

THE MELILITES OF THE EJECTA AND LAVAS FROM THE ALBAN HILLS (ROME, ITALY)

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RIASSUNTO. — Lo studio si prefigge di approfondire le conoscenze sulla variabilità chimica del minerale, anche in riferimento alla giacitura, e di definire le relazioni fra chimismo e proprietà fisiche.

Le meliliti « vulcaniche », che presentano una notevole omogeneità chimica, sono caratterizzate da un più elevato contenuto in Si e Fe e da un inferiore contenuto in Al nei confronti delle meliliti rinvenute nei proietti che mostrano viceversa una ampia variabilità chimica.

Delle meliliti esaminate sono state determinate le costanti ottiche e le dimensioni della cella elementare. L'analisi fattoriale condotta ha permesso di rilevare correlazioni positive fra a_0 , contenuti in Mg, (Na, K), Fe, Si e indici di rifrazione e fra c_0 e contenuti in Al e Ca. Sulla base di tali correlazioni si sono evidenziati due trends.

Delle note « peg structures », riscontrate nelle meliliti « vulcaniche » con maggiore frequenza, sono state definite le composizioni chimiche e ne è discussa l'interpretazione.

ABSTRACT. — Thirty-three samples of melilites from the lava flows and ejecta from Alban Hills volcanic region have been examined with the aim of establishing the variability of the chemical composition also in connection with the various field situations and of investigating the relationship between chemistry and physical properties.

The « volcanic » melilites, showing a narrow range of chemical composition, are characterized by higher Si and Fe contents and by a lower Al content as compared with melilites in ejected blocks, which in contrast show a wider compositional range.

Refractive indices and lattice parameters have been determined.

On the basis of factor analysis positive correlations between a_0 value, Mg, (Na, K), Fe, Si contents and refractive indices and between c_0 value and Al, Ca contents have been found.

Peg structures, more frequent in « volcanic » melilites, have been investigated as for their chemical composition and their origin is discussed.

Introduction

Interest in melilite over a wide range of genetic environments, both terrestrial and extra-terrestrial, has been reported by many workers, and its important role in various

mineralogical and petrographic problems has been highlighted by H.S. YODER (1973).

As is commonly known, the first finding of melilite was made by FLEURIEU DE BELLEVUE (1790) in the leucititic lava of Capo di Bove (Alban Hills) both in the ground-mass and in the cavities and cracks of the same lava. Later DAMOUR and DES CLOIZEAUX (1844), VON RATH (1866), ZAMBONINI (1906), MILLOSEVICH (1921), WASHINGTON (1927), SCHERILLO (1935) and, more recently, TILLEY and HENRY (1953) and TURI (1968) have examined not only the melilite of the above-mentioned locality but also the melilite found in ejecta of various kind, included in the well-known formations of « Peperino di Albano », « Villa Senni tuffs », « Middle Aniene Valley pozzolans » and « Corcolle black pozzolans ».

Although the melilite has been reported as one of the components of many ejecta from the « Welded scoriae » formation of Colle Cimino (Marino) studied by BARBIERI et al. (1970), FEDERICO (1976) and FEDERICO and GIANFAGNA (1980) its composition was not yet known. For this reason we proposed to study the mineral of this locality and, at same time, to re-examine all melilites of the Alban Hills, including the abundant material from the above-mentioned localities existing in the Museum of the Institute of Mineralogy and Petrography of Rome with the aim of establishing the range of variability of chemical composition and its relations with optical properties and lattice parameters.

Occurrence

The material examined here refers to the following occurrences:

— melilites found as crystals lining the

cavities of the Capo di Bove leucitite and those contained in the groundmass of the leucititic lava (Acqua Acetosa flow, km 8.5 of the Via Laurentina);

— melilites in the ejecta embedded in the following formations:

- a) « Peperino di Albano » (localities: Albano, Ariccia and Parco Chigi);
- b) « Black pozzolans » (locality: Corcolle);
- c) « Welded scoriae » (locality: Colle Cimino, Marino).

Melilite paragenesis is greatly varied; for the sake of brevity, table 1 shows the main mineralogical associations observed in blocks. As may be seen, melilite-kaliophilite and melilite-kalsilite associations are very frequent, while the melilite-kalsilite-kaliophilite association is only encountered in a single case. A detailed study of the Alban Hills kalsilite is in progress.

In the samples studied, the melilite varies in colour from almost colourless to yellowish, honey yellow, orange, brownish-red or brown, luster ranging from glassy to resinous. It is often found in short prismatic crystals with {100} and {001} dominant, both in the cavities of the ejecta (of which it also sometimes represents a fundamental component) and of the leucititic lava.

Samples studied and experimental procedure

In the present study eighteen samples of melilite from ejecta and fifteen of melilite crystals lining the lava cavities were examined.

Chemical analyses of mentioned samples were obtained at the University of Manchester using a Cambridge Geoscan Microanalyser fitted with a Link System quantitative energy dispersive system (E.D.S.) type 290 with ZAF-4/FLS quantitative software. The electron beam voltage was 15 kV with a 3 nA probe current.

Divalent iron was determined using the micro-analytic method reported by RILEY and WILLIAMS (1959) on sample quantities of approximately 5 mg.

Strontium has been determined by means of a Jeol JXA-50A electron microprobe (¹) fitted with a W.D.S. system. Glass G.S.E. of N.S.B. and melilites whose Sr content was known (BARBIERI et al., 1972) have been used as standards (Sr content ranging from 350 to 7400 ppm). Operating conditions: 20 kV, 30 nA.

Peg structure analyses were obtained using a

Jeol JXA-50A microprobe (¹) fitted with a Link system quantitative energy system (E.D.S.) type 860 with ZAF-4/FLS quantitative software. The electron beam voltage was 15 kV with 3 nA probe current.

Table 2 shows the averaged analyses of each examined melilite sample. Ferrous iron values reported in the analyses are those obtained with the above-mentioned procedure.

Elaboration of the analytical average data supplied the number of cations, calculated on the basis of 14 oxygens, shown in table 2 and plotted in diagrams and the percentages of end-members (²) reported in table 4.

Refractive indices have been determined by the Emmons double variation method, using phenylisothiocyanate and α -chloronaphthalene as immersion liquids.

Lattice parameters were obtained using the least-squares refinement program elaborated by DE ANGELIS et al. (1977). The values of spacings obtained from X-ray diffractometer data were corrected using reflexions (111), (220), (311) and (400) of silicon used as an internal standard.

Chemical composition

The data from the chemical analyses of all studied samples will be separately considered for melilites from both leucititic lava and ejected blocks. Details on symbols and occurrence are given in Appendix 1.

Melilites found as crystals lining cavities and in groundmass of leucitites

Two unpublished electron microprobe analyses carried out on melilite found in the groundmass of leucitite from Acqua Acetosa flow were made available to us by R. TRIGILA and reported in table 2. The chemical composition of the Acqua Acetosa and Capo di Bove leucitites is very similar, as shown by FORNASERI et al. (1963).

As for the melilite lining the cavities of the Capo di Bove lava flow DAMOUR and DES CLOIZEAUX (1844) and ZAMBONINI (1906) analyzed both the yellow and brown

(¹) Equipment set up at the Institute of Mineralogy and Petrography of Rome.

(²) In calculating the end-member percentages some calcium excess is frequently met. This « excess » calcium is usually interpreted as due to the presence of some Ca-rich phases such as rankinite, merwinite, larnite, monticellite in solid solution with melilite (YODER, 1973). Strontium has been included with calcium and potassium with sodium in the recalculation.

TABLE 1

Observed mineralogical associations in melilite bearing ejecta from the Alban Hills

FORMATION	"PEPERINO DI ALBANO"									"BLACK POZZOLANS"			"WELDED SCORIAE"					
Locality	Albano		Ariccia		Parco Chigi					Corcolle			Colle Cimino					
Samples	AL-39	AL-140	AR-KS	AR-52	PC-121	PC-126	PC-132	PC-134	PC-416	CO-T	CO-D	CO-K16	CC-A	CC-B	CC-X	CC-K2	CC-L1	CC-L2
Phases	AL-39	AL-140	AR-KS	AR-52	PC-121	PC-126	PC-132	PC-134	PC-416	CO-T	CO-D	CO-K16	CC-A	CC-B	CC-X	CC-K2	CC-L1	CC-L2
Mel	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Amph					X													
Cpx		X	X	X			X						X	X	X	X	X	
Phlog	X	X	X		X	X	X	X	X				X	X	X	X	X	X
Mt	X									X	X	X	X	X	X		X	
Kp			X			X						X		X	X		X	
Ka													X				X	X
Gr		X				X		X	X				X	X				
Lc		X		X									X		X			
Ne				X														
Ap							X						X			X		
Hauy						X	X									X		
Ol																		X
Perov										X	X				X			

varieties, while Washington (1927, an. 3; see also BUDDINGTON, 1922) examined a yellowish-brown melilite. The most distinctive features of the composition of the melilites analyzed by DAMOUR and DES CLOIZEAUX and ZAMBONINI is the unusual content in ferric iron which reaches a value of 11.34 percent, expressed as Fe_2O_3 .

The chemical composition resulting from forty-five microprobe analyses carried out on six samples of yellow and nine of brown melilite crystals from the Capo di Bove leucite, reported in table 2 as averaged analyses, shows great homogeneity in both varieties. In all analyses the iron content never reaches the values indicated by mentioned Authors. In fact, for the brown melilite the Fe_2O_3 contents never exceed 6.3 and FeO contents are below 0.6 percent, excepting one case in which its value reaches 1.9 percent, while for the yellow melilite the highest FeO value was around 5 percent; that of Fe_2O_3 was always extremely low and only in two cases were values of 2.4 and 2.7 percent respectively found. Moreover for all examined melilites the ratio (Na+K)/Al results minor than 1.

In the Na-melilite—gehlenite, Fe-gehlenite—akermanite, Fe-akermanite diagram in

fig. 1 the composition of the examined melilites is shown including also the analyses in the literature⁽³⁾. An inspection of this figure shows that the representing points fall in a small area near the Na-melilite—akermanite join while in the Al—Fe, Mn—Mg diagram (fig. 2) the greater part of dots clusters around a point corresponding to 34, 21, 45 percent respectively.

The analyses also confirm an enrichment of iron at the expense of magnesium at the rim (see fig. 3), as observed by SAHAMA (1967) in melilite phenocrysts of several volcanic rocks and of strontium at the expense of calcium, as reported by FITTON and HUGHENS (1981) for melilite microphenocrysts of a nosean-leucite-nephelinite lava from Etinde, Cameroon.

Lastly, the melilite contained in the leucite from the Acqua Acetosa flow and in the cavities of the Capo di Bove leucite show quite similar features with, in particular, very high values of the number of Si ions, never falling below 3.8 on the basis of 14 oxygen atoms, and sometimes reaching 3.9.

(³) Details on occurrence and references of literature analyses are given in Appendix 2.

TABLE 2

Averaged chemical analyses and structural formulae of melilites from the Alban Hills

	CC-12	CB-G53	CB-G63	CB-G76	CB-G85	CB-G86	CB-G98	CB-B31	CB-B61	CB-B71	CB-B74	CB-B75	CB-B78	CB-B83	CB-B84	CB-B87	LA-T
Xwt	n=3	n=3	n=3	n=3	n=3	n=3	n=3	n=4	n=3	n=3	n=3	n=3	n=3	n=3	n=2	n=3	n=2
SiO ₂	40.77	42.53	42.88	42.68	42.43	42.53	42.32	42.26	42.35	42.17	42.16	42.38	42.34	42.12	42.34	42.31	43.12
TiO ₂	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
Al ₂ O ₃	12.44	6.69	6.16	6.82	6.94	6.31	6.76	6.69	6.70	6.06	7.03	6.18	6.34	6.19	6.41	6.78	5.89
Fe ₂ O ₃	1.74	.48	.05	2.67	.38	1.32	2.38	5.63	4.84	3.53	5.11	5.12	5.35	4.49	6.32	5.00	nd
FeO	.86	5.11	4.17	3.30	4.54	3.77	4.21	.63	.07	1.88	.68	.26	.10	.41	.40	.12	4.90(.)
MnO	+	.09	+	+	+	+	.09	+	+	+	+	+	+	+	+	+	.04
MgO	6.21	6.66	7.84	6.49	7.02	7.27	5.95	6.38	7.42	7.41	6.52	7.44	7.18	7.63	6.33	7.31	8.16
CaO	35.23	34.92	36.37	34.65	35.25	35.51	34.54	34.40	35.55	35.51	34.52	35.36	35.52	36.05	35.08	35.25	35.32
Na ₂ O	3.66	2.70	2.30	2.92	2.80	2.55	2.95	2.86	2.45	2.66	2.95	2.67	2.60	2.57	2.85	2.79	2.69
K ₂ O	.21	.23	.36	.32	.30	.30	.24	.21	.31	.25	.27	.25	.27	.30	.29	.29	.10
SrO	.44	.99	.78	.85	.90	.95	.88	.88	.65	.85	.98	.95	1.17	.93	1.11	.92	nd
Total	101.56	100.44	100.91	100.70	100.56	100.51	100.32	99.94	100.34	100.32	100.22	100.61	100.87	100.69	101.13	100.77	100.22

Cations: calculated on the basis of 14 oxygens.

Si	3.632	3.905	3.904	3.892	3.882	3.893	3.891	3.867	3.846	3.860	3.848	3.854	3.846	3.834	3.847	3.835	3.934
Ti	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Al	1.306	.724	.661	.733	.748	.681	.733	.721	.717	.654	.756	.662	.679	.664	.686	.724	.634
Fe ³⁺	.117	.033	.003	.183	.026	.091	.165	.388	.331	.243	.351	.350	.366	.308	.432	.341	—
Fe ²⁺	.064	.392	.318	.252	.347	.289	.324	.048	.005	.144	.052	.020	.008	.031	.030	.009	.374
Mn	—	.007	—	—	—	—	.007	—	—	—	—	—	—	—	—	—	.003
Mg	.825	.912	1.064	.882	.958	.992	.816	.870	1.005	1.081	.887	1.009	.972	1.035	.857	.988	1.109
Ca	3.363	3.436	3.548	3.386	3.456	3.482	3.402	3.372	3.459	3.483	3.376	3.445	3.457	3.516	3.415	3.424	3.453
Na	.632	.481	.406	.516	.497	.453	.526	.507	.431	.472	.522	.471	.458	.454	.502	.490	.475
K	.024	.027	.042	.037	.035	.035	.028	.025	.036	.029	.031	.029	.031	.035	.034	.034	.010
Sr	.023	.053	.041	.045	.048	.050	.047	.047	.034	.045	.052	.050	.062	.049	.059	.048	—

^ = number of averaged analyses; + = below detection limit; (.) = total iron as FeO; Analyst: A. GIANFAGNA.

In terms of end-members, variations in composition may be represented in the yellow variety by Na-melilite 23-30, gehlenite 5-6, Fe-gehlenite⁽⁴⁾ 0-5, Fe-akermanite 13-21 and akermanite 44-55 percent and in the brown variety by respectively 26-31, 4-7, 7-13, 0-8 and 50-58 percent, as shown in table 4.

Melilites of ejected blocks

Twenty-eight analyses have been performed on nine samples from the « Peperino di Albano » formation, ten analyses on three samples⁽⁵⁾ from « Black pozzolans » and twenty analyses on six samples from « Welded scoriae » formation. Averaged

analytical data are reported in table 2.

As may be seen from the diagram in fig. 1 the variation of the chemical composition is greater in melilite from the ejecta. The representing points are scattered over a large area in which also the melilites from the Capo di Bove leucitite are included. The Al—Fe, Mn—Mg diagram (fig. 2) shows clear distinction between the melilites of the lavas and those of the blocks. Actually, the compositions of the samples CC-A, AR-K8 and AR-52 only overlap with those of the melilites on the lavas while all the remaining melilite samples in the blocks are distributed along the Mg—Al join and generally show high Al contents.

Lastly, it should be noted that the number of Si ions is far less than that calculated for the melilite from the leucitites, corresponding to a mean value of 3.69.

In terms of end-members, variations in composition may be represented by Na-me-

⁽⁴⁾ According to NURSE and MIDGLEY (1953), the composition of the Fe-gehlenite member corresponds to Ca₂Fe²⁺AlSiO₇. Instead, according to various Authors (SAHAMA, 1967; YODER, 1973; SEIFERT, 1974; HUCKENHOLZ and OTT, 1978) it corresponds to Ca₂Fe³⁺SiO₇.

⁽⁵⁾ One of the samples previously studied (TURI, 1968) has been kindly supplied by B. TURI.

TABLE 2

Averaged chemical analyses and structural formulae of melilites from the Alban Hills

	AL-39	AL-140	AR-K8	AR-52	PC-121	PC-126	PC-132	PC-134	PC-416	CO-T	CO-D	CO-K16	CC-A	CC-B	CC-Y	CC-K2	CC-L1
Xwt	n=3	n=4	n=3	n=3	n=3	n=3	n=3	n=3	n=3	n=2	n=4	n=4	n=4	n=3	n=2	n=4	n=4
SiO ₂	36.75	41.80	42.85	41.92	36.94	41.20	42.80	42.96	40.18	36.02	38.14	40.70	43.18	41.44	40.94	42.34	42.06
TiO ₂	+	+	+	+	+	+	.09	.02	+	+	+	+	+	+	+	+	+
Al ₂ O ₃	15.43	9.43	6.29	6.51	15.87	9.58	7.61	8.06	10.02	14.25	11.10	8.88	7.04	8.87	8.30	9.22	7.59
Fe ₂ O ₃	.85	1.59	1.33	6.84	1.04	2.14	1.14	.94	1.99	1.91	2.51	5.28	4.25	2.11	2.77	2.24	2.32
FeO	1.16	.65	2.73	.09	1.00	1.66	1.30	1.22	.79	1.10	.66	.60	1.25	2.50	.29	.71	.27
MnO	+	.13	+	.09	.14	.10	.17	.14	+	.11	+	.07	.19	.30	+	+	+
MgO	6.38	7.96	7.77	6.09	5.28	6.93	8.32	9.02	7.37	6.56	7.54	5.97	6.37	6.38	7.72	7.06	8.36
CaO	37.81	35.70	36.04	34.79	36.79	35.10	36.28	34.91	36.38	39.56	38.74	34.79	32.28	35.75	36.57	35.21	35.80
Na ₂ O	1.92	2.60	2.93	3.13	2.53	2.69	2.18	2.65	2.59	1.09	1.23	3.22	3.77	2.67	2.35	3.36	2.90
K ₂ O	.21	.19	.19	.21	.09	.40	.19	.23	.15	.26	.22	.40	.32	.34	.29	.18	.23
SrO	.50	.56	.69	.53	.50	.83	.77	.72	.76	.30	.40	.59	1.88	.83	.71	1.05	.98
Total	101.01	100.61	100.82	100.20	100.18	100.63	100.85	100.87	100.23	101.16	100.54	100.50	100.53	101.19	100.14	101.37	100.52

Cations: calculated on the basis of 14 oxygens.

Si	3.319	3.756	3.891	3.834	3.355	3.733	3.849	3.846	3.653	3.275	3.476	3.712	3.930	3.758	3.720	3.792	3.806
Ti	--	--	--	--	--	--	.006	.001	--	--	--	--	--	--	--	--	--
Al	1.642	.999	.673	.702	1.699	1.023	.807	.850	1.074	1.527	1.192	.955	.755	.948	.942	.973	.809
Fe ³⁺	.058	.108	.091	.471	.071	.146	.077	.063	.136	.131	.172	.362	.291	.144	.189	.151	.158
Fe ²⁺	.088	.049	.207	.007	.076	.126	.098	.091	.060	.084	.050	.046	.095	.190	.022	.053	.020
Mn	--	.010	--	.007	.011	.008	.013	.011	--	.009	--	.005	.015	.023	--	--	--
Mg	.859	1.066	1.052	.831	.715	.936	1.116	1.204	.999	.889	1.025	.812	.864	.863	1.046	.943	1.128
Ca	3.659	3.437	3.507	3.410	3.580	3.408	3.496	3.349	3.544	3.854	3.783	3.399	3.147	3.473	3.560	3.378	3.471
Na	.336	.453	.516	.555	.446	.473	.380	.460	.457	.192	.217	.569	.665	.469	.361	.583	.509
K	.024	.022	.022	.025	.010	.046	.022	.026	.017	.030	.026	.047	.037	.039	.034	.021	.027
Sr	.026	.029	.036	.028	.026	.044	.040	.037	.040	.016	.021	.031	.099	.044	.037	.055	.051

^ = number of averaged analyses; + = below detection limit; (.) = total iron as FeO. Analyst: A. GIANFAGNA.

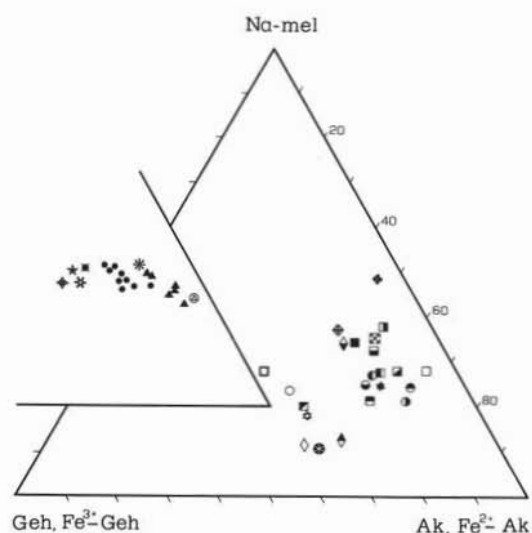


Fig. 1. — Melilite compositions in terms of « end-members ». Symbols as in the Appendixes.

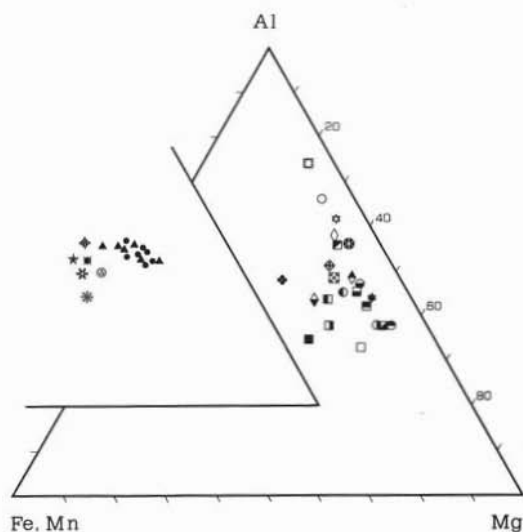


Fig. 2. — Melilite samples plotted in Al—Fe, Mn—Mg diagram. Symbols as in the Appendixes.

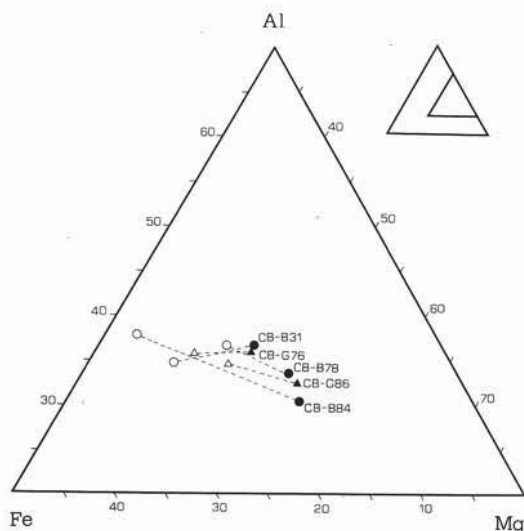


Fig. 3. — Variations of the Al:Fe:Mg ratio from the core (black symbols) to the rim (open symbols) of selected melilite crystals.

lilite 12-38, gehlenite 1-34, Fe-gehlenite 1-14, Fe-akermanite 1-11 and akermanite 37-60 percent, as shown in table 4.

Peg structures

Under the microscope, the well-known peg structures with their usual appearance, so-

TABLE 3
Electron microprobe analyses of some points of a peg compared with microprobe analysis of the host melilite

	M	a	b	c
SiO ₂	41.99	50.26	55.93	61.35
Al ₂ O ₃	7.19	7.82	10.52	11.25
FeO	5.14	6.53	6.21	9.38
MgO	7.96	3.12	1.95	1.40
MnO	0.08	0.32	0.08	0.32
CaO	35.47	12.37	11.00	3.99
Na ₂ O	2.75	0.97	0.96	0.26
K ₂ O	0.22	0.38	0.87	0.73
TiO ₂	0.18	—	0.09	0.06
Cr ₂ O ₃	—	—	0.19	—
TOTAL	100.98	81.77	87.80	88.75

M = brown melilite from Capo di Bove (CB-B61); a, b, c = analyses of three different spots of a peg.



Fig. 4. — Peg structures in lava melilite, brown variety. Length of the bar 0.05 mm.

metimes more or less lens-shaped, in the melilites from both the ejecta and leucite lava flows are displayed, as can be seen in fig. 4.

The microprobe analyses of some points of a lens-shaped peg are shown in table 3 and compared with the analysis of the host melilite. A remarkable progressive lowering of the Ca and Mg values may be seen, countered by an equal progressive enrichment of Si, Al and Fe. In the present investigation on the peg structures, points whose composition could be referred to nepheline or kalsilite are not found.

As for the interpretation of these structures various hypotheses have been formulated: according to NURSE and MIDGLEY (1953) they are due to an exsolution of a second phase, while YODER (1973) suggests that they record volume change resulting from polymorphic inversion. Later EL GORESY and YODER (1974) reported in some pegs the presence of three phases: a Ba-rich kalsilite, nepheline and a SiO₂-rich mineral which represents the major part. They consider kalsilite and nepheline as the product of exsolution of a high temperature akermanite rich in Na-melilite and K-melilite and the SiO₂-rich phase as an alteration product of the melilite. As shown by SAHAMA (1976), the formation of a single peg implies the removal of Ca and Mg, the introduction of

TABLE 4
Chemical compositions expressed in terms of end-members, optical constants and lattice parameters of melilites from the Alban Hills

SAMPLES	Na-mel.	Gehl.	Fe ³⁺ -gehl.	Fe ²⁺ -ak.	Akerm.	ω	ϵ	$\omega - \epsilon$	α_c	C_c
AL-39	18.24	32.42	1.46	4.43	43.45	1.6417	1.6398	0.0019	7.758 ± 0.002	5.035 ± 0.001
AL-140	24.79	13.68	2.81	3.07	55.67	1.6398	1.6352	0.0017	7.771 ± 0.001	5.026 ± 0.001
AR-88	28.16	3.54	2.38	10.85	55.07	1.6394	1.6358	0.0036	7.775 ± 0.005	5.026 ± 0.006
AR-52	33.69	3.55	13.68	0.81	48.27	1.6521	1.6473	0.0048	7.771 ± 0.002	5.036 ± 0.001
PC-121	23.81	32.46	1.86	4.53	37.34	1.6439	1.6417	0.0022	7.733 ± 0.002	5.036 ± 0.001
PC-126	27.12	13.18	3.81	6.97	48.92	1.6399	1.6356	0.0043	7.768 ± 0.001	5.031 ± 0.001
PC-132	21.51	10.83	2.06	5.92	59.68	1.6373	1.6360	0.0013	7.775 ± 0.001	5.025 ± 0.001
PC-134	24.24	9.08	1.58	5.08	60.02	1.6398	1.6372	0.0026	7.765 ± 0.001	5.027 ± 0.001
PC-416	24.94	15.77	3.58	3.16	52.55	1.6385	1.6365	0.0020	7.774 ± 0.002	5.024 ± 0.001
CO-T	11.57	33.95	3.40	4.79	46.28	1.6492	1.6512	0.0020	7.776 ± 0.002	5.033 ± 0.001
CO-D	12.93	25.27	4.58	2.68	54.54	1.6480	1.6470	0.0010	7.765 ± 0.002	5.029 ± 0.001
CO-R16	33.67	9.25	9.91	2.80	44.37	1.6577	1.6538	0.0039	7.759 ± 0.002	5.034 ± 0.001
CC-A	38.00	1.43	7.87	5.94	46.76	1.6410	1.6320	0.0090	7.772 ± 0.002	5.031 ± 0.001
CC-B	27.13	11.71	3.84	11.34	45.99	1.6455	1.6395	0.0060	7.763 ± 0.001	5.028 ± 0.001
CC-X	21.56	14.95	5.17	1.20	57.11	1.6510	1.6470	0.0040	7.770 ± 0.001	5.035 ± 0.001
CC-K2	32.48	9.93	4.06	2.86	50.68	1.6378	1.6369	0.0009	7.757 ± 0.003	5.030 ± 0.002
CC-L1	28.18	7.22	4.16	1.08	59.37	1.6431	1.6416	0.0015	7.773 ± 0.001	5.027 ± 0.001
CC-L2	34.03	16.86	3.02	3.32	42.77	1.6358	1.6305	0.0053	7.746 ± 0.001	5.033 ± 0.001
CB-G53	26.12	5.57	0.85	20.55	46.91	1.6412	1.6363	0.0049	7.788 ± 0.002	5.023 ± 0.001
CB-G63	23.11	5.50	0.09	16.39	54.92	--	--	--	7.789 ± 0.001	5.023 ± 0.001
CB-G76	29.62	4.80	4.90	13.47	47.21	--	--	--	7.789 ± 0.002	5.023 ± 0.001
CB-G85	27.16	5.53	0.67	17.74	48.90	--	--	--	7.791 ± 0.001	5.024 ± 0.001
CB-G86	25.52	5.06	2.38	15.11	51.93	--	--	--	7.788 ± 0.002	5.023 ± 0.001
CB-G98	29.59	4.77	4.40	17.67	43.57	--	--	--	7.791 ± 0.004	5.021 ± 0.002
CB-B31	30.59	5.45	11.15	2.77	50.04	--	--	--	7.771 ± 0.003	5.029 ± 0.003
CB-B61	26.44	7.07	9.36	0.30	56.84	--	--	--	7.765 ± 0.003	5.029 ± 0.004
CB-B71	27.03	4.11	6.56	7.76	54.53	--	--	--	7.781 ± 0.001	5.022 ± 0.002
CB-B74	31.28	5.73	9.92	2.93	50.14	--	--	--	7.775 ± 0.002	5.026 ± 0.001
CB-B75	28.01	4.56	9.82	1.11	56.52	1.6572	1.6515	0.0057	7.775 ± 0.001	5.027 ± 0.001
CB-B78	28.01	5.43	10.47	0.43	55.56	--	--	--	7.776 ± 0.002	5.025 ± 0.001
CB-B83	27.19	4.89	8.56	1.74	57.63	--	--	--	7.781 ± 0.002	5.023 ± 0.001
CB-B84	31.24	4.39	12.60	1.77	50.00	--	--	--	7.778 ± 0.001	5.023 ± 0.001
CB-B87	29.24	5.59	9.52	0.51	55.14	--	--	--	7.776 ± 0.000	5.027 ± 0.000

K and a probably passive enrichment of Si, Al and Fe. This Author concludes that the peg structures may represent the first phase of an alteration process later involving the whole crystal, and referable to a complex metasomatic process during which some points may reach the composition of nepheline and kalsilite or both, without necessarily involving exsolution processes.

In this context the above mentioned progressive enrichment of Si, Al and Fe observed in the melilite pegs from the Alban Hills may well represent the beginning of a large scale alteration process involving the whole crystal, which has already been described as originating the material indicated as *opal pseudomorph after melilite* observed both in ejecta and in leucites of the Alban Hills (FEDERICO and GIANFAGNA, 1980).

Relationship between chemical composition, refractive indices and lattice parameters

Table 4 shows the refractive indices of all melilite samples coming from the ejecta; for the melilite from Capo di Bove leucite the refractive indices of two samples only were measured, respectively for the yellow and brown varieties, because of their great chemical homogeneity. The optical sign was negative, except for the CO-T sample from Corcolle, which has been previously reported to be isotropic (TURI, 1968), and the birefringence ranging between 0.0009 and 0.0090.

A comparison of the refractive indices with the chemical composition as reported

in the same table shows that Fe-gehlenite⁽⁶⁾ appears to have a great influence on the raising of refractive indices, while the influence of the Fe-akermanite is less: this is particularly true for samples CB-G53 and CB-G75, representing respectively yellow and brown melilites from Capo di Bove leucite, whose Na-melilite⁽⁷⁾ and akermanite contents are comparable. In fact, CB-B75 sample, whose refractive indices are among the highest measured, has mean percentages of Fe-gehlenite, gehlenite and Fe-akermanite of 9.8, 4.6 and 1.1 respectively, while very low values are found in CB-G53 sample, whose percentages correspond to 1.0, 5.6 and 20.6 respectively.

Lattice parameters of examined melilites, reported in table 4, are shown in fig. 5 in which two areas may be identified, one, perfectly defined, occupied by the yellow variety and the other in which nearly all the brown melilites on lavas are contained. This area comprises also a few melilites found in the ejecta. Since analyses indicated that the yellow and brown melilites are very similar, only being different owing to the reversed Fe^{2+}/Fe^{3+} ratio, we should conclude that the higher a_0 values characterizing the yellow melilites are related to the presence of the Fe^{2+} cation and thus to the percentage of Fe-akermanite, whose lattice parameters, according to SEIFERT (1974), are $a_0 = 7.900$ (2), $c_0 = 5.002$ (3) Å.

As far as it is known (KIMATA, 1980), the a_0 value is effectively influenced by the iron radius of the divalent cations, while increase in the c_0 parameter depends significantly on the ion radius of the trivalent cations, both occupying the tetrahedral sites in the structure. In KIMATA's paper,

(⁶) The refractive indices are not known for this member since the values supplied by NURSE and MIDGLEY (1953) refer to a synthesized product whose composition, according to HUCKENHOLZ and OTT (1978), does not correspond to that reported by the above Authors. In the products synthesized by HUCKENHOLZ and OTT the solid solution of Fe-gehlenite in gehlenite reaches a maximum value of 37.5 percent under certain P, T conditions.

(⁷) Na-melilite refractive indices reported by NURSE and MIDGLEY (1953) actually refer to a product interpreted as a mixture of nepheline and pseudowollastonite as reported by YODER (1973). The maximum content of Na-melilite in melilite is about 39 percent by weight (EL GORESY and YODER, 1974).

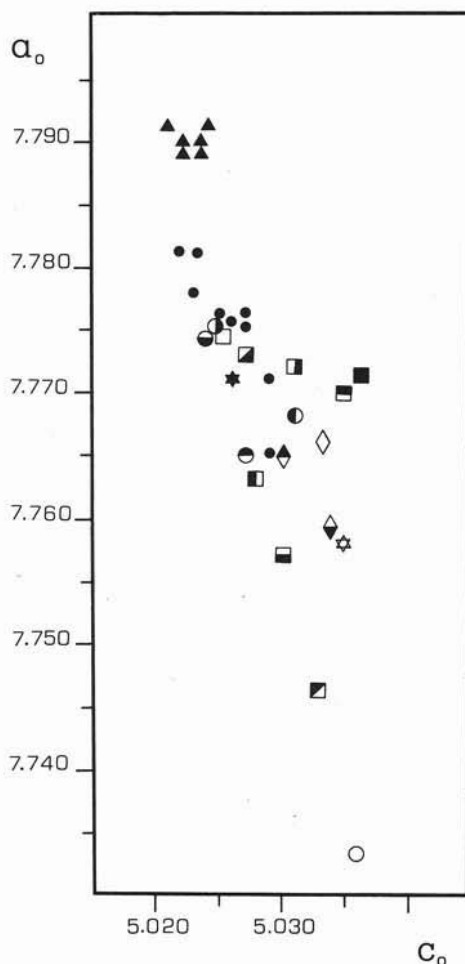


Fig. 5. — Relationship of a_0 versus c_0 of melilite samples. Symbols as in Appendix 1.

however, no iron-containing melilites were examined.

HUCKENHOLZ and OTT (1978) showed that the replacement of Al by Fe^{3+} in gehlenite—Fe-gehlenite solid solutions involves a double increase on a_0 with respect to c_0 . Fig. 5 shows a_0 values referring to the melilites found in the blocks, generally richer in aluminium and with low Fe^{3+} content, confirming the above results also for natural products.

In order to obtain a more complete and quantitative evaluation of the relationship between chemical and physical data, factor analysis according to J. FRANE and M. HILL (1974, 1976) has been applied to the chemical composition, expressed in terms

TABLE 5

Variance explained and cumulative proportion of total variance for first set of selected variables

FACTOR	VARIANCE EXPLAINED	CUMUL. PROPOR. OF TOTAL VARIANCE	SAMPLES
1	4.046	.450	AL-39
2	2.371	.713	AL-140
3	1.351	.863	AR-K8
4	.789	.951	AR-52
5	.258	.980	PC-121
6	.101	.991	PC-126
7	.068	.998	PC-132
8	.015	1.000	PC-134
9	.000	1.000	PC-416

of both (Na, K), Ca, Mg, Fe²⁺, Fe³⁺, Al and Si cations and end-member percentages, lattice parameters and refractive indices of studied melilite samples.

As first set of variables cation numbers and a_0 , c_0 values (9 variables) have been selected, as second set percentages of end-members and refractive indices (ϵ , ω values) (9 variables) have been selected and as third set cation numbers, lattice parameters and refractive indices (11 variables).

TABLE 6

Correlations among variables and factors

	FACTOR	FACTOR	FACTOR
	1	2	3
Al	2	-.863	-.438
a_0	8	.859	.000
c_0	9	-.831	.000
Si	6	.776	.600
Mg	5	.677	-.452
Na, K	1	.000	.956
Ca	7	.000	-.910
Fe ²⁺	4	.287	.000
Fe ³⁺	3	.000	.371
VP	3.361	2.670	1.738

Table 5 shows the variance explained and the cumulative proportion of total variance for the first set. Three « factors » accounting for 86 percent of the information are reported in table 6. On the basis of these

TABLE 7

Matrix of factor loadings for studied samples on the first three factor axes

	FACTOR	FACTOR	FACTOR
	1	2	3
AL-39	-1.983	-1.346	.660
AL-140	.279	-.489	-.134
AR-K8	.679	-.054	.610
AR-52	-.551	1.066	-1.524
PC-121	-3.107	-.195	.727
PC-126	-.544	.294	.270
PC-132	.920	-.972	.004
PC-134	.667	-.419	-.187
PC-416	.123	-.721	.079
CO-T	-1.303	-2.599	.273
CO-D	-.198	-2.515	-.412
CO-K16	-1.211	1.156	-.757
CC-A	-.435	2.247	-.274
CC-B	-.574	.358	.724
CC-X	-.109	-1.015	-.833
CC-K2	-.672	.726	-.154
CC-L1	.645	-.429	-.655
CC-L2	-1.971	1.188	.343
CB-G53	.616	.512	2.184
CB-G63	1.243	-.575	1.556
CB-G76	.557	.829	1.060
CB-G85	.692	.348	1.925
CB-G86	.962	-.016	1.274
CB-G98	.497	1.013	1.675
CB-B31	-.035	.810	-.955
CB-B61	.303	-.161	-1.317
CB-B71	1.007	-.113	-.048
CB-B74	.168	.750	-.696
CB-B75	.652	-.012	-1.204
CB-B78	.678	-.031	-1.189
CB-B83	1.062	-.429	-.913
CB-B84	.448	.635	-.989
CB-B87	.496	.158	-1.125

« factors » positive correlations between a_0 value and (Na, K), Fe²⁺, Si contents and between c_0 value and Al, Ca, Fe³⁺ contents are found. Table 7 gives the matrix of factor loadings for 33 samples on the first three factor axes which have been rotated according to the varimax procedure. Inspection

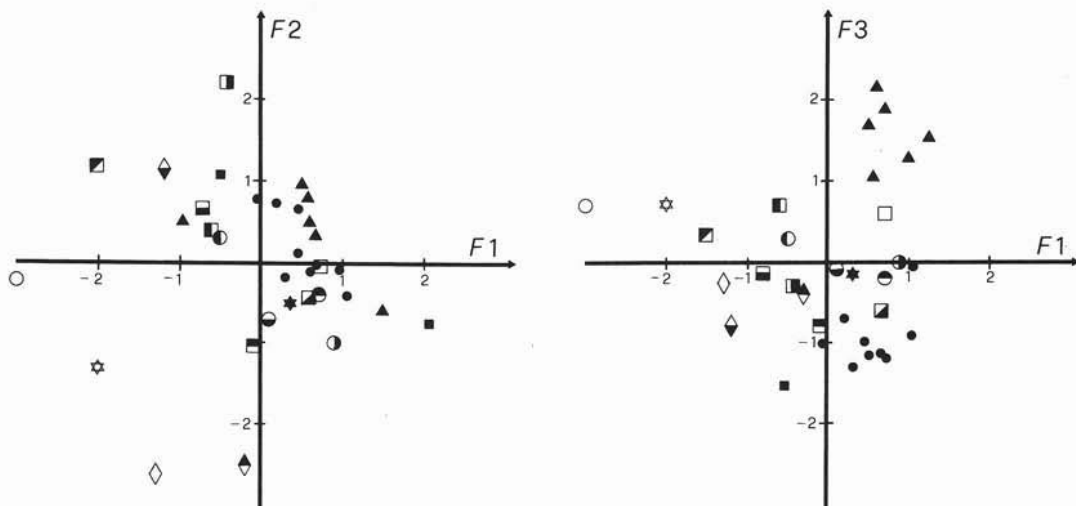


Fig. 6. — Plots of factor loadings on the three varimax factor axes. Symbols as in Appendix 1.

of fig. 6, where the factor loadings two axes at a time are plotted, allows to point out the different behaviour of lava melilites from the most part of ejecta melilites. In factor 1 vs. factor 3 diagram a trend appears from ejecta melilites, characterized by greater Al content and higher c_0 value (least factor 1 value), to lava melilites, characterized by greater Si content and higher a_0 value (highest factor 1 value).

The parallel analysis carried out on the second set, either selecting cation numbers

and refractive indices as variables or end-member percentages and refractive indices as variables, allows a less clear definition of factor loading trends.

From the third set the variance explained and the cumulative proportion of total variance are reported in table 8. The first four « factors » accounting for 94 percent of the information are found in table 9. On the basis of these « factors » positive correlations between a_0 values, Mg, Si, (Na, K), Fe contents and refractive indices as well as c_0

TABLE 8
Variance explained and cumulative proportion of total variance for third set of selected variables

FACTOR	VARIANCE EXPLAINED	CUMUL. PROPOR. OF TOTAL VARIANCE
1	4.234	.385
2	2.847	.644
3	2.228	.846
4	1.015	.939
5	.282	.964
6	.226	.985
7	.081	.992
8	.054	.997
9	.018	.999
10	.015	1.000
11	.000	1.000

TABLE 9
Correlations among variables and factors

		FACTOR 1	FACTOR 2	FACTOR 3	FACTOR 4
a_0	8	.864	.000	.000	.000
Mg	5	.859	.000	-.250	-.359
c_0	9	-.805	.000	.343	.000
Al	2	-.756	-.590	.000	.000
Na,K	1	.000	.970	.000	.000
Ca	7	.000	-.961	.000	.000
Si	6	.639	.745	.000	.000
ω	10	.000	.000	.968	.000
ϵ	11	.000	-.385	.886	.000
Fe ³⁺	3	.000	.445	.820	-.251
Fe ²⁺	4	.000	.000	-.279	.930
VP		3.172	3.155	2.753	1.244

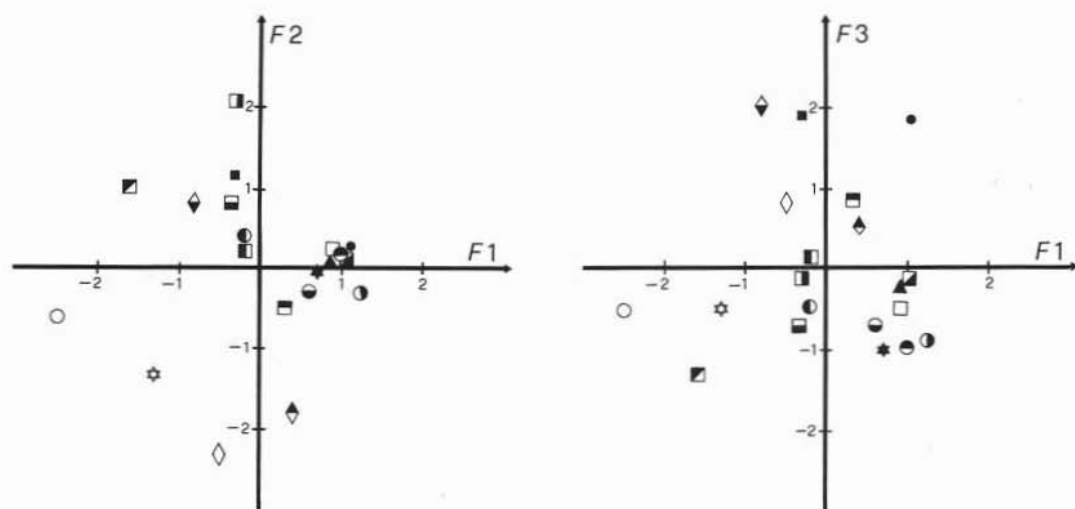


Fig. 7. — Plots of factor loadings on the four varimax factor axes. Symbols as in Appendix 1.

TABLE 10
Matrix of factor loadings for studied samples
on the first four axes

SAMPLES	FACTOR 1	FACTOR 2	FACTOR 3	FACTOR 4
AL-39	-1.294	-1.262	-0.531	.105
AL-140	.666	-.014	-.971	-.780
AR-K8	.933	.192	-.530	.901
AR-52	-.339	1.148	1.905	-.388
PC-121	-2.463	-.555	-.457	.089
PC-126	-.194	.374	-.471	.262
PC-132	1.246	-.328	-.862	-.358
PC-134	.987	.134	-.964	-.948
PC-416	.582	-.349	-.672	-.291
CO-T	-.539	-2.301	.792	.368
CO-D	.371	-1.803	.490	-.450
CO-K16	-.808	.781	2.016	.075
CC-A	-.284	2.055	-.118	.139
CC-B	-.222	.193	.101	1.250
CC-X	.319	-.502	.853	-.813
CC-K2	-.353	.799	-.709	-.631
CC-L1	1.015	.086	-.144	-1.097
CC-L2	-1.588	.982	-1.319	-.430
CB-G53	.923	.123	-.228	3.418
CB-B75	1.043	.248	1.820	-.422

value Al and Ca contents are found. Table 10 gives the matrix of factor loading on the

first four axes which have been rotated according to the varimax procedure.

From factor 1 vs. factor 3 diagram in fig. 7 two trends are found. The first is characterized by a continuous decrease of c_0 value and Al content and the second one by a continuous decrease of refractive indices and Fe^{3+} content. As indicated above yellow and brown varieties of lava melilites have a very similar composition but their different Fe^{2+}/Fe^{3+} can account for the anomalous location of the latter.

Conclusion

A brief treatment of the present knowledge of the Alban Hills ejecta may be a suitable introduction to the conclusion.

From both older (LACROIX, 1893, 1917; WASHINGTON, 1927) and more recent studies (FORNASERI, 1951; FORNASERI et al., 1963; FEDERICO, 1976) the ejecta embedded in the various formations of the Alban Hills may generally be classified in two groups. Some of them, due to their structure and mineralogical composition, are closely related to the leucititic magma itself, of which they represent the intratelluric crystallisation product. Others show characteristics processes within the framework of metamorphism between the leucititic magma and the basement rocks which are composed of units of Mesozoic pelagic sequen-

ces, represented essentially by limestones, overlain by neogenic and post-orogenic complexes, composed mainly of shales and pelitic units, present on the depressed parts of the basement. Although from the study of sedimentary inclusions in tuffs there is no direct proof of the existence of Triassic terms, there are several indirect indications of the existence in the series of such terms referable to evaporitic episodes (FUNICIELLO and PAROTTO, 1978).

In this work the melilites found on the lava as well as those contained in the ejected blocks have been examined. It was possible to establish the limited range of composition in terms of Fe/Mg ratio of the melilites lining the cavities or contained in the groundmass of the leucitite, which may be considered as « volcanic ». This term, put forward by SAHAMA (1967), is used for melilites originating from magmatic consolidation including thereto the late action of the fluid phases and excluding every interaction process between the magma and the wall rocks.

The chemical characteristics distinguishing the « volcanic » melilites are represented by a high number of Si, Fe²⁺ and Fe³⁺ cations up to 3.907, 0.393, 0.471 atoms respectively on the basis of 14 oxygens.

In contrast to the restricted compositional variation of the « volcanic » melilites, a large variation in the chemical composition as a whole of the ejecta melilites has been found whose main feature is represented by a generally high number of Al cations, sometimes reaching 1.699 on 14 oxygens, for the typically metasomatic melilites. As can be seen in the Al—Fe, Mn—Mg diagram in fig. 2 most of the samples plot near the Mg—Al join. Only a few of them fall in the area occupied by the « volcanic » melilites, namely the CC-A, AR-K8 and AR-52 samples. The blocks containing melilite of such a composition may be interpreted as products of intratelluric crystallization either of the leucitic magma itself or of some derived melts due to a limited assimilation of the wall rocks.

At present these results seem to outline a certain correlation between the nature of the blocks and chemical-physical features of the melilite. However for want of a detailed

petrogenetic study of the ejecta melilite cannot be considered a genetic tracer from a petrographic point of view.

The relation between chemical composition in terms of end-members and refractive indices shows the great influence on the raising of the refractive indices of the Fe-gehlenite.

Well-defined areas may be identified in the a_0 vs. c_0 graph, respectively for the yellow « volcanic » melilite — containing large amounts of Fe²⁺ — and the brown variety in which, on the contrary, Fe³⁺ prevails.

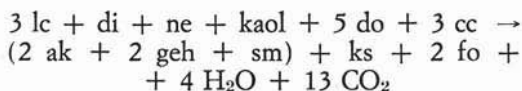
Factor analysis has shown positive correlation between a_0 value, Mg, Si, (Na, K), Fe contents and refractive indices as well as c_0 value and Al, Ca contents.

Peg structures, sometime lens-shaped, are often observed in Alban Hills melilites. Electron microprobe analyses allowed to follow the process which, starting from melilites, by progressive leaching out Ca and Mg, leads to an essentially silica-rich final product, which may be identified, although on a different scale, with the pseudomorphic opal after melilite already reported in a previous work (FEDERICO and GIANFAGNA, 1980). The peg structures would therefore seem to represent the initial stage of a peculiar transformation process involving gradually, in extreme cases, the whole crystal with complete alteration of its composition.

The paragenesis of melilite in the metasomatic ejecta is greatly varied. It may be better understood if we consider the interactions between the leucitic magma — itself having some of the characteristics of a melilitic magma — and the country rocks of the basement. YODER (1973) fully discussed the possibility of the reaction between diopside and calcite to form akermanite. The formation of gehlenitic molecules implies the presence of the necessary amount of aluminium, which could be made available by the clay minerals of the highest terms of the sedimentary series as described above. Desilicization of the leucitic magma fully justifies the formation of kalsilitic molecules and the observed associations of melilite and kalsilite and/or kaliophilite. In the most significant of these associations (e.g.

CC-L1 and CC-L2 samples), it is worth noting that the melilite in equilibrium with the kalsilite is rich in the Na-melilite member.

The formation of melilite in the metamorphic blocks may be represented, for an hypothetical transitional situation between pelitic terms and carbonaceous facies, by a chemical reaction somehow taking into account the composition observed for the melilites. Such a reaction may be expressed by:



where the most common minerals observed in association with melilite may easily be recognized.

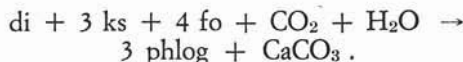
The occurrence of abundant phlogopite in

APPENDIX 1

Symbols, locality and occurrence of the samples examined in this work:

SAMPLE SYMBOLS	LOCALITY	OCCURRENCE
☆ AL-39	Albano	"Peperino di Albano" ejecta
★ AL-140	"	" " "
□ AR-X8	Ariccia	" " "
■ AR-52	"	" " "
○ PC-121	Parco Chigi	" " "
⊙ PC-126	"	" " "
⊙ PC-132	"	" " "
⊙ PC-134	"	" " "
⊙ PC-416	"	" " "
◇ CO-T	Corcolle	"Black pozzolans"
◇ CO-D	"	" " "
◇ CO-K16	"	" " "
■ CC-A	Colle Cimino	"Welded scoriae"
■ CC-B	"	" " "
■ CC-X	"	" " "
■ CC-K2	"	" " "
■ CC-L1	"	" " "
■ CC-L2	"	" " "
▲ CB-G: yellow variety	Capo di Bove	Melilite-leucitite
● CB-B: brown	"	" " "
⊙ LA-T	Acqua Acetosa Via Laurentina Km 8.5	Melilite-nepheline-leucitite

almost all the samples (excluding the ejecta from the «Corcolle black pozzolans» in which it has not yet been observed) may be ascribed to the combined action, in different proportions, of water and CO₂ on the diopside-kalsilite-forsterite association, as shown by YODER (1973) according to the equation:



This retrograde reaction, which implies transformation of the products formed through the previously described process, in presence of excess of diopside, may have taken place during a late phase of the metamorphic process.

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APPENDIX 2

Symbols, occurrence and references of literature analyses:

SAMPLE SYMBOLS	OCCURRENCE	REFERENCES
☆ yellow variety	Capo di Bove leucitite	Damour & Des Cloizeaux (1843)
◆ brown	" " "	" " "
★ yellow	" " "	Zambonini (1906)
★ brown	" " "	" " "
★ yellowish-brown variety	"Peperino di Albano" ejecta	Millosevich (1921)
◆	"Villa Senni Tuffa"	Washington (1927)
◆	"Middle Aniene Valley pozzolans"	Scherillo (1935)
□	"Peperino di Albano"	Tilley and Harry (1953)
⊙	"Black pozzolans"	Turi (1968)

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