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THE ORIGIN OF THE MILOS BENTONITE DEPOSITS

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ABSTRACT

The protoliths of the Milos bentonites are pillow lavas, pillow breccias and hyaloclastites which were produced during the Upper Pliocene by repeated voluminous extrusions of andesitic to quartzlatitic lava from subaqueous fissures within a E-W tending graben system in the northern part of the island.

Middle to Upper Pleistocene seems to be the reasonable time for the hydrothermal activity of the Trogalas-Aggeria-Voudia (TAV)-system. The most important factor controlling the formation of bentonite is the andesitic to latitic composition of the protolith, especially the contents of Si, Al and Mg. Substantial transport of Mg and Al by the hydrothermal fluids during bentonitization was not required. Leaching at temperatures between 160 and 230°C of Si, Ca, Na, K, Ba, Sr, Rb and LREE from the glass-rich pillow lavas, breccias and hyaloclastites were necessary, in order to achieve ideal montmorillonite compositions. Therefore, the highly differentiated, Al- and Mg-poor plagioliparites cannot be regarded as the original host rocks for bentonitization. A silica-rich glass with pumice fabrics, however, appears to be the most probable starting material for kaolinitization.

1. INTRODUCTION

Greece is the largest producer of bentonite in Europe with a production exceeding 1 Million tpa. These bentonites are mainly exploited on the island of Milos in the Aegean Sea.

Up to now, the common genetic interpretation of the bentonite deposits has been that the bentonites were formed through hydrothermal alteration of the glassy matrix of either plagiorhyolitic and dacitic composition (FRANZINI et al., 1964; CAILLERE & ECONOMOU, 1974; FYTIKAS & MARINELLI, 1976; LIAKOPOULOS, 1991; CHRISTIDIS & MARKOPOULOS, 1992) associated more andesitic tuffites (the "Brockentuffe" of WETZENSTEIN, 1972 and 1975) in a submarine environment .

The majority of the bentonite deposits occur within depressions filled with tuffitic material around upper Pliocene dacitic to rhyodacitic plugs and flows in the northeastern and eastern part of Milos island (Fig.1). These volcanoclastic deposits

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are also of uppermost Pliocene age (2 to 1 ma) since they are covered by dacitic and rhyolitic tephra derived from the Pleistocene volcanoes Trachilas and Phyriplaka in the western, and in the central parts of the island respectively (FYTIKAS et al. 1976, 1985, 1986; ANGELIER et al. 1977).

On Milos island, the formation of bentonite seems to be controlled by: the volcanologic environment, the nature and bulk chemistry of the volcanoclastic host-rocks, and by influx of hydrothermal fluids. The precise volcanic origin and chemistry of the protolith, as well as the composition of the hydrothermal fluids, the rock-fluid interaction, and the conditions of bentonite deposition remained unresolved.

A field petrographic and geochemical reinvestigation of these tuffites or "Brockentuffe" yielded the following observations and data which permit a more precise interpretation on the origin of the bentonite deposits.

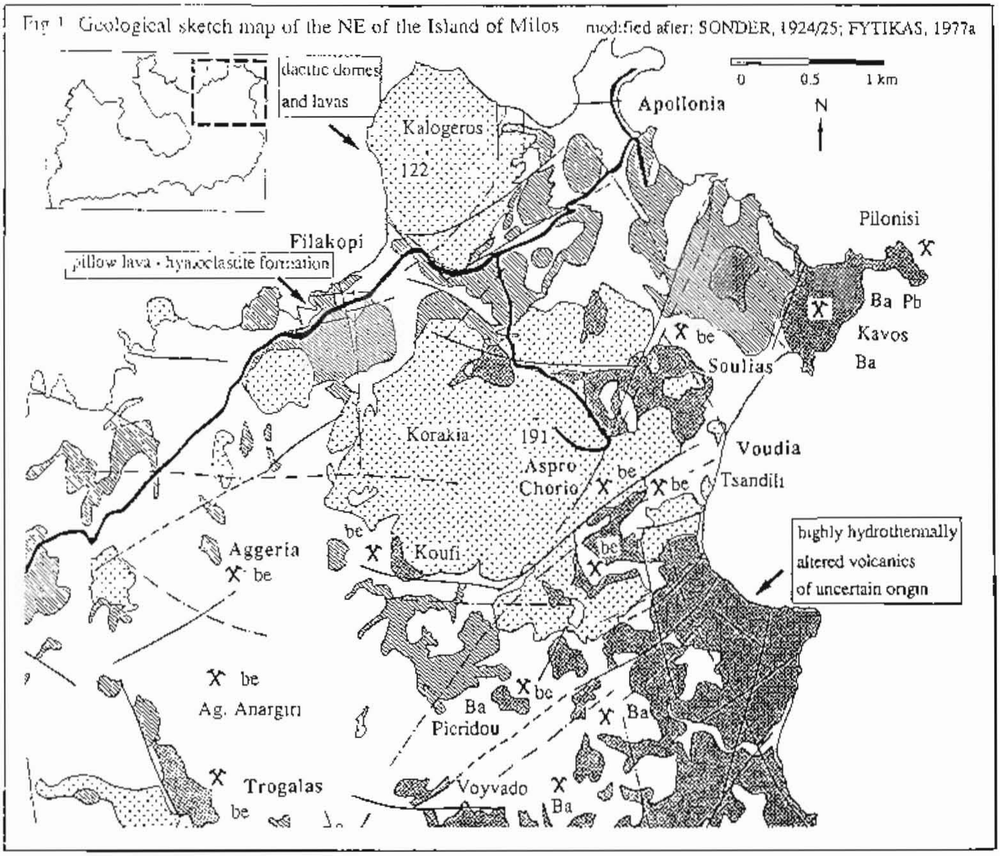

Fig.1 Geological sketch map of the northeastern part of the Island of Milos (modified after SONDER, 1924/25; FYTIKAS, 1977a)

2. THE BENTONITE PROTOLITH

2.1 Field petrographic observations

All of the tuffites are pillow lavas and hyaloclastites which were formed on a shallow submarine shelf. Hyaloclastic flows were produced by repeated voluminous extrusions of andesitic to quartzlatitic lava from subaqueous fissures (E-W and N-S) within a E-W tending graben system in the northern part of the island (Fig.1). This system is bordered by the Kalogeros volcano in the north and Chondro Vouno volcano in the south.

The narrow inlets and shores of **Filakopi** along the main route to Apollonia are a **type locality** for the pillow-hyaloclastite formation (Fig.1). There, unaltered andesitic pillow lava appears at the base of the hyaloclastites approximately at sea level and below (Fig.2 and 3). These andesitic lavas are overlain by reworked pillow lava and hyaloclastic flows with a poorly sorted hyaloclastic matrix grading upward into more fine-grained, well sorted and bedded hyaloclastites (Fig.4).

The irregular distribution of pumice in all sizes from ashes and lapilli to bombs, as well as the intercalation of marls is a common feature (Fig.2-4). The outcrops of this locality also show a rapid change in lithofacies due to repeated and rapid infill and to currents within the irregularly shaped topographic depressions.

Towards the northern part of the island, the rhyolitic tuffs on top of the hyaloclastites disappear and are replaced by Upper-Pliocene and Pleistocene marls. These relations can be studied along the main road from Filakopi to Apollonia. South of Apollonia however, a very limited alteration has effected the pillow lava and hyaloclastites. In contrast, further east bentonitization increases. In **Voudia**, **Soulias** and **Apro Chorio**, the entire hyaloclastite formation reaches a maximum thickness of 100 m, overlying brecciated metamorphic crystalline basement.

The following lithologic succession can be observed in the **mine of Aggeria** (Fig.1) from top to bottom: 2-5 m local Quaternary volcanic debris; 1-2 m reddish oxidation zone, rich in rhyolitic tuffs; 1-3 m milky bentonite, partly brecciated (originally reworked and stratified fine-grained hyaloclastite with intercalations of pillow breccia); 10-20 m yellowish and light grey bentonite (pillows and pillow breccia, structurally entirely preserved, see Fig.5) with intercalations of pumice and tuffitic horizons; 10-20 m greenish brecciated bentonite (rather reworked pillow breccia and hyaloclastite, rich in sulfides) in the lower levels of the 1991 pit. A completely bentonized feeder dike, progressively grading into pillow lava, has been recognized in the south walls of the pit. This demonstrates that the extrusion of these andesitic to latitic lavas took place at very high level into a very shallow marine environment. The maximum depth of the bentonites is not clear. In this area, the metamorphic basement contour-lines show a variation between 200 and 300 m below the actual surface (TSOKAS, 1985).

Field observations in the open **pit of Aggeria** clearly show, that certain volcanoclastics, e.g. blocks, boulders, and bombs of dacitic to rhyolitic composition and with hemi- to holocrystalline textures, are not affected by low-temperature alteration processes or hydrothermal influx. This material remains as hard waste within the soft bentonites.

Kaolinite-rich horizons and small lenses also occur in the bentonites. In detail, the original volcanic structures such as tephra layers (mainly rhyolitic and rhyodacitic in composition) can be recognized. Light grey and yellowish intercalated tuffites and marls are visible between the bentonites, often containig a high content of detrital shallow water fossils, such as *Chlamys sp.*, *Pectes sp.*, *Ostrea sp.* etc.

A similar situation occurs in the bentonite **pit of Trogalas** (Fig.1). According to WETZENSTEIN (1972), approx. 40-50 m of "Brockentuffe" (bentonized pillow breccia and hyaloclasitite) are covered by rhyolitic tuffs, which contain up to 50 vol.% alunite.

2.2 Bulk Chemistry of the bentonite protolith

The general chemical characteristics of the pillow lavas and hyaloclastites are andesitic to latitic with close similarities to low Ti-shoshonites. In the TAS-classification scheme (**T**otal **A**lkaline versus **S**ilica, LE BAS et al., 1986), the rocks lie along the high-potassic calcalkaline trend (FYTIKAS et al. 1976 and 1986). This observation is enforced by the relatively low contents of Ti, Nb and Y. The range of composition is given in Table 1. For comparison, the table also contains the bulk chemical composition of the altered pillow lava from Apollonia. These lavas seem to have an almost identical composition to the unaltered lavas of the Filakopi type-locality. Thus, low temperature seawater and surface alteration did not significantly effect the original bulk composition, although most of the glass has been altered to smectites. The completely bentonized pillow lavas from the mine of Aggeria show major differences in bulk chemical composition (Table 1) compared to the unaltered and altered pillow lavas.

In this respect, three groups of elements could be distinguished: the more-hygromagmatophile elements, which are strongly partitioned into a fluid with distribution coefficients $D \leq 0.01$ (Si, Ca, Na, K, Ba, Sr, Rb and LREE); the less-hygromagmatophile elements with $D \leq 0.1$ (Ti, Al, Mg, P, Zr, Nb, Y), and the transitional elements, such as Fe, Mn, V, Cr, Ni, Co, Cu, Zn and Sc.

Alteration effects on glass-rich basalts have been studied in great detail, and grouping between less- and more-stable elements during alteration processes has been discussed extensively by PEARCE and CANN (1973), HUMPHRIS and THOMPSON (1978), FLOYD and TARNEY (1979), WOOD et al. (1979), STAUDIGEL et al. (1981), etc. A drastic decrease of the more-hygromagmatophile elements, especially Ca, Na, and K, due to leaching by hydrothermal fluids during bentonitization of the Angeria pillow lavas and hyaloclastites is evident. However, Al and Mg, as well as the less-hygromagmatophile elements, in particular Ti, P, Zr, and V seem to have been quite immobile and record the original bulk composition of the lavas. Minor changes are visible in the compositional pattern of the transitional elements. Differences are obvious for Fe, Mn, Cu and Sc.

The most important factor controlling the formation of bentonite is the andesitic to latitic composition of the protolith, especially the contents of Si, Al and Mg. Substantial transport of Mg and Al by the hydrothermal fluids during bentonitization was not required. Leaching of Si, Ca, Na, and K from the glass-rich pillow lavas, breccias and hyaloclastites was necessary, in order to achieve ideal montmorillonite compositions.

	unaltered pillow lava		altered pillows	100% bentonized pillows	hyaloclastites		
Locality:	(Filakopi)		(Apollonia)	(Aggeria)	(Aggeria)		
No.	91-80	91-1	91-2	91-35	91-36	MB2	91-3
Major elements in (wt.%)							
SiO2	57.55	57.02	61.63	54.89	55.95	47.12	51.73
TiO2	.73	.78	.73	.82	.49	.66	.83
Al2O3	16.64	17.53	18.19	19.64	19.29	15.18	18.57
Fe2O3tot	5.54	5.65	4.62	3.98	2.50	3.97	3.34
MnO	.08	.10	.05	.06	.04	.13	.01
MgO	2.39	2.53	1.26	3.72	3.69	2.74	3.17
CaO	6.12	6.23	5.09	2.16	1.56	11.86	3.42
Na2O	3.66	4.17	3.03	.66	.35	.48	1.06
K2O	1.94	2.15	2.23	.15	.53	.33	.37
P2O5	.16	.18	.16	.27	.12	.12	.07
ign.loss	3.54	2.80	2.38	9.47	12.16	15.26	13.27
Total	98.35	99.14	99.37	95.76	96.19	97.85	95.84
Trace elements (ppm)							
Ba	632.	666.	839.	106.	486.	1191.	18.
Sr	610	611	574.	106.	70.	89.	97.
Rb	53.	54.	79.	3.	4.	28.	14.
Nb	3.	4.	3.	6.	3.	6.	4.
Zr	133.	138.	185.	209.	119.	152.	165.
Y	3.	8.	15.	12.	6.	5.	6.
Zn	49.	52.	116.	42.	71.	51.	51.
Cu	3.	5.	4.	8.	5.	10.	10.
Cr	14	15	18	8	18	12	9.
Ni	12.	12	8.	6.	20.	20.	3.
Co	5.	4	12.	5.	7.	10.	8.
V	163.	160	155.	183.	91.	220.	245.
Sc	17.	17.	16.	12.	9.	8.	15.
S	1932.	768.	278.	2560.	3341.	2840.	1485.

Table 1
Bulk chemical composition of selected samples from pillow lavas and hyaloclastites from Filakopi, Apollonia and Aggeria (Milos island), determined by X-ray fluorescence (methods described in NISBET et al., 1979 and DIETRICH et al., 1984). In a few cases, the pillow lavas and hyaloclastites contain remarkably high concentrations of Ba, up to 4000 ppm ,and Sulfur, up to 3000 ppm.

3. MINERALOGY AND CHEMISTRY OF THE BENTONITES

In general, the Milos bentonites represent mixtures of primarily Ca- and Ca-Na-montmorillonites WETZENSTEIN (1972) with some magnesium as exchangeable cation and minor amounts of calcite, quartz, cristobalite, alkali-feldspar, sanidine, plagioclase, kaolinite, illite, baryte, gypsum, alunite, jarosite, anatase and sulfides.

The chemistry of montmorillonite clays are best characterised by the Ca/Na exchange (GUEVEN, 1988). Normally, Ca-Montmorillonites have high Ca:Na ratios of (10-20):1. According to bulk bentonite analyses, the montmorillonites from Milos have much higher concentrations of Na_2O compared to CaO and therefore Ca:Na of approx. (2-3):1) WETZENSTEIN (1972) even designated the montmorillonites from the Kavos mine as Na-Montmorillonite. The large Si, Ca, Na, and K variation in the bulk chemical composition of the Milos bentonites show that such a designation is unrealistic. According to X-ray powder diffraction analyses, this variation can be attributed to the variations in the contents of quartz, cristobalite, calcite, alunite, kaolinite, sanidine, plagioclase, and rhyolitic glass. Especially the unaltered glass (SHELFORD, 1978: with a composition of SiO_2 = 75 to 80 wt.%, Al_2O_2 ca.12.9 wt.%, Na_2O ca.3.7 wt.%, K_2O ca.3.3 wt.%), contributes largely to the high variation in SiO_2 and the alkalis in the bentonites. This has already been indicated by SHARMA (1970).

In many cases, the plagioclase macro- and microphenocrysts are preserved and are andesine and oligoclase in composition. Only the glassy matrix of the andesitic to latitic pillow lavas and breccias, as well as all the reworked hyaloclastic material (Table 1: SiO_2 = 55 to 60 wt.%, Al_2O_3 = 16-20 wt.%, MgO = 2-3 wt.%, CaO = 2-4 wt.%, Na_2O = 0.5 to 1 wt.%.) has been bentonized. The greenish bentonites from the lower part of hyaloclastite formations, the brecciated pillow lavas and hyaloclastites contain relatively high abundances of baryte, pyrite, marcasite, sphalerite, and covellite.

Two different sulfates have been found in the bentonites: alunite: $KAl_3[(OH)_6/(SO_4)_2]$ and Jarosite $KFe_3[(OH)_6/(SO_4)_2]$. SHARMA (1970) showed that the Na content of the sulfates rather low with ratios of $K_2O:Na_2O$=6:1.

4. ORIGIN OF THE TAV-BENTONITES

4.1 Submarine weathering of hyaloclstites (Halmyrolysis)

Halmyrolytic processes have transformed major proportions of the basaltic glass into smectites (for review see SCOTT & HAJASH, 1976. However, this alteration process, at temperatures below 20°C, is time dependent and at least several thousand years are necessary to produce smectite from fresh basaltic glass out of a pillow rim. This process, well known from the basaltic oceanic crust and volcanic seamounts (eg. ROBINSON et al.,1977; HATHAWAY, 1979), does not seem to have influenced the andesitic to latitic pillow and hyaloclastite formation in Milos (see Table 1) for the following reasons:

- The glasses are too rich in silica.
- Rapid coverage of the hyaloclastic formation took place by deposition of shallow-water marls.
- Rapid uplift and coverage with rhyolitic airfall ashes.
- The seawater temperatures at the Pliocene/Pleistocene boundary were - too low, as indicated by the appearance of cold water nannoplankton and by the deposits of diatomites.

4.2 Hydrothermal processes in the hyaloclastites

The formation of bentonites was the result of interaction of hydrothermal fluids on the glass-rich pillow lavas, breccias and hyaloclastites. However, this process was dependent on several effects: the type of hydrothermal system; permeability of the host rock; composition and temperature of the hydrothermal fluids; the lithostatic overpressure; hydrostatic pressure, and the time of influx, and total discharge of fluids.

Certainly the permeability and grain composition of the host-rocks are ideal for the penetration of hydrothermal fluids. In addition, the migration of fluids must have been favoured by reactivation of complex, mainly two SW-NE and E-W striking fault systems in the northern part of the island which will be called here the **"Trogalas-Aggeria-Voudia (TAV)"-hydrothermal system**.

Bentonization is limited by fault scarps towards north in the Aggeria area, whereas to the south fault scarps and rapid termination of the hyaloclastic formation along the original depositional channels control the hydrothermal alteration.

The TAV-hydrothermal system, as indicated by the distribution of the bentonite ocurrences and excavated areas, was mainly a liquid-dominated convective system, limited to major fault zones.

Hydrothermal activity, now absent in the northeastern part of Milos, has shifted to the central and southern parts of the island and seems to be limited by a major N-S striking fault system. The spring water, appearing at the lower level in the Aggeria mine is entirely meteoric in composition and its temperatures are consistent with the annual temperature changes.

At present, two hydrothermal systems are active in Milos island (for extensive discussion, see LIAKOPOULOS ,1987). In the eastern part, a smaller closed system exists with temperatures between 100 and 150°C and probably extending only 100-200 m depth as indicated by the negative values of $\partial^{18}O$ and ∂D. The water is mainly meteoric water, slightly mixed with sea water and of low salinity. The limited thickness of only 200-300m of the volcanic edifice overlying metamorphic basement (FYTIKAS, 1977a and b; TSOKAS, 1985), is the major reason for the shallow level of the meteoric water system. In the central and southern part of the island, a major hydrothermal system reaching more than 1000 m depth is presently active. The waters have high salinity with 1.4 to 1.6 mol Cl/l, pH between 3.8 and 5.3, high contents of alkalis and trace elements, but low values of Mg, Al and SO_4. They reach temperatures over 300°C at 1000 m depth. $\partial^{18}O$ values are positive. The hydrothermal fluids are regarded as results of seawater evaporation following a Raleigh-distillation process (LIAKOPOULOS and BAOULEGUE (1987)).

The **composition of the hydrothermal fluids of the TAV-system** during Pleistocene time must have been similar to the present-day major hydrothermal system for several reasons: the regional distribution of bentonite deposits in the whole northeastern part of the island; the existence of at least two larger phreatic craters southwest of the Aggeria/Trogalas area (SONDER, 1924/25; FYTIKAS et al., 1986) and to the chemistry of the present-day hydrothermal waters, which in greater depth of the geothermal wells are highly depleted in Mg and Al. However, the existence of two hydrothermal convection systems, an upper system richer in meteoric water and a lower system enriched in evaporated seawater, has also been envisaged during Pleistocene times.

In the **TAV-system**, alunite is generally very abundant in the cristobalite-quartz and kaolinite-rich tuffaceous cover on top of the bentonites. In the reddish oxidation horizon below, alunite occurs in paragenesis with gypsum, goethite, and hematite. Alunite is also present in the bentonized hyaloclastites and pillow breccias but less abundant and occurrs always in a paragenesis with quartz, calcite and kaolinite.

Taking all of the field-petrographic evidence into consideration ... During the hydrothermal bentonitization in the Milos TAV-system is feasible. Neither laumontite, prehnite, epidote nor wairakite are present in the lowest levels of the Milos mines. The temperatures of formation of Ca- and Ca-Na- montmorillonites together with the alunite paragenesis must therefore have been below 230°C (GIGGENBACH, 1984) The paragenesis of kaolinite, alunite, and cristobalite in the caprocks indicate temperatures lower than 160°C. These two temperature limitations bracket the range of the montmorillonite crystallization between 160 and 230°C. The stability of Ca-montmorillonite, however, is also a function of CO_2 fugacity and temperature (GIGGENBACH, 1984). At log f_{CO2} near to 0, montmorillonite is stable between 180 and 220°C.

Since the bentonite deposits occur in Milos at a very high level, their formation seems to have been independent of the pressures in the TAV-system. However, the HAAS (1971) equation shows that pressure is, in terms of height of a water column, equivalent to the equilibrium vapor pressure at a given temperature. Therefore, the temperature change of the boiling point with depth has a fundamental impact on the fluid-rock interaction, mass transport and mineral deposition. Deep drilling into the New Zealand hydrothermal systems, especially into the liquid-dominated Wairakei system (GRANT, 1981), have shown that the pressures are close to the hydrostatic curves of HAAS.

5.THE LIFETIME OF THE TAV-SYSTEM

The exact regional distribution of lower part of the TAV-system and its exact timing is not yet clear, but the system certainly must have been active during Pleistocene times. The lifetime of the TAV-hydrothermal system, as well as the influx and discharge rates are at present entirely unconstrained. To provide a crude estimate of the lifetime and of the system during the Pleistocene, a few field observations are relevant. In the 1991 exploration pit at the western extension of the Aggeria bentonite mine, the fault scarps were visible in the southern walls. Towards south, bentonization of the hyaloclastite formation terminates sharply at the fault plane. Only small irregular veins, a few centimeters in width and penetrating the unaltered hyaloclastites, are rich in bentonite. This structural pattern suggests that the influx time of hot waters near the surface must have been short, maybe in the order of a few thousand years. Over a long time interval one would expect a more voluminous penetrative alteration of the hyaloclastites beyond the fault scarps.

Middle to Upper Pleistocene seems to be the reasonable time for the hydrothermal activity of the TAV-system. According to radiometric dating, the youngest volcano Phyriplaka erupted rhyolitic lavas and tephra between 140 000 to 90 000 years (FYTIKAS et al., 1986).

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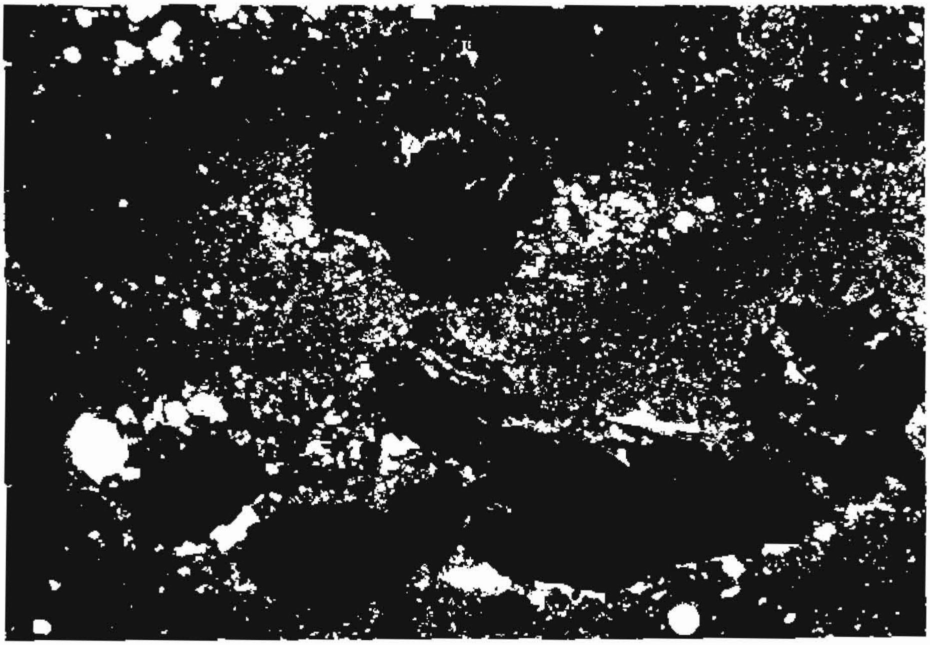

Fig.2. Type locality of Filakopi: loose packed, unaltered, andesitic pillow lava in a hyaloclatic matrix (broken glass-rich pillows) at the base of the hyaloclastite formation approximately at sea level.

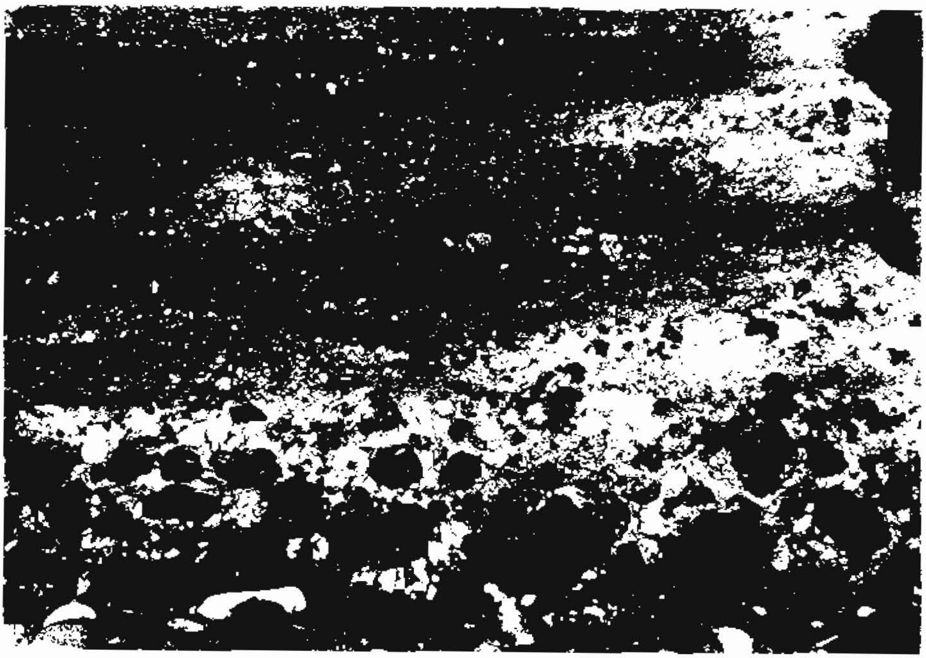

Fig.3. At Filakopi, the pillow lavas lavas are overlain by reworked pillow lava and hyaloclastic flows with a poorly sorted hyaloclastic matrix.

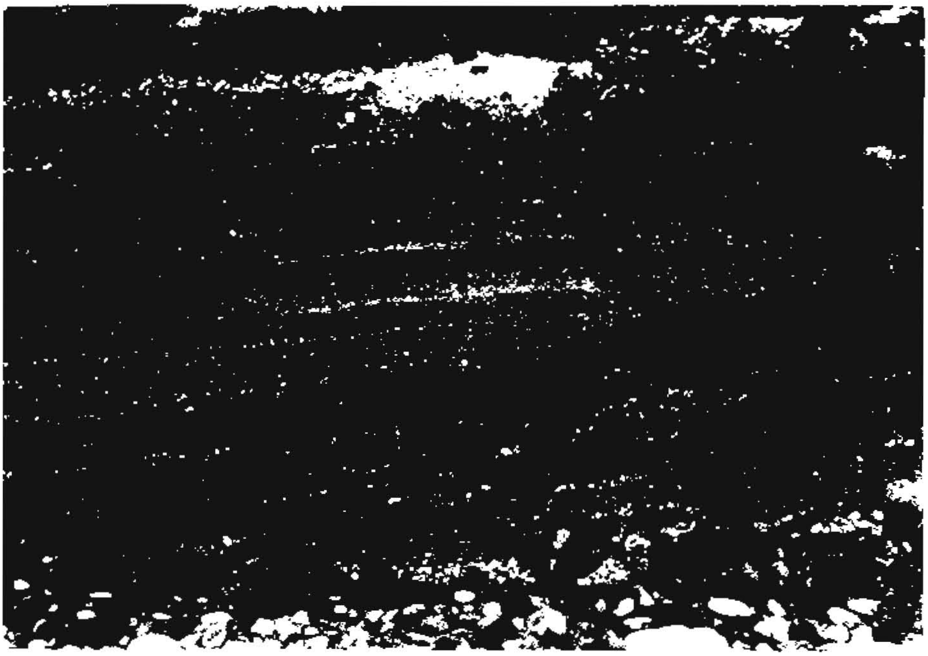

Fig.4. Hyaloclastites upward grading into more fine-grained, well sorted and bedded hyaloclastites Note the irregular distribution of pumice in all sizes from ashes and lapilli to bombs, as well as the intercalation of marls.

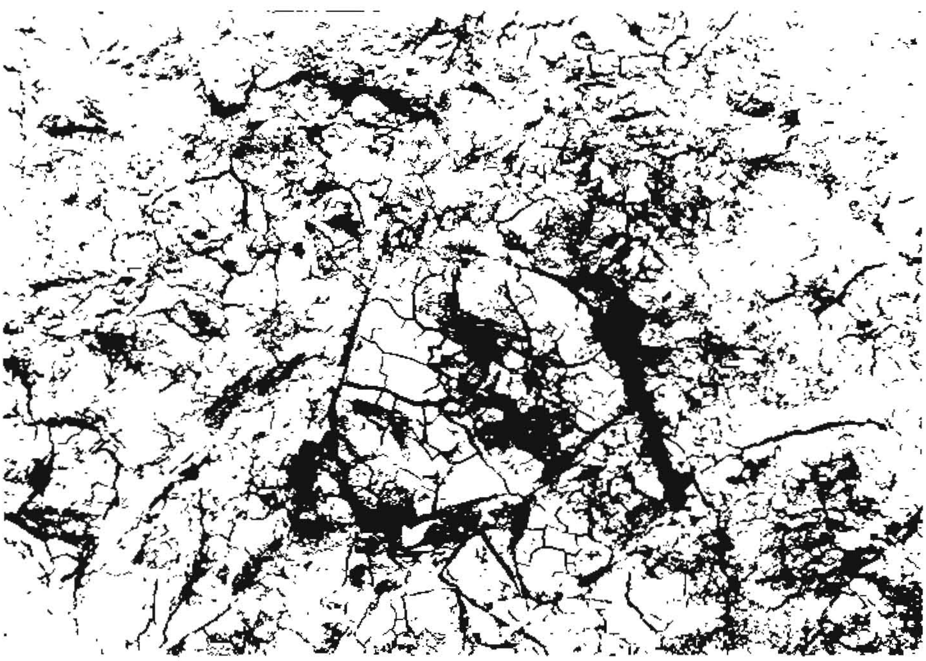

Fig.5. Mine of Aggeria, yellowish and light grey bentonite zone with structurally entirely preserved pillows and pillow breccia.