ARFVEDSONITE AND AEGIRINE-AUGITE FROM SEAL LAKE, LABRADOR

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ABSTRACT

The chemical composition and optical and physical properties of co-existing arfvedsonite and aegirine-augite are described. The arfvedsonite has a composition expressed by $(Na_{2.09}K_{0.72}Ca_{0.22})(Fe_{2.71}{}^{+2}Fe_{1.08}{}^{+3}Mn_{0.30}Mg_{0.21}Ti_{0.20}Al_{0.15}Nb_{0.01})(Si_{7.89}Be_{0.06}Al_{0.05})$-$O_{22.09}(OH)_{1.72}F_{0.19}$, and the following properties: unit-cell parameters—$a = 9.968$ Å, $b = 18.034$ Å, $c = 5.309$ Å, $\beta = 104°\,14'$; refractive indices—$\alpha = 1.685$, $\beta = 1.695$, $\gamma = 1.698$; extinction angle $X \wedge c = 29°$; $2V = -69°$; ellipticity of vibration 0.47; specific gravity 3.38; and specific magnetic susceptibility 60×10^{-6}e.m.u. The aegirine-augite has a composition expressed by $(Na_{0.77}Ca_{0.14}K_{0.02})(Fe_{0.53}{}^{+3}Fe_{0.24}{}^{+2}Al_{0.08}Ti_{0.08}$-$Mn_{0.03}Mg_{0.02}Nb_{0.02})(Si_{1.95}Al_{0.03}Be_{0.02})O_{5.85}(OH)_{0.09}F_{0.06}$, and the following properties: unit-cell parameters $a = 9.681$ Å, $b = 8.793$ Å, $c = 5.303$ Å, $\beta = 105°\,06'$; refractive indices $\alpha = 1.737$, $\beta = 1.769$, $\gamma = 1.787$; extinction angle close to zero; $2V = -74°$; specific gravity 3.517; and specific magnetic susceptibility 50×10^{-6} e.m.u.

The distribution of major cations between the two minerals is consistent with the theory that increasing polymerization of silicon-oxygen terahedra favours the incorporation of connecting ions with lower electronegatives.

INTRODUCTION

The Seal Lake area in Labrador has aroused considerable interest within the past few years because of the occurrence of unusual beryllium- and niobium-bearing minerals. The geology of the area has been described by Brummer & Mann (1961), and that of the beryllium deposit by Evans & Dujardin (1961). According to these authors, the beryllium concentrations occur in the Letitia Group of rocks that lie near the base of the Seal Lake synclinal trough. The principal beryllium concentration is found in a paragneiss that occupies a position between a syenite intrusive body and a series of porphyritic lavas and fragmental rocks. Some of the more unusual minerals from the beryllium deposit have been described by Heinrich and co-workers (1962, 1963), and by Nickel and co-workers (1962, 1963, 1964). Some of these papers include references to the occurrence of soda amphibole and pyroxene as major rock constituents, but no optical or chemical data have been published to enable these minerals to be properly defined.

The amphibole and pyroxene described in this paper were obtained from a single hand-sample (No. 718), which was one of a suite of samples from the beryllium-bearing paragneiss provided by officials of Rio Tinto Canadian Exploration Limited.

GENERAL DESCRIPTION

Samples of the paragneiss examined to date are all rather prominently banded. The light bands consist chiefly of albitic feldspar, the dark chiefly of arfvedsonite. Aegirine-augite is relatively rare, occurring as a major constituent in only a few of the samples, one of them being sample No. 718.

The arfvedsonite and aegirine-augite, in common with the other mineral constituents, are quite fine-grained, the majority of the grains being between 0.1 and 0.3 mm. in greatest dimension. The arfvedsonite and aegirine-augite occur predominantly in layers up to several millimetres thick, although they are also found as individual grains and small aggregates in feldspar. It is rather difficult to distinguish the two minerals in the hand specimen since both are fine-grained and practically black. In thin sections they are also somewhat similar in appearance, both being dark and highly pleochroic. However, they can be distinguished by a number of features, as follows: The arfvedsonite tends to occur as prismatic grains, whereas the aegirine-augite grains tend to be equi-dimensional. The arfvedsonite is almost completely opaque in its position of maximum absorption, whereas the aegirine-augite is dark green in its position of maximum absorption. The arfvedsonite in certain orientations (sections close to the 010 plane) does not extinguish between crossed nicols, whereas aegirine-augite exhibits complete extinction in all orientations. Finally, in sections approximately normal to the *c*-axis, the minerals exhibit the characteristic amphibole and pyroxene cleavages respectively.

PROCEDURES

The arfvedsonite and aegirine-augite were concentrated from sample No. 718 by a combination of heavy-liquid and magnetic separations. This method proved to be quite effective because of the appreciable differences in specific gravity and magnetic susceptibility of the two minerals.

The chemical analyses were performed by standard wet chemical procedures, including the use of the flame photometer for the alkalis.

X-ray powder diffraction data were obtained by means of a 114.6 mm. Debye–Scherrer camera and a Philips diffractometer, using nickel-filtered copper radiation. The *d*-values were obtained from the film measurements, corrected for shrinkage, and these values were used to calculate the cell parameters by means of a least-squares method. The diffracted intensities were measured from peak heights on the diffractometer tracings, and recalculated to a scale of 100.

The optical determinations were made on single grains mounted on a spindle stage constructed from a hypodermic syringe (Oppenheim, 1962), following the procedure suggested by Joel (1963). The ellipticity of vibration of the arfvedsonite was measured according to the general method described by Shoda (1958). However, instead of using Shoda's triple-stage microscope, a petrographic microscope, fitted with a simple device for inserting and rotating a quarter wavelength plate below the microscope stage was used, together with an elliptical mica compensator in the normal accessory slot. The positions of minimum light intensity, necessary for a measure of the ellipticity, were estimated visually.

The specific gravities of the analyzed concentrates were measured by pycnometer. The specific magnetic susceptibilities were determined by means of a calibrated Frantz iosdynamic separator according to the procedure outlined by McAndrew (1957).

The responsibility for the work reported in this paper is divided between the authors as follows: Miss Mark is responsible for the chemical analyses shown in Tables 1 and 4 and E. H. Nickel is responsible for the remainder of the investigation and for the preparation of this paper.

ARFVEDSONITE

Chemical composition

The chemical composition of the arfvedsonite is shown in Table 1.

The half-cell formula of this arfvedsonite, based on a total of 24 oxygen, hydroxyl and fluorine ions, and dividing the aluminum to give 8.00 (Si + Be + Al) ions, is: $(Na_{2.09}K_{0.72}Ca_{0.22})(Fe_{2.71}{}^{+2}Fe_{1.08}{}^{3+}Mn_{0.30}Mg_{0.21}Ti_{0.20}Al_{0.15}Nb_{0.01})(Si_{7.89}Be_{0.06}Al_{0.05})O_{22.09}(OH)_{1.72}F_{0.19}$. The formula weight is 942.6. This formula is in fair general agreement with the ideal arfvedsonite formula of $Na_{2.5}Ca_{0.5}(Fe^{+2},Mg,Fe^{+3},Al)_5(Si_{7.5}Al_{0.5})O_{22}(OH, F)_2$, proposed by Deer, Howie & Zussman (1963).

Arfvedsonite can form isomorphous series with eckermannite through the replacement of Fe^{+2} by Mg^{+2}, and with riebeckite by an increase in the $Fe^{+3}:Fe^{+2}$ ratio. The dividing line between arfvedsonite and eckermannite has been set at a composition of approximately 70 mole per cent of the magnesium end-member (Deer, Howie & Zussman, 1963), which corresponds to a MgO:FeO mole ratio of less than 2.3 for arfvedsonite. The MgO:FeO mole ratio of the Seal Lake arfvedsonite is 0.213:2.709 = 0.079, which is well within the arvedsonite field.

The dividing line between arfvedsonite and riebeckite has not yet been clearly defined. Deer, Howie & Zussman (1963) give the riebeckite

Table 1. Chemical Compostiion of Arfvedsonite
(Analyst: Miss E. Mark)

Analyzed Components	Weight Per cent	Ions	Number of ions on the basis of 24 (O, OH, F)	
SiO_2	50.16	Si	7.887	8.000
BeO	0.18	Be	0.063	
Al_2O_3	1.10	Al	0.050	
		Al	0.153	4.660
TiO_2	1.64	Ti	0.194	
Nb_2O_5	0.14	Nb	0.010	
Fe_2O_3	9.12	Fe^{+3}	1.079	
FeO	20.60	Fe^{+2}	2.709	
MnO	2.27	Mn	0.302	
MgO	0.92	Mg	0.213	
CaO	1.32	Ca	0.221	3.033
Na_2O	6.85	Na	2.087	
K_2O	3.61	K	0.725	
H_2O(+110° C)	1.64	(OH)	1.722	24.000
H_2O(−110° C)	0.04			
F	0.39	F	0.194	
		O	22.084	
	99.98			
−O≡F	0.16			
	99.82			

end-member formula as $Na_2Fe_3^{+2}Fe_2^{+3}Si_8O_{22}(OH, F)_2$. This differs from the arfvedsonite end-member in having two alkali ions in place of three, and in having a relatively high $Fe^{+3}:Fe^{+2}$ ratio (0.67). The Seal Lake amphibole has approximately three alkali ions in its formula, and an $Fe^{+3}:Fe^{+2}$ ratio of 0.40, both compositional features corresponding to arfvedsonite rather than riebeckite.

X-ray diffraction analysis

Single-crystal *x*-ray diffraction investigations were made by Bell (1963), employing Weissenberg and Buerger precession cameras. According to Bell, the arfvedsonite is monoclinic and can be assigned to the standard space group *C*2/*m*. Following the original work on tremolite by Warren (1930), monoclinic amphiboles were generally referred to space group *I*2/*m*, which is structurally equivalent to *C*2/*m* (Zussman, 1955; Whittaker, 1960; Heritsch *et al.* 1960). In more recent publications, the standard space group of *C*2/*m* has been generally adopted (Ghose, 1961; Ernst, 1962; Frost, 1963; Kawahara, 1963), and will also be used in this paper.

The unit-cell dimensions obtained by Bell (1963) were refined by a least-squares method using the powder diffraction data. The cell parameters thus obtained (a = 9.968 Å, b = 18.034 Å, c = 5.309 Å, β = 104° 14′) were used to calculate a series of *d*-values, which are in good agreement with the measured ones (Table 2).

TABLE 2. X-RAY POWDER DIFFRACTION DATA FOR ARFVEDSONITE

hkl	*d*(calc.) $a = 9.968$ Å $b = 18.034$ $c = 5.309$ $\beta = 104°\ 14'$	Arfvedsonite, Seal Lake		Synthetic riebeckite-arfvedsonite solid solution Sample No. 37 (Ernst, 1962)	
		*I**	*d*(meas.)	*I*	*d*(meas.)
020	9.02Å			4	9.07 Å
110	8.52	100	8.57Å	100	8.52
001	5.15	2	5.12		
$\bar{1}11$	4.90	3	4.89	4	4.882
200	4.83			5	4.819
040	4.51	5	4.51	17	4.551
111	4.04	w	4.04		
$\bar{1}31$	3.894	w	3.886	12	3.888
$\bar{2}21$	3.698	w	3.723		
131	3.410	5	3.412	37	3.441
240	3.296	8	3.299	22	3.307
310	3.171	44	3.179	77	3.160
$\bar{3}11$	3.049	w	3.049	16	3.008
221	2.977	1	2.984		
330	2.839	16	2.843	18	2.837
$\bar{3}31$	2.751	2	2.750		
151	2.720	7	2.718	99	2.747
061	2.596	4	2.595	41	2.619
$\bar{2}02$	2.546	4	2.545	51	2.530
—	—	w	2.486		
—	—	5	2.421		
350	2.402	w	2.402	4	2.408
$\bar{3}51$	2.348	4	2.349	25	2.339
$\bar{4}21$	2.345				
$\bar{3}12$	2.288	2	2.291	16	2.263
331	2.285				
—	—	w	2.217		
261	2.177	4	2.180	33	2.196
$\bar{3}32$	2.157	w	2.150		
—	—	w	2.116		
202	2.071	2	2.074	12	2.091
351	2.039	2	2.042	12	2.053
$\bar{4}02$	2.028	w	2.034	7	2.004
		w	1.964		
		w	1.945		
		w	1.927		
		w	1.889		

*Relative intensities on a scale of 100. The intensities marked w are for weak lines that were not resolved on the diffractometer tracing but were detected on film.

A comparison of the *x*-ray powder diffraction pattern of the Seal Lake arfvedsonite and a synthetic arfvedsonite-rich member of the arfvedsonite-riebeckite series (Table 2) shows a general similarity. Some of the intensity differences can be attributed, at least in part, to preferred orientation due to the pronounced (110) amphibole cleavage. The differences in corresponding *d*-values, particularly noticeable at lower values, is due to their different cell parameters, the Seal Lake arfvedsonite having somewhat greater *a* and β parameters, and a smaller *b* parameter than the synthetic mineral. The *c* parameters of the two minerals are quite similar.

As Whittaker (1960) has pointed out, increasing ionic radius of the interchain ions in the M_4 position (generally occupied by the alkali ions) in the amphibole structure tends to increase the monoclinic angle β, which is a function of translation parallel to the silicate chains (in the *I*-centred cell used by Whittaker the angle β actually decreases; this corresponds to an increase in β of the *C*-centred cell used in this paper). Since the Seal Lake arfvedsonite contains a significant proportion of calcium and potassium, both of which are larger than sodium, the only alkali in the synthetic amphibole, the larger β angle of the former conforms to expectations. The larger alkali ions would also be expected to increase the distance between the amphibole chains, i.e. in the value of $a\sin\beta$. Since $\sin\beta$ decreases with increasing values of β, the value of a must increase so as to bring about an increase in $a\sin\beta$. This, therefore, explains the larger *a* parameter of the Seal Lake arfvedsonite. The value of *c* can be expected to remain relatively unchanged since this direction is parallel to the silicate chains, which are considered to be relatively rigid structural units, not affected by the interchain cations. The reason why the *b* parameter of the naturally-occurring arfvedsonite is smaller than that of the synthetic amphibole is not clear, particularly since *b* would also be expected to increase with increasing size of the interchain cations.

Optical and physical properties

The optical properties of arfvedsonite are very difficult to determine accurately because of extremely high absorption and because it does not extinguish completely between crossed nicols in certain orientations, even with monochromatic light. Under these conditions the spindle stage proved to be extremely useful, since partial extinction curves could be plotted and the missing portions interpolated. The results of the optical determinations are given in Table 3.

Different explanations have been proposed for the failure of arfvedsonite to extinguish between crossed nicols. Eskola & Sahlstein (1930) and

TABLE 3. OPTICAL AND PHYSICAL PROPERTIES OF ARFVEDSONITE

Refractive indices: $\alpha = 1.685$; $\beta = 1.695$; $\gamma = 1.698$
Pleochroism: X = dark green
Y = brown
Z = dark green
Extinction: $X \wedge c = 29°$
Optical angle: $2V = -69°$
Axial ratio of elliptical vibration: 0.47
Specific gravity: 3.382 (meas.); 3.384 (calc.)
Specific magnetic susceptibility: 60×10^{-6} e.m.u.

Sahama (1956) suggested that it is due to a submicroscopic intergrowth of two amphiboles, but there is no evidence for this in the single-crystal or powder x-ray diffraction patterns of the Seal Lake arfvedsonite. Shoda (1958) attributes the phenomenon to elliptical light vibration caused by strong absorption. This appears to be a reasonable explanation for the phenomenon in the Seal Lake arfvedsonite, since it was possible to measure the ellipticity of vibration. A crystal mounted with its (010) plane parallel to the microscope stage was found to have a value of $\epsilon = 0.47$. This corresponds to the value obtained by Shoda on a sample of heikolite, a variety of soda amphibole from Kansenri, Japan. Unfortunately, the latter mineral has not been chemically analyzed (Shoda, personal communication), and therefore the chemical composition responsible for the strong elliptical polarization is not known.

The refractive indices fall within the range of values given by Miyashiro (1957) for arfvedsonite from a number of localities, as well as those given by Borley (1963) for some Nigerian alkali amphiboles. However, they are appreciably lower than any of the values reported for synthetic riebeckite-arfvedsonite solid solutions (Ernst, 1962). Why the naturally-occurring amphiboles should have lower refractive indices than the synthetic varieties is rather obscure, since most of the observed compositional deviations from the ideal formula of the naturally-occurring amphiboles would be expected to increase, rather than decrease, the refractive indices.

The specific magnetic susceptibility of the arfvedsonite is 60×10^{-6} e.m.u. From the relationship between magnetic susceptibility and total $FeO + MnO$ content in amphiboles established by Vernon (1961), the total amount of $FeO + MnO$ (the Fe_2O_3 is calculated as FeO) should be 31.6 per cent. If the Fe_2O_3 in the Seal Lake arfvedsonite is recalculated to FeO, and this is added to the FeO and MnO determined analytically, a value of 31.1 per cent is obtained, which is in good agreement with the value deduced from its magnetic susceptibility. This supports Vernon's conclusion that magnetic susceptibility provides a good measure of total iron plus manganese content.

The specific gravity of the arfvedsonite, determined on a 13-gram sample, is 3.382. This is in excellent agreement with the value of 3.384 calculated from the chemical analysis and unit-cell parameters.

AEGIRINE-AUGITE

Chemical composition

The chemical composition of the aegirine-augite is given in Table 4.

The formula, calculated on the basis of $6(O + OH + F)$ ions, and apportioning the aluminum so as to give a total of $2.00\ (Si + Al + Be)$

TABLE 4. CHEMICAL COMPOSITION OF AEGIRINE-AUGITE
(Analyst: Miss E. Mark)

Analyzed Components	Weight Per cent	Ions	Number of ions on the basis of 6 (O, OH, F)	
SiO_2	50.96	Si	1.951	
BeO	0.20	Be	0.018	2.000
Al_2O_3	2.52	Al	0.031	
		Al	0.083	
TiO_2	2.70	Ti	0.078	
Nb_2O_5	1.04	Nb	0.018	
Fe_2O_3	18.53	Fe^{+3}	0.534	1.006
FeO	7.56	Fe^{+2}	0.242	
MgO	0.31	Mg	0.018	
MnO	1.01	Mn	0.033	
Na_2O	10.34	Na	0.768	
CaO	3.46	Ca	0.142	0.935
K_2O	0.51	K	0.025	
H_2O(+110° C)	0.36	(OH)	0.092	
H_2O(−110° C	0.04			
F	0.51	F	0.062	6.000
		O	5.846	
	100.05			
−O≡F	0.21			
	99.84			

ions, is: $(Na_{0.77}Ca_{0.14}K_{0.02})(Fe_{0.53}{}^{+3}Fe_{0.24}{}^{+2}Al_{0.08}Ti_{0.08}Mn_{0.03}Mg_{0.02}Nb_{0.02})$-$(Si_{1.95}Al_{0.03}Be_{0.02})O_{5.85}(OH)_{0.09}F_{0.06})$, which corresponds quite closely to the ideal pyroxene formula XYZ_2O_6.

Deer, Howie & Zussman (1963) give the formulae of aegirine and aegirine-augite as $NaFe^{+3}Si_2O_6$ and $(Na,Ca)(Fe^{+3},Fe^{+2},Mg,Al)Si_2O_6$, respectively. These minerals form a complete isomorphous series, chiefly by the partial replacement of Fe^{+3} by Fe^{+2}. Deer, Howie & Zussman note that there is no general agreement on the nomenclature within this series, and suggest that the name aegirine be restricted to those members containing from 0.7 to 1.0 Fe^{+3} atoms per formula unit, whereas the name aegirine-augite be used for members containing fewer than 0.7. The Seal Lake pyroxene has 0.53 Fe^{+3} atoms per formula unit, which places it well within the aegirine-augite field.

The name "acmite" has also been commonly used for the $NaFe^{+3}Si_2O_6$ end-member, and there is no apparent chemical or crystallographic distinction between it and aegirine. In the discussion that follows, the terms acmite and aegirine will therefore be used interchangeably.

X-ray diffraction analysis

The *x*-ray powder diffraction data for the Seal Lake aegirine-augite and for synthetic acmite (Nolan & Edgar, 1963) are given in Table 5. Com-

TABLE 5. *X*-RAY POWDER DIFFRACTION DATA FOR AEGIRINE-AUGITE AND ACMITE

hkl	*d*(calc.) $a = 9.681$ Å $b = 8.793$ $c = 5.303$ $\beta = 105°\,06'$	Aegirine-Augite, Seal Lake, Labrador		Synthetic Acmite (Nolan & Edgar, 1963)	
		*I**	*d*(meas.)	*I*	*d*(meas.)
110	6.38 Å	100	6.38 Å	90	6.37 Å
020	4.40	22	4.41	45	4.40
—	—	w	3.74		
111	3.614	w	3.627	10	3.606
021	3.319	4	3.319		
220	3.180	w	3.185	20	3.180
$\bar{2}21$	2.988	32	2.984	90	2.983
$\bar{3}31$	2.908	61	2.907	100	2.899
310	2.905				
$\bar{1}31$	2.545	9	2.546	30	2.544
002	2.530	6	2.532	20	2.526
221	2.474	12	2.478	45	2.458
311	2.258	w	2.264		
112	2.200	w	2.198	10	2.197
040	2.198				
$\bar{3}31$	2.120	13	2.116	30	2.118
$\bar{4}21$	2.099	5	2.092	15	2.095
041	2.016	4	2.018		
240	1.985	w	1.983		
$\bar{2}41$	1.935	w	1.936		
$\bar{5}11$	1.887	w	1.880		
$\bar{4}22$	1.837	w	1.836		
510	1.807	3	1.807		
222	1.807				
150	1.728	8	1.730	20	1.728
312	1.686	w	1.689		
042	1.659	w	1.659		
$\bar{2}23$	1.637	3	1.631		
$\bar{5}31$	1.613	6	1.612	25	1.610
440	1.592	8	1.591	20	1.591
$\bar{6}02$	1.535	4	1.533		
$\bar{4}42$	1.494	w	1.503		
$\bar{5}13$	1.467	4	1.468		
260	1.397	5	1.398	17	1.394
531	1.396				

*Relative intensities on a scale of 100. The intensities marked w are for weak lines that were not resolved on the diffractometer tracing but were detected on film.

parison of the two sets of *x*-ray data shows that they are quite similar. The unit-cell parameters calculated from the indexed Seal Lake powder pattern are: $a = 9.681$ Å, $b = 8.793$ Å, $c = 5.303$ Å and $\beta = 105°06'$. The *d*-values calculated from these parameters are in good agreement with the measured values.

No systematic study has yet been made of the relationship of cell parameters to the compositions of members in the aegirine–aegirine-augite series. Investigation of the synthetic acmite-diopside system (Nolan & Edgar, 1963) has shown that, as the composition moves away from acmite toward diopside, a and b increase, whereas c and β decrease. Changes in composition toward aegirine-augite should also affect the cell parameters in the same direction, since both changes involve replacing the relatively small Fe^{+3} ion by larger ions (Mg in the case of diopside; Fe^{+2} in the case of aegirine-augite), and in the replacement of Na by Ca. Furthermore, the changes resulting from the replacement of Fe^{+3} by Fe^{+2} in aegirine-augite should be appreciably greater than those resulting from the replacement of Fe^{+3} by Mg in diopside because of the greater ionic radius of Fe^{+2}. However, this simple relationship does not appear to exist. The aegirine-augite does have a larger value of a and a smaller angle β than acmite, as expected, whereas b remains about the same, and c is appreciably greater. The reasons for this are not yet clear, since not enough is known about the detailed pyroxene structure.

Optical and physical properties

The optical and physical properties of the aegirine-augite are summarized in Table 6. The optical properties, referred to the diagram in Deer, Howie & Zussman (1963, p. 87) relating optical properties to composition in the aegirine–aegirine-augite series, correspond to an aegirine-augite with about 0.6 Fe^{+3} atoms per formula unit. Considering the scatter of points about the curves drawn by Deer, Howie & Zussman, this is in fair agreement with the 0.53 Fe^{+3} atoms actually determined.

TABLE 6. OPTICAL AND PHYSICAL PROPERTIES OF AEGIRINE-AUGITE

Refractive Indices: $\alpha = 1.737$; $\beta = 1.769$; $\gamma = 1.787$
Pleochroism: X = green
  Y = green
  Z = brown
Optical angle: $2V = -74°$
Extinction angle: $X \wedge c$ = small ($\sim 0°$)
Specific gravity: 3.517 (meas.); 3.541 (calc.)
Specific magnetic susceptibility: 50×10^{-6} e.m.u.

The magnetic susceptibility is 50×10^{-6} e.m.u., which, according to Vernon (1961), indicates the presence of 26.4 per cent FeO + MnO. This is in fair agreement with the actual value of 25.2 per cent determined analytically (with all the Fe_2O_3 calculated as FeO).

CRYSTAL CHEMICAL CONSIDERATIONS

Before entering upon a discussion of the stability relations between co-existing mineral pairs, it is essential to know whether the phases in question are in equilibrium or were in equilibrium at the time of crystallization. In the case of the arfvedsonite and aegirine-augite from Seal Lake, the evidence is fairly clear that these two minerals were most likely in equilibrium at the time of their formation. In the sample from which both minerals were concentrated for analysis, the two minerals occur as grains of similar size that are frequently adjacent, and there is no evidence of reaction between them.

The stability relations of synthetic riebeckite-arfvedsonite solid solutions have been investigated by Ernst (1962) working with a bulk composition of $Na_2O.5FeO_x.8SiO_2$ plus excess water. He found that the range of amphibole stability is governed by temperature, oxygen fugacity, and total pressure. At total fluid pressures in the range 500 to 2000 bars, riebeckite-arfvedsonite solid solutions are stable at temperatures up to about 700 °C, and at oxygen fugacities below 10^{-17}. Compositions toward the arfvedsonite end of the series are stable at lower oxygen pressures and higher temperatures than those toward the riebeckite end.

Ernst's work in the synthetic system shows that riebeckite-arfvedsonite solid solutions are not in stable equilibrium with acmite (i.e. aegirine), except along a univariant curve. The fact that arfvedsonite does occur in apparent equilibrium with aegirine-augite at Seal Lake, as well as in other occurrences (Deer, Howie & Zussman, 1963) indicates that the stability range of one or both of these minerals is greater than that exhibited by the synthetic minerals. This can be attributed to the relatively large numbers of "impurities" in the naturally-occurring minerals.

The distribution of ions between co-existing mineral phases is also a matter of some interest. The fractionation of the major cations between arfvedsonite and aegirine-augite is shown in Table 7. The figures given are the fraction of possible sites occupied, assuming the ideal formula for arfvedsonite to be $X_3Y_5Z_8O_{22}(OH, F)_2$, and for aegirine-augite, XYZ_2O_6.

Considering the competing ions in the X sites (i.e. the alkali ions), sodium and calcium are preferentially concentrated in aegirine-augite, whereas potassium is more abundant in arfvedsonite. With regard to

TABLE 7. THE DISTRIBUTION OF MAJOR CATIONS BETWEEN ARFVEDSONITE AND AEGIRINE-AUGITE

Site	Cation	Electronegativity (Pauling, 1960)	Fraction of Possible Sites Occupied	
			Arfvedsonite	Aegirine-Augite
X	Na	0.9	0.69	0.77
	Ca	1.0	0.07	0.14
	K	0.8	0.24	0.03
Y	Fe^{+3}	1.9	0.22	0.53
	Fe^{+2}	1.8	0.54	0.24

iron, which is the major element in the Y sites of both minerals, the ferrous ion predominates in arfvedsonite and the ferric in aegirine-augite. This distribution can be related to cation electronegativity. The ions with the higher electronegativities (Na and Ca in X and Fe^{+3} in Y) are preferentially concentrated in aegirine-augite, and the ions with lower electronegativities (K in X and Fe^{+2} in Y) predominate in arfvedsonite. As Ramberg (1952) has pointed out, increasing polymerization of silicon-oxygen tetrahedra in silicates favours the incorporation of connecting ions with lower electronegativities. The amphibole structure, consisting of double chains of silica tetrahedra, is more highly polymerized than the pyroxene structure, which contains only single chains of silica tetrahedra. Consequently, amphibole should have a higher proportion of ions with low electronegativities than pyroxene, which is in agreement with the observed facts.

The distribution of these elements between arfvedsonite and aegirine-augite is similar to that noted by McLachlan (1951) between a soda amphibole formed by alteration of an aegirine-augite and the aegirine-augite itself. DeVore (1957) found an apparently anomalous distribution of cations in his study of co-existing amphiboles and pyroxenes, which he attributed to the relatively high proportion of aluminum replacing silicon in four-fold coordination, thereby effectively reducing the polymerization. That the distribution of cations between the Seal Lake amphibole and pyroxene conforms to expectations may be due to the relatively minor replacement of silicon by aluminum in both minerals. In conclusion, therefore, it is evident that the distribution of major cations between arfvedsonite and aegirine-augite is consistent with the theory that increasing polymerization of silicon-oxygen tetrahedra favours the incorporation of connecting ions with lower electronegativities.

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