

THE SYSTEM ACMITE-DIOPSIDE AND ITS BEARING
ON THE STABILITY RELATIONS OF
NATURAL PYROXENES OF THE
ACMITE-HEDENBERGITE-DIOPSIDE
SERIES

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ABSTRACT

Phase-equilibrium relations in the system acmite-diopside have been studied at 1 atm. At lower temperatures there is a complete series of solid solutions between the two end members, but at higher temperatures acmitic pyroxenes melt incongruently to hematite and liquid. Since small part of the iron is always present as ferrous iron, the system is not truly binary even in the diopside-rich portion, and there is always a small amount of glass present in the region labelled pyroxene solid solutions. Optical properties of the synthetic pyroxenes of this system show continuous variation with composition.

Reinvestigation of the pyroxenes from the Morotu alkalic rocks shows that aegirine-augites whose composition lies within the immiscibility region of the acmite-hedenbergite-diopside system recently proposed by Aoki do exist under some igneous conditions. When natural alkalic pyroxenes are plotted on the Na-Fe²⁺-Mg diagram, pyroxenes from volcanic rocks and those from pegmatites have a rather well-defined area of distribution, while aegirine augites from other kinds of rocks are distributed between the two groups of pyroxenes.

It has been shown experimentally that the stability field of acmite is increased with increasing P₀₂ and that higher P₀₂ favors the crystallization of pyroxenes high in acmite. From these experimental and petrological data a model is proposed to show the relation between the prevailing P₀₂ and the composition of the pyroxenes; *i.e.*, the low P₀₂ in volcanic rocks favors the crystallization of Ca-rich pyroxenes of diopside-hedenbergite series, whereas the high P₀₂ in pegmatites or related rocks favors the crystallization of acmite-rich pyroxenes or nearly pure acmite. It seems likely, therefore, that there are continuous solid solutions in the pyroxene system acmite-hedenbergite-diopside. The Morotu alkalic rocks may be regarded as an example in which this continuous crystallization of pyroxenes took place.

INTRODUCTION

From petrochemical studies on the alkalic rocks of the Morotu district, Sakhaline, the present author established the following course of crystallization of monoclinic pyroxenes in the alkalic rocks: diopsidic augite→soda augite→aegirine augite→aegirine. Here soda augite contains Na_{0.1}-Na_{0.2}; aegirine-augite, Na_{0.2}-Na_{0.7}; and aegirine, Na_{0.7} or more, in the pyroxene formula on the basis of O=6. Yagi (1953) concluded that diopside, hedenbergite and acmite may form complete solid solutions with some complications caused by the incongruent melting of acmite and by the transformation of hedenbergite into wollastonite solid solutions.

As a first step in the experimental study of the ternary system acmite-hedenbergite-diopside, the binary system acmite-diopside had been

studied previously (Yagi, 1958, 1962). In the present paper the results of the phase-equilibria studies in this binary system are given, with special reference to the stability relations of natural aegirines or aegirine-augites.

PREPARATION OF GLASSES

The glasses were prepared by Dr. J. F. Schairer by the method commonly employed at the Geophysical Laboratory. Quartz, ferric oxide, sodium carbonate, calcium carbonate and magnesia were the raw materials. In practice sodium disilicate made from sodium carbonate and quartz was used as the source of soda instead of sodium carbonate, in order to prevent the loss of soda by volatilization at high temperatures.

The glasses were made in 10 weight per cent intervals between the two end members, acmite and diopside. Only the mixture acmite 75, diopside 25 per cent was made by mixing equal amounts of the glasses acmite 80, diopside 20 per cent, and acmite 70, diopside 30 per cent.

The crushed glasses, heated at subsolidus temperatures of 800°–1000° C. in experiments ranging from several days to 39 days, were found to consist of minute granular crystals of pyroxene. These minute crystals of pyroxene were used as starting material for all the quenching experiments, with the exception of those in which glass was used to check the results obtained from crystalline material. The refractive indices of the glasses are given in Fig. 1 and Table 1, together with the initial treatment for crystallizing glasses. The thermocouple was calibrated frequently at fixed melting points as follows: NaCl, 800.4° C.; Au, 1062.6°; diopside, 1391.5° C.

EXPERIMENTAL RESULTS

The results of the determination of liquidus and solidus temperatures by the method of quenching are given in Table 2, and the equilibrium diagram of the system acmite-diopside is shown in Fig. 2. The incongruent melting point of acmite was determined by Bowen *et al.* (1930) at $990^{\circ} \pm 5^{\circ}$ C. with separation of hematite, and the temperature at which hematite completely disappeared was about 1287° C. Recently Bailey and Schairer (1963) gave revised values of $988^{\circ} \pm 5^{\circ}$ C. and $1310^{\circ} \pm 10^{\circ}$ C., respectively, and these values are shown in Fig. 2.

Attention is called to the presence at low temperatures of a field of complete solid solution of pyroxenes between the two end members, acmite and diopside. In the acmite-rich portion of the system, however, a wide field of hematite+liquid is present at higher temperatures and a field of pyroxene solid solutions+hematite+liquid at lower temperatures. Consequently the system seems to be truly binary only in the diopside-rich portion, where separation of hematite is not observed. The

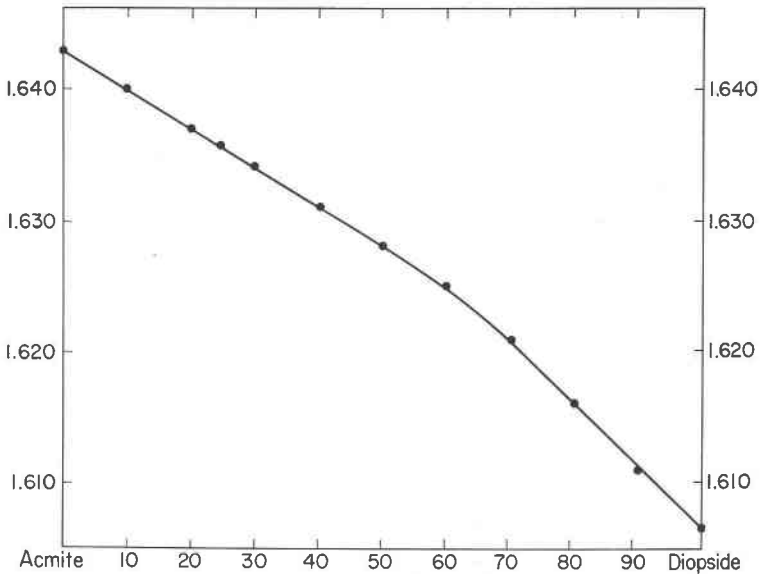


FIG. 1. Refractive indices of glasses of the system acmite-diopside (weight per cent).

solidus in this portion could not be accurately determined, however, for the following reason.

The presence of small amounts of glass within the fine granular crys-

TABLE 1. INITIAL TREATMENT FOR CRYSTALLIZING GLASSES USED IN QUENCHING EXPERIMENTS

Composition wt %		Refractive index of glass	Temperature, ° C.	Time, days
Acmite	Diopside			
100	0	1.643	800	39
90	10	1.640	850	29
80	20	1.637	850	5
75	25	1.636	800	25
70	30	1.634	850	5
60	40	1.631	900	5
50	50	1.628	900	5
40	60	1.625	900	5
30	70	1.621	1000	7
20	80	1.616	1000	7
10	90	1.611	1000	7
0	100	1.607	—	—

TABLE 2. RESULTS OF QUENCHING EXPERIMENTS

Composition wt %		Temperature, ° C.	Time, hrs	Results
Acmite	Diopside			
90	10	1240	24	All glass
90	10	1235	48	Very rare hm in glass
90	10	1080	48	Lots hm in glass
90	10	1075	72	Moderate Amount hm and very rare px in glass
90	10	1070	24	Moderate amount hm and rare px in glass
90	10	1000	24	Lots px, rare hm, and glass
90	10	990	96	Lots px and small amount glass
90	10	900	168	Lots px and very small amount glass
80	20	1215	4	All glass
80	20	1210	4	Very rare hm in glass
80	20	1150	48	Moderate amount hm in glass
80	20	1145	48	Moderate amount hm and rare px in glass
80	20	1005	72	Lots px, very rare hm, and small amount glass
80	20	1000	24	Lots px and very small amount glass
80	20	900	168	Well fritted cake
75	25	1190	24	All glass
75	25	1185	24	Very rare hm in glass
75	25	1175	24	Rare hm in glass
75	25	1170	67	Moderate amount px and small amount hm in glass
75	25	1030	96	Lots px and rare hm in glass
75	25	1025	48	Lots px, very rare hm, and small amount glass
75	25	1020	96	Lots px and very small amount glass
75	25	925	48	Barely fritted cake
75	25	915	48	Loose powder
70	30	1200	72	All glass
70	30	1195	72	Very rare px in glass
70	30	1165	48	Lots px in glass
70	30	1160	48	Lots px and very rare hm in glass
70	30	1035	48	Lots px, very rare hm, and small amount glass
70	30	1030	96	Lots px and very small amount glass
70	30	900	168	Well fritted cake
60	40	1225	24	All glass
60	40	1220	72	Small amount px in glass
60	40	1090	24	Lots px and small amount glass
60	40	1080	48	Lots px, very rare hm, and small amount glass
60	40	1075	96	Lots px and small amount glass
60	40	925	72	Slightly glazed cake
50	50	1265	5	All glass
50	50	1260	19	Very rare px in glass
50	50	1100	168	Lots px and small amount glass
50	50	925	72	Slightly glazed cake
40	60	1290	20	All glass
40	60	1285	21	Very rare px in glass
40	60	950	168	Slightly glazed cake
30	70	1315	5	All glass
30	70	1310	22	Moderate amount px in glass
30	70	950	168	Slightly glazed cake
20	80	1335	3	All glass
20	80	1330	21	Moderate amount px in glass
20	80	950	168	Well fritted cake
10	90	1365	16	All glass
10	90	1360	4	Moderate amount px in glass
10	90	1150	192	Slightly glazed cake
10	90	1055	336	Well fritted cake
10	90	950	168	Barely fritted cake

hm, hematite; px, pyroxene solid solution.

tals is usually very difficult to detect, and the method of fritting is generally adopted as an indication of the presence of liquid. The charge in the Pt envelope usually remains completely powdery if there is no glass present but becomes fritted if even a small amount of liquid is present. Therefore, the degree of fritting is used to locate the solidus temperatures. This method is not very accurate, however, and proved to be very unsatisfactory for mixtures richer in diopside than 40 per cent. As shown in Table 2 many mixtures formed well fritted cakes at temperatures as

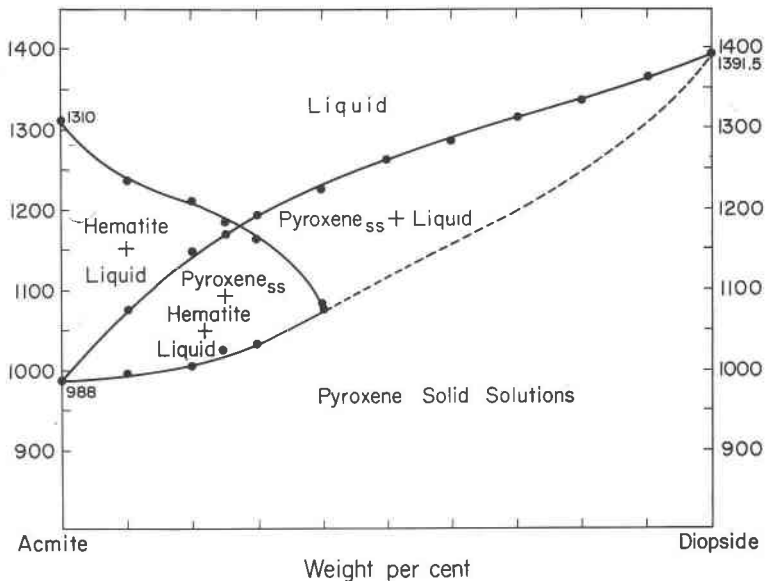


FIG. 2. Equilibrium diagram of the system acmite-diopside.

low as 900°–925° C., suggesting the presence of small amounts of liquids. The behavior of iron oxides in silicate melts studied by Yagi (1964) shows that ferrous iron is always present, even when heated in air, and increases with increasing temperature above 1100° C. Some ferrous iron, therefore, is assumed to be present in the mixtures in addition to the prevailing ferric iron, and the beginning of melting at temperatures as low as 900° C. is probably due to the presence of this ferrous iron. The system can be considered binary only when the small amount of ferrous iron is arbitrarily regarded as ferric iron.

In mixtures richer in acmite than 60 per cent, however, the temperature of the disappearance of hematite, which coincides with the solidus, could be accurately determined.

OPTICAL PROPERTIES OF SYNTHETIC PYROXENES

It is generally observed in silicate melt studies that Fe^{3+} or Fe^{2+} ions favor the crystallization of an iron-bearing silicate, *e.g.* fayalite, which forms fibrous crystals even during quenching. This is not the case in the present system, however, since pure diopside crystallizes into much better developed crystals than pure acmite or acmite-rich pyroxenes. Usually the prisms (110), (100), (010), (111) or ($\bar{1}\bar{1}\bar{1}$) are well developed, resulting in stout prismatic crystals. Cleavages are well developed parallel to (110), and twinned crystals are rarely observed.

TABLE 3. OPTICAL PROPERTIES OF PYROXENE SOLID SOLUTIONS

Composition, wt. %		α	β	γ	$2V_z$, degrees	$c \wedge Z$, degrees
Acmite	Diopside					
100	0 ¹	1.776	1.816	1.836	120	98
90	10	1.767	—	1.823	117	94
80	20	1.759	—	1.810	114	88
70	30	1.746	—	1.796	111	85
60	40	1.736	—	1.782	104	80
50	50	1.727	—	1.769	98	72
40	60	1.710	—	1.749	87	65
30	70	1.696	—	1.730	75	56
20	80	1.682	1.693	1.712	69	45
10	90	1.671	1.683	1.700	61	42
0	100 ²	1.664	1.672	1.695	59	38

¹ Determined by Merwin (Washington and Merwin, 1927).

² Determined by Barth (1931).

Under the microscope diopside is colorless, acmite is brownish yellow, and the intermediate members show colors intermediate between the two. The pleochroism in acmite is: X , yellow; Y , slightly greenish yellow; Z , slightly brownish yellow; absorption, $Z > Y > X$. The beautiful grass green or bluish green color common in natural aegirines was not observed in the synthetic acmitic pyroxenes. Refractive indices were determined using liquids of high refractive index devised by Meyrowitz and Larsen (1951). The determination of β was difficult except in favorable cases (Table 3). The refractive indices increase with increase in acmite, as shown graphically in Fig. 3.

The optic angle was determined on a universal stage using crystals cemented on a glass slide. The dispersion of the optic angle is $v > r$ in crystals varying from pure diopside to diopside 70, acmite 30 per cent, and $r > v$ in crystals varying from diopside 70, acmite 30 per cent, to pure

acmite. The extinction angle $c \wedge Z$ was determined on well-formed crystals. The z axis always lies within the obtuse angle of β ; the x axis lies within the obtuse angle of β in crystals varying from acmite to acmite 90, diopside 10 per cent, but in crystals richer in diopside than acmite 80, diopside 20 per cent, it lies within the acute angle of β .

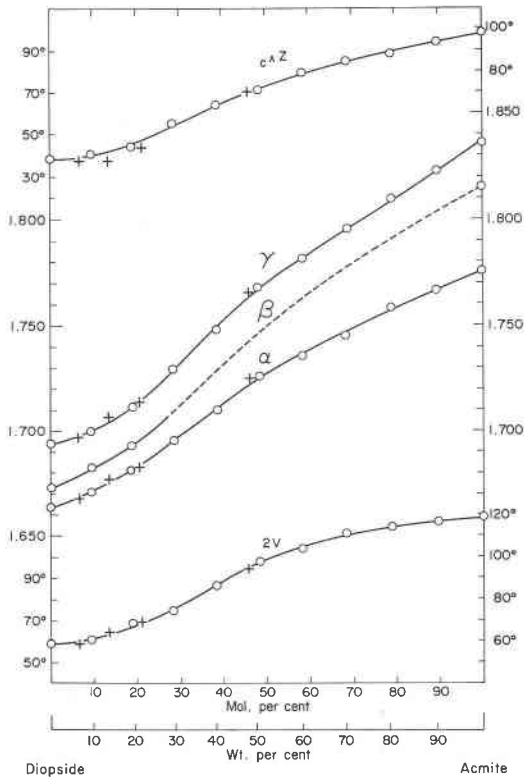


FIG. 3. Optical properties of synthetic pyroxenes of the system acmite-diopside. Circles, present data; crosses, Ostrovsky's data.

Ostrovsky (1946) also determined the optical properties of synthetic pyroxenes of this series having 6.59, 13.64, 20.89, and 45.68 mole per cent of acmite. His results, when plotted on the diagram, show close agreement with the present determinations. Nolan and Edgar (1963) conducted an x-ray investigation of synthetic pyroxenes in the system acmite-diopside, crystallized at 750° C. and 1000 kg/cm² water-vapor pressure. Their results showed a continuous variation in the lattice parameters of these pyroxenes.

AEGIRINE-AUGITES FROM MOROTU ALKALIC ROCKS

Recently Aoki (1964) made a detailed genetic investigation of clinopyroxenes from alkalic rocks of Japan and its surrounding regions. He showed the presence under igneous conditions of a wide region of im-

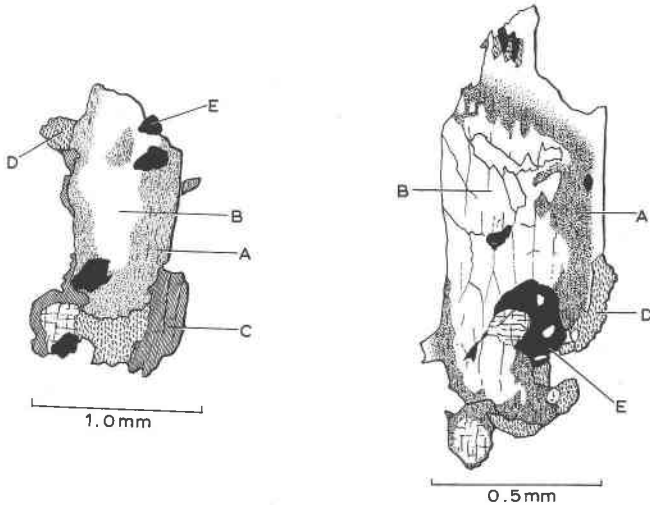


FIG. 4. Zoned structure of aegirine-augite in the syenite (8203) from the Morotu River, Morotu district. A, aegirine-augite 8203A; B, soda augite 8203B; C, arfvedsonite; D, secondary alteration product; E, iron ores.

miscibility, separating aegirines or aegirine-augites from calcium-rich pyroxenes. Since aegirine-augite No. 8203 of the Morotu alkalic rock studied by Yagi (1953) falls in the central portion of this suggested immiscibility region, a more detailed investigation of this pyroxene was made, with the following results.

The pyroxene in question forms an important constituent of a syenite (8203) from the middle reaches of the Morotu River in the central portion of the Morotu district. The rock is composed mainly of micropertite, surrounded by a clear mantle of albite, and the alkali pyroxene in question, associated with small amounts of arfvedsonite, hastingsite, lepidomelanitic biotite and iron ores. The chemical composition of the micropertite, recalculated as water-free, is given in Table 6. The alkali pyroxene shows distinct zonal structure, composed of a pale-colored core of soda augite and a greenish or deep greenish aegirine-augite mantle (Fig. 4). The boundary between the two parts is very sharp, especially under crossed polars, but narrow intermediate zones of transitional op-

tical properties are locally present, as shown below, and no resorption phenomenon is observed between them.

		Core	Margin
Extinction angle	$c \wedge Z$	48°→55°→60°→69°	
Optic angle	$2V(+)$	45°→58°→65°→81°	

The pyroxene sometimes shows parallel intergrowth with arfvedsonite in

TABLE 4. CHEMICAL COMPOSITION OF AEGIRINE-AUGITE 8203
FROM A SYENITE, MOROTU DISTRICT

	1	2	3		1a	2a
SiO ₂	50.49	50.10	44.26	Si	1.959	1.947
TiO ₂	0.63	0.76	2.38	Al	0.041	0.037
Al ₂ O ₃	0.99	0.79	4.76	Ti	—	0.016
Fe ₂ O ₃	12.75	4.60	15.48			
				Al	0.006	—
FeO	11.58	13.78	11.53	Ti	0.019	0.007
MnO	0.54	0.59	0.70	Fe ³⁺	0.372	0.135
MgO	4.60	8.07	4.10	Fe ²⁺	0.374	0.445
CaO	13.78	19.03	10.64	Mn	0.019	0.019
				Mg	0.268	0.471
Na ₂ O	4.92	1.91	3.76	Ca	0.572	0.793
K ₂ O	tr.	tr.	0.85	Na	0.368	0.145
H ₂ O ⁺	0.00	0.46	n.d.			
H ₂ O ⁻	0.17	0.27	2.12	Z	2.000	2.000
P ₂ O ₅	0.09	0.11	n.d.	WXY	1.998	2.015
				0	6.000	6.000
Total	100.54	100.47	100.58			

1. Aegirine-augite 8203A; outer part of zoned aegirine-augite from syenite 8203, Morotu River, Morotu district. Analyst, M. Chiba.
 2. Soda augite 8203B; inner core of zoned aegirine-augite from syenite 8303, Morotu River, Morotu district. Same rock as above. Analyst, M. Chiba.
 3. Bulk composition of aegirine-augite 8203 as a whole, with a considerable amount of impurities. Same locality (Yagi, 1953). Analyst, K. Yagi.
- 1a and 2a. Atomic proportions of 8203A and 8203B, respectively.

the marginal parts and usually carries inclusions of iron ores. Rarely a deep reddish brown alteration product is enclosed, which is believed to be derived from aenigmatite. This mineral occurs rather abundantly in some facies of the syenites in these environs. Therefore, the following paragenetic relation is noted: aenigmatite (in the core)→aegirine-augite→arfvedsonite→aegirine. The bulk composition of these zoned pyroxene crystals (No. 8203) is given in Table 4.

When carefully examined under the microscope the analyzed sample

was found to contain considerable amounts of impurities of microperthite and iron ores. Therefore, the host rock was crushed into finer powder, less than 150 mesh in size, and the pyroxene fraction was further separated by an isodynamic separator. Then this fraction was further separated into a denser aegirine-augite fraction (8203A) and a less dense soda-augite fraction (8203B) by means of Clerici solution. The samples were now very pure, and the impurities in both fractions were estimated less than 1 per cent under the microscope. The optical properties of these samples are given in Table 5. The results of the chemical analysis of these fractions are shown in Table 4, together with the atomic proportions based on $O=6$. A considerable difference is noted between the two frac-

TABLE 5. OPTICAL PROPERTIES OF AEGIRINE-AUGITE 8203A AND SODA AUGITE 8203B

Aegirine-augite 8203A	Soda augite 8203B
α 1.715~1.720 \pm 0.003	1.706~1.709 \pm 0.003
β 1.720~1.725	1.718~1.721
γ 1.737~1.742	1.735~1.739
2V(+) 81°-82°	45°-56°
$c \wedge Z$ 69°	48°
α max and γ min are calculated from α min, β range and γ max.	
Pleochorism	
X pale bluish green	pale green
Y pale green	pale green
Z yellowish green	pale yellowish green

tions. Especially remarkable is the increase of Na_2O and Fe_2O_3 from 8203B to 8203A: Na_2O , 1.91 \rightarrow 4.92; Fe_2O_3 , 4.60 \rightarrow 12.75. That the fractions of these pyroxenes do not add up to the bulk analysis is ascribed mainly to the presence of impurities and possibly also to some errors in the bulk analysis, as shown by the following calculation.

Since the two fractions obtained by the separation are nearly equal in amount, the bulk composition will be the mean of the two analyses, as calculated water free and P_2O_5 free in Table 6. The chemical composition of the microperthite enclosed is also given in Table 6. The exact chemical composition of the iron ores in these rocks is unknown. These iron ores are usually composed of an intimate intergrowth of magnetite, ilmenite and hematite, and the cleanest sample of iron ores obtained from a monzonite of this district shows 26 per cent of TiO_2 (Yagi, 1953). Therefore the bulk composition of these iron ores enclosed in the pyroxene can probably be represented by a titanomaghemite, for instance the one from an olivine-titanaugite-analcite dolerite from Atumi district, Japan, given in Table 6 (Katsura and Kushiro, 1961). Since the exact amounts of these

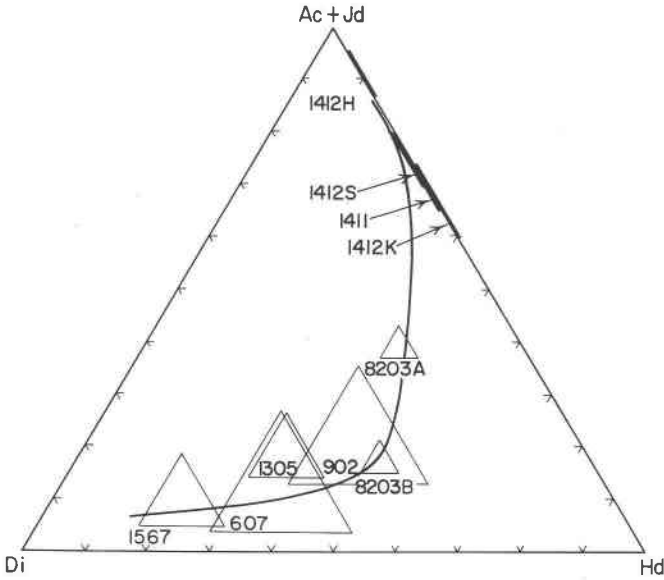


FIG. 5. Plot of pyroxenes from alkalic rocks of Morotu district in Ac+Jd-Hd-Di diagram using a calculation method proposed by Yoder and Tilley (1962). Numbers are those given by Yagi (1953).

TABLE 6. CHEMICAL COMPOSITION OF AEGIRINE-AUGITE, MICROPERTHITE, AND TITANOMAGHEMITE IN SYENITE 8293, MOROTU RIVER, MOROTU DISTRICT. RECALCULATED TO 100.00 WT %

	1	2	3	4	5
SiO ₂	50.32	65.94	—	46.85	44.96
TiO ₂	0.69	0.14	24.85	3.04	2.42
Al ₂ O ₃	0.89	19.71	—	2.68	4.83
Fe ₂ O ₃	8.68	0.75	62.09	13.23	15.72
FeO	12.69	—	13.06	11.46	11.71
MnO	0.56	—	—	0.45	0.71
MgO	6.33	0.05	—	5.07	4.16
CaO	16.42	0.68	—	13.21	10.81
Na ₂ O	3.42	7.80	—	3.52	3.82
K ₂ O	tr.	4.93	—	0.49	0.86

1. Mean value of 8203A and 8203B in equal amounts.
2. Chemical composition of microperthite (Yagi, 1953).
3. Chemical composition of titanomagemite from an olivine-titanaugite-analcite dolerite, Atumi, Japan (Katsura and Kushiro, 1961).
4. Chemical composition of a mixture composed of aegirine-augite (1) 80%, microperthite (2) 10%, and titanomagemite (3) 10%.
5. Chemical composition of bulk aegirine-augite 8203 (Yagi, 1953).

impurities are not known, it is estimated roughly that micropertthite and titanomaghemite are present to the amount of 10 per cent each in the analyzed samples. Then the bulk composition of these mixtures is calculated as shown in the table.

When this value is compared with the composition of the bulk pyroxene, the correspondence is not satisfactory, and some discrepancy is still noted in the contents of SiO_2 , Al_2O_3 and CaO . This is probably due to an error in the original bulk analysis, which was made on a very small amount of sample.

Since the pyroxenes from the alkalic rocks are free from "enstatite" or "ferrosilite" molecules, Yoder and Tilley (1962) proposed a calculation method modified after the method adopted by Yagi (1953). The result of this calculation of the pyroxenes from the Morotu alkalic rocks, including the two new analyses, is shown in Fig. 5. Note that the "error of closures" for the aegirine-augite 8203A and soda augite 8203B is quite small. When the new diagram is compared with the original one (Yagi, 1953) or the modified one (Yoder and Tilley, 1962), no substantial change is observed in the trend of pyroxene crystallization in the Morotu rocks. Therefore, the following trend, already established, is supported by this reinvestigation: diopsidic augite \rightarrow soda-augite \rightarrow aegirine-augite \rightarrow aegirine.

The new analyses were also calculated by the method proposed by Aoki (1964), with the result shown in Fig. 6, in which the immiscible region suggested by him is also given. It is remarkable that the composition of the aegirine-augite 8203A, which itself is homogeneous as shown by the optical properties and by the x -ray diffraction pattern, lies in the central portion of this immiscible region, whereas the soda augite 8203B lies within the Ca-rich pyroxene field and close to the boundary curve. Aoki tacitly suggested that actual pyroxene compositions of the Morotu magma lie within either field *A* (aegirines) or field *B* (diopside-hedenbergite solid solutions), although he suggested that the average pyroxene composition of the Morotu magma changes continuously from augite to aegirine. Therefore, aegirine-augite 8203A distinctly shows that pyroxenes whose composition lies within the proposed immiscibility region do exist under some igneous conditions.

OCCURRENCE OF AEGIRINE-AUGITES, AEGIRINES AND ACMITES IN NATURE

It was found that some natural aegirine-augites or related alkali pyroxenes have chemical compositions that plot in Aoki's immiscibility region. Brief descriptions are given below on such pyroxenes, which plot in the immiscibility region of the Na- Fe^{2+} -Mg diagram, with the numbers in parentheses corresponding to those in Fig. 6.

Vanadiferous aegirine (1) and aegirine (2), both from syenitic pegmatite, Libby, Montana (Goranson, 1927), represent the products of a later hydrothermal solution. A similar origin is also attributed to an aegirine-augite (9) from a gneissose nepheline syenite of Iron Hill, Colorado (Larsen, 1942). It has a chemical composition very similar to the aegirine-augite 8203A. Aegirine-augite (10) from a feldspar nepheline pyroxenite

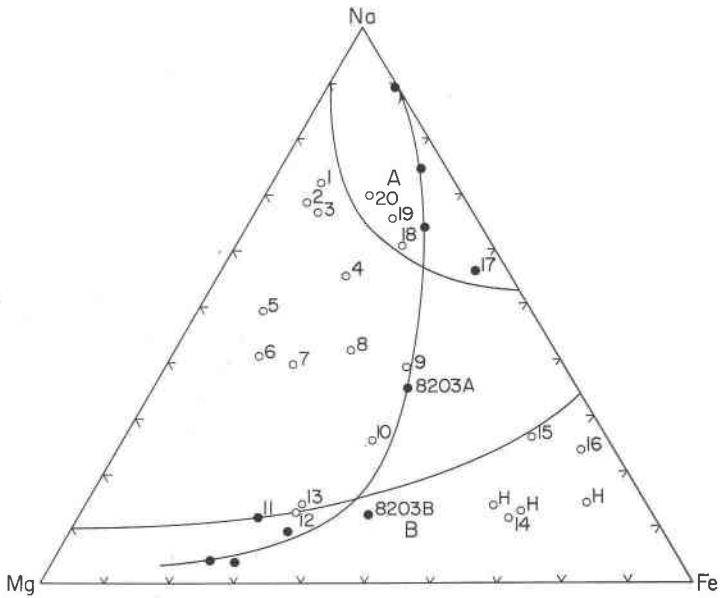


FIG. 6. Composition of some aegirine augites or related pyroxenes plotted on the Na-Fe-Mg diagram. Aoki's immiscibility region separating A- and B-fields is indicated by two curves. Numbers refer to descriptions in text. Solid circles and arrow indicate pyroxenes from alkalic rocks of Morotu district. H, pyroxenes from nepheline syenites of Haliburton-Bancroft district; A, field of acmite-rich pyroxenes; B, field of diopside-hedenbergite solid solutions.

of Iron Hill seems also to be of hydrothermal origin, although the evidence is less convincing. Larsen noticed that similar aegirine-augites are common in the hydrothermally altered carbonatite dikes of the same district.

Aegirine-augite (8) from syenite pegmatite of Ilmen Mountain (Zavaritzky, 1946) forms characteristic graphic intergrowth with micropertthite, sometimes more than 10 cm in size. It is believed that aegirine-augite was formed from micropertthite by hydrothermal solution. Blanfordite, a manganese-rich aegirine-augite (3), from Chikla Bhandara is found in the pegmatite, which also contains manganese ores (Bilgrami, 1956). Its

origin is probably related to an assimilation phenomenon. Aegirine-augite (4) of Glen Lui (McLachlan, 1951) is a unique example of such pyroxene in granulite and is evidently the product of soda metasomatism on the pre-existing schist by highly sodic pegmatitic solutions.

Aegirine-augites are also found as essential constituents of some crystalline schists from Shikoku, Japan. Aegirine-augite from Besshi (5) comes from magnesioriebeckite-aegirine-augite-garnet-hematite-albite-quartz schist of epidote amphibolite facies, and that from Bizan (6) is present in the magnesioriebeckite-aegirine-augite-muscovite-garnet-hematite-chlorite-quartz schist of glaucophane schist facies (Banno, 1959). Note that diopside and acmite molecules are nearly equal in these aegirine-augites and the hedenbergite molecule is exceedingly low. Actually, of all aegirine-augites so far known throughout the world, these have a chemical composition closest to the binary system acmite-diopside. Aegirine-augite from Laacher See (7) is found in the schist xenoliths enclosed in nepheline basaltic magma and is believed to have been derived by pyrometasomatism with addition of soda (Brauns, 1923).

Soda augites from a monzonite of Morotu (11; Yagi, 1953), from an ijolite pegmatite of Homa Bay (13; Pulfrey, 1950), and from a hornblende-biotite diorite of Rossie, Adirondack (12; Agar, 1923), are all similar in composition and are regarded as igneous in origin, although the last may have been formed as a result of contact metamorphism in this region.

Some soda-rich pyroxenes within the Ca-pyroxene field and close to its boundary should be mentioned. Acmitic diopsidic hedenbergite from a nepheline syenite (14) of Salem Neck (Washington and Merwin, 1927) has a composition similar to that of the aegirine-augite from Iron Hill (10) and is probably of igneous origin. It has a high soda content but not enough ferric oxide to make acmite, and a considerable amount of jadeite is calculated. It is suspected that alkali feldspar or nepheline contaminated the analyzed sample. An aegirine-hedenbergite from alkali granite of Camas Malag (15) probably formed as a result of contact metamorphism between dolomite and granite (Tilley, 1949). A hedenbergitic aegirine from alkali granite of Ambasibitika, Madagascar (16), is a pseudomorph after riebeckite (Lacroix, 1922) and has a composition similar to that of the sample from Camas Malag.

In the following some aegirine-augites in the *A*-pyroxene field and close to its boundary will be briefly mentioned. Aegirine-augite from a syenite of Morotu (17) forms the core of a zoned aegirine crystal in the arfvedsonite-aegirine syenite (Yagi, 1953). Aegirine-augites (18 and 19) come from the nepheline syenite pegmatite of Låven Island, Norway. The analysis (19) was made by Dr. Aoki for this study on the sample col-

lected by the author, which is similar to the sample (18) analyzed by Washington (Washington and Merwin, 1927). Aegirine-augite (20) from Magnet Cove is also from nepheline syenite pegmatite (Washington and Merwin, 1927).

Most of the aegirines or acmites are reported from syenitic or granitic rocks or their pegmatites and are associated with riebeckite or arfvedsonite as described below. They are believed to be of igneous origin.

Aegirine-riebeckite granite: Quincy, Massachusetts (Washington and Merwin, 1927; Aoki, 1964); Kigom Hills, Nigeria (Greenwood, 1951; Jacobson, Macleod, and Black, 1958).

Nepheline syenite: Kangerdluarsuk, Greenland (Washington and Merwin, 1927); Alter Pedroso, Portugal (Burri, 1928); Assynt, Africa (Sabine, 1950).

Aegirine syenite: Morotu, Sakhalin (Yagi, 1953); Kogan-zan, Korea (Ito, 1955); Iwagishima, Japan (Ishibashi, 1964).

Aegirines or aegirine augites are also found in crystalline schists. Suzuki (1934) reported occurrences of aegirine-augites and aegirines from soda-rich crystalline schist of the glaucophane schist facies in Hokkaido, Japan, but no chemical data were available. In this category belongs the aegirine found in the aegirine-riebeckite schist, Gout Creek, New Zealand, which has the highest acmite content (97.5 per cent) of any aegirine ever reported (White, 1962). An aegirine-augite associated with arfvedsonite in the paragneiss from Seal Lake, Labrador, seems to have a similar origin (Nickel and Mark, 1965). Acmites with very high acmite contents were also found within veins cutting the iron formations of Cuyuna Range, Minnesota. They are hydrothermal in origin (Grout, 1946). Rather perplexing is the presence of authigenic acmite and riebeckite in the Green River formation, deposited at or near room temperature and at low P_{O_2} (Milton and Eugster, 1959).

On the other hand, aegirines are rarely reported from the salic alkali volcanic rocks. Tomita (1935) mentioned the occurrence of aegirines in some rhyolitic rocks of Oki Island, Japan, but no chemical data were available. In this respect the occurrence of aegirine-arfvedsonite comendite from Oga peninsula, Japan, should be mentioned (Yagi and Chihara, 1963). The aegirine has the following optical properties: $\alpha = 1.752$, $\gamma = 1.810 \pm 0.003$, $c \wedge X = 5^\circ$, $2V(-) = 66^\circ$. Its composition is estimated to be about 80 per cent acmite.

A chemical analysis was not made because of the scarcity of the sample. A pantellerite from Pantelleria and a trachyte from Sardinia were reported to contain acmite (Chayes and Zies, 1962), but all pyroxenes in the pantellerites from Pantelleria were found to be soda hedenbergites by the chemical analysis of Carmichael (1962). There is a possibility that some of the small pyroxene crystals in such volcanic rocks were identified

as "acmite" only from their color, pleochroism or extinction angles (Chayes, pers. comm., 1965). Additional careful observations on the pyroxenes in these volcanic rocks are necessary to solve the problem of the stability of aegirine or acmite.

Suzuki (1938) reported that aegirine-augite is formed from augite adjacent to the later veins of natolite in the analcite dolerite of Nemuro district, Japan.

EXPERIMENTAL STUDIES ON SYNTHETIC ALKALIC PYROXENES

Schüller (1958) conducted experimental studies on the formation of solid solutions of acmite with other pyroxene molecules such as diopside, hedenbergite, $\text{NaCr}^{3+}\text{Si}_2\text{O}_6$, $\text{NaV}^{3+}\text{Si}_2\text{O}_6$, $\text{CaCoSi}_2\text{O}_6$, $\text{CaZnSi}_2\text{O}_6$. From the strong zonal structure and the regular change of extinction angles with compositions, he concluded that acmite and diopside form extensive solid solution. He found, however, that limited amounts of hedenbergite are soluble in acmite and concluded that they formed only limited solid solution. He did not give a definite range of miscibility in the acmite-hedenbergite system, since his experiments covered only the region from acmite 100 per cent to acmite 87.6, hedenbergite 12.4 weight per cent. The *x*-ray investigation of Nolan and Edgar (1963) on hydrothermally synthesized pyroxenes of the acmite-diopside series shows continuous variation of the unit-cell sizes of these pyroxenes, supporting the occurrence of complete solid solution of this series under high P_{O_2} and low temperatures.

Carmichael (1962), from the distribution of pyroxenes plotted on an Na-Fe²⁺-Mg diagram, suggested that solid solutions of the acmite-hedenbergite series do occur and are more common in natural pyroxenes than between diopside and acmite.

The stability relations of acmite were recently studied experimentally by Bailey (1963) at 2 and 5 kb, using cold-seal bombs with unbuffered charges, and with Ni/NiO and $\text{Fe}_3\text{O}_4/\text{Fe}_2\text{O}_3$ buffers. It was found that unbuffered runs yielded results closely similar to those obtained with Ni/NiO buffers, whereas the high P_{O_2} provided by the $\text{Fe}_3\text{O}_4/\text{Fe}_2\text{O}_3$ buffer resulted in a rise of about 60° C. in the melting curve of acmite. This fact supports the inference that high P_{O_2} favors the crystallization of acmite from the melts. No difference was noticed in the optical properties and the unit-cell sizes of the acmite formed hydrothermally and that formed at 1 atm. It was also found that the incongruent melting of acmite appears to persist under pressures up to 10 kb, indicating that the primary crystallization of acmite is difficult from magmas at higher temperatures, even at considerable depths in the crust.

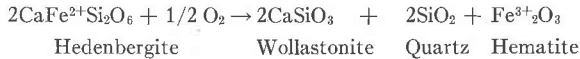
On the other hand, Schairer *et al.* (1954), in their study on the system

$\text{Na}_2\text{O}-\text{FeO}-\text{SiO}_2$ in equilibrium with iron crucibles, found "acmite," by x-ray powder patterns, in a small area labelled "several fields" on their diagram although the presence of acmite was not definitely confirmed. If the crystals are really acmite, it would suggest that acmite could be formed even at very low P_{O_2} in some cases (Yoder, pers. comm., 1965). More detailed study is expected on this problem.

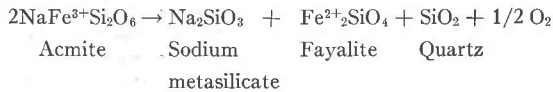
The experimental study by Ernst (1962) on the stability relations between riebeckite-arfvedsonite solid solutions, acmite, and aenigmatite can be best applied to the Morotu rocks, where aegirine or aegirine-augite is intimately associated with arfvedsonite and rarely also aenigmatite. From the field occurrence of these rocks it is evident that they were solidified in a rather shallow place, probably not more than 2 or 3 km in depth. Since the total pressure in such cases is expected to be of the order of 10^3 bars, the isobaric $f_{\text{O}_2}-T$ diagram at 500 bars presented by Ernst (1962, Fig. 11B) could be applied to the present study.

The colored minerals in the syenite 8203 are zoned aegirine-augite with small amounts of arfvedsonite, hastingsite, lepidomelanitic biotite and iron ores. Rarely, deep reddish brown alteration products are enclosed in the aegirine-augite. From their modes of occurrence and features, they are interpreted to have been probably derived from aenigmatite. Actually this mineral is present in many other syenites in this district, and therefore a paragenetic relation soda augite \rightarrow aenigmatite \rightarrow aegirine-augite \rightarrow arfvedsonite \rightarrow aegirine may be explained by Ernst's diagram as follows. At the time of crystallization of soda augite and aenigmatite, the temperature was probably about $700^\circ-750^\circ$ C. and the f_{O_2} was about 10^{-21} bars, which is, according to Krauskopf (1959), a reasonable lower limit for most magmatic gases. With decreasing temperature aenigmatite was followed by aegirine-augite, which formed the bulk of these pyroxenes, and finally arfvedsonite formed stably at about 600° C. or less. In the syenite 8203 the crystallization ended by the separation of arfvedsonite. In most of the syenites, however, which have higher contents of alkalis, the crystallization continued with increasing water and other volatile components. Since some iron ores have become hematite, it is expected that the boundary curve between hematite and magnetite should have been crossed. Consequently f_{O_2} increased to about 10^{-15} or higher and the acmite field was attained, separating aegirine or nearly pure acmite in the latest stage.

The above-mentioned experimental and petrological evidence is sufficient to show the stability of acmite or aegirine at high P_{O_2} . Although the stability of hedenbergite at various P_{O_2} 's has not been determined, it is unstable at high P_{O_2} , and one possible reaction showing the dissociation is as follows:



The lower limit of P_{O_2} for acmite is also not known, but it is possible that acmite is unstable at lower P_{O_2} where hedenbergite is stable up to about 965°C . (Bowen *et al.* 1933). Under such conditions acmite may dissociate as follows:



In both cases the wollastonite and sodium metasilicate molecules are expected to go into pyroxene solid solutions, or with Al_2O_3 available, into plagioclase solid solutions in the natural rocks.

GENETIC CONSIDERATIONS

As shown in the preceding sections, the pyroxenes of the system acmite-hedenbergite-diopside cover a wide range with respect to both composition and mode of occurrence. Vogt (1924) plotted all the analyses of pyroxenes available at that time in the acmite-hedenbergite-diopside diagram, without regard for their origin, and he was led to the conclusion that there should be a complete solid solution in this ternary system. Since it seems imperative to discriminate between the modes of occurrence for the present discussion, all chemical analyses of pyroxenes of this series are plotted in the Na- Fe^{2+} -Mg diagram (Fig. 7) in four groups according to their host rocks as follows: volcanic rocks, plutonic rocks, crystalline schists, and pegmatites.

It is clear from this diagram that pyroxenes of different kinds of rocks have their own fields of composition. Those from alkali volcanic rocks have very limited composition, low in soda. Even the phenocrysts in the pantellerites high in soda are sodic ferrohedenbergite with less than 20 per cent of Na component (Carmichael, 1962), and they represent the goal of such volcanic pyroxenes. Within this narrow area the following trend, as discussed by Wilkinson (1956) and by Aoki (1964), is observed: diopside-augite \rightarrow augite \rightarrow ferroaugite \rightarrow hedenbergite.

All the pyroxenes from syenites, alkali granites, or other plutonic alkali rocks have higher Na component than those from volcanic rocks; and most of these pyroxenes are usually aegirine-augites and rarely aegirines or soda augites. In this connection nepheline syenites of Haliburton-Bancroft province, Canada, are remarkable in having only sodic hedenbergites low in acmite. According to Tilley and Gittins (1961) these rocks have a magmatic origin and their temperatures were probably higher and their P_{O_2} lower than the ordinary nepheline syenites.

Pyroxenes from pegmatites of alkali syenites or granites are always rich in Na component, and sometimes nearly pure acmite or aegirine is found. Therefore, a rather regular shift of the compositional fields of alkali pyroxenes according to their parentage is noticed beginning from those of volcanic rocks, through plutonic rocks, to pegmatites.

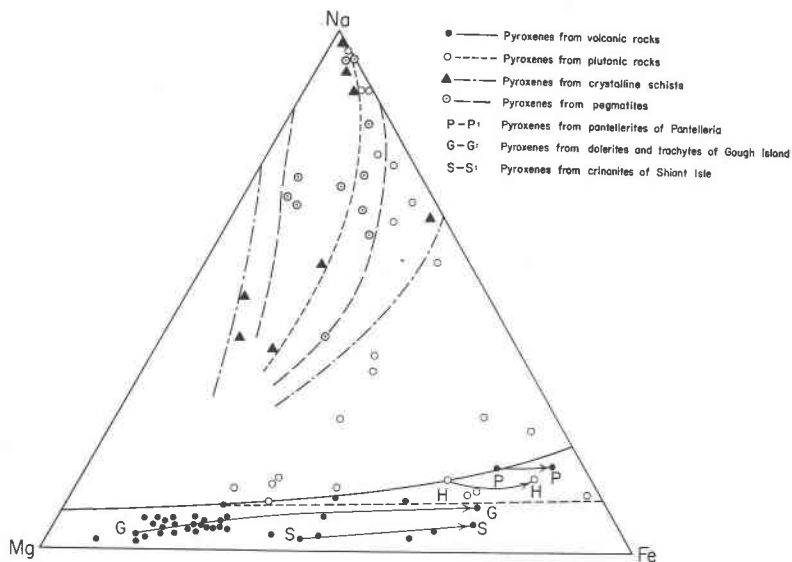


FIG. 7. Compositions of all pyroxenes of the acmite-hedenbergite-diopside system plotted in the Na-Fe-Mg diagram. Pyroxenes of different origin are indicated by different symbols, and their fields are bounded by different lines. H, the same as in Fig. 6.

Usually P_{O_2} is low in the volcanic magmas as compared with the pegmatitic liquids, which are rich in water or other volatile components. This is supported by the predominance of Fe^{2+} -rich minerals such as hedenbergite, fayalite or, ferropigeonite in the volcanic rocks, which are stable only at low P_{O_2} . Experiments show that the P_{O_2} will be 10^{-18} bars or less at $900^\circ C$. with buffers containing fayalite in the basaltic melts (Hamilton, Burnham, and Osborn, 1964). The experimental data show that the stability field of acmite is enlarged with increasing P_{O_2} . Therefore the very high content of acmite in pyroxenes from some pegmatites may be ascribed to the high P_{O_2} , and the low content of acmite in the volcanic pyroxenes, to the low P_{O_2} , even when the original liquid is sufficiently high in soda.

In contrast to these pyroxenes of volcanic origin, alkali pyroxenes of the metamorphic rocks, such as crystalline schists or xenoliths in volcanic

rocks, are generally higher in Na components, and their field partly overlaps those of the pyroxenes from pegmatites or from plutonic rocks. It is especially worthy of note that there are some pyroxenes from crystalline schists which are very close to the binary solid solution of acmite-diopside, for example those from crystalline schists of Shikoku, Japan (Banno, 1959). This is probably due to the chemical composition of the host rocks, rich in both MgO and Na₂O. The absence of such pyroxenes in the igneous rocks may be ascribed to the rarity of the rocks, which are rich in both Na₂O and MgO. The diversity of the composition of alkali pyroxenes from crystalline schists or other metamorphic rocks, therefore, may be related to the wide variation in chemical composition of the original rocks, as well as the wide ranges in the physical environment.

This relation can be now expressed by a solid model (Fig. 8), in which the horizontal plane gives the composition of pyroxenes and the vertical line gives the approximate measure of P_{O_2} , increasing from the bottom to the top. Temperature decreases also from the bottom to the top, but since the relation between temperature and P_{O_2} is not clear, the temperature is not indicated in this diagram.

As compared with acmite and hedenbergite, both of which are sensitively controlled by the prevailing P_{O_2} , diopside is stable throughout the whole range of variation of P_{O_2} during the crystallization of magmas or metamorphism of the rocks. It is expected, therefore, that the diopside molecule plays such a role in the pyroxene solid solutions as a "solvent," in which either hedenbergite or acmite molecule is present as "solute," and consequently a continuous solid solution relation is possible, as indicated by a continuous volume extending through this model.

As previously mentioned, however, acmite has been synthesized at various P_{O_2} 's, and it may be possible that the range of P_{O_2} tolerable for acmite is fairly wide. Because of insufficient knowledge of accurate values of P_{O_2} for acmite formation and of the absolute values of P_{O_2} in various rocks, and also because of the scarcity of pyroxenes in the central part of the Na-Fe²⁺-Mg diagram, the author does not claim that the diagram presented here is quantitative. Nevertheless, it will give some explanation of the crystallization course and stability relations of the pyroxenes of the acmite-hedenbergite-diopside series under igneous and metamorphic conditions.

As a consequence of plotting the different suites of volcanic and pegmatitic pyroxenes in one diagram, Aoki (1964, Fig. 3) concluded that Na-rich pyroxenes and Ca-rich pyroxenes are separated by an apparently large region of immiscibility. In the temperature ranges studied during the present experiments, however, no such immiscibility region was found, and it seems most likely that continuous pyroxene solid solutions

exist as indicated by a volume extending obliquely through this model. The crystallization of pyroxenes usually comes to an end at the intermediate stage, but an ideal case is represented by a continuous crystallization of pyroxenes in the Morotu alkalic rocks throughout this volume,

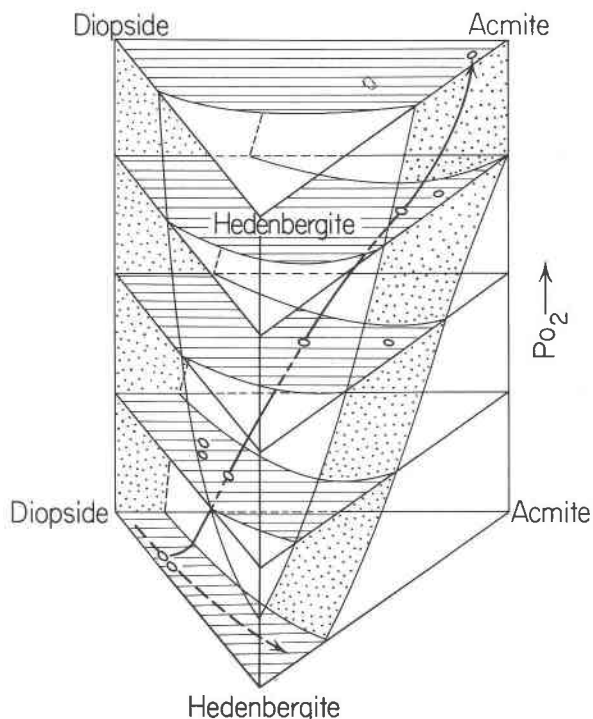


FIG. 8. Stability relations of pyroxenes of the acmite-hedenbergite-diopside system. Horizontal plane, composition; vertical line, P_{O_2} (increasing upwards). The bottom plane represents the condition of volcanic rocks and the top represents that of pegmatitic rocks.

Solid line with white circles: crystallization course of diopside augite→soda-augite→aegirine-augite→aegirine (in the pyroxenes from the alkalic rocks of Morotu district).

Broken line: crystallization course of diopside augite→augite→ferroaugite→hedenbergite (in the pyroxenes from the volcanic rocks).

as indicated by the heavy curve in the diagram. The drastic changes in chemical composition between the soda augite 8203B and the aegirine-augite 8203A may also be ascribed to a great difference in physical condition as well as the chemical composition of the liquids from which they were formed. The rare occurrence of pyroxenes similar to the aegirine-augite 8203A is probably due to the rarity of such a physical environment.

It is concluded that, if low P_{O_2} is maintained throughout, the course of crystallization of pyroxenes from volcanic magma will remain on the plane with low P_{O_2} and will proceed from diopsidic augite toward soda hedenbergite, as indicated by the broken line in Fig. 8. This includes the course diopsidic augite→augite→ferroaugite→hedenbergite or diopsidic augite→augite→ferropigeonite, both of which were discussed by Aoki (1964). If the P_{O_2} increases gradually with crystallization, as shown by the pyroxenes of the Morotu rocks, then the course goes from diopsidic augite through soda augite and aegirine-augite to aegirine, and finally pure acmite may be formed in the latest stage (Yagi, 1953).

CONCLUSION

The phase-equilibria studies in the acmite-diopside system show the presence at lower temperatures of a complete series of solid solutions between the two end members, and at higher temperatures acmite-rich pyroxenes melt incongruently to hematite and liquid. In the temperature ranges studied, no evidence of immiscibility is found in this system, although the possibility of immiscibility at much lower temperatures cannot be ruled out. In natural rocks magnetite is usually present in place of hematite. Aegirine high in acmite or pure acmite does not occur as phenocrysts even in the salic volcanic rocks rich in soda at high temperatures. In the volcanic rocks the pyroxenes are usually represented by diopsidic augite to ferroaugite, and rarely by soda ferrohedenbergite in the latest stage, all of which are low in acmite.

It was shown experimentally, however, that aegirines or acmites are stable at high P_{O_2} and at lower temperatures and consequently most of them are present in the nepheline syenites, alkali granites or their pegmatites. Sometimes similar acmitic pyroxenes are found in some crystalline schists, in which similar physical environments are probable. Aegirine-augites moderately rich in acmite are formed mostly in the syenitic rocks and also as products of hydrothermal alteration of pre-existing minerals.

From these occurrences and experimental data, a solid model is presented to explain the course of crystallization and the stability relations of the pyroxenes in the acmite-hedenbergite-diopside system. They form continuous solid solutions, which are not separated by an immiscible region into Ca-rich pyroxenes and acmite-rich pyroxenes. Since they are controlled by P_{O_2} and temperature, the course of crystallization will trend from diopsidic augite to ferroaugite in alkalic volcanic rocks, but rarely the course extends continuously from diopsidic augite through aegirine-augite to aegirine when the physical and chemical conditions are favorable for the formation of pyroxenes moderately rich in acmite.

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REFERENCES

- AGAR, W. M. (1923) Contact metamorphism in the Western Adirondacks. *Proc. Am. Phil. Soc.* **62**, 95-174.
- AOKI, K. (1964) Clinopyroxenes from alkaline rocks of Japan. *Am. Mineral.* **49**, 1199-1223.
- BAILEY, D. K. (1963) The stability relations of acmite. *Carnegie Inst. Wash. Year Book* **62**, 131-133.
- AND J. F. SCHAIRER (1963) Crystallization of the rock-forming silicates in the system $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{Fe}_2\text{O}_3-\text{SiO}_2$. *Carnegie Inst. Wash. Year Book* **62**, 124-131.
- BANNO, SHOHEI (1959) Aegirine-augites from crystalline schists in Sikoku. *Jour. Geol. Soc. Japan* **65**, 652-657.
- BARTH, T. F. W. (1931) Pyroxen von Hiva Oa, Marquesas Inseln und die Formel titanhaltiger Augite. *Neues Jahrb. Mineral. Geol., Beil. Bd. A*, **64**, 217-224.
- BILGRAMI, S. A. (1956) Manganese silicate minerals from Chikla, Bhandara District, India. *Mineral. Mag.* **31**, 236-244.
- BOWEN, N. L., J. F. SCHAIRER AND E. POSNJAK (1933) The system, $\text{CaO}-\text{FeO}-\text{SiO}_2$. *Am. Jour. Sci.* **26**, 193-284.
- J. F. SCHAIRER AND H. W. V. WILLEMS (1930) The ternary system $\text{Na}_2\text{SiO}_3-\text{Fe}_2\text{O}_3-\text{SiO}_2$. *Am. Jour. Sci.* **20**, 405-455.
- BRAUNS, R. (1923) Injizierte metamorphe Schiefer aus dem Laach-See Gebiet. *Centralbl. Min.* **1923**, 449-463.
- BURRI, C. (1928) Zur Petrographie der Natron-Syenit von Alter Pedroso. *Schweiz. Mineral. Petrol. Mitt.* **8**, 374-437.
- CARMICHAEL, I. S. E. (1962) Pantelleritic liquids and their phenocrysts. *Mineral. Mag.* **33**, 86-113.
- CHAYES F. AND E. G. ZIES (1962) Sanidine phenocrysts in some peralkaline volcanic rocks. *Carnegie Inst. Wash. Year Book* **61**, 112-118.
- ERNST, W. G. (1962) Synthesis, stability relations, and occurrence of riebeckite and riebeckite-arfvedsonite solid solutions. *Jour. Geol.* **70**, 689-736.

- GORANSON, R. W. (1927) Aegirine from Libby, Montana. *Am. Mineral.* **12**, 27-29.
- GREENWOOD, R. (1951) Younger intrusive rocks of Plateau Province, Nigeria, compared with the alkalic rocks of New England. *Bull. Geol. Soc. Am.* **62**, 1151-1178.
- GROUT, F. F. (1946) Acmite occurrences in the Cuyuna Range, Minnesota. *Am. Mineral.* **31**, 125-130.
- HAMILTON, D. L., C. W. BURNHAM AND E. F. OSBORN (1964) The solubility of water and effects of oxygen fugacity and water content on crystallization in mafic magmas. *Jour. Petrol.* **5**, 21-39.
- ISHIBASHI, K. (1964) Aegirine from Iwagi-shima, Ehime Prefecture, Japan. *Jour. Min. Soc. Japan* **6**, 361-367 (in Japanese).
- ITO, J. (1955) Recueil Mineralogique III (5), Aegirine from Hoansan in Fukushin-san district, Korea. *Jour. Min. Soc. Japan* **1**, 252-254 (in Japanese).
- JACOBSON, R. R. E., W. N. MACLEOD AND R. BLACK (1958) Ring-complexes in the Younger Granite Province of Northern Nigeria. *Geol. Soc. London, Mem.* **1**.
- KATSURA, T. AND I. KUSHIRO (1961) Titanomaghemite in igneous rocks. *Am. Mineral.* **46**, 134-145.
- KRAUSKOPF, K. B. (1959) The use of equilibrium calculations in finding the composition of a magmatic gas phase. In, *Researches in Geochemistry*, edited by P. H. Abelson John Wiley and Sons, New York, pp. 260-278.
- LACROIX, A. (1922) *Minéralogie de Madagascar*, Vol. 1 A. Challamel, Paris, p. 516.
- LARSEN, E. S., JR. (1942) Alkali rocks of Iron Hill, Gunnison County, Colorado. *U. S. Geol. Surv. Prof. Paper* **197A**
- MCLACHLAN, G. R. (1951) The aegirine granulites of Glen Lui, Braemar, Aberdeenshire. *Mineral. Mag.* **29**, 476.
- MEYROWITZ, R. AND E. S. LARSEN, JR. (1951) Immersion liquids of high refractive index. *Am. Mineral.* **36**, 746-750.
- MILTON, C. AND H. P. EUGSTER (1959) Mineral assemblages of the Green River formation. In, *Researches in Geochemistry*, edited by P. H. Abelson, John Wiley and Sons, New York, pp. 118-150.
- NICKEL, E. H. AND E. MARK (1965) Arfvedsonite and aegirine-augite from Seal Lake, Labrador. *Can. Mineral.* **8**, 185-197.
- NOLAN, J. AND A. D. EDGAR (1963) An x-ray investigation of synthetic pyroxenes in the system acmite-diopside-water at 1000 kg/cm² water-vapor pressure. *Mineral. Mag.* **33**, 625-634.
- OSTROVSKY, I. A. (1946) Optical properties of synthetic aegirine-diopside. *Acad. Sci. USSR, Belyankin Vol.* p. 505.
- PULFREY, W. (1950) Ijolitic rocks near Homa Bay, Western Kenya. *Quart. Jour. Geol. Soc.* **105**, 425.
- SABINE, P. A. (1950) The optical properties and composition of the acmitic pyroxenes. *Mineral. Mag.* **29**, 113-125.
- SCHAIRER, J. F., H. S. YODER, JR. AND A. G. KEENE (1954) The systems Na₂O-MgO-SiO₂ and Na₂O-FeO-SiO₂. *Carnegie Inst. Wash. Year Book* **53**, 123-128.
- SCHÜLLER, K. H. (1958) Das Problem Akmit-Aegirin. *Beitr. Mineral. Petrog.* **6**, 112-138.
- SUZUKI, J. (1934) On some soda-pyroxene and amphibole-bearing quartz schists from Hokkaido. *Jour. Fac. Sci. Hokkaido Univ., Ser. 4*, **2**, 339-353.
- (1938) On the occurrence of aegirine-augite in natrolite veins in the dolerite from Nemuro, Hokkaido. *Jour. Fac. Sci. Hokkaido Univ., Ser. 4*, **4**, 183-191.
- TILLEY, C. E. (1949) An alkali facies of granite at granite-dolerite contacts in Skye. *Geol. Mag.* **86**, 81-93.

- AND J. GITTINS (1961) Igneous nepheline-bearing rocks of the Haliburton-Bancroft Province of Ontario. *Jour. Petrol.* **2**, 38–48.
- TOMITA, T. (1935) On the chemical compositions of the Cenozoic alkaline suite of the Circum-Japan Sea Region. *Jour. Shanghai Sci. Inst., Sect. 2*, **1**, 227.
- VOGT, J. H. L. (1924) The physical chemistry of the magmatic differentiation of igneous rocks. *Norsk. Vid. Akad. Oslo, Math. Nat. Kl. Skr.* **15**.
- WASHINGTON, H. S. AND H. E. MERWIN (1927) The acmitic pyroxenes. *Am. Mineral.* **12**, 233–252.
- WHITE, A. J. (1962) Aegirine-riebeckite schists from South Westland, New Zealand. *Jour. Petrol.* **3**, 38–48.
- WILKINSON, J. F. G. (1956) Clinopyroxenes of alkali olivine-basalt magmas. *Am. Mineral.* **41**, 724–743.
- YAGI, K. (1953) Petrochemical studies on the alkalic rocks of the Morotu district, Sakhalin. *Bull. Geol. Soc. Am.* **64**, 769–810.
- (1958) Synthetic pyroxenes of the acmite-diopside system. *Jour. Min. Soc. Japan* **3**, 763–769 (in Japanese).
- (1962) A reconnaissance of the systems acmite-diopside and acmite-nepheline. *Carnegie Inst. Wash. Year Book* **61**, 98–99.
- (1964) Experimental petrology on pyroclastic flows. *Bull. Volcanol. Soc. Japan, Ser. 2*, **8**, 134–143 (in Japanese).
- AND K. CHIHARA (1963) Occurrence of arfvedsonite comendite from Oga peninsula, Japan. *Jour. Japan. Assoc. Mineral., Petrol., Econ. Geol.* **49**, 22–28.
- YODER, H. S., JR. AND C. E. TILLEY (1962) Origin of basaltic magmas: An experimental study of natural and synthetic rock systems, *Jour. Petrol.* **3**, 342–532.
- ZAVARITZKY, A. N. (1946) An interesting example of a syenite-pegmatite from Ilmen Mountains. *Acad. Sci. USSR, Fersman Vol.* pp. 319–325.

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