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Some aluminous clinopyroxenes from Vesuvius and Monte Somma, Italy

S. RAHMAN[1]
Department of Geology, Dacca University, Bangladesh

SUMMARY. Aluminous Ca-rich clinopyroxenes (5·95–7·63 wt % Al_2O_3) are next in abundance to leucite in the basic alkaline potassic lavas from Monte Somma and Vesuvius, Italy, and occur as phenocrysts (1 to 7 mm), microphenocrysts (< 1 mm), and groundmass granules. Zoning of various types is a conspicuous feature in these pyroxenes. Optical and chemical data are presented and a comparison is made between these pyroxenes and similar ones from alkali basalts. It is known from published data that Al_2O_3 fluctuates strongly in the oscillatory zones of Vesuvian pyroxenes. This can be explained as due to temperature variation in the magma, to magma variation in silica content due to contamination, to oscillations in leucite precipitation, or to a combination of these factors. Of these three factors, oscillations in leucite precipitation, as it appears, would be more effective than the others since it would have a greater control over Si/Al availability in the magma.

VESUVIUS (with Monte Somma), a typical strato-volcano dominating the Bay of Naples, Italy, is one of the three main volcanic centres that form the petrographic province of Campania. Clinopyroxene is one of the most abundant minerals in the Somma-Vesuvius basic alkaline lavas. The results of chemical and optical studies on clinopyroxenes from nine rocks (see Table I) are presented. No orthopyroxenes have been found. The commonly associated minerals in the Vesuvius rocks are leucite, calcic plagioclase (An_{70-85}), and opaque minerals (mainly iron oxides), whereas the clinopyroxene in Somma lavas is associated with minor olivine and occasional sanidine. The abundance of the phenocrystic clinopyroxenes can be seen from Table I.

The study of the pyroxenes from Somma-Vesuvius lavas has a long history. The most notable contributions, however, were made by Lacroix (1917), Washington and Merwin (1921), Alfani (1934), Zambonini (1936), and Muller (1936). Recently Savelli (1967) has given some analyses of these pyroxenes.

In hand-specimen and separated grains the clinopyroxenes are dark green in colour. The depth of greenish tinge decreases slightly towards the outer margin of each crystal. The clinopyroxenes form large (usually between 1 and 7 mm), euhedral or subhedral, prismatic or tabular phenocrysts. In thin section, the grains are pale yellowish in colour and almost non-pleochroic. In sample no. 22 (see Table I) the rims of the pyroxene phenocrysts and the groundmass granules are pleochroic from yellowish-brown to green.

Microphenocrysts are fairly common, occasionally forming glomerocrysts typically resembling xenolithic pyroxene accumulates. The amount of groundmass granules varies between samples. The almost identical optical nature of the various generations

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[1] Present address: Dept. of Geology, Makerere University, Kampala, Uganda.

TABLE I. *Modal (vol. %) analyses of Somma-Vesuvian rocks*

Sample	Leucite*		Clinopyroxene*		Feldspar†	Rest‡
	P	G	P	G		
7	10	32	6	8·5	14	29·5
9	31	9·5	5	8·5	15	31
12	20	7·5	10	5	9	49·5**
14	28	8·5	7	4·5	21	31**
16	25	3	11	15	20	26**
17	30	7	3	1·5	11	47·5**
19	25	6	6	7	9§	47
22	36‖		20	6	23·5¶	14·5**
38	25	11	10	4	40	10††

* P = phenocryst and microphenocryst; G = groundmass.
† Unless stated represents plagioclase feldspar (An_{85-70}).
‡ Includes glassy material, iron oxide, apatite, occasional olivine and biotite, some groundmass feldspar and pyroxene granules.
§ contains about 6 % phenocrystic plagioclase.
‖ distinction between phenocryst, microphenocryst, and groundmass is not possible.
¶ contains approx. 4 % sanidine (Or_{69}).
** olivine present.
†† olivine, Fo_{67}, present.

Nos. 7 to 19, *Vesuvian lavas*: all leucite tephrite.

7: Just west of Boscotrecase; probably 1906 flow. Medium-grey, compact, porphyritic rock containing numerous pale-grey leucite and dark-green pyroxene phenocrysts, which give a speckled appearance to the rock.

9: Sea-shore outcrop of Torre del Greco; a small castle is built on the headland formed by this flow of A.D. 1794 or 1861. Similar to sample 7 but contains a greater number of leucite phenocrysts.

12: West of Caprari and north of Boscoreale; a ropy variety, probably 1764 flow. The dark-grey hand-specimen clearly shows the ropy surface with gaps between the twisted cords. It is perforated by numerous flow-oriented vesicles and has a speckled surface, due to the spots of leucite and pyroxene.

14: Town of Massa di Somma; probably 1855 flow. Similar to 9 but the leucite phenocrysts are bigger in size.

16: On road to the summit of Vesuvius; north side of the road at San Vito; probably 1767 flow. Similar to 9 and 14, but leucite phenocrysts are more numerous.

17: Lower slope of Vesuvius; flow of 1944. This is a compact, dark-grey, porphyritic rock with some irregular vesicles. Pale-grey leucite phenocrysts cover almost a third of the surface.

19: Higher up than no. 17; 1944 flow. This is so vesicular that it looks rather spongy with globules of leucite standing out on the rough surface. On a cut surface, however, the typical speckled appearance due to the presence of leucite and pyroxene is present.

Nos. 22 and 38, *Somma lavas*: both phonolitic tephrites.

22: Scree at the foot of the rim of Monte Somma; prehistoric lava. A coarse-grained, porphyritic rock having a lighter shade due to the abundance of leucite and feldspar. Slightly pinkish due to the oxidation of iron.

38: Scree at the foot of the rim of Monte Somma; prehistoric lava flow. Highly porphyritic, light-grey, compact rock. The large phenocrysts that can be seen without any optical aid are greyish, waxy leucite, dark-green pyroxene, and some rare grains of yellowish olivine.

of pyroxenes suggests that the phenocrysts are probably non-accumulative in these rocks.

Myriads of inclusions are observed in these pyroxenes. Besides grains of leucite, the other common inclusions in the clinopyroxenes are granular and acicular apatite, small prismatic and lath-shaped plagioclase, granular iron ore, and patches of glass.

One of the most interesting characteristics of these clinopyroxenes is the zoning of the phenocrysts and microphenocrysts. Most common is the oscillatory type in which the zonary bands appear as alterations of very narrow, closely spaced, light and dark striae parallel to the crystal margins. Occasionally as many as 60 zones may be seen in one crystal. Hour-glass structure is very often associated with this type of zoning (fig. 1).

Less commonly, the zones are picked out by mineral inclusions, oriented in rows (very often only one row) parallel to the crystal outline. Small, rounded grains of leucite lying along the zones (fig. 1) probably indicate major breaks during the growth of the larger phenocrysts of clinopyroxenes.

In rare cases zoning is revealed by several isolated patches, generally anhedral, with outlines that bear no apparent crystallographic relationship to the final crystal outlines. This type of zoning is reported by Wilkinson (1957) in clinopyroxene from the Black Jack teschenite sill.

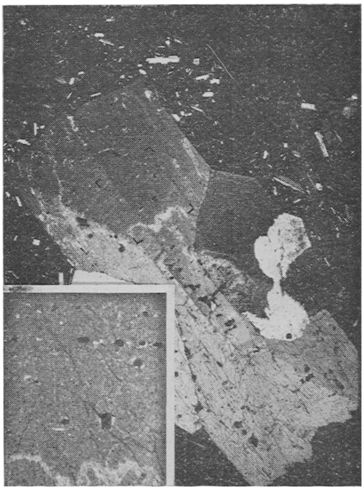


FIG. 1. Clinopyroxene crystal showing twinning, hour-glass structure, and oscillatory zoning embedded in glass in specimen 12. Photographic inset in the lower left is an enlargement of the area marked by brackets and shows the rhythmic precipitation of leucite blebs along some of the zones. Length of the clinopyroxene crystal is 6 mm. (X Polars.)

Method of separation and analysis. The separation and purification of the pyroxenes for chemical analysis was carried out with a magnetic separator and repeated centrifuging in Clerici solution. Purity of the sample was estimated to be not less than 99–99·5 %. The main impurity was acicular inclusions of apatite, and a correction to the composition of analysed pyroxene has been made from the determination of P_2O_5 (Table II) assuming no phosphorus in the pyroxene structure.

Total iron, TiO_2, MnO, and P_2O_5 were determined colorimetrically; CaO and MgO by atomic-absorption spectrophotometer, the precision being checked by gravimetric determination of CaO and MgO in four samples; Na_2O and K_2O were determined with the flame photometer. Total iron was also determined by titration with standardized ceric sulphate solution after reducing the ferric to ferrous iron in a silver reductor column. Ferrous iron was determined by decomposing the sample in HF in the presence of ammonium metavanadate and then titrating with standardized ferrous ammonium sulphate solution.

Optical data. The methods employed were essentially those of Hess (1949). Only the β-index was measured on the crushed pyroxene grains (Table II), using (100) parting tablets. All refractive index measurements were made in monochromatic sodium light with careful control of the temperature of the liquids. Since the pyroxenes were zoned, it was not possible to determine the exact index from grain to grain. However, it is believed that the index recorded in Table II gives a fair idea of the average β-index of the pyroxene in question.

Optic axial angle measurements were made on a Zeiss four-axis universal stage, using the orthoscopic method. The angles are believed accurate to within ±1°, but represent an average of zoned areas. The composition of the pyroxenes, estimated from measurements of the optic axial angle and β-refractive index (Hess, 1949; Muir, 1951), deviate considerably from the compositions obtained by chemical analyses. This discrepancy may perhaps be due to the presence of considerable amounts of Al, Fe^{3+}, and Ti in octahedral co-ordination (Table II), which are thought to increase β and lower 2V of the pyroxenes (Hori, 1954; Deer *et al.*, 1963).

Chemical data. Seven complete and two partial analyses of clinopyroxenes from Somma-Vesuvius lavas are presented in Table II. All the complete analyses are recalculated into the standard formula $(WXY)_2Z_2O_6$. The distribution of the cations between the (WXY) and the (Z) groups was made according to the method of making $Z = 2$ by alloting an arbitrary amount of Al to substitute for deficient Si in the tetrahedral site. The distribution of cations between the two sites has also been calculated according to the method described by Hess (1949) and for reasons discussed by Brown (1957). This second method was adopted mainly to check the quality of the present analyses. In both cases the formulae calculated on the basis of 6 oxygens are seen to fit closely to the theoretical structural formulae for clinopyroxenes. The number of Si together with Al atoms is always sufficient to make the Z group equal to 2. Al in the Z group is always greater, by as much as three to four times, than Al in the Y group.

One characteristic feature of the analyses of these pyroxenes is their general uniformity in bulk composition, which contrasts with the intense variation within the zones of which they are composed (Thompson, 1972). This reflects the limited range in composition of the rocks containing them (Table III). The amount of silica is fairly constant in these pyroxenes and is comparable with that of clinopyroxenes from some alkali basalts (Murray, 1954; Wilkinson, 1957; Aoki, 1964). Their Al_2O_3 contents are high (see discussion below), as is usual in pyroxenes from strongly alkalic rocks. They also contain notable amounts of Fe_2O_3, which, from the known purity of the minerals separates, must be held within the pyroxene structure in $CaFe_2SiO_6$ (Huckenholz, Schairer, and Yoder, 1969) and $CaFeAlSiO_6$ (Hijikata and Onuma, 1969) substitutions, rather than as Fe oxide inclusions. Although titania is not as high as is usually found in the pyroxenes from undersaturated feldspathoidal rocks (Kushiro, 1960; Le Bas, 1962), it is present in significant amount in these pyroxenes and increases with increasing % Al^{iv} (fig. 2). Analyses by Savelli (1967) of some Vesuvian clinopyroxenes show considerably more potash, silica, alumina, and iron oxide, and less lime and titania than the present analyses. The high potash and silica and low lime in Savelli's analyses cast some doubt on the purity of his pyroxene samples. On similar grounds Washington and Merwin (1921) classified the analyses of Vesuvian clinopyroxenes by Casoria (1907) and Zambonini (1910) as unacceptable. The high alumina content in Savelli's analyses will be discussed later.

Compositional variation of the pyroxenes. In fig. 4, compositions of the clinopyroxenes from Somma-Vesuvius rocks are plotted on the pyroxene quadrilateral. Their compositions fall in the sahlite field (Poldervaart and Hess, 1951). The crystallization trend

TABLE II. *Chemical analyses of Somma-Vesuvian pyroxenes*

	7	9	12	14	16	17	19	22	38
SiO_2	47·05	48·91	47·27	47·12	—	48·17	46·74	—	47·75
TiO_2	1·55	1·27	1·52	1·46	1·41	1·26	1·70	—	1·43
Al_2O_3	7·19	5·95	7·40	7·63	—	6·72	7·35	—	6·43
Fe_2O_3	4·13	2·74	3·07	3·01	2·94	3·13	3·78	2·47	2·85
FeO	3·98	4·01	4·91	5·02	4·44	4·31	4·88	3·44	5·78
MgO	12·25	13·69	12·26	12·83	13·30	13·03	12·23	14·52	13·63
CaO	23·30	23·18	22·88	22·82	22·77	22·69	22·67	23·08	21·49
MnO	0·11	0·14	0·15	0·14	0·14	0·14	0·14	—	0·12
Na_2O	0·43	0·41	0·47	0·46	0·42	0·46	0·49	—	0·49
K_2O	0·04	0·08	0·06	0·03	0·05	0·08	0·03	—	0·03
Total	100·03	100·38	99·99*	100·52	—	99·99*	100·00*	—	100·00*
Optical data									
β	1·716	1·709	1·713	1·713	1·713	1·715	1·718	1·709	1·709
2V	57·50°	54·75°	55·50°	54·75°	55°	55·65°	55·70°	50°	52°
Numbers of ions on the basis of 6 oxygens									
Si	1·755	1·806	1·763	1·749	—	1·789	1·747	—	1·779
Al^{iv}†	0·245	0·194	0·237	0·251	—	0·211	0·253	—	0·221
	(0·245)	(0·186)	(0·230)	(0·232)	—	(0·207)	(0·244)	—	(0·202)
Al^{vi}†	0·071	0·065	0·088	0·082	—	0·083	0·071	—	0·062
	(0·071)	(0·073)	(0·095)	(0·101)	—	(0·087)	(0·080)	—	(0·080)
Ti	0·044	0·035	0·043	0·041	—	0·035	0·048	—	0·040
Fe^{3+}	0·116	0·076	0·086	0·084	—	0·088	0·106	—	0·080
Fe^{2+}	0·124	0·124	0·153	0·156	—	0·134	0·053	—	0·180
Mn	0·003	0·004	0·005	0·004	—	0·004	0·004	—	0·004
Mg	0·681	0·753	0·681	0·710	—	0·721	0·681	—	0·757
Ca	0·931	0·917	0·914	0·907	—	0·903	0·908	—	0·858
Na	0·031	0·029	0·034	0·033	—	0·033	0·036	—	0·035
K	0·002	0·004	0·003	0·001	—	0·004	0·001	—	0·001
Z	2·000	2·000	2·000	2·000	—	2·000	2·000	—	2·000
	(2·000)	(1·992)	(1·993)	(1·981)	—	(1·996)	(1·991)	—	(1·981)
WXY	2·003	2·007	2·007	2·018	—	2·005	1·908	—	2·017
	(2·003)	(2·015)	(2·014)	(2·037)	—	(2·009)	(1·917)	—	(2·035)
Per cent cations									
	7	*9*	*12*	*14*	*16*	*17*	*19*	*22*	*38*
Ca	50·18	48·92	49·70	48·75	48·54	48·81	49·01	48·39	45·67
Mg	36·70	40·18	37·04	38·12	39·44	38·99	36·78	42·33	40·29
$Fe^{3+}+Fe^{2+}+Mn$	13·12	10·90	13·26	13·13	12·02	12·20	14·21	9·28‡	14·04
% Al_z§	12·27	9·71	11·86	12·57	—	10·58	12·65	—	11·03

* recalculated to 100 % after subtracting the values of P_2O_5, in samples 12, 17, 19, and 38 the amount (wt %) of P_2O_5 being 0·11, 0·21, 0·23, and 0·19 respectively.

† Values quoted within parentheses are obtained by the method of Hess (1949).

‡ without Mn.

§ % $Al_z = (Al^{iv} \times 100)/z$, where $z = 2$.

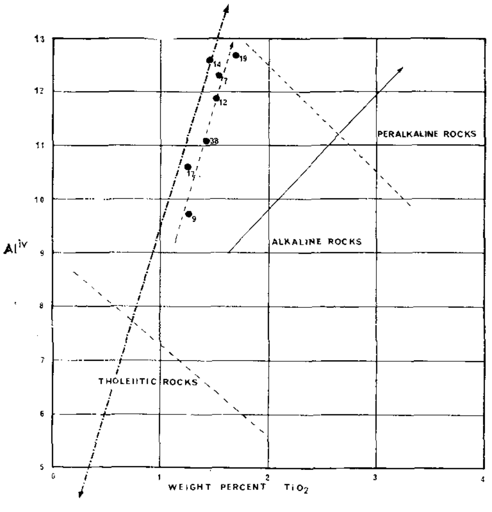
of these clinopyroxenes is almost parallel to the diopside–hedenbergite join, and is limited to the compositional field Fs_9–Fs_{15}, Wo_{45}–Wo_{50}. Similar restricted compositional trends of clinopyroxenes of alkali-basalt magmas analysed by classical means are illustrated by the pyroxenes of the Garbh Eilean sill, Shiant Isles (Murray, 1954), Black Jack sill, New South Wales (Wilkinson, 1957), monzonite complex at Mount Dromedary, New South Wales (Boesen, 1964), and Square Top intrusion, New South Wales (Wilkinson, 1966). However, in the more differentiated members of some alkali volcanic series (e.g. in certain mugearites, trachytes, and pantellerites), the clinopyroxenes, while still calcium-rich, reveal more extensive Fe^{2+}/Mg^{2+} replacements, their compositions extending towards hedenbergite (Muir and Tilley, 1961; Carmichael, 1962; Aoki, 1964). Gibb (1973) has recently used microprobe analyses to show that this is also the case in the Garbh Eilean sill.

TABLE III. *Chemical analyses of the Somma-Vesuvian rocks. Key to analyses see Table I*

	7	9	12	14	16	17	19	22	38
SiO_2	47·62	48·22	47·26	48·27	47·07	48·52	47·53	48·47	51·11
TiO_2	1·18	1·06	0·95	0·97	1·03	0·98	0·83	1·10	0·93
Al_2O_3	18·93	17·33	19·11	18·60	18·33	18·55	20·43	16·86	18·45
Fe_2O_3	3·19	4·48	2·23	4·37	3·38	5·51	3·07	5·12	1·25
FeO	4·82	3·16	5·62	3·14	5·33	2·19	4·45	2·72	5·30
MnO	0·19	0·19	0·21	0·19	0·23	0·20	0·21	0·18	0·16
MgO	3·56	4·69	4·50	3·88	3·97	3·62	3·54	5·23	4·83
CaO	9·02	9·45	9·78	8·59	8·91	8·19	8·10	9·98	8·42
Na_2O	2·82	2·56	2·72	2·73	2·84	2·58	2·69	2·15	1·95
K_2O	7·16	7·28	6·63	7·78	7·28	7·80	7·93	6·68	6·50
H_2O^+ H_2O^-	0·35	0·41	0·37	0·26	0·29	0·44	0·34	0·34	0·56
CO_2	0·14	0·28	0·15	0·20	0·17	0·77	0·04	0·09	0·12
P_2O_5	0·91	1·01	0·86	0·97	0·97	0·89	0·82	0·93	0·71
Total	99·89	100·12	100·39	99·95	99·80	100·24	100·08	99·85	100·29

The limited Fe^{2+}/Mg^{2+} variation in the Square Top pyroxenes has been attributed to the increasing oxygen fugacity in the successive liquids (Wilkinson, 1966). The high Fe_2O_3/FeO ratios in the analysed Somma-Vesuvius rocks and their pyroxenes indicate a high fo_2 within the magma, which is probably responsible for the restricted Fe^{2+}/Mg^{2+} variation in the clinopyroxenes. High Fe_2O_3/FeO ratios in Vesuvian pyroxenes are also shown by the analyses quoted by Alfani (1934), Muller (1936), and Savelli (1967).

Bulk chemical analyses of some clinopyroxenes of trachytic rocks from Ischia, a volcanic island in the vicinity of Monte Somma and Vesuvius, were made by Khalil (1966), and are plotted in fig. 4 for comparison. The compositions of Ischian pyroxenes are similar to pyroxene sample 38, and are slightly poorer in the wollastonite component than the other Somma-Vesuvian pyroxenes.



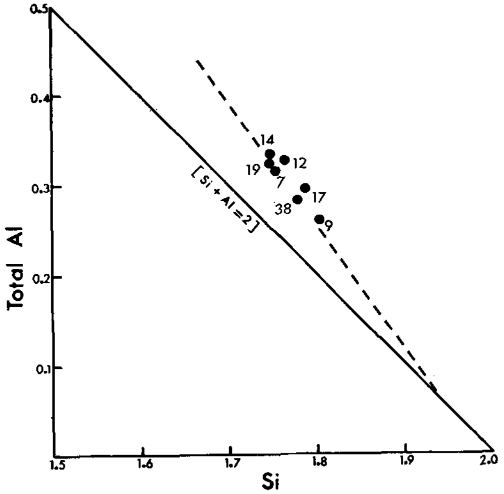


FIG. 2.

FIG. 3.

FIG. 2. Plot of Al_z against wt % TiO_2 of the pyroxenes given in Table II. Broken arrow indicates the trend of TiO_2 variation in the Somma-Vesuvian pyroxenes; solid arrow shows the differentiation trend in alkaline rocks suggested by Le Bas (1962). Dot-and-dash line indicates the range of Al_z and TiO_2 within a single oscillatory-zoned phenocryst (Thompson, 1972).

FIG. 3. Relation between the atomic proportions of Si and Al in the Somma-Vesuvius clinopyroxenes. The dashed line indicates the range of Si and Al within a single oscillatory-zoned phenocryst (Thompson, 1972).

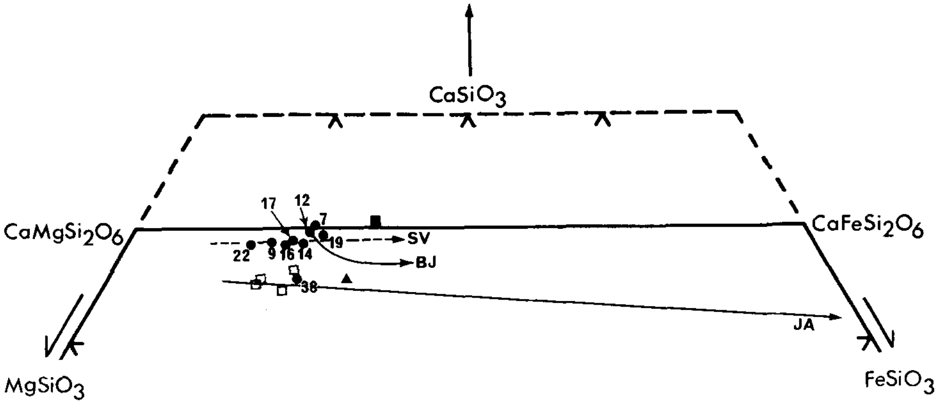


FIG. 4. Composition of some clinopyroxenes, in terms of the components $CaSiO_3$, $MgSiO_3$, and $FeSiO_3$ (mole %). Solid circles represent the composition of Somma-Vesuvius pyroxenes (present analyses); solid square and solid triangle that of Vesuvian pyroxenes obtained by Alfani (1934) and Muller (1936) respectively; open squares represent Ischian pyroxenes (Khalil, 1966). The pyroxene crystallization trend S-V is for Somma-Vesuvius, BJ for the Black Jack teschenite sill (Wilkinson, 1957), and JA for Japanese alkali basalt series (Aoki, 1964).

Discussion. The compositions of the pyroxenes are plotted in fig. 5 (after Le Bas, 1962). From this diagram it can be seen that all the Vesuvian pyroxenes fall inside or close to the field of 'peralkaline'[1] rocks, as defined by Le Bas (1962). The two Somma clinopyroxenes plot well inside the field of alkaline rocks. This may be due to the slightly higher amount of silica in the Somma lavas (Table III; see also Savelli, 1967).

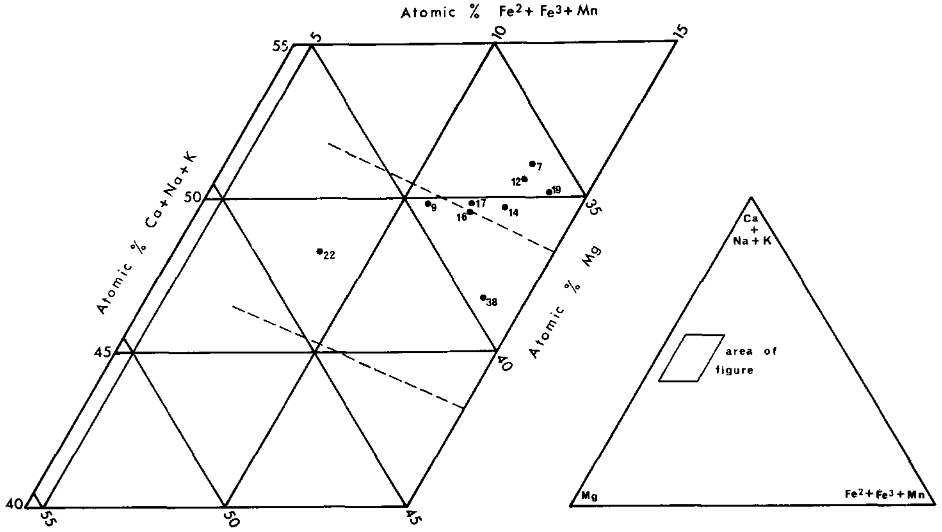


FIG. 5. Plot of clinopyroxenes on part of the triangular diagram (Ca+Na+K):Mg:(Fe^{2+}+Fe^{3+}+Mn). Dashed lines are obtained from Le Bas (1962) which define the field of strongly alkaline rocks (top), normal alkaline rocks (middle), and non-alkaline rocks (bottom).

Kushiro (1960) and Le Bas (1962) have independently published compilations of the compositions of clinopyroxenes from various major rock types in order to estimate the relationship between these and the parent magma types. They have found that clinopyroxenes from tholeiitic, high alumina, and calc-alkaline rocks have high silica and low alumina, those from undersaturated, feldspathoid-bearing alkaline rocks are poor in silica and rich in alumina, and those from normal alkaline (non-feldspathoidal) rocks are intermediate. The importance of paragenesis on the stability of aluminous pyroxenes has earlier been pointed out by Tilley (1938) and is recently emphasized by a number of workers including Brown (1967) and Bence and Papike (1972).

It was mentioned earlier that oscillatory and sector zoning is a most interesting and conspicuous feature in the Somma-Vesuvius pyroxenes. A range of between 1·0 and 9·6 wt % Al_2O_3 has recently been recorded from the zones within a single phenocryst by Thompson (1972). High Al_2O_3 (13·8, 13·1, 7·5, 11·2, 8·1 wt %) in bulk analyses of the Vesuvian pyroxenes has also been shown by Savelli (1967, Table III). Comparatively small figures for Al_2O_3 in the present bulk analyses indicate a considerable range of Al_2O_3 contents in pyroxenes from the various lavas.

Opinions differ on the effect of temperature and pressure in the control of Al in the

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[1] Undersaturated, feldspathoid-bearing alkaline rocks (Le Bas, 1962).

Ca-rich pyroxenes. This problem has been discussed by Brown (1967, pp. 123–4). Experimental work by Boyd and England (1963), by Clark *et al.* (1962), and by Kushiro and Yoder (1964) shows that higher Al in the pyroxenes is favoured by higher pressures. A high alumina (up to 6·6 %) content of augite phenocrysts from apparently tholeiitic basalts (Muir and Tilley, 1964) has been suggested as due to high crystallization pressures. The effect of temperature on Al_2O_3 contents of the pyroxenes is not yet firmly established. Hytönen and Schairer (1961) demonstrate by experiments that diopside crystallizing from mixtures of Diopside–Ca–Tsch. and of Diopside–Enstatite–Anorthite are enriched in Al with falling temperatures, while for the Skaergaard augite series (Brown, 1957; Brown and Vincent, 1963) the reverse holds true.

Any suggestion put forward to explain the complex nature of zoning in the Somma-Vesuvius pyroxenes, as described by Thompson (1972) and illustrated in fig. 1, will obviously be very conjectural. It seems that varying total pressure is unlikely to explain such vast variations in Al. However, the effect of varying P_{H_2O} on the control of Al distribution in the magma cannot be ignored. Zoning in the Somma-Vesuvian pyroxenes involving Al variation may be accounted for by one or more of the following reasons: temperature variation in the magma; magma variation in silica content possibly due to contamination (limestone assimilation); and oscillations in leucite precipitation (as evidenced by the inclusion zones within pyroxene phenocrysts and generations of leucite in the lavas). The writer, however, prefers oscillations in leucite precipitation to the other factors and considers that this factor would have a greater control over Si/Al availability in the magma and thus would vary the available Al for the pyroxene to cause the zoning. It may be added here that crystallization and solution of leucite would be sensitive to P_{H_2O} variations in the magma.

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