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On aluminous and edenitic hornblendes

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SUMMARY. Based on nearly 1500 published amphibole analyses the maximum possible Al^{VI} in hornblendes is shown to increase with increase of Al^{IV} . New analyses of hornblendes from amphibole-coriundum rocks, with and without anorthite, are given and after critical examination of the available data it is concluded that the maximum verified Al^{VI} -rich calciferous amphibole that approaches the closest to hypothetical tschermakite comes from a kyanite-bearing aluminous high-pressure-crystallized schist from Lukmanier, Switzerland. Pure natural edenite or ferroedenite is unknown, but a new analysis of the nearest known natural edenite, from Mysore, India, agrees with the postulated view that extraordinarily low temperatures are needed for edenite-ferroedenite crystallization, much below that possible in magmas and only rarely achieved in metamorphic rocks containing amphiboles. The limit of the approach of igneous hornblendes to edenite-ferroedenite and tremolite-ferroactinolite is outlined.

At least 1100 °C is required for complete expulsion of water from some amphiboles.

Aluminous hornblendes

A FEW years ago, after examining 936 published amphibole analyses, Leake (1965) suggested that the maximum possible Al^{VI} in calciferous and subcalciferous amphiboles increases regularly as Al^{IV} increases, where calciferous amphiboles are defined as possessing $Ca \geq 1.50$ in the half unit cell and subcalciferous amphiboles have Ca between 1.00 and 1.50. A number of amphiboles that appeared to contain more than the suggested maximum Al^{IV} for their Al^{IV} values were re-analysed and the previous values were shown to be erroneous. At that time samples of rocks containing hornblende with either corundum or kyanite were not available to the writer and the line limiting the maximum possible Al^{VI} was perspicaciously extrapolated to values close to Si 6.00, Al^{IV} 2.00, Al^{VI} 1.40, mainly based on the re-analysis of an amphibole from an anorthite-vesuvianite rock. If this extrapolation is confirmed, then the above value would be the nearest that natural amphiboles approach to the hypothetical pure tschermakite $Ca_2Mg_3Al_2Si_6Al_2(OH)_2O_{22}$, which contains 2.00 Al^{VI} .

Recently, Kostyuk and Sobolev (1969, p. 79) have claimed that 550 chemical analyses of amphiboles 'reveal a negative correlation between Al^{IV} and Al^{VI} ', 'in contrast to the data of Leake (1965)'. However, Saxena and Ekstrom (1970) using the data given by Leake (1968a) have shown a positive correlation of Al^{IV} with Al^{VI} .

The present account presents new analyses of amphiboles from ruby-rich amphibolites and re-examines the original suggestion of a relationship between the maximum possible Al^{vi} and Al^{iv} in the light of over 500 more published amphibole analyses than were available in 1965, making a total of nearly 1500 post-1890 amphibole analyses all with at least 1.00 Ca in the half unit cell. Of these analyses 1217 have been published in a catalogue (Leake, 1968a) and it is convenient to use the catalogue number to refer to these analyses.

Fig. 1 is a plot of the 500 new analyses obtained largely from the literature, from which it is again suggested, as it was with the previous 936 analyses referred to, that the maximum possible Al^{vi} increases as Al^{iv} increases. This does not imply that, nor has it been suggested by the author that there is a positive correlation between Al^{iv} and Al^{vi} in any particular amphibole series. Quite clearly, from both figs. 1 and 2, although increase of Al^{vi} is statistically accompanied by increase in Al^{iv} there are many analyses with little or no Al^{vi} even at maximum Al^{iv} (2.0–2.5), but there are no samples with high contents of Al^{vi} and no Al^{iv} , a situation only found in glaucophanes. It is, however, apparent that a negative correlation of Al^{iv} and Al^{vi} is completely ruled out. The line limiting the suggested maximum Al^{vi} is copied from that previously given (Leake, 1965) and in the writer's opinion, analyses that plot appreciably above this line are generally erroneous, unless their Ca values approach 1.00. In order to support this it is necessary to briefly consider the numbered analyses that plot near or above the line.

The analyses considered are, in catalogue number order:

951. S. H. Kranck (1961). *Journ. Petrology*, **2**, 177. With analysed clinopyroxene and quartz, in a meta-ironstone, Quebec. Total (96.3 %) very low even allowing for non-determination of water.

962. A. Osann (1916). *Beitr. Chem. Petrographie*, **3**, Leipzig (Borntraeger), 331. Amphibole, Siberia. H_2O+ (0.99 %) is low, CaO extremely low (7.41 %), and with low Na_2O (0.06 %) giving a sum for the X group of only 1.28.

963. C. W. V. Gumbel (1891). *Geogn. Beschr. Bayern*, **4**, 210 Diorite, Bavaria. H_2O+ (0.98 %) low giving extremely high Y group total, 5.54.

964. L. Lokka (1950). *Bull. Comm. Géol. Finlande*, **25**, 69. Pegmatite with quartz, K-feldspar, muscovite, tourmaline, and wiikite. Silmapuolivaara, Finland. In view of the reputation of the late Dr. Lokka attempts have been made to locate this amphibole by the Geological Survey of Finland, unfortunately without success. It seems almost certain that the H_2O+ (0.59 %) is seriously low, especially in a paragenesis with muscovite, but this alone is not an adequate explanation and attempts are continuing to obtain some of this amphibole.

991. J. V. Lewis (1896). This is dealt with below.

996. H. Sustschinsky (1912). *Trav. Soc. Imp. Natural. St. Petersbourg*, **36**, 203. Al_2O_3 ; TiO_2 not determined; H_2O+ low (0.74 %) and alkalis low.

1002. F. Loewinson-Lessing (1900). *Trav. Soc. Imp. Natural. St. Petersbourg*, **30**, 169. 'Amphibole trap granulites.' Salaja, S. Urals, U.S.S.R. TiO_2 not determined, H_2O+ low (1.05 %), MnO low, probably a poor analysis.

1024. K. Tomita (1962). *Mem. Coll. Sci. Univ. Kyoto, Ser. B*, **28**, 553. Volcanic agglomerate, Gaspan, Parao Island, Japan. Analysis repeated in K. Tomita (1965), *Mem. Coll. Sci. Univ. Kyoto, Ser. B*, **32**, 57, with H_2O+ increased from 0.52 to 1.46 resulting in movement of the plotted point as shown in Fig. 1.

1028. C. Trenzen (1902). *Neues Jahrb. Min.* **2**, 37. Phenocrysts in basalt, Seigertshausen, Hesse, Germany. Extraordinarily low H_2O+ (0.24 %) and high Y group total (5.97).

1054. R. G. Coleman *et al.* (1965). *Bull. Geol. Soc. Amer.* **76**, 500. Eclogite, Amos stream, New Caledonia. Ca 1.03 in the half-cell contents is so low that this barroisite is excluded from consideration.

1060. T. Gjelsvik (1947). *Norsk. Geol. Tids.* **26**, 42. Amphibolized garnet-bearing gabbro, Heidal, Gudbrandsdalen, Norway. Said to be between an amphibole and a pyroxene. Is almost certainly contaminated with pyroxene; CaO 14.10 % and Y group total only 4.28.

1069. T. Strand (1951). *Norges Geol. Unders.* no. 178, 105. Quartz–biotite–muscovite–hornblende–garnet schist. Sel, Pillargurippiggen, Otta River, Norway. Analysis made for Goldschmidt in 1915. Because of the rather aluminous environment this amphibole deserves re-study but as yet the Mineralogical Museum in Oslo have not been able to locate the material.

1089. B. G. Lutz (1962). *Mem. All-Union Min. Soc. (U.S.S.R.)*, **91**, 332. Diopside–hypersthene–plagioclase–hornblende granulite. River Anabar, Berega, Siberia, U.S.S.R. H_2O+ low (1.04 %), with no determination of F.

1091. M. S. Naik (1963). *Journ. Karnatak Univ. Sci.* **8**, 49. Dyke-like band in granite. Molakalmuru, Mysore State, India, H_2O+ low (0.82 %) and CaO too high (14.25 %).

1092. M. Ishii and T. Yamada (1962). *Journ. Geol. Soc. Japan*, **68**, 115. Gneissose granodiorite. Koshibu River, Nagano Pref. Japan. The analyses given by these authors for a range of differing granodiorites and quartz diorites all show high Al_2O_3 although the samples are from differing localities. Probably a systematic analytical error.

1102. As 1092.

1164. B. G. Lutz (1962). *Mem. All-Union Min. Soc. (U.S.S.R.)*, **91**, 332. Orthopyroxene–diopside–hornblende granulite. River Anabar, Berega, Siberia, U.S.S.R. H_2O+ low (0.50 %) and excessive alumina.

The remaining analyses are not included in the catalogue referred to above and they are identified by their silica contents, arranged in order of decreasing silica:

55.46 % S. P. Das Gupta (1967). *Min. Mag.* **36**, 26. With a little albite, quartz, sphene, and epidote in a schist. Salwari, Sikar, Rajasthan, India. H_2O+ (0.65 %) low.

52.84 % S. Roy and F. N. Mitra (1964). *Proc. Nat. Sci. India*, **30A**, 413. With winchite and rhodonite, Madhya Pradesh, India. H_2O+ 0.16, H_2O- 0.64, F 0.05 % seems impossible for an amphibole.

52.5 % P. Bearth (1967). *Beitr. Geol. Karte Schweiz, N.F.* **132**, 89. Epidote–hornblende schist lens in glaucophane schist. Rimpfischen, Südwall, Switzerland. Semi-quantitative analysis reported to first decimal place only. Plots just below the line but is rather low in Ca (1.44 in half unit cell).

50.86 % G. Troll. (1968) *Bayer. Akad. Wiss. Mat.-Nat. Kl. Abhandl., N.F. Heft* **133**, 73. Quartz–diorite, Bavaria, Germany. From unpublished water determinations made by Leake on some amphiboles received from Dr. Troll from this region the water content of this analysis (1.10 %) is certainly low. Calculated to 23 (O) the analysis plots below the line.

49.99 % N. Bakun-Czubarow (1968). *Archiwum Mineral.* **28**, 305. In an eclogite with kyanite, omphacite, garnet, and rutile. Nowa Wies, Snieznik klodzki, Poland. H_2O+ (0.69 %) and the total (99.34 %) are low but otherwise analysis appears satisfactory. Calculated on the basis of 23 (O) gives Si 6.90, Al^{VI} 0.97.

49.88 % N. Bakun-Czubarow (1968) as above. Ca (1.39) is rather low in this analysis and so is H_2O+ (1.18 %). The analysis plots just above the line in fig. 1, within experimental error of the line.

49.68 % R. A. Binns (1967). *Journ. Petrology*, 8, 361. In an eclogite with garnet, omphacite, rutile, and apatite. This is believed to be a good analysis and it plots just below the line despite the low Ca (1.18). It should be noted that $H_2O+F = 1.85\%$.

49.00 % B. V. Govinda Rajulu and A. Shariff (1966). Amphibolite. Khushalnagar, Coorg, Mysore State. H_2O+ (0.72 %) low. Re-analysis of this amphibole, Table II, GR, changes its position on fig. 1 markedly and shows how pronounced is the influence of analytical error.

47.10 % M. Novotny (1954) quoted by D. Némec (1970). *Neues Jahrb. Min., Abh.* 113, 52. Amphibole-garnet skarn. Pernstejn, Western Moravia, Czechoslovakia. Recalculated to exclude impurities. H_2O+ (0.76) low. Unreliable.

46.4 % P. Bearth (1967). *Beitr. Geol. Karte Schweiz. N.F.* 132, 89. Epidote amphibolite vein in serpentinite. Stocknubel, Switzerland. Semi-quantitative analyses with 1.5 % H_2O . Is well within experimental error of plotting below the line.

43.5 % and 43.4 % (MF228). M. Frey (1969). These are dealt with below and in Table J.

41.32 % S. P. Korikovskiy (1967). *Acad. Nauk SSSR*, 66. Hornblende-biotite-epidote-plagioclase-quartz schist. Chdokano-Stanovoi Region, U.S.S.R. H_2O+ (0.65 %) too low. Plots below the line on the basis of 23 (O).

Analyses that plot immediately below the line and are considered reliable have previously been considered (Leake, 1965), but some additional ones marked on fig. 1 include 1116, 1121, 1128, and 1129, all by Raychaudhuri (1960) from amphibolites, 1074 from an eclogite in Japan (Banno, 1964), 41.58, 39.66, and 39.60 % SiO_2 from biotite-chlorite amphibolites in New Zealand (Cooper and Lovering, 1970. These three are probe analyses without water and calculated to 23 (O). Correction for the probable Fe_2O_3 content (4.50 %) using wet-determined values given by the authors on other similar amphiboles lowers the Si and Al^{VI} values but they are still rich in Al^{VI}).

Amphiboles in amphibole-corundum rocks. Particularly interesting in the search for analyses with high contents of Al^{VI} and therefore approaching tschermakite is an old amphibole analysis by J. V. Lewis (1896) from a corundum-amphibole rock, Corundum Hill, North Carolina (999; table I and fig. 1). Although this has only moderately high Al^{VI} (0.84) this is primarily because the analysis is reported to contain 4.63 % H_2O , a value difficult to believe. If the analysis is recalculated on the basis of 23 (O) it plots almost on the line in fig. 1 with Si 6.35, Al^{IV} 1.65, Al^{VI} 1.27. It is easy to predict that amphiboles in corundum-bearing rocks will contain the maximum possible Al allowable by the conditions of crystallization. Accordingly it seemed important to investigate the amphiboles co-existing with corundum in North Carolina especially as the analysis by J. V. Lewis did not report Ti or Fe^{3+} , while the MnO value is unusually high in view of the low iron content.

By the kindness of Dr. J. R. Butler of Chapel Hill, North Carolina, two samples of amphibole-corundum rocks were obtained and the amphiboles analysed (table I). Most surprising was the complete absence of titania, a very rare circumstance in hornblende. The complete absence was carefully verified. Allowing for the Fe_2O_3 correction of the alumina reported by Lewis, the analyses are close to that of Lewis except for the H_2O+ and MnO. The half cell contents of the new analyses plot rather below the suggested maximum possible Al^{VI} for their Si or Al^{IV} value. It is significant that despite the presence of 2.00 and 2.44 % Na_2O in these hornblendes, the plagioclase is anorthite

TABLE I. *Chemical analyses of aluminous amphiboles*

	1 991	2 AB	3 13A	4 AY	5 MF228	6 MF228	7 5A	8 7A
SiO ₂	44·38	44·81	44·51	42·47	43·5	42·70	45·80	45·70
Al ₂ O ₃	17·32	16·35	17·10	17·81	17·8	18·26	15·02	16·30
TiO ₂	—	0·00	0·00	0·26	0·45	0·45	0·03	0·09
Cr ₂ O ₃	0·38	0·73	0·06	0·89	—	—	—	—
Fe ₂ O ₃	—	0·41	1·45	0·92	3·1	3·14	0·58	0·68
FeO	3·83	4·48	4·80	4·49	11·9	12·07	4·07	4·39
MgO	15·48	16·45	15·68	16·33	8·3	8·42	18·68	18·60
CaO	11·51	12·19	12·14	11·87	10·5	10·65	11·25	10·39
Na ₂ O	1·24	2·44	2·00	2·40	1·4	1·42	1·65	2·06
K ₂ O	0·38	0·35	0·55	0·69	0·48	0·49	0·10	0·08
MnO	0·90	0·04	0·04	0·15	0·07	0·07	tr	tr
P ₂ O ₅	—	0·02	0·01	0·05	—	—	0·07	0·21
H ₂ O+	4·63	1·76	1·87	1·78	2·1	2·13	2·79	2·17
H ₂ O—	—	0·00	0·00	0·00	—	—	—	—
	100·05	100·11	100·21	100·19	99·60	99·80	100·04	100·67
	<i>Atoms to 24(O,OH)</i>				<i>Atoms to 23(O)</i>			
Si	6·05	6·34	6·29	6·05	6·34	6·22	6·44	6·33
Al ^{iv}	1·95	1·66	1·71	1·95	1·66	1·78	1·56	1·67
Al ^{vi}	0·84	1·07	1·13	1·04	1·40	1·35	0·92	0·99
Ti	—	0·00	0·00	0·03	0·05	0·05	0·00	0·01
Cr	0·04	0·08	0·01	0·10	—	—	—	—
Fe ³⁺	—	0·04	0·15	0·10	0·34	0·34	0·06	0·07
Fe ²⁺	0·44	0·53	0·57	0·53	1·45	1·47	0·47	0·50
Mn	0·10	0·01	0·01	0·02	0·01	0·01	0·00	0·00
Mg	3·14	3·47	3·30	3·46	1·80	1·83	3·94	3·87
Ca	1·68	1·84	1·83	1·81	1·63	1·66	1·69	1·54
Na	0·33	0·67	0·54	0·66	0·39	0·40	0·45	0·55
K	0·07	0·06	0·10	0·13	0·09	0·09	0·02	0·01
OH	4·20	1·66	1·76	1·69	2·04	2·06	(2·00)	(2·00)
O	19·80	22·34	22·24	22·31	21·96	21·94	22·00	22·00
Σ Z	8·00	8·00	8·00	8·00	8·00	8·00	8·00	8·00
Σ Y	4·56	5·20	5·17	5·28	5·05	5·05	5·39	5·44
Σ X	2·08	2·57	2·47	2·60	2·11	2·15	2·16	2·10
mg	0·85	0·86	0·82	0·84	0·50	0·50	0·88	0·87
α	—	1·634	1·636	1·643	—	—	1·633	1·635
γ	—	1·657	1·655	1·662	—	—	1·651	1·654

- 991, Hydro-tschermakite associated with corundum. Corundum Hill, North Carolina, U.S.A. J. V. Lewis (1896, p. 65).
- AB, Alumino-pargasitic hornblende in a pink corundum-amphibole rock. Clay Co., North Carolina, U.S.A. Precise location unknown. Total includes NiO 0·04, Cl 0·05, and F 0·00 determined by B. W. Evans by microprobe analysis giving O ≡ Cl 0·01 and Ni 0·00 and Cl 0·01 in the cell contents. Remainder of analysis by A. Kemp and B. E. Leake.
- 13A, Alumino-tschermakitic hornblende in a corundum-amphibole-anorthite rock with veins of ruby corundum and An_{90·5}Ab_{8·8}Or_{2·7} (mol. %). Macon Co., North Carolina, precise locality unknown. Analyst, A. Kemp.
- AY, Alumino-pargasite in a ferrian zoisite (Ca₂Al₂AlSi₃O₁₃OH + Fe³⁺) rock with ruby porphyroblasts (1 cm) without feldspar. A little chromitic spinel is present in the amphibole in the rock. From Merkerstein, 45 miles north of Arusha, Tanzania. Collector, D. S. Tyrwhitt. Analyst, A. Kemp.

TABLE I (cont.)

- 5: MF228, Alumino-tschermakitic hornblende with kyanite, garnet, biotite, An_{25-35} , quartz, chlorite, and a little staurolite, epidote, ilmenite, apatite, and tourmaline. Frodaler, Lukmanier, Switzerland. M. Frey (1969, p. 68).
- 6: MF228, Alumino-tschermakite, same material as 5. SiO_2 and Al_2O_3 by microprobe analysis by C. H. Emeleus using amphibole 2 and 95052 (Leake, 1968b) as standards. Remaining oxides by Frey (1969, p. 68) recalculated by Leake to allow for quartz inclusions. See text.
- 7: 5A, Tschermakitic hornblende in an anorthite amphibolite rich in sapphirine with a little corundum. 0.7 Km ESE of Chantel, Vallée du Haut-Allier, Massif Central, France. F. H. Forestier and B. Lasnier (1969, p. 208). H_2O+ is loss on ignition.
- 8: 7A, Tschermakitic hornblende in an anorthite amphibolite with corundum, zoisite, spinel, and sapphirine. Locality and reference as 5. H_2O+ is loss on ignition.

with less Na_2O (1.62 % Na_2O in 13A) than in the hornblende, being in agreement with previous analyses of highly aluminous amphiboles from North Carolina (Leake, 1965). Anorthite is the most aluminous of all the feldspars.

Another corundum-amphibole rock, a specimen of anyorite from Tanzania, made of ferrian zoisite with ruby porphyroblasts and hornblende, was also investigated (table I). The amphibole is very similar to those from North Carolina and plots fairly close to these (fig. 1). The bright green colour of all these amphiboles associated with corundum appears to reflect the high Mg, low iron, and appreciable Cr content of the amphiboles.

Recently, Forestier and Lasnier (1969) have given five analyses of amphiboles associated with corundum and anorthite from Chantel, Vallée du Haut-Allier, Massif Central, France. The rocks are amphibolites, often with spinel and sapphirine, and are clearly a highly aluminous environment. Unfortunately the water contents of these amphiboles were not determined, loss on ignition being substituted instead. Two of these analyses are cited in table I and are plotted as 7A and 5A in fig. 1. Allowing for the half cell contents being calculated on the basis of 23 (O) they plot reasonably close to the new analyses presented in this account of amphiboles associated with corundum.

Water contents of amphiboles. The water contents of these amphiboles have been particularly critically determined using a tube furnace, dried nitrogen, and the method of Riley (1958). With a temperature reading of 1170 °C sample AB gave 1.26 % H_2O ; 13A gave 1.47, and AY gave 1.28 % H_2O , all rather low values for amphiboles though within the range commonly reported, and the analysis totals ranged from 99.61 to 99.81. Because of the particularly low value of water in AB this was repeated several times with consistent results between 1.20 and 1.30 %. Professor B. W. Evans kindly analysed this sample on the microprobe for F and Cl but only trivial amounts of Cl (0.05 %) were detected. Precise temperature determinations were then made inside the tube furnace using a thermocouple and this showed that the maximum temperature inside the tube was only 1050 °C when 1170 °C was recorded outside the tube. The temperature was therefore raised to 1350 °C on the outside of the tube giving a maximum temperature inside the tube of 1205 °C. The determinations were then repeated and from 0.40 to 0.50 % more water obtained than before, these results being given in table I. Quite clearly a very high temperature is essential to remove

all the water in some amphiboles, certainly over 1100 °C. This was confirmed with some other amphiboles, which gave 1.42 and 1.55 % H₂O at 1035 °C but 1.91 and 2.19 at 1150 °C, both temperatures being inside the tube.

This problem of accurate water determination is of paramount importance in amphibole studies for, as Borg (1968) has shown, it cannot be assumed that a calculation of cell contents on the basis of 23 (O) is necessarily correct. There is no doubt that a great many amphibole analyses are deficient in water resulting in spuriously high cation contents, especially Al^{vi} because of the method of allocation to the Y group of Al that is in excess of that needed to fill the Z group. The possibility of using loss on ignition at 1200 °C as a measure of the H₂O+Cl+F content of an amphibole has been investigated. The usual objection to the use of loss on ignition values is that the uncertain degree of oxidation of the FeO renders the determination of little value. This is easily overcome by determining the FeO content of the powder after heating and then correcting for the weight of oxygen gained. Sample AB was heated for 5 hours at 1200 °C and gave a corrected loss on ignition of 2.16 %, a value appreciably in excess of the determined H₂O+F+Cl. Analyses showed, however, that this excess was not primarily due to water loss but to loss of Na₂O and possibly silica. Loss on ignition values cannot therefore be used as a measure of H₂O+F+Cl as they may give too high values—a circumstance suggested for analysis 7, Table I.

Maximum possible Al^{vi} in hornblendes. It is clear from previous analyses of amphiboles and from theoretical studies that the content of Al^{vi} in amphiboles is primarily controlled by rock composition and pressure, with high pressure favouring high Al^{vi}. It is easy to anticipate that maximum Al^{vi} will occur in amphiboles crystallized in highly aluminous environments with moderate or low alkali contents and under high pressures. Such an environment should give higher Al^{vi} than corundum-bearing assemblages crystallized under lower pressure conditions. Assemblages including kyanite and garnet are likely to be of particular value and thus attempts have been made to obtain some of amphibole 49.99 % SiO₂ (Bakun-Czubarow, 1968) and MF 228 (Frey, 1969). Although samples of eclogites from the same locality as that studied by Dr. Bakun-Czubarow were kindly sent to me they do not contain kyanite and thus are unlikely to contribute to the search for amphiboles that plot appreciably above the line in fig. 1.

Dr. Frey kindly sent some of the impure concentrate of amphibole MF228 (43.5 % SiO₂). Attempts to purify this for check analysis were defeated by the presence of numerous droplets of quartz. Frey (1969, p. 58) records that the amphibole is sieved with quartz inclusions. This raised the possibility that the wet analysis might be too high in silica, which would not only raise the Si content of the calculated cell contents but also the Al^{vi}, because of the method of computation. On the other hand, because this amphibole co-exists with garnet, kyanite, and staurolite, all highly aluminous minerals, and occurs in a region of high-pressure metamorphism known to contain amphiboles particularly rich in Al^{vi} (Steiger, 1961, reports 15 analyses which contain up to 1.24 Al^{vi}) it seemed this specimen might well contain the maximum possible Al^{vi}. Dr. C. H. Emeleus kindly analysed a grain mount for SiO₂ and Al₂O₃ using an

electron microprobe, a 3μ electron spot, and using two amphiboles analysed by the author as standards. The SiO_2 was reduced by 0.80 % to 42.70 %. If it is assumed that this difference is due to quartz inclusions in the wet-chemically analysed powder then the remaining oxides need to be slightly increased to restore the original total and this gives the values given in table I, analysis 6, except that the calculated Al_2O_3 of 18.06 has been replaced by the electron microprobe determination of 18.26, the two values being in satisfactory agreement. Calculation of analysis 6, table I, shows that it plots only just above the line drawn on fig. 1; certainly the experimental error of the water determination alone could account for the displacement above the line. It is suggested that this revised analysis with Si 6.22, Al^{VI} 1.35 contains the highest verified Al^{VI} content of any calciferous amphibole yet investigated and thus approaches the closest to hypothetical tschermakite $\text{Ca}_2\text{Mg}_3\text{Al}^{\text{VI}}_2\text{Si}_6\text{Al}^{\text{IV}}_2(\text{OH})_2\text{O}_{22}$ of any analysed amphibole whose composition is reliably known.

Fig. 2 is a plot of all the amphibole analyses available to the writer with $\text{Ca} \geq 1.00$ in the half unit cell. All the analyses plotting above the line have been considered either in this account or in the previous one (Leake, 1965) and either shown to be erroneous by re-analysis (e.g. Leake, 1968*b*) or thought to be on reasonable grounds, such as low $\text{H}_2\text{O}+$ values. Fig. 2 also shows the analyses, plotting close to the line, that are regarded as reliable and upon which the precise position of the line is based. It seems that new analyses that plot above the line should be carefully verified before publication and any such analyses probably indicate a high-pressure aluminous environment. It is most important for future work to ascertain the lowest Ca content in the half unit cell for which the limitation holds of Al^{VI} to less than $0.6 \text{ Al}^{\text{IV}} + 0.25$, the equation of the line in fig. 2.

The results suggest that there is a definite limitation to filling the octahedral sites in hornblendes with Al without increasing the Al^{IV} whereas Al^{IV} can range between about 5.50 and 8.00 with quite variable Al^{VI} . Conditions of crystallization are clearly important because there is marked restriction of igneous amphiboles to lower Al^{VI} contents than metamorphic ones. This probably indicates that lower temperatures and higher pressures favour higher Al^{VI} . Lower temperatures will favour more highly ordered cation distributions and so on the whole it is predictable that higher Al^{VI} will be statistically accompanied by greater cation ordering, a view in agreement with Saxena and Ekstrom's (1970) statistical results. It does not necessarily follow, however, that increased Al^{VI} is the cause of the greater ordering though in practice the preference of Al^{VI} for M_2 (Ghose, 1965) will reduce the available choice of position for the remaining octahedral cations. The evidence clearly indicates that it is increasingly difficult to fill more than about half of the M_2 sites with Al^{VI} . As yet no natural tschermakites with complete occupancy of M_2 by Al^{VI} are known, amphibole 228 having only 67 % of these sites filled with Al^{VI} . Ghose's (1965) results suggest that amphiboles with high contents of Al^{VI} in M_2 should tend to be more magnesium-rich than usual, because Fe^{2+} , which is usually concentrated in these positions, will be displaced (M_2 sites favour Fe^{2+} over Mg^{2+}). There is some evidence to support this as some of the Al^{VI} -rich amphiboles in table I are Mg-rich. It seems likely that the difficulty of achieving charge balance when 2Fe^{2+} are replaced by

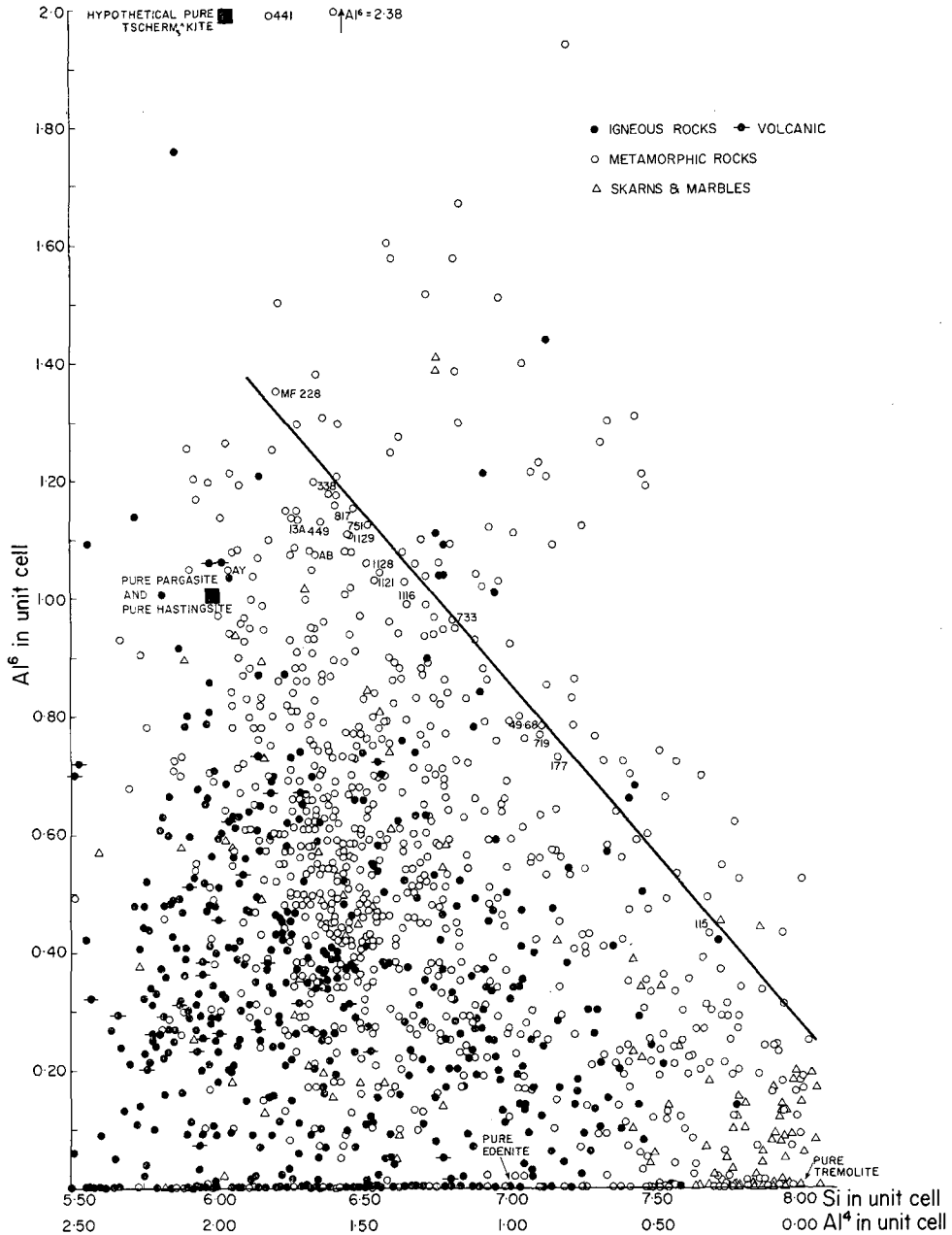


FIG. 2. Plot of octahedral Al in the half unit cell against tetrahedral Al (or Si) for all available (about 1500) analysed amphiboles containing Ca 1.00 or more in the half unit cell. Analyses plotting above the diagonal line and those labelled below the line are discussed in the text or have previously been considered (Leake, 1965).

2Al^{3+} is at least as critical in requiring substantial replacement of Si^{4+} by Al^{3+} as structural considerations, and may be partly responsible for the rather low Ti^{4+} and Fe^{3+} in many Al^{VI} -rich calciferous amphiboles.

The existence of edenitic amphiboles

A plot of the half unit cell contents of Si against $\text{Ca} + \text{Na} + \text{K}$ for amphiboles from igneous rocks (fig. 3) is based on the available calciferous amphibole analyses obtained from Leake's (1968a) catalogue of 1217 amphibole analyses together with about 250 analyses obtained since the catalogue was compiled in 1965. Analyses classified in the catalogue as inferior in quality have not been plotted. The criteria used to identify these inferior analyses are discussed in detail in Leake (1968a). Analyses from the catalogue upon which attention is focused are numbered with the numbers used in the catalogue while any newer analyses of particular interest are identified by their silica contents, or by lettered numbers.

It is apparent from fig. 3 that there are no reliable analyses known of calciferous igneous amphiboles that are close to edenite or ferro-edenite $\text{NaCa}_2(\text{Fe}, \text{Mg})_5\text{Si}_7\text{AlO}_{22}(\text{OH})_2$, tremolite or ferroactinolite $\text{Ca}_2(\text{Fe}, \text{Mg})_5\text{Si}_8\text{O}_{22}(\text{OH})_2$, or tschermakite and ferrotschermakite $\text{Ca}_2(\text{Fe}, \text{Mg})_3\text{Al}_2\text{Si}_6\text{Al}_2\text{O}_{22}(\text{OH})_2$. It has long been known that there are no igneous tremolites or actinolites, but the present data define the limit of the approach of the igneous amphiboles to tremolite-actinolite much more precisely than hitherto. Wright and Bowes's (1968) interesting observation of actinolite-tremolite in explosion breccias at Kentallen, Argyllshire, Scotland has not yet been verified chemically, but seems possible under the extreme conditions of high water-vapour pressure and low temperature such as these explosion breccias clearly crystallized under. The amphibole analyses available from igneous rocks do not exceed Si 7.50 in the half unit cell, the largest Si value being 7.48 for an analysis with 50.46 % SiO_2 given by Haslam (1968) from the Outer Quartz diorite of Ben Nevis, Scotland. The water content of this analysis (1.37 %) is probably rather low as it is the lowest of 11 amphibole analyses given by Haslam and consequently the sum of the Y group (5.42) is rather high and so the Si and $\text{Ca} + \text{Na} + \text{K}$ are also presumably a little high. Calculated on the basis of 23 (O), Si is 7.38 and the true value probably lies between 7.38 and 7.48, close to Si 7.45 given by another of Haslam's analyses.

Most important is Haslam's (1968) observation that the amphiboles in the Ben Nevis Quartz diorites did not crystallize primarily from the magma but replaced magmatic pyroxenes at some temperature below the liquidus of the magma, a conclusion earlier pointed out for many Caledonian igneous diorites, tonalites, and granodiorites by Nockolds and Mitchell (1948). Obviously, then, the field of true igneous hornblende does not even extend as far as Si 7.50. Unfortunately most of the analyses plotted on fig. 3 are not accompanied by sufficiently detailed textural observations to distinguish magmatic and post-magmatic crystallization. If a separate plot (fig. 4) is made of the analyses recently given by Haslam (1968) and Troll (1968) of post-magmatic hornblendes and by Dodge *et al.* (1968) of magmatic hornblendes from Sierra Nevada granodiorites and quartz monzonites, it is clear that magmatic hornblendes extend at

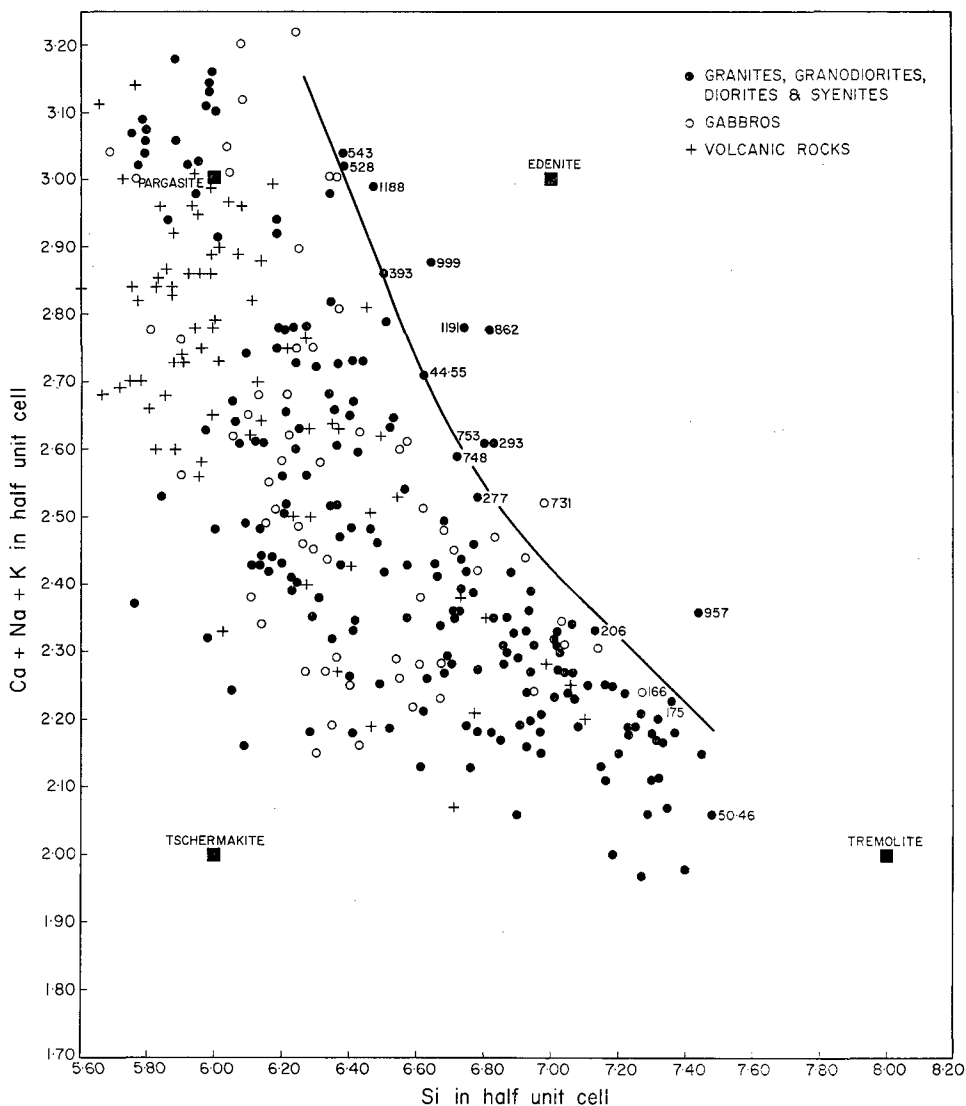


FIG. 3. Plot of Si against Ca+Na+K in the half unit cell for the available igneous calciferous amphiboles excluding inferior analyses. Curved line indicated suggested limits of igneous amphiboles. Numbered analyses are discussed in text.

least to Si 7.30 while the post-magmatic ones overlap the igneous field and extend to some value below Si 7.50.

Fig. 3 also suggests that there is some definite control over the maximum content of Ca+Na+K that can be incorporated into any igneous hornblende at any particular Si content. Thus the field of the analysis is not arbitrarily ended but is almost linearly

ended, a circumstance strongly supported by the recent data in fig. 4. This suggests that this edge is defined by the limit of crystallization of magmatic hornblende and it is significant that Haslam's post-magmatic amphiboles lie on the upper side of the igneous amphiboles of Dodge *et al.* (1968). It is suggested that igneous hornblendes are limited to the field below the line drawn on figs. 3 and 4, and that hornblendes crystallizing from gabbros and volcanic rocks do not exceed Si about 7.0, the lower Si and higher Al^{iv} being promoted by the higher temperature of crystallization as pointed out by Harry (1950).

It is necessary to examine critically the 10 analyses that plot above this line and one or two of the analyses that plot on or immediately below the line. In order of catalogue number these analyses are:

166. Spinel-bearing gabbro, Kazi Island, Ehime Pref., Japan. T. Yoshimura (1940) *Journ. Geol. Soc. Japan*, **47**, 304. Total 100.87 is rather high: Ca 2.14 and H_2O 2.87 are very high for a gabbroic amphibole. This analysis is therefore probably significantly erroneous for the present purpose.

175. Pegmatite with no feldspar in a hybrid rock. Cairnsmore of Carsphairn, Kirkcudbrightshire, Scotland. W. A. Deer (1937) *Geol. Mag.* **74**, 360. This is believed to be a superior analysis and the amphibole probably crystallized from water-rich residual magma at the limit of igneous hornblende crystallization.

206. Mixed syenitic hybrid, Glenelg-Ratagain igneous complex, Inverness-shire, Scotland. G. D. Nicholls (1950) *Quart. Journ. Geol. Soc.* **106**, 331. This is also believed to be a superior analysis.

277. Corona around quartzite xenolith in orthoclase olivine gabbro (Kentallenite). Kentallen Quarries, Argyllshire, Scotland. I. D. Muir (1953) *Geol. Mag.* **90**, 416. A superior analysis with crystallization at the transition between igneous and metamorphic conditions.

293. Granodiorite, Morven-Strontian complex, Inverness-shire, Scotland, S. R. Nockolds and R. L. Mitchell, 1948. *Trans. Roy. Soc. Edinburgh*, **61**, 560. This analysis is essentially a superior analysis but its position on the plot, combined with the rather low water content of 1.40, suggests that this amphibole may lie exactly on the line, a position it adopts when calculated on the basis of 23 (O) and due to the slightly low water content it plots above the line though it is within experimental error.

393. Reference as 293 but this amphibole plots on the line.

528. Nepheline syenite, Stavarnsjö, Norway. W. Kunitz (1930) *Neues Jahrb. Min. Abt. A*,

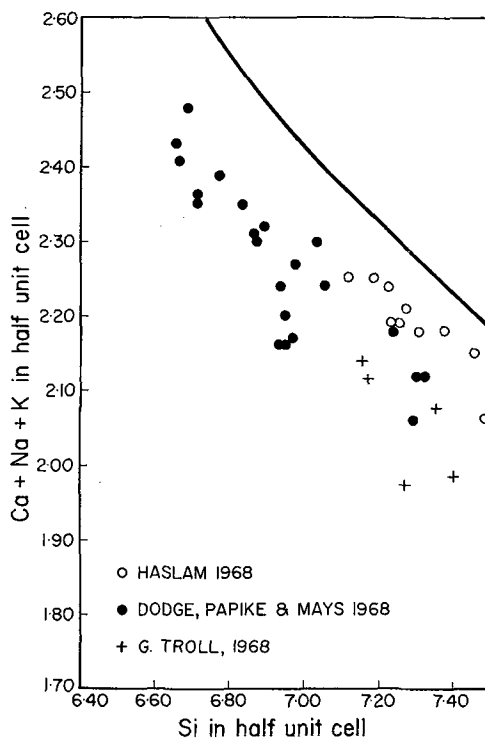


FIG. 4. Plot of Si against Ca+Na+K in the half unit cell for some recently analysed hornblendes considered to be reliable and which plot close to the limit of the igneous field.

60, 244. This appears to be a moderate analysis and the line has been drawn close to this analysis.

543. Nepheline syenite, Skuttersundskjar, Norway. W. Kunitz (1930) *Neues Jahrb. Min. Abt. A*, **60**, 244. Total a little high and water (1·16) a little low.

731. Albite-rich diabase of igneous origin. Vähä-Kurkkio, Enontekiö, N.W. Finland. K. Merilainen (1961) *Bull. Comm. Géol. Finlande*, no. 195, 43. Has insufficient Si+Al to fill the 8 Z positions; probably crystallized entirely in the solid state by replacement of pyroxene. The albite suggests an unusually low temperature of crystallization.

748. Granite, Osnitsk, Ukraine, U.S.S.R. O.I. Matkovsky (1960) *Min. Sborn. Lvov Geol. Soc.* **14**, 374. Total (100·76) is rather high but the analysis otherwise seems to be excellent.

753. Diorite with biotite and An₁₁. Holyczowka, Korzec, Ukraine, U.S.S.R. I. Kardymowicz (1957) *Roczn. Pol. Tow. Geol.* **27**, 92. H₂O— (0·99) astonishingly high and the Y group total rather low (4·70) but the albite-rich nature of the feldspar suggests an unusually low temperature of crystallization and the analysis is within analytical error of the line drawn.

862. Granite with albite and fluorite. Pankshin Complex, Jos Plateau, Nigeria. G. D. Borley and M. T. Frost (1963) *Min. Mag.* **33**, 651. H₂O+ not determined so cell contents uncertain and plotted point based on 23 (O). Probably plots near to the line as the occurrence of albite and fluorite suggest a low temperature of crystallization.

957. Granite. Pre-de-Fouchon, Vosges. J. Jung (1927) *Mem. Service Carte géol. Alsace Lorraine*, no. 2, 350. H₂O+ (1·38) rather low and total rather high (100·72) especially as MnO not determined. Analysis is thought to be faulty.

999. Grandiorite, Ilmen Mountains, Urals, U.S.S.R. D. Beljankin (1910) *Nachr. Petersburg Polyt.* **17**, 150. H₂O+ (0·70) too low and this results in the analysis plotting above the line.

1188. Nepheline syenite. Godavari River, Rudramkota, Khammam district, Andhra Pradesh, India. K. V. Subbarao (1965) Ph.D. thesis, Sri Venkateswara University College, Tirupati, A.P., India. Plots within analytical accuracy of the line. Analysis is good but water may be a little low (1·32).

1191. Granite. Peabody, Salem Quadrangle, Massachusetts, U.S.A. P. Toulmin III (1964). *Bull. U.S. Geol. Survey* **1163-A**, A33. Recalculated after subtracting impurities. H₂O+ (1·06) is low. Probably plots near or on line.

44·55 % Granodiorite, Shomyogawa, Hida, Japan. H. Shibata, N. Oba, and N. Shimoda (1966) *Sci. Rep. Tokyo, Kyoiku Daigaku, Sec. C* **9**, 103. Line is drawn through this analysis although the Si+Al (7·90) does not quite fill all the Z sites.

The line on fig. 3 has been drawn rather conservatively and particular magmas, at particular total pressures and partial pressures of water and oxygen, will each be limited to different positions on the low Si side of the line, as suggested by fig. 4, but clearly igneous edenites do not exist and it is important to discover why.

As the composition of edenite-ferroedenite, like tremolite-actinolite, has no extraordinary element requirements that typical magmas could not supply the absence of these amphibole varieties is ascribed to the absence of suitable conditions of magma crystallization combined with the limited stability fields of these amphiboles. The general absence of volcanic amphiboles (crystallized under high-temperature and low-pressure conditions) near to the Si-rich edge of the igneous amphibole field suggests that this edge is populated by rather low-temperature amphiboles or high-pressure ones. The latter suggestion is disproved by the number of amphiboles plotting near to the line that come from high-level intrusions, e.g. Scottish Caledonian and Sierra

Nevada diorites and granodiorites. Further light on this problem should be obtained by examining metamorphic calciferous amphiboles.

A plot of metamorphic calciferous amphiboles (fig. 5) shows a much wider spread than the igneous amphiboles, probably reflecting mainly the wider lower thermal stability of many amphiboles. Nevertheless, there is a distinct thinning of the plots towards both edenite and tschermakite. Previous re-analysis of a number of amphiboles supposed to plot very near to edenite showed these to be completely erroneous and the nearest verified analysis to edenite-ferroedenite is the Eganville amphibole (Leake, 1962) but this has only 1.49 Ca and strictly should not appear on the plot, which otherwise has a lower limit at 1.50 Ca.

It is necessary therefore to critically examine some of the analyses that approach edenite. Certainly, also some of the amphiboles plotting between edenite and pargasite with X group totals near to or exceeding 3 are erroneous. For example, two analyses given by Sen (1966) and plotted as D2 and 41A on fig. 5 are reproduced in table II. It will be noted that 41A has X and Y group totals of 3.17 and 4.64, with $\text{Fe}^{3+} > \text{Fe}^{2+}$ and $\text{Ca} > 2.00$ while D2 also has rather high Fe^{3+} relative to Fe^{2+} . Re-analysis of these amphiboles (table II), kindly supplied by Dr. Sen, gives substantially different values and moves the analysis points markedly on both figs. 1 and 5, repeating previous gloomy experiences with amphibole analyses. The analysis labelled 43.71 on fig. 5 was also given by Sen (1966) and this too is believed to be erroneous.

The nearest published analysis to $\text{NaCa}_2(\text{Fe}, \text{Mg})_3\text{Si}_7\text{AlO}_{22}(\text{OH})_2$ (edenite-ferroedenite) is labelled GR on fig. 5 and in table II, being published by Govinda Rajulu and Shariff (1966). Re-analysis of this amphibole, as generously supplied by Dr. Govinda Rajulu, gave similar values (table II) to the previous ones except for the $\text{H}_2\text{O}+$ and Al_2O_3 . The higher Al_2O_3 of the original analysis is partly because of undetermined Cr_2O_3 , partly because of lower total iron, and possibly because some silica had been carried over into the R_2O_3 precipitate, a common difficulty. The new analysis although departing more from the theoretical Si_7Al_1 of edenite than the original analysis is in other ways nearer to edenite for the Al^{VI} is markedly reduced from 0.80 to 0.21. From the original account of this amphibole, which occurs in an amphibolite, there is no indication of the metamorphic conditions, but in a personal communication Dr. Govinda Rajulu notes that plagioclase has not been found in these amphibolites but epidote is sometimes present, suggesting very low temperatures. He also notes evidence of 'deuteric action' probably indicative of high pH_2O , which could be consistent with an unusually low temperature of crystallization.

The next nearest analysis to edenite is 1100 (fig. 5) given by Watanabe (1965) from an amphibolite, but the extremely low $\text{H}_2\text{O}+$, which together with $\text{H}_2\text{O}-$ is only 0.57 %, makes this analysis unacceptable and is partly the cause of the high X group total. Analysis 228 comes from a gabbroic amphibolite, Urundi, central Africa (Vande Putte, 1939) and unsuccessful attempts have been made to locate this sample in Belgium by Professor Bartholomé. As K_2O was never determined, although Na_2O cannot be determined alone by the classical method of alkali analysis, it seems the high Na_2O (2.92 %) cannot be reliable. Analysis 46.4 (Bearth, 1967) is semi-quantitative and has been noted in connection with its Al^{VI} content.

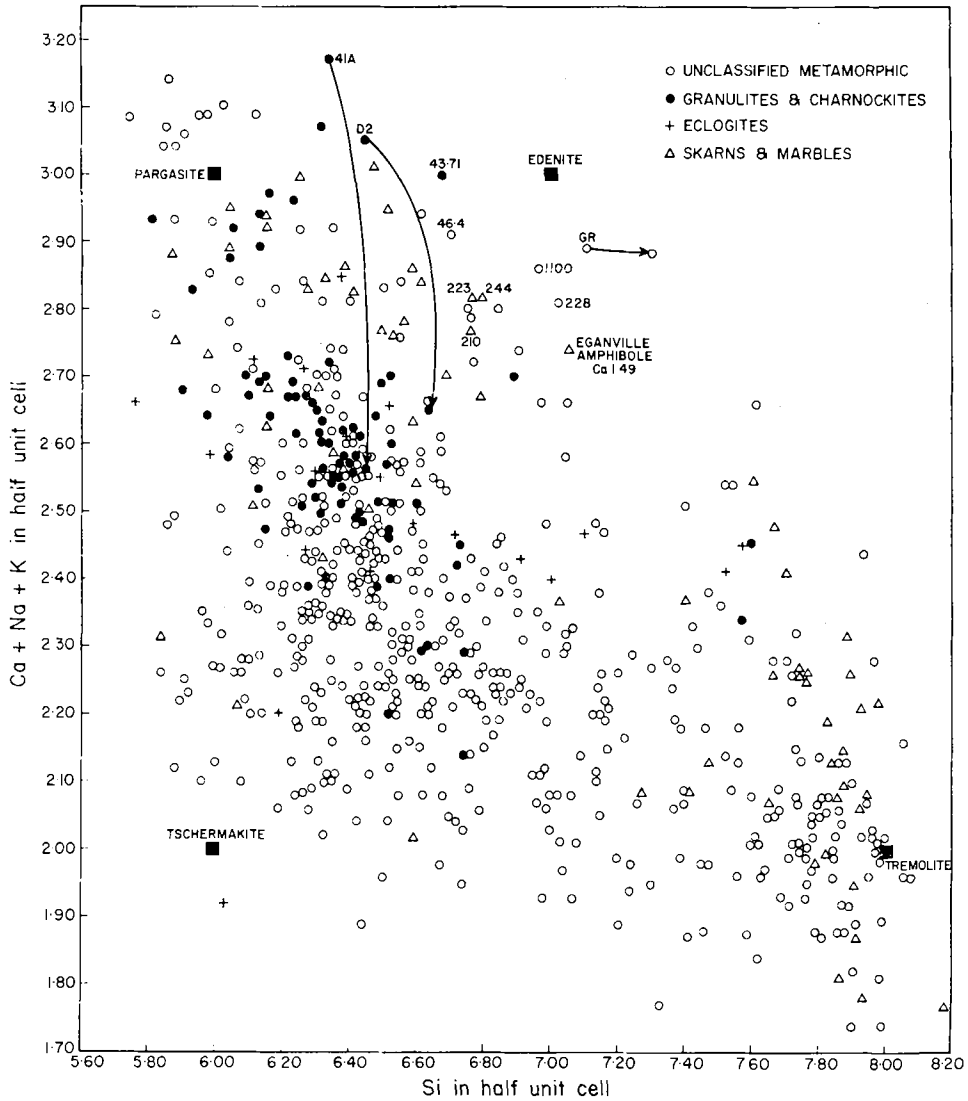


FIG. 5. Plot of Si against Ca + Na + K in the half unit cell of the available metamorphic calciferous amphiboles, excluding inferior analyses. Numbered analyses are discussed in text.

TABLE II. *Analyses of amphiboles approaching edenite*

	1 41A	2 41A	3 D ₂	4 D ₂	5 GR	6 GR
SiO ₂	41.00	42.49	42.40	44.76	49.00	50.70
Al ₂ O ₃	11.02	12.40	14.73	12.37	10.00	5.45
TiO ₂	0.81	1.84	0.80	1.82	0.26	0.32
Cr ₂ O ₃	—	0.01	—	—	—	0.48
Fe ₂ O ₃	12.82	3.01	7.01	1.80	1.82	1.81
FeO	10.48	19.08	11.87	14.68	5.93	6.59
MgO	5.72	5.39	5.34	8.37	15.56	15.89
CaO	12.80	11.25	11.47	11.72	12.85	12.22
Na ₂ O	2.59	1.49	2.83	1.69	2.83	2.80
K ₂ O	0.91	0.97	1.20	1.06	0.54	1.23
MnO	0.25	0.32	0.22	0.18	0.20	0.17
P ₂ O ₅	—	—	—	—	—	0.00
H ₂ O+	1.03	1.86	1.13	1.70	0.72	2.08
H ₂ O—	0.05	0.00	0.08	0.00	—	0.00
	99.48	100.12	99.08	100.20	99.71	99.80
<i>Atoms to 24 (O,OH)</i>						
	1	2	3	4	5	6
Si	6.34	6.45	6.44	6.64	7.10	7.29
Al ^{iv}	1.66	1.55	1.56	1.36	0.90	0.71
Al ^{vi}	0.35	0.67	1.08	0.79	0.80	0.21
Ti	0.09	0.21	0.09	0.20	0.03	0.03
Fe ³⁺	1.48	0.35	0.80	0.20	0.20	0.20
Fe ²⁺	1.36	2.43	1.51	1.82	0.72	0.79
Mn	0.04	0.04	0.03	0.03	0.02	0.02
Mg	1.32	1.22	1.20	1.84	3.36	3.40
Ca	2.12	1.83	1.86	1.86	1.99	1.88
Na	0.78	0.44	0.83	0.49	0.79	0.78
K	0.27	0.28	0.36	0.30	0.10	0.22
OH	1.06	1.88	1.14	1.66	0.70	1.99
O	22.94	22.12	22.86	22.31	23.30	22.01
Σ Z	8.00	8.00	8.00	8.00	8.00	8.00
Σ Y	4.64	4.92	4.71	4.88	5.13	4.71
Σ X	3.17	2.55	3.05	2.65	2.89	2.88
mg	0.31	0.30	0.34	0.47	0.78	0.77
α	1.652	1.672	1.663	1.663	1.625	1.630
γ	1.677	1.690	1.688	1.687	1.667	1.651

- 1: 41A, Ferri-magnesian hastingsitic hornblende with An₂₈, iron ore, and quartz in a granulite, Saltora (86° 56' E, 23° 31' N), West Bengal, India. S. K. Sen (1966, p. 128). H₂O+ is loss on ignition at 600 °C. Optics by personal communication.
- 2: 41A, Magnesian hastingsitic hornblende as above. Analysis total includes F 0.02 and O ≡ F 0.01. Analyst, A. Kemp. Optics, B. E. Leake.
- 3: D₂, Magnesian hastingsitic hornblende with An₄₈, ortho- and clinopyroxene, iron ore, biotite, and apatite in a granulite, Saltora (86° 56' E, 23° 31' N), West Bengal, India. S. K. Sen (1966, p. 128). H₂O+ is loss on ignition at 600 °C. Optics by personal communication.
- 4: D₂, Ferro-edenitic hornblende as above. Analysis total includes F 0.08 and O ≡ F 0.03. Analyst, A. Kemp. Optics, B. E. Leake.
- 5: GR, Edenite in an amphibolite with secondary dahllite, apatite, colophonane, and quartz, Khushalnagar, Coorg District, Mysore State, India. B.V. Govinda Rajulu and A. Shariff (1966, p. 363).
- 6: GR, Edenite as above. Analysis total includes NiO 0.06 giving Ni 0.01 in the cell contents while Cr is 0.05. Analyst, A. Kemp. Optics, B. E. Leake.

On the other hand, analysis 210 from a marble at Pargas, Finland (Kreutz, 1908) is confirmed by 223 from the same locality (Laitakari, 1920) and is also very similar to 244, from marble at Noble Mine, Sterling Hill, New Jersey, U.S.A. (Palache, 1935), in a very similar assemblage with chondrodite. These analyses are believed to be reliable.

It is concluded that as yet no analysis of a natural amphibole very close to edenite-ferroedenite is available, that of amphibole GR being the nearest, but the field of metamorphic amphiboles encroaches appreciably nearer to edenite-ferroedenite than does the field of igneous amphiboles. It should be pointed out that the method of representing amphibole analyses used by Colville *et al.* (1966) and reproduced by Ernst (1968) is misleading in the present context as it shows many analyses plotting between edenite and ferroedenite but this is based only on the content of $\text{Fe}^{3+} + \text{Al}^{\text{VI}}$ and ignores Si and $\text{Ca} + \text{Na} + \text{K}$.

The reason for the scarcity of edenite amphiboles must be speculative in view of the absence of experimental work on the phase relationships of edenite but the natural assemblages do suggest that low temperatures are essential and that the upper thermal stability of edenite-ferroedenite is well below that at which magmas can exist. There is no evidence to support the view that extraordinary pressures are required.

It is notable that whereas fig. 5 shows eclogite amphiboles to have a wide scatter, granulites and charnockites are much more confined (most of the few scattered analyses are certainly erroneous) and fall into the field of igneous amphiboles, thus confirming recent views on the importance of temperature and the relative unimportance of pressure in the evolution of charnockitic rocks.

The existence of two separated fields of amphiboles from skarns and marbles is again confirmed by fig. 5 with one group near to tremolite and the other near to pargasite.

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