occurs in an aegirine-nepheline-syenite pegmatite dike cutting the ultrabasic rock of the Cnoc-na-Sròine mass. The dike, which is a member of the great post-Cambrian sill and dike suite of Assynt, outcrops in the Bad na h'Achlaise 700 yards south of Ledmore. It is about 5 feet 6 inches thick and is collinear with a 2-foot dike of variable nepheline-syenite in the adjoining Cambrian quartzite. Dr. J. Phemister (Macgregor and Phemister, 1937) has pointed out that this suggests that the pyroxenite is intrusive in Cambrian quartzite, and not in limestone as thought by Shand (1910).

Petrologically, the dike-rock is of variable composition. Parallel, half-inch-long prisms of aegirine occur in narrow streaks set in coarsely crystalline grey felspar with pink micaceous pseudomorphs representing original euhedral nepheline. In thin section (Geological Survey slice no. S. 37399) the felspar is seen to be predominantly of perthite akin to soda-orthoclase and containing only a little albite. Chequer-albite forms selvedges along the margins of the felspar crystals. The nepheline is now represented by pinitic mica. Scarce magnetite crystals are present, moulded upon felspar, and there are occasional anhedra of biotite. Euhedral aegirine, which occurs in both the felspar and in the nepheline pseudomorphs, in places has a fairly robust habit, yet elsewhere forms slender prisms.

To separate the aegirine the rock was crushed to pass a 50 mesh sieve and then separated in methylene iodide. The heavy fraction, consisting almost entirely of aegirine, was passed through a Frantz isodynamic separator and finally the sample was hand picked. The chemical analysis and optical properties of the mineral are given in table I.

It may be noted that if $H_2O(+105°C.)$ is excluded the analysis may be expressed as $(Na,Ca,K)_{1\cdot007}(Fe^{+3},V,Ti,Fe^{+2},Mn,Mg,Al)_{1\cdot009}(Si,Al)_{2\cdot000}O_{6\cdot000}$, which corresponds very closely with the ideal formula $XYSi_2O_6$, and perhaps indicates that the water is not really constitutional. The weight percentage of the 'acmite' molecule present is 73·4.

The refractive indices of the mineral were determined by the immersion method and the optic axial angle on a universal stage. The pleochroism is α deep green, β deep green, γ yellowish-brown, and the absorption $\alpha > \beta = \gamma$. Owing to the high refractive indices, intense

TABLE I. Analysis of aegirine from Assynt, Sutherland.

Number of atoms when (O + OH) = 6

SiO$_2$	51·35	Si^{+4} ...	1·961	⎫ 2·000
Al$_2$O$_3$	2·88*			⎰ 0·039 ⎱
Fe$_2$O$_3$	25·43	Al^{+3} ...	0·129	⎰ 0·090
FeO	3·70			
MgO	0·58	Fe^{+3} ...	0·728	
CaO	3·29	V^{+3} ...	0·001	
Na$_2$O	11·65	Ti^{+4} ...	0·018	⎱ 0·994
K$_2$O	0·14	Mg^{+2} ...	0·033	⎰ 1·997
H$_2$O $-$105° C. ...	0·09†	Mn^{+2} ...	0·006	
H$_2$O $+$105° C. ...	0·24†	Fe^{+2} ...	0·118	
TiO$_2$	0·65			
MnO	0·18	Na^{+1} ...	0·861	
V$_2$O$_3$	0·02	Ca^{+2} ...	0·135	⎱ 1·003
ZrO$_2$	not detected	K^{+1} ...	0·007	
(Ce,Yt)$_2$O$_3$ ...	„ „			
	———	OH^{-1} ...	0·061	⎱ 6·000
	100·20	O^{-2} ...	5·939	

Analysis by Geochemical Laboratories, Wembley.

* Corrected for V$_2$O$_3$.
† Determined in the Chemical Laboratory, Geological Survey and Museum.

α 1·751, β 1·786, γ 1·800, 2V$_\alpha$ 65–73°, $\alpha : c = 1°$.

A quantitative spectroscopic determination carried out by Dr. J. A. C. McClelland of the Government Laboratory showed V$_2$O$_3$ 0·02–0·03 % and the Al$_2$O$_3$ figure has been corrected accordingly.

colour, and moderate to strong dispersion the determination of the optic axial angle was attended by considerable difficulty, and a single value has not therefore been quoted. The range of figures determined was 65–73° and the mean 68½°, but as it is not possible to appraise the errors, this figure should be regarded with caution.

Structure and composition.

The structure of acmite was studied by Warren and Briscoe (1931) who showed that it had the same structure as the other monoclinic pyroxenes, typified by diopside, and consisted of chains of silicon–oxygen tetrahedra linked by the cations Na^{+1} and Fe^{+3} in 8-fold and 6-fold co-ordination respectively. They gave the general formula for the monoclinic pyroxenes $X_m Y_{2-m}(\text{Si,Al})_2(\text{O,OH,F})_6$, where m tends to be 1·0 or 0, $X =$ Na,Ca,K,Mn, and $Y =$ Mg,Fe,Al,Ti,Mn.

In the past, attempts to display the variation of physical properties with chemical composition of the acmitic pyroxenes have mainly been based upon interpretations of analyses in terms of theoretical oxides, or 'minals' as they have been termed by Alling (1921). Winchell (1933) presented a double triangular diagram to show the optical properties of

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the pyroxenes, plotting diopside and hedenbergite as the ends of the common side of the triangles and acmite and jadeite as the two vertices. He remarked, however, that the diagram was based upon insufficient data and was therefore not accurate. An additional reason for the inaccuracy lies, in the opinion of the writer, in the variable chemical composition of the minerals themselves. Larsen (1942) published a graph showing the various optical properties of the acmitic pyroxenes plotted against Fe_2O_3 content, with an interpolated scale of acmite. He did not give the sources of the figures he used. Ostrovsky (1946) has given data for synthetic aegirine-diopside mixtures.

In table II are shown the calculated percentages of acmite, hedenbergite, and diopside for thirteen minerals quoted by Washington and Merwin (1927) together with the sum of the remaining constituents in each mineral.

TABLE II. Calculated percentages of acmite, hedenbergite, diopside, and remaining constituents in thirteen analysed specimens.

No.	Acmite, $NaFeSi_2O_6$.	Hedenbergite, $CaFeSi_2O_6$.	Diopside, $CaMgSi_2O_6$.	Remaining constituents.
1.	89·17	2·48	0	8·35
2.*	86·36	7·19	0	6·55
3.	82·70	1·24	0	16·06
4.	82·70	5·71	0	11·59
5.	81·31	7·19	0	11·50
6.	73·46	9·67	8·64	8·23
7.	67·91	18·35	6·70	7·04
8.	65·60	15·87	9·07	9·46
9.	65·60	19·09	1·08	14·23
10.	62·83	7·19	14·90	15·08
11.	55·44	23·81	12·31	8·44
12.	36·04	14·13	34·78	15·05
13.	9·70	59·77	17·28	13·25

* Calculated from rock analysis by Washington and Merwin.

1, Quincy, Mass., U.S.A. 2, Rockall Island. 3, Rundemyr, Norway. 4, Kangerd-luarsuk, Greenland. 5, 'Brevik', Norway. 6, Låven, Langesund Fjord, Norway, 7–8, Magnet Cove, Ark., U.S.A. 9, Arö Skjär, Norway. 10, Libby, Mont., U.S.A. 11, Låven, Langesund Fjord, Norway. 12, Libby, Mont., U.S.A. 13, Salem Neck. Mass., U.S.A.

In calculating the composition in terms of acmite, hedenbergite, and diopside, these authors allotted firstly the greatest possible percentage of Na_2O and Fe_2O_3 to form acmite. Hedenbergite and diopside were then calculated. If $FeO+MgO+(MnO)$ was in excess of CaO an arbitrary choice had then to be made. Washington and Merwin gave preference to FeO, first calculating as much hedenbergite as possible and then assigning the remaining CaO to MgO to form diopside. This proce-

TABLE III. Atomic proportions of analysed acmitic pyroxenes.

No.	Si.	Al.	[Al].	Ti.	Fe^{+3}	Mg.	Fe^{+2}	Mn^{+2}	Ca.
1.	1·989	0·045	0·034	0·017	0·911	0·008	0·029	0·011	0·023
2.	1·959	none	none	0·022	0·893	0·016	0·059	0·009	0·070
3.	2·003	0·094	0·094	0·028	0·835	0·020	0·007	0·005	0·012
4.	1·968	0·080	0·048	0·007	0·833	0·020	0·099	0·010	0·054
5.	1·995	0·060	0·055	0·016	0·811	0·025	0·070	0·003	0·065
6.	1·977	0·044	0·021	0·016	0·773	0·125	0·086	0·003	0·179
7.	1·957	0·082	0·039	0·003	0·682	0·086	0·170	0·003	0·246
8.	1·969	0·050	0·019	0·057	0·647	0·129	0·136	0·008	0·242
10.	1·976	0·017	none	0·025	0·622	0·176	0·047	0·019	0·226
11.	1·944	0·126	0·070	0·015	0·550	0·131	0·219	0·016	0·350
12.	2·115	0·065	0·065	0·011	0·368	0·042	0·122	0·015	0·518
13.	1·886	0·207	0·093	0·041	0·100	0·186	0·564	none	0·755
14.	1·989	0·071	0·060	0·037	0·754	0·081	0·076	0·007	0·106
15.	1·982	0·146	0·129	0·010	0·840	0·039	0·020	none	0·007
16.	1·913	0·192	0·105	0·023	0·345	0·220	0·316	0·013	0·548
17.	2·052	0·057	0·057	0·073	0·630	0·022	0·051	0·014	0·212
18.	2·02	0·17	0·17	0·002	0·77	0·01	0·02	0·01	0·03
19.	1·923	0·099	0·022	0·010	0·400	0·304	0·238	0·020	0·548
20.	1·961	0·129	0·090	0·018	0·728	0·033	0·118	0·006	0·135

No.	Na.	K.	Zr.	V.
1.	0·868	0·018	0·008	n.d.
2.	0·884	none	0·052	n.d.
3.	0·952	0·009	0·004	n.d.
4.	0·946	0·018	0·003	n.d.
5.	0·868	0·011	0·025	n.d.
6.	0·713	0·013	n.d.	n.d.
7.	0·739	0·012	0·002	n.d.
8.	0·688	0·004	n.d.	n.d.
10.	0·770	0·011	n.d.	0·121
11.	0·592	0·007	0·001	n.d.
12.	0·481	0·013	n.d.	0·091
13.	0·249	0·022	n.d.	n.d.
14.	0·840	0·017	0·003	n.d.
15.	0·850	0·026	n.d.	n.d.
16.	0·405	0·020	n.d.	n.d.
17.	0·764	0·009	n.d.	none
18.	0·93	0·01	n.d.	n.d.
19.	0·394	0·019	n.d.	0·001
20.	0·861	0·007	none	0·001

n.d. = not determined.
1–13. See table II for localities (Washington and Merwin, 1927).
14. Bearpaw Mountains, Mont., U.S.A. (Pecora, 1942).
15. French River, Ontario, Canada (Walker and Parsons, 1926).
16. Iron Hill, Colo., U.S.A. (Larsen, 1942).
17. Libby, Mont., U.S.A. (Goranson, 1927).
18. Norra Kärr, Sweden (Adamson, 1944) (atomic proportions given).
19. Ilmen Mountains (Zavaritzky, 1946).
20. Bad na h'Achlaise, Assynt, Sutherlandshire (new analysis).

dure they adopted because they found the hedenbergite molecule more abundant than that of diopside. Remaining constituents were then allotted to the theoretical molecules $Na_2O.Al_2O_3.4SiO_2$, $Na_2O.V_2O_3$. $4SiO_2$, $Na_2O.SiO_2$, $MgO.SiO_2$, $FeO.SiO_2$, $Fe_2O_3.3SiO_3$, $Al_2O_3.3SiO_2$, Al_2O_3, and SiO_2. It will be seen from the table that the sum of these various remaining constituents may amount in some cases to nearly one-sixth of the molecule, even in those minerals with high acmite content. It is in the variable quantity and composition of these remaining constituents that one of the principal causes of the variability in the optical properties appears to lie: the acmitic pyroxenes are of too complicated composition to be readily expressible in terms of the three 'end-members'.

The chemical composition may, however, be recalculated as atomic proportions. Atomic proportions for the pyroxenes studied by Washington and Merwin, together with a number based on analyses published since the date of their paper, are presented in table III. These have been calculated, as in Berman's paper (1937) on the basis $O+OH = 6$, if necessary making $Si = 2$ by utilizing a proportion of the Al present. For convenience in the sequel, the aluminium in 6-fold co-ordination, which remains after allotting the amount (in 4-fold co-ordination) necessary to make $Si = 2$, is denoted as [Al] and referred to as 'excess aluminium'. The almost exact equation, after excluding water, of the X and Y groups in the Assynt aegirine was not found to be repeated in the other analyses, and they have all therefore been computed without first excluding water $(+105°$ C.).

The various rules governing the distribution of elements in minerals were deduced by Goldschmidt, whose results have been confirmed many times, as, for example, in the recent careful study by Nockolds and Mitchell (1948) of the trace elements of the Caledonian plutonic rocks. In the present study the association of elements of similar ionic size is clearly displayed. The mutual relationships of the various elements in the acmitic pyroxenes are best seen from the graphs (figs. 1a, b, c). The validity of the following statements is apparent:

1. Sodium and potassium together vary directly with ferric iron, titanium, excess aluminium, and vanadium (fig. 1b). If Fe^{+3} alone is plotted against $Na^{+1}+K^{+1}$ the approximation $Fe^{+3} = \Sigma Na^{+1}$ is not so good, corroborating the postulated ionic replacements.
2. Magnesium and ferrous iron together vary directly with calcium (fig. 1c).
3. Sodium and potassium together vary inversely with calcium (fig. 1a).

Fig.1a

Fig.1b

Fig.1c
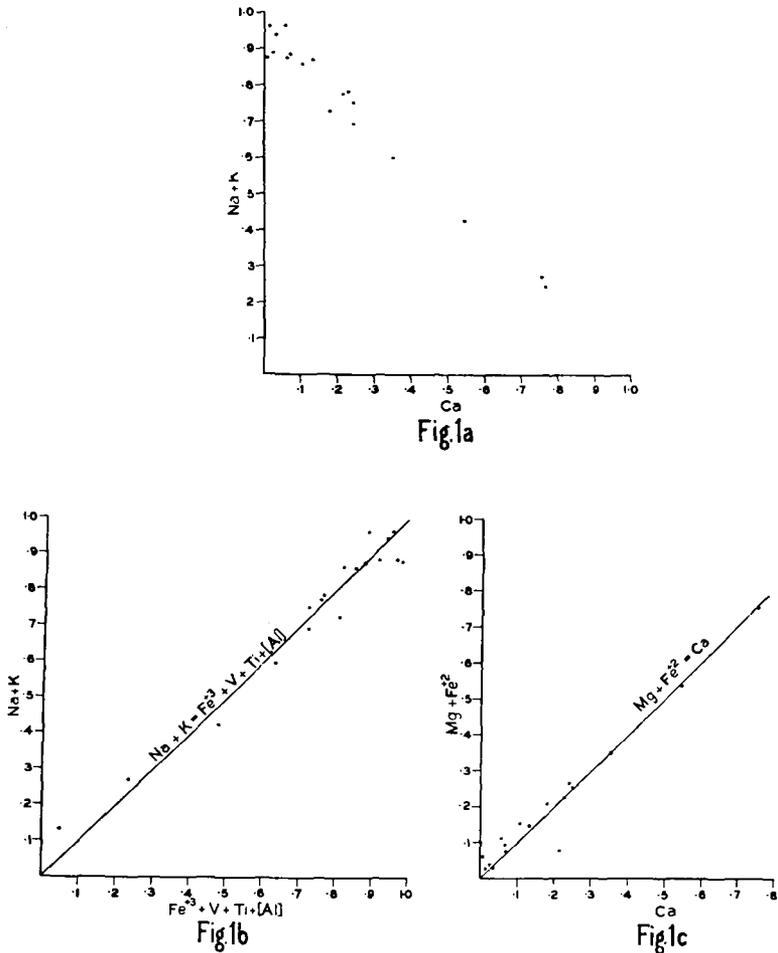
FIG. 1. Graphs showing the relative atomic proportions in the acmitic pyroxenes.

TABLE IV. Ionic radii (Evans, 1939).

Si^{+4}	0·39	V^{+3}	0·65	Fe^{+2}	0·83	Na^{+1}	0·98 Å.
Al^{+3}	0·57	Fe^{+3}	0·67	Zr^{+4}	0·87	Ca^{+2}	1·06
Ti^{+4}	0·64	Mg^{+2}	0·78	Mn^{+2}	0·91	K^{+1}	1·33

The ionic radii of the elements concerned are shown in table IV. It will be seen that Fe^{+3}, V^{+3}, Ti^{+4}, and Al^{+3} are of similar size, Mg^{+2} and Fe^{+2} are of similar size, whilst Ca^{+2} is intermediate in size between Na^{+1} and K^{+1}. The introduction of the divalent calcium ion in place of

Na^{+1} immediately requires a balancing of the electrostatic charges by a concomitant introduction of a second divalent ion in place of Fe^{+3}. Since, moreover, there is a direct relationship between ΣNa^{+1} and ΣFe^{+3}, Ca^{+2} would be expected to vary directly with the sum of Mg^{+2} and Fe^{+2}.

Optical properties.

The optical properties of the acmitic pyroxenes whose atomic ratios are presented in table III are shown in table V.

TABLE V. Optical properties of the acmitic pyroxenes.

	α.	β.	γ.	2V.	α : c.
1.	1·767	1·806	1·823	−60°	6°
2.	1·765	—	1·815	—	3–5
3.	1·770	1·811	1·825	—	3
4.	1·769	1·805	1·824	—	4
5.	1·760	1·790	1·810	—	0
7–8.	1·763	1·780	1·812	—	−3–+3
10.	1·745	1·770	1·782	−69°	1–2
11.	1·744	1·768	1·782	−70° −80°	10
12.	1·720	—	1·747	—	24
13.	1·711	1·729	1·759	+75° +80°	27–32
14.	1·757	1·786	1·797	−62°	low
15.	1·752	1·788	1·801	−63°	—
16.	1·730	1·748	1·768	90°	14
17.	1·742	1·768	1·787	−81°	2
18.	—	—	—	−58°	3
19.	1·720	1·740	1·757	−82° −88°	14–15
20.	1·751	1·786	1·800	−68½°	1
21.	1·776	1·819	1·836	−61°	8–10

1–20, as tables II–III. 21, synthetic acmite (Washington and Merwin, 1927).

The relationships between the composition and optical properties have been expressed by means of a triangular diagram (fig. 2). In using triangular co-ordinates it is necessary to assume the cations to sum to 2, and the vertices have therefore been taken to denote $(Na^{+1}+K^{+1}) = 2$, $(Fe^{+3}+V^{+3}+Ti^{+4}+[Al]^{+3}) = 2$, and $(Mg^{+2}+Fe^{+2}+Zr^{+4}+Mn^{+2}+Ca^{+2}) = 2$. As the ideal summation to 2 is but rarely attained in nature each analysis appears not as point but as a small triangle. These triangles, representing the atomic proportions given in table III, are shown in fig. 2, and it will be seen that they are mainly quite small. An exception is analysis no. 12 which is not shown as the triangle is too large to permit of its use. This doubtless arises from the high Si^{+4} content (2·115) which casts doubt on the veracity of the analysis. It should be noted that in plotting the diagrams Ca has not been included with Na + K as the points would not then lie on an altitude.

To plot the optical properties the centre of each of these triangles has been taken as representing the idealized composition. All such points lie very close to the line A–B representing $\Sigma Na^{+1} = \Sigma Fe^{+3}$ and for the present purposes may be considered to lie on the line. The refractive
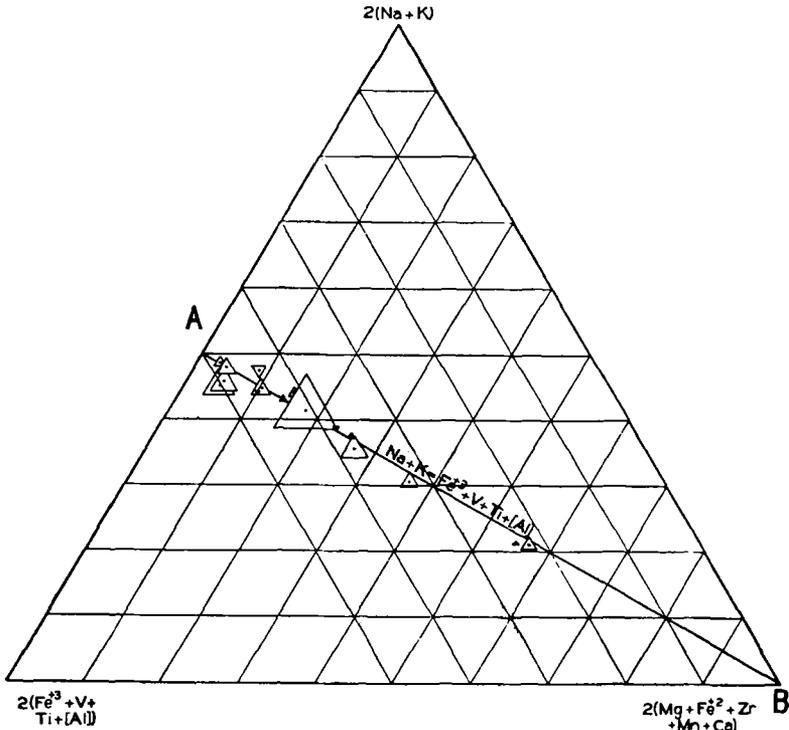
FIG. 2. The chemical composition of the acmitic pyroxenes.

indices and optic axial angle have been plotted in fig. 3 using this line as abscissa of composition so that for any given value of α, β, γ, or $2V$ a theoretical composition is readily obtained from the corresponding point on AB. Having obtained the theoretical composition in terms of ΣNa^{+1}, ΣMg^{+2}, and ΣFe^{+3}, the approximate content of Ca^{+2} may be obtained from fig. 1b, since Ca^{+2}, Mg^{+2}, and Fe^{+2} together constitute the majority of $(Mg^{+2}+Fe^{+2}+Zr^{+4}+Mn^{+2}+Ca^{+2})$ present.

The range of compositions shown by minerals having similar optical properties appears to arise principally from replacement of the Mg^{+2} and Fe^{+2} by each other. This cannot be detected from the optical properties

and remains a source of considerable error. Other properties of the minerals were plotted in a similar manner, but showed too great irregularities to be of much value. It may, however, be remarked that within the range of minerals now described the extinction angle varies rather
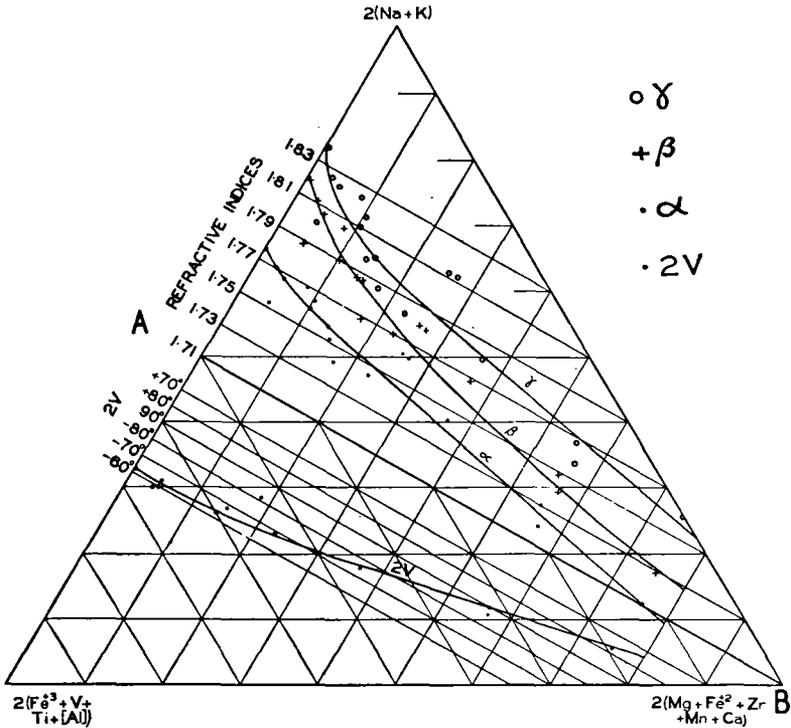
FIG. 3. The relationships of the optical properties to the chemical composition of the acmitic pyroxenes.

irregularly from 8–10° (in the obtuse angle) for pure acmite through 0° to 27–32° (in the acute angle) for aegirine-augite (acmitic-diopsidic hedenbergite) of β 1·729; the specific gravity ranges irregularly from 3·59 to 3·42 and the birefringence varies rather irregularly from 0·069 for pure acmite to 0·038 for the mineral with β 1·748.

Nomenclature.

Washington and Merwin adopted the following nomenclature. (1) Where there was present 80 % or more of one of the 'end-members', the mineral was called by the name of the end-member—acmite,

diopside, or hedenbergite. (2) If only two end-members were present such names as acmitic diopside or diopsidic acmite were used, but members intermediate between diopside and hedenbergite, with 10 % or less of acmite were called augite. (3) The terms acmitic augite or augitic acmite were applied when all three end-members were present in large amount. When first describing aegirine-augite, Rosenbusch (1892) gave the extinction $(\gamma:c)$ as 55° and, comparing diopside, basaltic augite, aegirine-augite, and aegirine, gave aegirine-augite as having the extinction $(\gamma:c)$ 60° and aegirine 94°. This figure for aegirine-augite corresponds roughly to a mineral containing only about $Na_{0.25}Fe^{+3}_{0.25}$.

The restriction by Washington and Merwin of the term 'acmite' to minerals containing 80 % of the theoretical molecule, roughly equivalent to $Na_{0.8}Fe^{+3}_{0.8}$, appears to the writer rather to limit the use of the name. On the other hand, limiting 'aegirine-augite' to minerals containing up to $Na_{0.25}Fe^{+3}_{0.25}$ also appears to be too rigid. Many minerals lie in the range between $Na_{0.8}Fe^{+3}_{0.8}$ and $Na_{0.25}Fe^{+3}_{0.25}$. It is therefore suggested that a convenient division of the aegirine–aegirine-augite series might be taken at $Na_{0.45}Fe^{+3}_{0.45}$, at or about which $2V = 90°$. Negative minerals would then continue to be denoted as aegirine and positive ones as aegirine-augite. This division corresponds to an extinction angle of about 15° $(\gamma:c\ 75°)$, the lower figure quoted by Rogers and Kerr (1933) for aegirine-augite.

It might be objected that the aegirine group proposed has rather a long range of refractive indices (β 1·744 to 1·819). The refractive indices of aegirine-augites of small acmite content, not here considered, may, however, be low. The typical mean refractive index of aegirine-augite is commonly quoted as 1·687 and within the range 1·687–1·819, the division now suggested is approximately central. The extinction angle of 15° is similarly placed. A further possible objection is that within the group, between $Na_{0.45}Fe^{+3}_{0.45}$ and $Na_{0.50}Fe^{+3}_{0.50}$ the acmite molecule would not exceed the sum of the other molecules. It is, however, very unlikely that any other single molecule would be present in greater quantity than the acmite. The advantages of the proposed division thus appear to outweigh the disadvantages, and for most acmitic pyroxenes the division affords an easy method of classification.

The two names 'aegirine' and 'aegirine-augite' are adequate for general descriptive purposes. When greater precision is required and the mineral has been analysed, the nomenclature proposed by Washington and Merwin, or better, the calculated formula may be used. 'Aegirine' is to be preferred to 'aegirite', since the mineral was first described, in

German, as 'Aegirin'. It may be remarked that although in the later editions of Dana's 'System of mineralogy' the mineral is recorded as 'ægirite' and 'aegirite', in the earlier editions in which the name first appeared (3rd edit., 1850; 4th edit., 1854) it was given as 'ægirine'.

It is suggested that in the present state of our knowledge, the choice of the name 'acmite' or 'aegirine' should depend upon the optical properties of the mineral: thus, green pleochroic varieties should be termed aegirine and brownish weakly pleochroic varieties acmite. The distinction should not, as has sometimes been the case, be based upon chemical composition.

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