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THE RELATIONSHIP BETWEEN TETRAHEDRAL ALUMINUM AND THE MAXIMUM POSSIBLE OCTAHEDRAL ALUMINUM IN NATURAL CALCIFEROUS AND SUBCALCIFEROUS AMPHIBOLES

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ABSTRACT

Critical examination of a plot of 936 available analyses of calciferous and subcalciferous amphiboles leads to the conclusion that the maximum possible Al^6 increases regularly as the Al^4 increases. The maximum allowed Al^6 at any Al^4 value is empirically obtained. New chemical analyses, together with new optical determinations, are given for three previously described amphiboles, from Madras and North Carolina, which appeared to contain more than the maximum allowed Al^6. The new analyses show that the two Madras amphiboles contain much less than the maximum possible Al^6, whereas the North Carolina sample contains the most Al^6 allowed for its Al^4 content.

INTRODUCTION

It is well-known that temolite and actinolite are poor not only in tetrahedrally coordinated aluminum but also in octahedrally coordinated aluminum, whereas the silica-poor pargasite, hastingsite and the hypothetical tschermakite are rich in both tetrahedral and octahedral aluminum. It seems therefore that in these amphiboles increasing amounts of tetrahedral Al permit increasing amounts of octahedral Al and it follows that at any one value of Al^4 there is certain maximum amount of Al^6 which is feasible. This paper tries to show what the maximum possible amount of Al^6 is for each Al^4 value of natural calciferous and subcalciferous amphiboles. Calciferous amphiboles are defined as those with at least 1.50 Ca in the half unit cell, referred to 24 (O, OH, F, Cl), while subcalciferous amphiboles contain more than 1.00 and less than 1.50 Ca in the half unit cell.

Obviously the maximum allowable Al^6 will only be obtained in amphiboles whose host rocks have a suitable chemistry and which have crystallized under the most propitious physical conditions. It has been argued elsewhere (Leake, 1965) that the most suitable conditions are those of high pressures and moderate, not high, temperatures, for igneous amphiboles generally have low amounts of Al^6 whereas amphiboles with the highest Al^6 values are often associated with kyanite, jadeite or glaucophane, which are often considered indicative of high pressure conditions. This agrees with the general view formed from other mineral groups that high Al^6 is favoured by high pressures during crystallization. Examples include clinopyroxenes (LaBas, 1962), orthopyroxenes (Boyd and

England, 1960,) and the Al_2SiO_5 polymorphs, while it is significant that the only amphibole which contains really large amounts of Al^6 is glaucophane, which is excluded from this account as it is neither calciferous nor subcalciferous.

PREVIOUS ANALYSES

In Fig. 1 936 post-1890 chemical analyses of calciferous and subcalciferous amphiboles have been plotted. Those which lie above the line drawn across the figure are believed to be unreliable analyses and so the line shows the maximum amount of Al^6 at each Al^4 value assuming that the relationship is linear. While further analyses will undoubtedly slightly modify the precise position of the line there seem to be enough reliable analyses immediately below the line to ensure that it is substantially correct.

It is important to appreciate that a plot of Al^4 against Al^6 for amphiboles is hypersensitive to analytical error. It is now well-known that by both classical methods of chemical analysis and by the more recent colorimetric methods, the accuracy of alumina determination is among the poorest of the major constituents of rocks and minerals. The accuracy of combined water determinations is also very poor when, as with amphiboles, the material being analyzed does not part with all its water below 1000°–1200° C. Most determinations of combined water have been, and still are, made by the Penfield method (Penfield, 1894) and this is known to give low results (Riley, 1958a) unless a silica tube and a temperature in excess of 1000° C is used. (Glass tubes melt at such a temperature). A great many amphibole analyses are therefore seriously deficient in water. Low OH in the cell content calculation results in spurious excessive oxygen, a situation aggravated by the common absence of F and Cl determinations. This excessive oxygen is the cause of erroneously high cation values, especially high Si and Al. As Al^6 is obtained by subtracting $8(Si+Al)$ from the total $Si+Al$, when the calculation is on the basis of $24(O, OH)$, the resulting Al^6 is often grossly excessive. It is therefore necessary to appraise critically those analyses which have high contents of Al^6 for their Al^4 value.

The analyses which plot above the line in Fig. 1 have been numbered with the same numbers as they have in a compilation of amphibole analyses, which it is hoped are to be published shortly. Accordingly these analyses are not reproduced here and only their sources are given together with brief notes on the analyses to indicate why they are regarded as unreliable. Obviously many, but not all, of the analyses which plot below the line are also unreliable but they are not of interest at present. A few of the numbered points, above the line are analyses which have

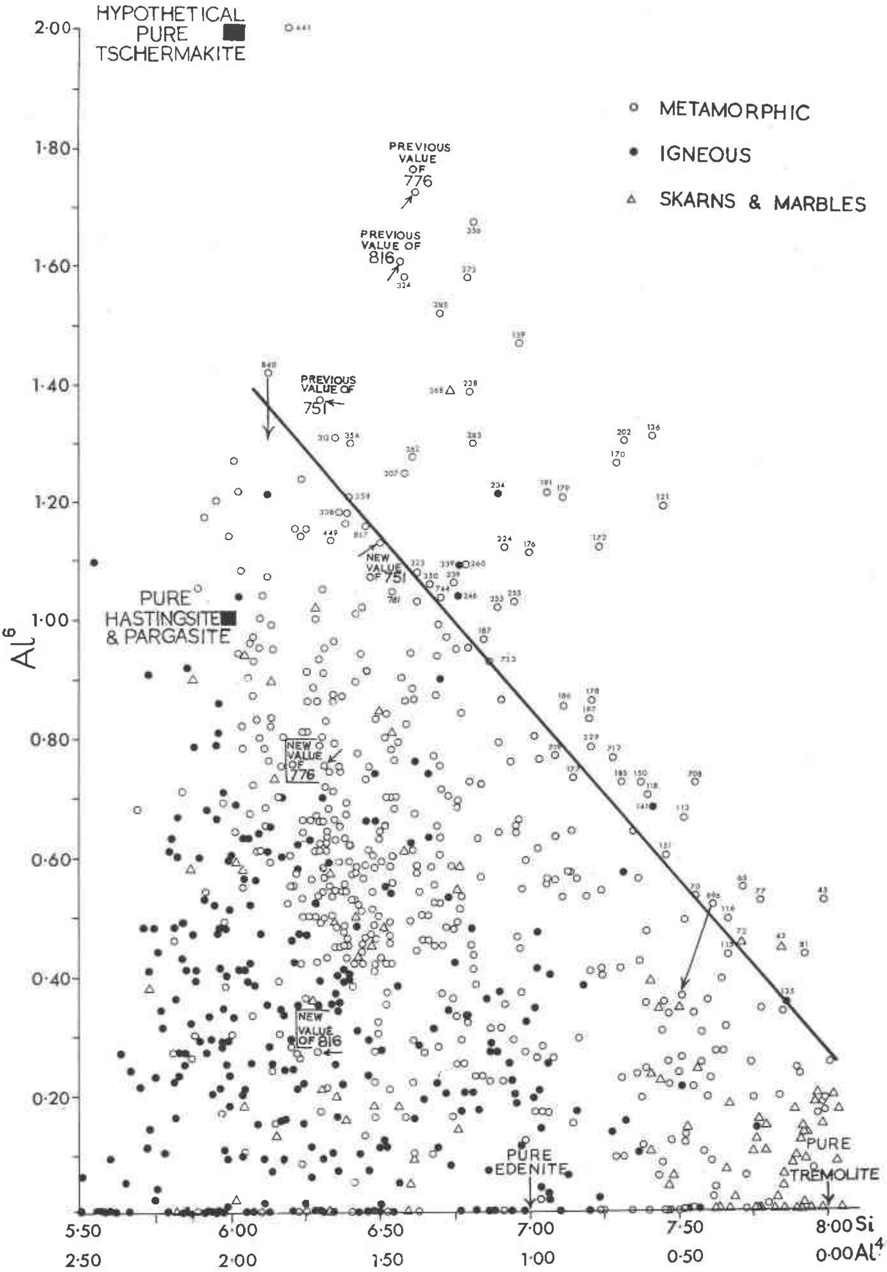

FIG. 1. Plot of octahedral aluminum in the half unit cell against tetrahedral aluminum (or silicon) for 936 analysed calciferous and subcalciferous amphiboles. The numbered analyses are discussed in the text. The suggested maximum possible Al^6 is shown by the line drawn across the figure. Analyses with Si greater than 8.00 are excluded from the plot as they are obviously erroneous.

no certain deficiencies and although they are believed to be erroneous, it has not been possible to conclusively show this. These are also listed below. It is generally accepted that amphibole compositions calculated from a chemical analysis of a rock and its modal constitution are not reliable unless the amphibole forms a very large proportion of the rock and so such compositions are not discussed in detail. Some of the analyses which plot just below the line and which are considered to be trustworthy are numbered and brief notes on these are given separately.

Analyses which plot above the line in Fig. 1.

43 Peltola, E. (1960) *Bull. Comm. Geol. Finlande* **192,** 53. Analysed material contained 3.95% C. as graphite, 0.21% S as pyrrhotite and 1.64% CO_2 as calcite. Analysis was re-calculated to 100% to remove these impurities. OH+F+Cl in cell contents is low, being only 0.83.

45 Yen, T. P. (1959) *Bull. Geol. Surv. Taiwan* **11,** 23. H_2O+ is far too low, being only 0.17.

65 Andreatta, C. and R. Pirani (1950) *Min. Abst.* **11,** 330. H_2O+ is rather low (1.24%) especially as only a trace of Fe_2O_3 is reported and the host rock is a serpentine.

70 Rondolino, R. (1936) *Min. Abst.* **7,** 26. TiO_2 and FeO not reported; Fe_2O_3 said to be a trace, analysis total, 99.14, is low.

72 Dolar-Mantuani, L. and Gr. Gagarin (1936) *Min. Abst.* **7,** 256. OH+F in cell contents is low, being only 0.90.

77 Krenner, A. (1900) *Neues Jahr. Min. Ref.* **1,** 341. Old analysis with TiO_2 and MnO not determined, H_2O+ low, being 0.51, analysis total, 100.97, is high.

81 Parsons, A. L. (1930) *Univ. Toronto Studies, Geol. Ser.* **29,** 31. OH in cell contents is low, being only 0.98.

113 Mikkola, E. and T. G. Sahama (1936) *Bull. Comm. Geol. Finlande* **115,** 367. Recalculated after subtracting about 2½% carbonate. H_2O+ is very low, being 0.42.

116 Phillips, A. H. and H. H. Hess (1936) *Am. Mineral.* **21,** 346. H_2O+ is very low, being 0.39.

118 Novotny, M. (1949) *Min. Abst.* **11,** 199. Recalculated to 100% to remove impurities; TiO_2 and Fe_2O_3 not reported.

121 Jure, A. E. (1930) Unpub. Ph.D. thesis, Wisconsin University. Al_2O_3 and Na_2O probably wrong.

135 Heron, A. M. (1924) *Rec. Geo. Surv. India* **56,** 192. OH+F in cell contents is low being only 0.77.

136 As 121.

141 Jones, W. A. (1930) *Univ. Toronto Studies, Geol. Ser.* **29,** 68. OH in cell contents is low, being 0.92.

150 Eskola, P. (1950) *Am. Mineral.* **35,** 730. Recalculated from an analysis containing 25% of cummingtonite. $H_2O+0.87$, is low.

151 Van de Putte, R. (1939) *Bull. Musee royal d'Hist. nat. Belgique* **15,** (31), 8. K_2O and MnO not determined, $H_2O+1.00$, is low.

159 Yen, T. P. (1959) *Bull. Geol. Surv. Taiwan* **11,** 23. Although the CaO is rather low and the Fe_2O_3 is greater than the FeO, it is not certain that this is a poor analysis.

170 As 121, also the total, 99.30, is low.

172 Ramachandra Rao, M. B. (1937) *Bull. Mysore Geol. Dept.* **16,** 31.

176 Foslie, S. (1945) *Norsk Geol. Tidd.* **25,** 83. Calculated from a rock analysis and a mode with 55% hornblende

178 Lacroix, A. (1942) *Min. Abst.* **9,** 274. May contain some glaucophane impurity like 186.

179 As 121.

185 Eskola, P. (1914) *Bull. Comm. Geol. Finlande* **40,** 110. Analysis total, 99.20, CaO 7.99, and H_2O+ 0.56, are all low.

186 Van der Plas, L. (1959) *Leidse Geol. Meded.* **24,** 457. Excellent analysis but the material is zoned and heterogeneous with glaucophane cores.

187 Shevchenko, E. V. (1959) *Min. Sb. Geol. Soc. Lvov* **13,** 301.

191 Kreutz, S. (1908) *Sitz.-ber. Akad. Wiss, Wien* **117,** Abt I, 948. Analysis mostly by Rammelsberg (1858) and is rather old. Total, 99.04 and OH+F in cell contents of 0.39, are both low.

197 As 172.

202 Richarz, S. (1930) *Am. Mineral.* **25,** 65. TiO_2 not determined.

224 As 176 but the rock contains 66% hornblende.

229 Palmunen, M. K. (1925) *Fennia* **45,** (9), 9. $H_2O+0.16$, is far too low.

234 Yakovleva, M. E. (1941) *Compt. Rend. Dokl. Acad. Sci. U.S.S.R.* **31,** 794. $H_2O+0.91$, is rather low.

238 As 176 but the rock contains 61% hornblende.

239 As 176 but the rock contains 54% hornblende.

246 Morozewicz, J. (1928) *Neues Jahr. Min. Ref* II, 145.

253 As 151 but H_2O+ is 1.32 and analysis total is 99.46.
255 Naidu, P. R. J. (1942) *Current Sci. Bangalore* **11,** 192. TiO_2 and MnO not determined, $H_2O+0.70$ and the analysis total, 99.41, are low.
256 Ramberg, H. (1943) *Norsk. Geol Tidd.* **23,** 20. Calculated from a rock analysis and a modal analysis with 75% amphibole.
260 As 176 but the rock contains 60% hornblende.
263 Hezner, L. (1903) *Tsch. Min. Pet. Mitt.* **22,** 562. $H_2O+0.54$ is low.
273 As 121.
285 Sugi, K. (1935) *Jap. Jour. Geol. Geogr.* **12,** 133. Calculated from a rock analysis and a mode containing only 35% hornblende.
307 Pantin. H. M. (1956) *Trans. Geol. Soc. Glasgow* **22,** 57. $H_2O+1.28$, is rather low.
312 As 45 but calculated from a rock and modal analysis.
323 As 285 but the rock contains 64% hornblende.
324 As 285 but the rock contains 73% hornblende.
339 Rice, H. M. A. *Am. Mineral.* **20,** 308. $H_2O+0.35$ is low because it was determined at only 300° C. Analysis total, 99.13, is also low.
350 As 307 but H_2O+, 0.59, is even lower.
354 Serdiuchenko, D. P. (1954) *Dokl. Acad. Sci. U.S.S.R.* **96,** 1234, OH+F+Cl in cell contents is only 0.33.
358 Gustavson, M. and A. Grønhaug (1959) *Norsk Geol. Under.* **211,** 42. H_2O+, 0.45, is low.
362 As 307 but H_2O+, 0.95, is even lower.
368 Sustschinsky, P. P. (1912) quoted by Niggli, P. (1922) *Zeit. Krist.* **56,** 546. TiO_2 not determined; Al_2O_3 included MnO, $H_2O+0.74$, is low.
441 Hezner, L. (1908) *Neues Jahr. Min* **27,** 167. $H_2O+0.97$, is low and was determined by loss on ignition. Al_2O_3, 22.73%, is undoubtedly too high for 15 new analyses of hornblendes from the same series of schists in the same area (Steiger, R. (1961) *Schweiz. Min. Petr. Mitt.* **41,** 151) contain a maximum of 18.1% Al_2O_3 giving Al^6 1.24, Si 6.24, Al^4 1.76.
696 Hietanen, A. (1963) *U. S. Geol. Surv. Prof. Paper* **344-B,** 21. $H_2O+0.94$, is believed by Hietanen to be low and is the cause of the low analysis total, 98.78. If the cell contents are recalculated assuming that OH+F is 2.00 (instead of 1.20) then the analysis plots below the line on Fig. 1, as is shown.
708 Rajasekaran, K. C. (1961) *Indian Mineral.* **2,** 43. $H_2O+0.61$, is low.
712 Iwasaki, M. (1963) *Jour. Fac. Sci. Univ. Tokyo Sec.* II, **15,** 57.
744 Layton, W. (1963) *Jour. Geol. Soc. Australia* **10,** 263. CaO 14.59 and Al_2O_3 are probably both too high.
751
776
816
840 These are dealt with below.

Apparently reliable analyses for the present purpose, which plot immediately below the line in Fig. 1.

115 Compton, R. R. (1958) *Am. Mineral.* **43,** 893.
177 Joplin, G. A. (1939) *Jour. Roy. Soc. New South Wales* **73,** 95.
338 Larsen, E. S. Jnr. and W. M. Draisin (1948) *Proc. 18th Inter. Geol. Cong. Part* **II,** 71.
449 Rosenweig, A. and E. Watson (1954) *Am. Mineral.* **39,** 586.
719 Kaufman, H. (1963) *Geol. Studies. Brigham Young Univ.* **10,** 155.
761 Raychaudhuri, B. (1964) *Am. Mineral.* **49,** 199.
733 Siroshtan, R. I. and N. I. Polovko (1959) *Ukr. Acad. Sci. Geol. Ser.* **19,** 49.
817 Raychaudhuri, B. (1964) *Am. Mineral.* **49,** 199.

New Analyses

Of particular interest in Fig. 1 are those analyses which have very large amounts of Al^6. There is no doubt that 441 is an erroneous analysis (see above) whereas 256, 324 and 285, which are calculated from rock and modal analyses, are highly dubious. Two analyses marked "Previous value of 776 and 816" in Fig. 1 have had an interesting history. Both these amphiboles come from Madras State, India, and were originally described by Naidu (1960, p. 18) who gave the analyses reproduced in Table 1 as I and IV. Somewhat curiously changed figures were later

TABLE 1. CHEMICAL ANALYSES OF RE-INVESTIGATED AMPHIBOLES

	I	II	III	IV	V	VI	VII	VIII
SiO_2	45.91	45.91	44.42	43.41	43.91	42.78	45.34	46.84
Al_2O_3	18.25	18.35	14.30	19.04	19.04	11.40	18.66	15.96
TiO_2	0.05	0.05	0.13	0.11	0.89	1.35	0.02	0.04
Fe_2O_3		0.00	2.51		0.00	6.23	0.41	0.49
FeO	6.07	6.07	7.33	8.65	8.65	10.10	3.14	3.32
MgO	12.10	12.10	16.10	12.96	12.96	12.45	16.36	17.81
CaO	15.42	15.92	10.30	12.13	12.13	10.43	12.21	11.70
Na_2O	0.32	0.32	1.65	1.29	1.29	1.65	1.19	1.25
K_2O	0.05	0.05	0.02	0.17	0.17	0.55	0.16	0.10
MnO	0.08	0.08	0.13	tr.	tr.	0.25	0.06	0.04
P_2O_5			0.01			0.01	0.01	0.01
H_2O+		0.54	2.65		0.60	2.50	1.99	2.06
H_2O-		0.15	0.03		0.11	0.00	0.00	0.00
Total	98.30	99.54	99.58	97.76	99.75	99.70	99.89	99.96

NUMBER OF IONS ON THE BASIS OF 24(O, OH)

	I	II	III	IV	V	VI	VII	VIII
Si	6.44	6.61	6.32	6.18	6.36	6.29	6.31	6.52
Al^4	1.56	1.39	1.68	1.82	1.64	1.71	1.69	1.48
Al^6	1.45	1.72	0.72	1.38	1.61	0.27	1.37	1.13
Ti	0.00	0.00	0.01	0.01	0.10	0.15	0.00	0.00
Fe^3		0.00	0.26		0.00	0.69	0.04	0.05
Fe^2	0.71	0.73	0.87	1.03	1.05	1.23	0.36	0.38
Mn	0.01	0.01	0.02	0.00	0.00	0.03	0.01	0.00
Mg	2.53	2.60	3.41	2.75	2.79	2.72	3.39	3.69
Ca	2.31	2.46	1.57	1.85	1.88	1.64	1.82	1.74
Na	0.09	0.09	0.45	0.36	0.37	0.47	0.33	0.33
K	0.01	0.01	0.00	0.03	0.03	0.10	0.03	0.02
OH	(2.00)	0.52	2.51	(2.00)	0.57	2.44	1.84	1.90
O	(22.00)	23.48	21.49	(22.00)	23.43	21.56	22.16	22.10
ΣZ	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.00
ΣY	4.70	5.06	5.29	5.17	5.55	5.09	5.21	5.28
ΣX	2.41	2.56	2.02	2.24	2.28	2.21	2.18	2.09

I Amphibole from anorthite gneiss, Tiruchengodu, Sittampundi, Madras State, India. P. R. J. Naidu (1960) p. 18. $2V\alpha 95°$. Total includes Cr_2O_3 0.05.

II Same amphibole as I, P. R. J. Naidu (1963) p. 121. $2V_\alpha$ 85–90°. This is plotted on Fig. 1 as the 'Previous value of 776.'

III Same amphibole as I. Analyst, A. Kemp. $\alpha 1.637$, $\beta 1.648$, $\gamma 1.660$ $2V_\alpha 92 \pm 3°$. Associated feldspar, An_{88}. Plotted on Fig. 1 as 'New value of 776.'

IV Amphibole from biotite gneiss, Tiruchengodu, Sittampundi, Madras State, India. P. R. J. Naidu (1960) p. 18. $2V_\alpha$ 71°.

V Same amphibole as IV, P. R. J. Naidu (1963) p. 121. $2V_\alpha$ 72–78°. This is plotted on Fig. 1 as the 'Previous value of 776.'

VI Same amphibole as IV, analysts A. Kemp and B. E. Leake. $\alpha 1.661$, $\gamma 1.684$, $2V_\alpha 68 \pm 2°$. Associated felspar, An_{38}. Plotted on Fig. 1 as 'New value of 816.'

VII Amphibole from Clay County, North Carolina, U.S.A. H. Kaufman (1963) p. 152. $\alpha 1.626$, $\beta 1.643$, $\gamma 1.671$, $2V_\alpha 96$. Total includes Cr_2O_3 0.22, NiO 0.11, Li_2O 0.01, V_2O_5 0.00, F 0.00. Cell contents include Cr 0.03, Ni 0.01, Li 0.00, V 0.00, F 0.00.

VIII Same amphibole as VII. Analyst, B. E. Leake. $\alpha 1.626$, $\gamma 1.646$, $2V_\alpha 98$. Associated with anorthite, An_{94} and vesuvianite. Total includes Cr_2O_3 0.22, NiO 0.11, Li_2O 0.01, V_2O_5 0.00 and F 0.00 determined by C. O. Ingamells. Cells contents include Cr 0.02, Ni 0.01, Li 0.00, V 0.00, F 0.00.

published (Naidu, 1963, p. 121) and these are also reproduced in Table 1 (II and V). While the original analyses have extraordinarily high Al^6, the revised analyses, which are plotted in Fig. 1, have even more Al^6. It therefore seemed advisable to re-investigate these two amphiboles especially as one of the amphiboles was said to be associated with anorthite, and as this is the most aluminous plagioclase it therefore seemed feasible that this amphibole might be unusually aluminous. Professor Naidu kindly sent some of the analyzed amphiboles to the writer.

After purification of the powders new chemical analyses were carefully carried out in triplicate and appreciably different results were obtained (Table 1, III and VI) using the methods of Riley (1958b). The new analyses are plotted on Fig. 1 as "New values of 776 and 816." The new H_2O, Fe_2O_3, CaO and Al_2O_3 values are much more satisfactory. It is apparent that both amphiboles contain less than the suggested maximum amount of Al^6.

New 2V measurements on the amphiboles have been made and the refractive indices determined. The associated feldspar of 776 has γ-1.582, indicating An_{88} using the curves of Smith (in Hess, 1960) whereas the felspar with 816 has γ-1.555, indicating An_{38}.

Amphibole 840 (Hashimoto, 1961, p. 118) also appears to contain more than the allowed amount of Al^6. The host rock is an epidote-An_{48} amphibolite with minor magnetite and comes from Kyushu, Japan. The remaining piece of the hand specimen from which the amphibole was separated was kindly sent to the writer by Dr. Hashimoto as none of the originally analyzed powder was left. Unfortunately, despite several days work on this rock it has proved impossible to completely separate the hornblende from the epidote. There is a considerable overlap of density, magnetic and shape characteristics of the two minerals. A grain mount of the purest amphibole fraction obtained gave about 8% epidote by point-counting and analysis of this powder gave 16.86% Al_2O_3 compared with 19.26% reported previously. Since epidotes usually contain between 20 and 30% Al_2O_3 and epidotes with less than 20% Al_2O_3 are distinctly unusual (Deer *et al.* 1962, p. 199) it seems that the epidote impurity will enhance the Al_2O_3, and thus it can be deduced that the hornblende must contain less than 16.86% Al_2O_3. Probably the high alumina previously reported was at least partly the result of epidote contamination. Certainly, if the amphibole contains less than 16.86% Al_2O_3 then it will plot below the line drawn across Fig. 1.

Analysis 751, from Clay County, North Carolina, was recently published by Kaufman (1963, p. 152) and this plots well above the line in Fig. 1. As this analysis was made by C. O. Ingamells (Table 1, VII) it seemed unlikely that it would be significantly erroneous. Nevertheless, a number of disquieting features suggested a re-examination of this amphibole was necessary. The recorded refractive indices and birefringence of this amphibole, and several others published with it, seemed impossible for the compositions recorded. The mineral assemblage in the rock from which this amphibole came was not recorded but the analyzed powder was reported (Kaufman, 1963, p. 140) to contain "0.3% inclusions of actinolite, tremolite and hornblende (?)"

A piece of the rock was kindly sent to the writer by Professor W. R. Phillips. Thin sections of the rock show that only one, apparently homogeneous, amphibole is present associated wth vesuvianite and anorthite and with a trace of chromite. The anorthite has α-1.573, γ-1.585, indicating An_{94}, a surprisingly calcic composition. A new chemical analysis (Table 1, VIII) of carefully purified material shows significant differences from the previous result. These differences are consistent with the view that the originally analyzed powder was appreciably contaminated with anorthite, and possibly vesuvianite in addition, for this would explain the slightly increased SiO_2, TiO_2, Fe_2O_3, FeO, H_2O and the considerably increased MgO while also explaining the lower Al_2O_3 and CaO. The new analysis plots exactly on the line of maximum possible Al^6 (Fig. 1) which is not surprising in view of the aluminous nature of the rock, combined with what must be a low alkali content. The original powder was not pure simply because the rock was not crushed finely enough to separate the constituent minerals, for Kaufman (1963, p. 137) ground only to 60 mesh, whereas at least 120 mesh is essential to separate this amphibole.

New determination of γ gives 1.646, which agrees with the composition satisfactorily and also gives a reasonable birefringence.

It seems therefore that there are substantial reasons to believe that the maximum possible Al^6 at each Al^4 value is fairly well shown by the line drawn on Fig. 1. It remains a matter of very serious concern that so many amphibole analyses are not reliable. From Fig. 1 it is apparent that there are no amphiboles known which even approach the composition of the hypothetical tschermakite. The conclusions drawn certainly do not apply to amphiboles with less than 1.00 Ca in the half unit cell (*e.g.* glaucophane) and may not be valid for those containing just a little more than 1.00 Ca.

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