MINERALOGICAL EVIDENCE FOR LIQUID IMMISCIBILITY IN MAGMATIC ROCKS FROM WESTERN SICILY

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Abstract: Magmatic rocks of Western Sicily display spheroidal coarse-grained aggregates (enclaves) of leucocratic minerals, identical in both composition and zoning to minerals in the surrounding rock. Liquid immiscibility phenomena are indicated by immiscible glass globules in each other and confirmed by exploratory melting experiments. Moreover, detailed mineralogy indicates that the Sicilian enclaves originated as immiscible felsic liquids that separated from a basic magma, and rule out the other possibility that they are formed by mixing of two liquids generated independently one from the other. This study provides a basis for interpreting the cooling history of the two-liquid Sicilian magma. Our results strongly support Lucido's (1990) theory according to which, in time, as the temperature of a critical basic magma decreases, a liquid-liquid phase separation in an acidic sense occurs.

Introduction

Most researchers hold that silicate liquid immiscibility is not a very significant phenomenon in the petrogenetic processes of eruptive rocks. This limited support for liquid immiscibility is due essentially to two reasons. The first reason is connected to some articles brought out by Greig (1927, 1928) and Bowen (1928). In fact, these authors agree in maintaining that there is not any well-grounded reason to suppose unmixing processes during the consolidation of melts having the composition of magmas. Generally, Greig (1927, 1928) and Bowen (1928) tend to exclude that silicate liquid immiscibility can be the cause of the differentiation process. As a result, experimental investigations were not encouraged in this direction, and the process of fractional crystallization became the fundamental theory of magmatic differentiation. The second motive for the poor credit given to liquid immiscibility is the scarcity of evidence found in natural systems during the last half century. The first indisputable proof of the existence of liquid immiscibility in magmas was found in the Apollo 11 samples brought back from the Moon (Roedder & Weiblen 1970a). Subsequently, glasses containing immiscible globules in each other were discovered in various terrestrial rocks (e.g. De 1974; Philpotts 1978, 1979). These clear evidences of liquid immiscibility are supported by several experimental studies (e.g. Ferguson & Currie 1971, 1972; Massion & Koster van Groos 1973; Philpotts 1971, 1976; Dixon & Rutherford 1979). During research carried out on basaltic rocks of the Sicanian Mts. (Western Sicily), the study of enclaves (see Didier & Barbarin 1991) and their relative host-rock appeared of considerable interest because it may provide valuable information on the role of liquid immiscibility in the differentiation of magmas (Lucido 1981). In this paper, detailed mineralogical and petrological data on the Sicilian rocks are presented and their genetic significance is discussed.

Petrographic features pertaining to felsic enclaves and their enclosing mafic rock

The Sicilian igneous outcrop in which the enclaves occur is one of a number of basaltic manifestations which originated from fissure activity in the Sicanian Basin (Lucido et al. 1978). In the Meso-Cenozoic period, this basin represented the southwest termination of the great Pindos seaway which cut through the southern continental margin of the Tethys (Scandone 1975). The outcrop was first reported on by Baldacci (1886) who considered it a basaltic dike. More recently, Broquet (1968) considered the outcrop to be a diabase. In agreement with Broquet (1968), the outcropping petrographic types are essentially alkali diabases containing felsic enclaves. Diabases and their enclaves occur as a dike. The outcrop is poorly exposed, making it extremely difficult to determine the relationships between the magmatic rocks and the country rocks. The alkaline rocks form three small hillocks which crop out upon a probable erosion surface of Paleocene, Eocene and Oligocene sediments. Lacking geochronological data and because of the poor field-relationships the actual age of the igneous rocks is uncertain. A geological sketch map of the Sicilian igneous outcrop is given in Fig. 1.

Tab. 1 shows the major-oxide compositions (wt. %) of the Sicilian rocks. The contrast between the chemical compositions enables us to divide them into two groups. The first group consists of felsic enclaves (analyses 1-6), the second consists of enclosing mafic rocks (analyses 7-12). The Sicilian felsic enclaves are clearly richer in Si, Al, Na, K, and are poorer in calcium, magnesium, iron, manganese, titanium and phosphorus than the mafic rocks. In this manner, the elements normally forming framework structures are concentrated in the felsic portions, whereas the less polymerizing ones are partitioned into the mafic rocks.
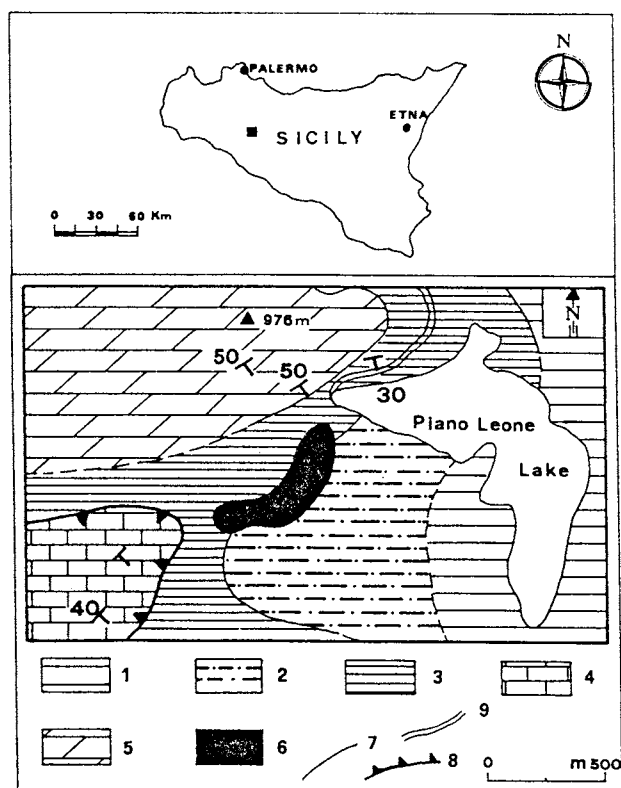
Fig. 1. Geological sketch map. 1 - Grey bluish marls from the Middle to the Upper Miocene; 2 - Sandy marls of the Upper Oligocene; 3 - Calcareous clays and red marls, limestones and white marly limestones (Paleocene-Upper Eocene); 4 - Light siliceous limestones of the Upper Trias; 5 - Dolomitic limestones and dolomites of the Upper Trias; 6 - Alkaline mafic rocks enclosing felsic enclaves; 7 - Geologic boundaries; 8 - Anomalous contacts; 9 - Road (after Broquet 1968). The small square in the inset indicates the location of the outcropping magmatic rocks.

Although enclaves are found throughout the outcrop, their abundance and size increase towards the north-eastern part of the outcrop; most of them are clearly visible because of their much lighter colour. The Sicilian enclaves commonly range from a few centimetres to some decimetres in size; however, several enclaves measure less than 1 cm in diameter and are too small for simple separation. The largest enclave found is nearly 25 cm long and appears flattened. As indicated by their cuspate borders the small enclaves coalesce between themselves to form larger enclaves. The enclosing rock of the fresh samples is black and fine-grained, whereas the rock of the weathered ones is of a greenish colour which is locally light, tending to be greyish. At times, mafic spherules are found at the cuspate margins of the felsic portions (Lucido 1981). Zoning is much developed in minerals common to both host-rock and enclave. Finally, lenses and veinlets a few millimetres wide are sometimes scattered in the host-rock.

Mineralogy and crystal-chemistry

Modal analyses in volume percent of two representative Sicilian rock-specimens are presented in Tab. 2. The minerals occurring in the mafic host-rock are the same as those in the felsic enclave, but they occur in differing proportions, with leucocratic and hydrous phases concentrated in the felsic fraction. Electron microprobe analyses of the minerals were carried out at the University of Modena by an ARL-SEMQ instrumentation operating at 20 Kv and 20 nA. Microprobe analyses have been made of all the phases, except ilmenite and rarer accessory minerals, in over 10 representative rock-specimens. For each mineral species taken into account, phases occurring in both felsic and mafic fractions were analysed. The analyses were corrected for matrix effects using the calculations of Bence & Albee (1968). The results were averaged as necessary and are presented in the appropriate tables. In a single-crystal, for descriptive purposes, we will distinguish a border zone, a middle zone, and a core zone.

Table 1: Chemical analyses of felsic enclaves (1-6) and mafic rocks (7-12) from Western Sicily (Lucido 1981).

| Elements (wt. %)	1 core	2 border	3	4	5	6	7	8	9	10	11	12
SiO_2	51.08	54.42	51.86	52.41	52.00	53.15	43.38	43.49	43.61	43.04	43.88	42.42
Al_2O_3	15.66	17.16	15.68	15.70	16.84	17.19	11.29	11.46	11.31	11.30	11.92	11.10
FeO_{tot}	8.78	6.47	8.47	7.67	7.73	7.03	14.11	13.30	13.90	13.57	13.95	14.51
MgO	4.00	3.17	3.90	4.59	3.10	2.58	8.99	8.65	8.85	8.90	8.68	9.45
CaO	4.60	1.78	4.64	3.20	3.10	2.40	6.35	6.70	6.55	6.18	6.58	7.06
Na_2O	5.14	4.83	5.06	4.43	5.59	6.02	2.78	2.96	2.82	2.84	2.39	2.04
K_2O	3.78	5.87	3.82	4.70	4.29	5.10	2.03	2.05	2.13	2.13	2.05	1.79
TiO_2	2.22	1.23	2.22	1.84	1.82	1.69	3.41	3.17	3.36	3.29	3.39	3.69
P_2O_5	0.29	0.17	0.44	0.24	0.32	0.16	1.58	1.52	1.44	1.50	1.05	0.79
MnO	0.16	0.12	0.16	0.13	0.14	0.12	0.19	0.18	0.18	0.18	0.18	0.18
H_2O^-	0.84	0.71	1.16	1.19	1.20	0.86	2.20	3.00	2.03	1.99	2.18	2.67
H_2O^+	4.20	3.58	3.22	4.01	4.56	4.39	4.30	4.15	4.01	5.06	4.24	4.87
Total	100.75	99.51	100.63	100.11	100.69	100.69	100.61	100.63	100.19	99.98	100.49	100.57
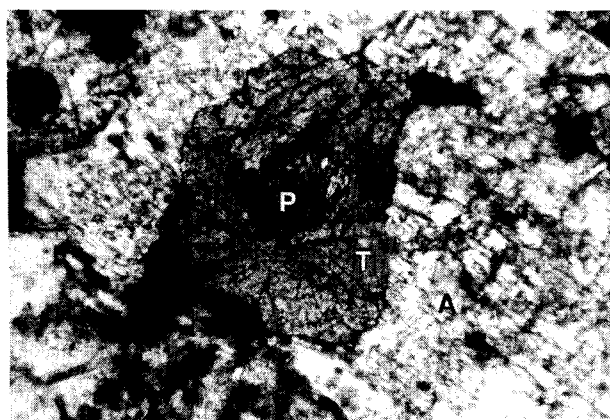
Fig. 2. Felsic enclave from Western Sicily showing andesine crystal (A) enclosing a titanaugite crystal (T), which in turn contains a subhedral pseudomorph (P) after presumed olivine. Width of Fig. 2 is 800 μm (parallel nicols).

Main mineralogical phases

Clinopyroxene is an essential phase of all rock-samples. Both host-rock and enclaves contain some pyroxene phenocrysts whose core zone is constituted by subhedral to euhedral pseudomorphs after olivine (Fig. 2). Unaltered olivine was never observed, but its pseudomorphs were also found as isolated accessory phases. A consistent feature of all the clinopyroxenes examined is the variability in composition. Fig. 3 illustrates the range of composition, expressed as atomic percentages of Ca, Mg and (Fe_{tot} + Mn), of 15 pyroxene minerals. Under the microscope, several pyroxene crystals exhibit both sector and core-to-border compositional zoning. In plane polarized light, the (100) and (010) sectors are pinkish, whereas the (111) sector is distinctly less pink. Similar zoning in pyroxene has been described in detail by numerous researchers (e.g. Bence et al. 1970; Hollister &

Hargreaves 1970; Hollister & Gancarz 1971; Wass 1973; Leung 1974; Dowty 1976). These compositional differences between the different sectors are in some cases evidenced by hourglass zoning (see e.g. Wass 1973). The pleochroism is fairly marked with α = pink, β = dark pink, γ = light pink. The average of the measurements carried out on several crystals with the Fedoroff universal stage gave: c^γ = 45° and $2V_γ$ = 47°. From the optical characteristics it follows that they are *titanaugite* crystals. Titanaugite analyses are presented in Tab. 3. In agreement with the optical characteristics, various analyses carried out on single-crystals show a clear zoning. An important feature of the chemistry of this pyroxene is that the titanium and aluminium values are depleted in the border zone, where they are reduced to nearly half. In fact, the analyses given in Tab. 4 display progressive Si, Fe, Mn, Na enrichment and concomitant Ti and Al depletion with distance from the core zone to the border zone.

Amphibole occurs as phenocrysts up to 5 mm long, but it is more frequently 1-2 mm in length. It is decidedly prevalent in the felsic portions, whereas it is rarer or absent in the mafic ones. A concentric zoning from pleochroic dark brown centres to much paler brown rims is displayed by all examples. Fig. 5 is a schematic representation of the composition range of 21 amphibole minerals in the system $Ca_2Mg_5Si_8O_{22}(OH)_2$-$Ca_2Fe_5Si_8O_{22}(OH)_2$-$Mg_7Si_8O_{22}(OH)_2$-$Fe_7Si_8O_{22}(OH)_2$. In several instances it is clearly evident that the amphibole has grown on a pre-existing titanaugite crystal (Fig. 4), locally having the cleavage planes in continuity (or parallel) with those of the pyroxene. In thin section its colour varies (α = pale brown, β = brown, γ = dark reddish brown); the extinction angle c^γ is found to be 9°. The optic axial angle directly measured by universal stage is $2V_α$ = 77°. From the above optical evidence it may be concluded that the amphibole is *kaersutite*. Kaersutite analyses are shown in Tab. 3. The kaersutite crystals display significant compositional variations; FeO_{tot} increasing and MgO decreasing from core to border (Tab. 4).

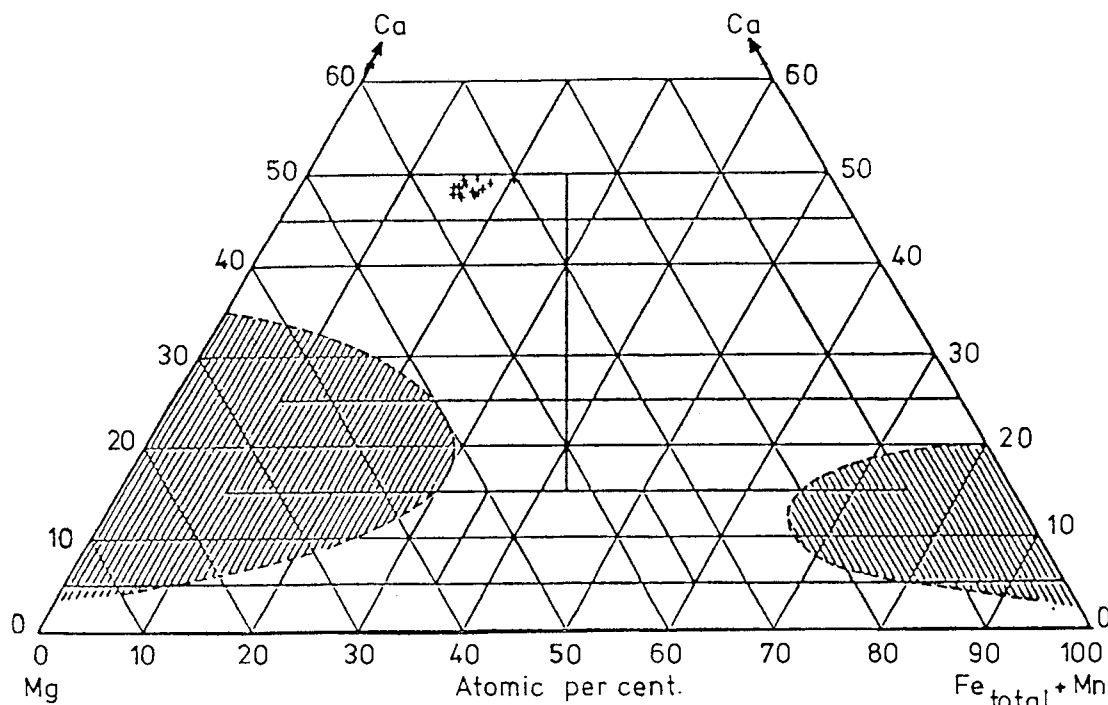*Feldspar* crystals are restricted to the groundmass of both

Fig. 3. The pyroxene quadrilateral showing 15 pyroxene analyses (crosses - the average of these analyses is represented in Tab. 3) plotted as atomic percentages. Shaded areas represent compositions not represented by natural minerals.

Table 2: Modal data (vol. %) of two representative rock-specimens from Sicily.

	Felsic enclave	Mafic host-rock		Felsic enclave	Mafic host-rock
Titanaugite	4.00	12.05	Magnetite	2.46	18.23
Kaersutite	8.50	3.99	Albite	1.83	—
Andesine	16.60	32.42	Anorthoclase	0.92	—
K-Feldspar	4.34	0.97	Ti-aegirinaugite	0.30	0.12
Analcite	5.48	0.62	Baerkevikite	0.20	—
Natrolite	9.06	0.51	Titanite	0.19	0.24
Biotite	1.45	1.18	Apatite	0.08	0.08
Augite	1.51	1.62	Groundmass	38.86	13.91
Ilmenite	3.15	0.13	Alteration products (mainly chlorites and glauconite)	1.07	13.93

Table 3: Main minerals common to Sicilian felsic enclaves and their mafic host-rock. Asterisk indicates minerals which are absent in some mafic rock-samples. N indicates the number of spot analyses.

N	15	21	6	4	5	5
Elements (wt.%)	Titanaugite	Kaersutite *	Andesine	K-feldspar *	Analcite	Natrolite
SiO2	45.64	40.02	56.90	64.06	53.49	48.90
TiO2	3.33	6.25	0.15	0.05	0.05	0.03
Al2O3	6.70	11.50	26.10	19.10	23.89	29.55
Cr2O3	0.00	0.02	0.14	0.00	0.00	0.00
FeOtot	9.67	15.41	0.32	0.55	0.12	0.02
MnO	0.20	0.23	0.00	0.01	0.01	0.00
MgO	11.37	10.27	0.04	0.02	0.00	0.00
CaO	22.17	11.46	7.59	0.25	0.45	0.45
Na2O	0.76	2.93	6.82	5.15	9.66	7.88
K2O	0.00	0.98	0.76	9.84	0.35	0.01
Total	99.84	99.07	98.82	99.03	88.02	86.84

Fig. 4. Felsic enclave from Western Sicily showing kaersutite (K) growing on a previous crystal of titanaugite (T). Width of Fig. 4 is 2.2 mm (parallel nicols).

felsic and mafic rocks. Microscopic analysis shows that the plagioclase often has a concentric zoning. The composition of the vast majority of the plagioclases is *andesine*, with the An-content highest ($\sim An_{50}$) in cores and lowest ($\sim An_{30}$) in grain

margins. The plagioclase crystals normally contain inclusions of pyroxene, amphibole and opaque minerals. Along with other light phases, plagioclases have the tendency to form clusters. Andesine analyses are compiled in Tab. 3. Confirming previous optical characters, the andesine crystals are strongly zoned. In fact, analyses carried out on a single crystal (Tab. 4) evidenced that Si, Na, and K increase, while Ca and Al decrease from the core to the rim. More exactly, the core zone has a composition at the border-line with labradorite (An_{50}) and the rim zone a composition at the boundary with oligoclase (An_{30}). The other major oxides do not show significant variations. Fig. 6 illustrates the variations inside the andesinic compositional range. Although in the enclaves andesine contains proportionally more of the albite component than that in the basic host-rock, it shows the same compositional range (chemical overlap) as plagioclase in the enclosing rock (see e.g. Philpotts & Hodgson 1968; Mc Sween et al. 1979). *K-feldspar* is prevalent in the felsic portions in the form of small laths, and as minute patches of fine-grained to crypto -crystalline material. Its chemical analysis is represented in Tab. 3.

Analcite is present in both the host-rock and enclaves. This mineral occurs as an interstitial phase in the feldspar-dominated lining of enclaves or in segregations, and in the groundmass areas. Under the microscope most crystals are isotropous, but in some cases they have a relatively low birefringence. In some
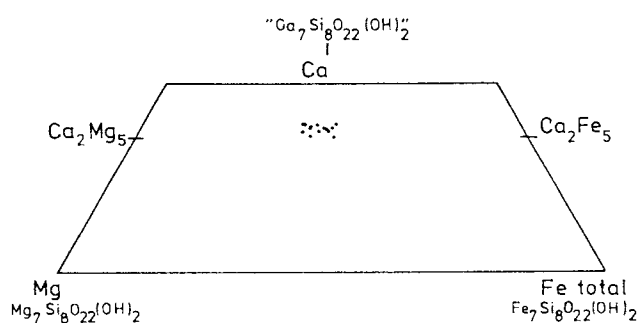
Fig. 5. Distribution of 21 amphibole analyses (solid circles - the average of these analyses is represented in Tab. 3) with respect to Ca, Mg and Fe_{tot} atoms. Note the variation of the Fe_{tot}/Mg ratio due to zoning.

instances analcite crystals fill vugs or pseudomorph feldspar. However, the possibility that some of the analcite represents a primary magmatic precipitate cannot be excluded on the evidence available.

Natrolite is fibrous, but locally exhibits relatively large crystals elongated parallel to the z axis. It is volumetrically major and shows anomalous interference colours. Natrolite is normally associated with analcite. The presence of analcite and natrolite may also suggest that the felsic patches represent vugs in the mafic rocks lately filled up with minerals crystallized from some sodic fluid phases. As characterizing phases, analcite and natrolite are also included in Tab. 3.

Minor and accessory phases

Biotite occurs in small anhedral to euhedral grains in both host-rock and enclaves. However, this phyllosilicate is distinctly more concentrated in the fine-grained felsic portions. Microscopic observations clearly show that, in some cases, biotite grows on the margins of the pre-existing kaersutite crystals. *Augite* may be present as granules in both host-rock and enclaves. Augite crystals of middle-size in the fine-grained portions have the same chemical composition as the rims of the titanaugite grains (see Tabs. 4 and 5). In the basic host-rock the usual opaque phase is *magnetite*. This mineral is commonly
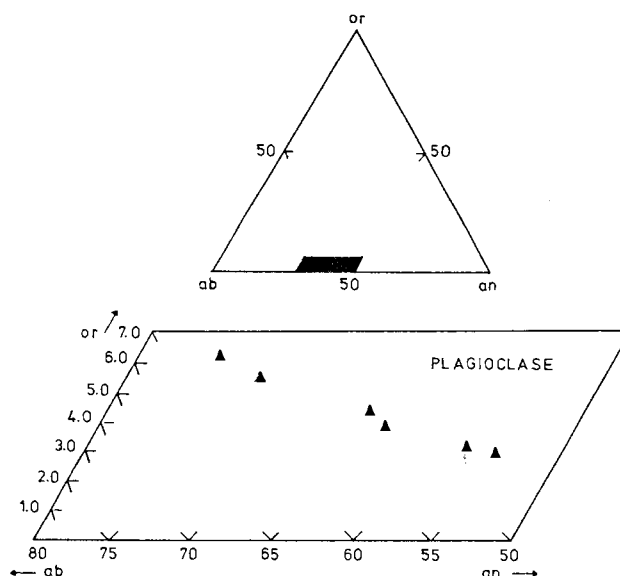

Fig. 6. Compositions of plagioclases (triangles) from Sicilian rock-samples as determined by electron microprobe. The figure illustrates the Ca enrichment of plagioclases from the core zone to the border one.

present in euhedral to subhedral forms, but an amount of rod-like material is also present. Within the enclaves *ilmenite* is the dominant mineral forming very elongated rods, often having a pronounced parallel alignment. A small amount of ilmenite occurs as fine-grained exsolution lamellae in magnetite. The last stage of feldspar crystallization is shown by replacement of previously crystallized feldspars by K-feldspar and nearly pure *albite*. Some plagioclase crystals are mantled with relatively thin and often discontinuous overgrowths of *anorthoclase*. In some instances, the titanaugite phenocrysts appear corroded and may have pleochroic rim-areas of light-to-dark green pyroxene. According to its optics, it is *aegirinaugite* with $\alpha > \beta > \gamma$ and $c^{\wedge}\alpha = 13°$; it is a Ti-rich term (see Tab. 5). In some rocks, this later crystallized titaniferous variety has a noticeably more calcic composition (CaO = 3.49 wt. %). At times, the kaersutite crystals show very notable compositional changes: in the border-zones a *baerkevikite* phase, having the chemical

Table 4: Zoning microprobe analyses of titanaugite, kaersutite and andesine crystals from enclaves.

Elements (wt.%)	Single crystal of titanaugite			Single crystal of kaersutite		Single crystal of andesine	
	core	middle	border	core	border	core	border
SiO_2	45.79	48.85	49.26	39.65	40.49	55.00	60.27
TiO_2	3.67	2.08	1.45	6.41	6.16	0.12	0.10
Al_2O_3	6.85	4.19	2.65	11.64	11.09	28.08	24.70
Cr_2O_3	0.00	0.00	0.00	0.00	0.06	0.00	0.00
FeO_{tot}	9.15	10.56	11.44	13.57	16.44	0.52	0.30
MnO	0.19	0.29	0.38	0.19	0.26	0.03	0.00
MgO	12.13	11.08	11.43	11.28	8.82	0.07	0.01
CaO	22.51	22.37	21.80	11.42	11.41	9.71	5.18
Na_2O	0.71	0.82	1.02	2.90	2.79	5.91	7.73
K_2O	0.00	0.00	0.00	0.97	0.98	0.49	1.30
Total	101.00	100.24	99.43	98.03	98.50	99.93	99.59

Table 5: Minor and accessory minerals common to Sicilian felsic enclaves and their mafic host-rock. Asterisk indicates minerals occurring in the felsic enclaves only. N indicates the number of spot analyses. Ilmenite, titanite and apatite were not analysed.

N	4	4	3	3	2	2	3	4	4
Elements (wt. %)	Biotite	Augite	Titano-magnetite	Albite *	Anorthoclase *	Titaniferous aegirinaugite	Baerkevikite *	Chlorite Fe/Mg ≅ 2.78	Chlorite Fe/Mg ≅ 0.98
SiO₂	36.18	48.78	5.90	66.63	64.95	51.92	43.46	29.43	29.65
TiO₂	7.06	1.31	23.48	0.03	0.09	5.96	4.67	0.17	0.12
Al₂O₃	13.27	2.68	2.63	19.57	19.78	0.75	7.30	14.73	22.93
Cr₂O₃	0.00	0.00	0.00	0.00	0.00	0.00	0.69	0.00	0.00
FeO$_{tot}$	21.16	11.53	63.66	0.25	0.18	26.14	22.35	30.76	15.80
MnO	0.22	0.40	1.71	0.00	0.05	0.37	0.38	0.15	0.12
MgO	11.02	11.26	1.31	0.00	0.02	0.07	7.50	11.05	16.09
CaO	0.00	21.95	0.43	0.25	1.02	0.22	8.90	0.82	0.52
Na₂O	0.84	0.90	0.02	9.70	7.13	14.24	3.93	0.07	0.10
K₂O	8.54	0.00	0.05	1.57	5.39	0.00	1.32	0.00	0.00
Total	98.29	98.81	99.19	98.00	98.61	99.67	100.50	87.18	85.33

composition represented in Tab. 5, is found. *Apatite* often occurs in elongated idiomorphic prisms, but the pseudo-hexagonal habit is also found. Occasionally, small inclusions of apatite may occur in the kaersutite crystals. Among the alteration minerals *glauconite* and especially chlorites are present. Two types of *chlorites* are evident: a phase having a Fe/Mg ≅ 2.78 ratio and the other one having a Fe/Mg ≅ 0.98 ratio (see Tab. 5). The accessory minerals are aegirinaugite, baerkevikite, *titanite*, apatite, albite and anorthoclase. Biotite, augite, titanomagnetite, albite, anorthoclase, titaniferous aegirinaugite, baerkevikite and chlorites microprobe analyses of the Sicilian enclaves and their host-rock are presented in Tab. 5.

Glass

The presence of glass at grain boundaries in both mafic and felsic Sicilian fractions is evidenced by the occurrence of an isotropic base, and by the presence of included feathery quench dendrites. In many cases the glass contains crystals of apatite and an opaque phase, very probably magnetite. Often also interstitial phases between laths of plagioclase contain residual isotropic glass. In many instances the glass phase is characterized by the presence of immiscible dark-coloured globules of glass (high-iron) enclosed in a base of lighter coloured or colourless glass (high-silica). The dark-coloured high-iron globules are generally spherical, rarely elliptical, irregular blob or dumb-bell shaped. When these globules are attached to plagioclase crystals they are flattened or hemispherical in form. Textures of the Sicilian rocks are shown in Figs. 7 to 9. These figures exhibit characters which are closely comparable to those found in the immiscible lunar samples (see Roedder & Weiblen 1970a, 1970b; Weiblen & Roedder 1973).

Eight electron microprobe analyses of glass were carried out (Tab. 6). Four analyses are of *acidic glasses* (Fe-poor/SiO₂-rich) and four analyses are of *basic glasses* (Fe-rich/SiO₂-poor). According to Furnes et al. (1981), the low analytical totals are, in general, due to the presence of water and the decrease in the totals can be used as a measure of progressive alteration. In our case, however, the low totals of the Sicilian glasses are essen-

tially due to the high water content (Lucido 1981) and only the basic glasses having the lowest totals show significant alteration effects. Also considering the alteration effects, in agreement with the textural and geochemical data (Lucido 1981, 1983; Lucido & Triolo 1983, 1984), acidic glasses are clearly richer in silica, alumina and alkalis than the basic glasses. Differently, these latter glasses are characterized by less polymerizing elements, in particular high Fe- and Mg-contents. When the analyses of the Sicilian glasses are plotted on the Greig diagram [SiO₂ - (Na₂O + K₂O + Al₂O₃) - (CaO + MgO + FeO + TiO₂ + P₂O₅)], they fall near the field of liquid immiscibility (Fig. 10). In particular, the tielines between acidic and basic glasses are essentially parallel to those in the system K₂O-FeO-Al₂O₃-SiO₂ (Roedder 1951), but the extent of immiscibility differs. However, they fall approximately near some Monteregian ocelli pairs (Philpotts 1976) and near varioles and matrices of Archean variolites (Gélinas et al. 1976), both considered to represent immiscibility. The above difference probably results from the fact that the pseudo-ternary diagram is a grossly oversimplified representation of the multicomponent natural systems (Roedder 1979).

Experiments

Experiments were carried out using the high-T and high-P equipment of the Department of Geology and Geophysics at the University of Connecticut (Storrs, U.S.A.). Thin wafers (1 × 3 × 5 mm) across the contact between the felsic and mafic fractions of one of the samples were cut. These were then supported on a platinum wire and sealed in a glass capsule along with an oxygen buffer. The charges were then heated till melting just began. It is very interesting to know that immiscible globules do form in between the crystals on first melting. A clear glass forms at grain boundaries in the felsic part of the rock. A similar clear glass forms at grain boundaries in the mafic fraction, but here small globules of an iron rich melt are also formed (see Fig. 11). The immiscible liquids in this figure were formed by heating the uncrushed sample at 1030 °C for 240 hours under an oxygen fugacity controlled by the quartz-fayalite-magnetite

Table 6: Analyses 1, 2, 3, 4 come from interstices in a mafic rock-sample, while analyses 5, 6, 7, 8 come from interstices in a felsic enclave.

Elements	Acidic glasses				Basic glasses			
(wt.%)	1	2	3	4	5	6	7	8
SiO2	62.69	62.08	61.47	60.58	40.00	35.64	38.60	36.67
TiO2	0.11	0.08	0.06	0.17	5.42	0.27	0.31	0.76
Al2O3	19.34	19.08	18.83	19.09	9.93	14.48	17.77	8.55
Cr2O3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
FeOtot	0.47	0.57	0.68	0.45	16.35	26.67	11.55	22.71
MnO	0.00	0.00	0.00	0.00	0.29	0.19	0.22	0.32
MgO	0.00	0.12	0.24	0.02	8.84	10.25	12.95	11.45
CaO	0.37	0.28	0.20	0.16	10.69	0.62	0.55	0.63
Na2O	5.38	4.98	4.59	4.60	2.75	0.91	1.56	1.09
K2O	8.05	8.76	9.48	8.59	1.27	1.32	2.94	2.46
Total	96.41	95.95	95.55	93.66	95.54	90.35	86.45	84.64
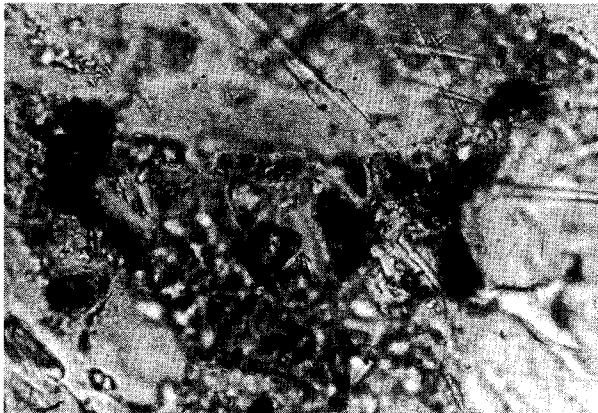
Fig. 7. Emulsion-like texture in a felsic enclave from Western Sicily. Immiscible high-iron globules (dark) and high-silica ones (light) in each other. Mineral occurring as needles is apatite. Width of Fig. 7 is 167 μm (parallel nicols).
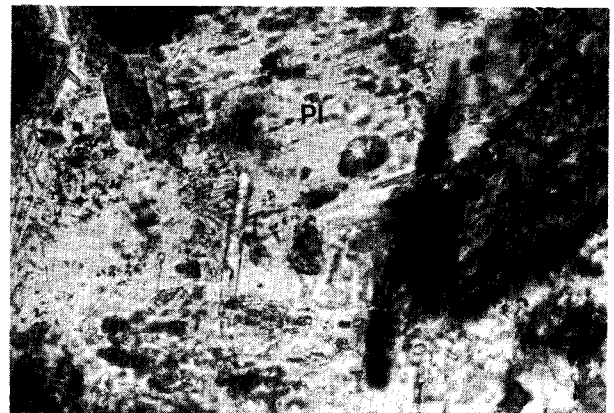
Fig. 8. Felsic enclave from Western Sicily. This figure shows a large iron-rich globule in a plagioclase crystal (Pl) and high-silica glass containing very small iron-rich globules, which in turn contain smaller droplets of the high-silica glass. Needle of apatite is evident in the central part of the figure. Width of Fig. 8 is 167 μm (parallel nicols).

buffer. These liquids result from the melting of phosphorus-rich patches of interstitial material. Apatite must have been involved. The question arises as to whether immiscibility would have developed if these immiscible patches could have been thoroughly mixed throughout the rock. The fact that immiscibility can be produced in the uncrushed rock, and particularly in the mafic phase, shows that an immiscibility field exists near the bulk composition of the rock at phosphorus-rich compositions. The presence or absence of immiscibility is commonly dependent on subtle differences in pressure or composition. So, it is possible that immiscibility could occur in the whole rocks under pressure, or under partial pressures of H_2O, CO_2, O_2, or sulphur. It was noticed that a very strong smell of sulphur was given off every time the silica glass tubes were broken open following an experiment. It is possible that if the sulphur fugacity could be controlled, this could have an effect on immiscibility. Further experimental work is in progress to find out what happens when other possible variables are investigated.
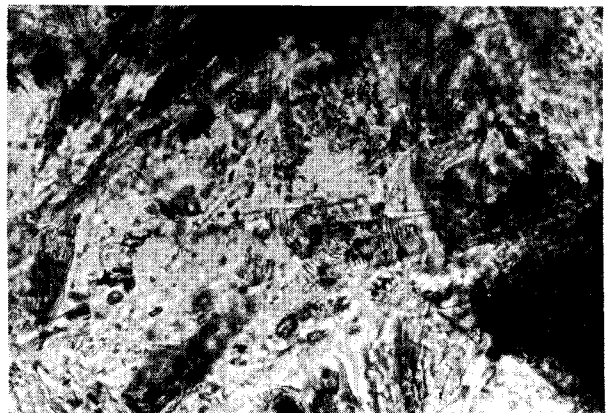
Fig. 9. Felsic enclave from Western Sicily. Typical odd ribbing in a glassy mesostasis as a result of liquid immiscibility. The elongated idiomorphic prisms are crystals of apatite. Width of Fig. 9 is 167 μm (parallel nicols).
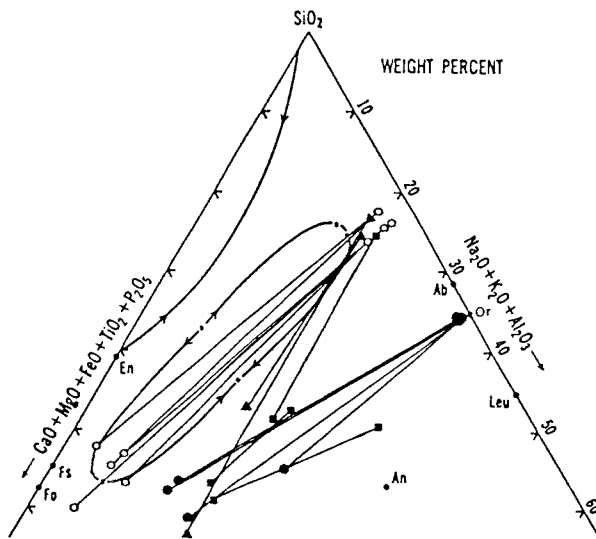
Fig. 10. Pseudo-ternary diagram showing field of low-temperature immiscibility in the system leucite-fayalite-silica, adapted from Weiblen & Roedder (1973) and tielines for some coexisting lunar glasses and rock pairs. Other lunar glasses are similar but have been omitted for clarity. All compositions recalculated on the basis of plotted oxides only. The phosphorus pentoxide of the Sicilian glasses was not detected by electron microprobe. **Open circles:** coexisting lunar glasses; data from Roedder & Weiblen (1970b, 1971, 1972), Weiblen & Roedder (1973) and Switzer (1975). **Solid circles:** Sicilian glasses. **Solid squares:** Monteregian ocelli pairs (Philpotts 1976). **Solid triangles:** varioles and matrices of Archean variolites (Gélinas et al. 1976).

Results and discussion

From all the above-reported data the following results appear evident for the Sicilian rocks.

1 - Surface tension presumably made the enclaves round, and their tendency to flatten indicates that they were in a liquid state to be deformed.

2 - Globules of glass of one composition occur in a glass of a different composition; according to Roedder (1978, 1979) this feature is one of the best proofs of immiscibility. Moreover, the occurrence of globules in each other (Figs. 7 and 8) proves the existence of two liquid phases (see e.g. De 1974; Philpotts 1978, 1979).

3 - Mineralogical phases occurring in the mafic rock are the same as those in the enclaves, but they occur in differing proportions (see Tab. 2), with leucocratic and hydrous minerals concentrated in the felsic fractions. This feature is clear evidence for a very similar stage of evolution, and suggests that the mafic and felsic fractions were two truly (thermodynamically, that is) immiscible liquids. Then, the fact that hydrous minerals are especially abundant in the felsic portions indicates that these portions crystallized in the presence of a considerable amount of water vapour. The presence of water causes changes in the melt structure. In particular, the presence of water changes the solubilities of the other components (Carstens 1979) and, therefore, it may be an important factor in evoking immiscibility by lowering liquidus temperatures (see e.g. Philpotts & Hodgson 1968; Philpotts 1971; Khitarov & Pugin 1978; Carstens 1979).
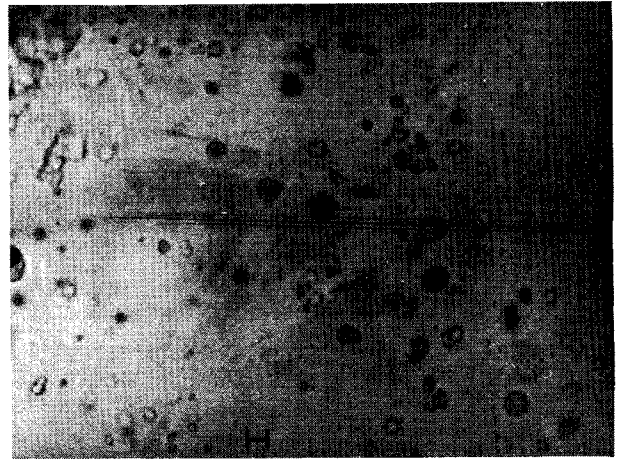

Fig. 11. Experimental liquid immiscibility in the Sicilian rocks produced by heating the rock at 1030 °C for 240 hours under an oxygen fugacity controlled by the Qz-Mt-Fa buffer. Immiscible iron-rich globules are scattered in a clear glass of the mafic fraction. Scale bar is 10 μm.

4 - Electron microprobe analyses of minerals occurring in both enclaves and host-rock indicate that these minerals had the same compositions in both liquids.

5 - Laboratory melting experiments show that an immiscibility field exists near the rock bulk-composition.

Liquid immiscibility appears to be the process responsible for the origin of the Sicilian rocks. This process is also supported by the following points:

a - The volume ratio of the mafic and felsic phases is difficult to estimate but it is enormously in favour of the mafic fraction (very roughly 97 % against 3 %).

b - In some instances, mafic spots occur at the cuspate borders of the felsic enclaves (Lucido 1981). This feature is very important: it is not only clear evidence for immiscibility, but also suggests that the mafic and felsic liquids had the same liquidus temperatures. This is possible because in our samples the separation into mafic and felsic portions occurred in a "critical liquid phase" preceding the solidification (Lucido & Triolo 1983, 1984). In fact, it is a well known and documented phenomenon (e.g. Cahn 1969; Simmons et al. 1970; Goldburg & Huang 1975; James 1975; Langer 1975) that a liquid phase undergoing critical fluctuations will eventually go into a phase separation region. The net effect will be the formation of two liquid phases having completely different compositions.

c - Unmixing is a thermal equilibrium process. Therefore, in some cases immiscibility from a common parent must be rejected because the mafic magma is chilled against the felsic magma. In the Sicilian samples, chilled margins against the felsic rocks were never observed.

d - There is no evidence of intermediate mineralogy, glassy rims, interdigitated layers or wrapping of the included bodies at the contact between felsic and mafic rocks. One would expect to see some of these gradational contacts if the mixing model were applicable.

e - Diffusion between coexisting miscible liquids, should lead to intermediate compositions along the contact of the two liquids and simple reduction of chemical gradients. In this regard, analyses 1 and 2 (Tab. 1) represent respectively the core and the rim of a felsic enclave. The rim is clearly richer in silica, alumina and potassium and poorer in Ca, Mg, Fe, Mn, Ti and P than the core. This distribution is the reverse of that which

one would expect to see if the two liquids were miscible. Several microprobe spot analyses, carried out with defocused beam, confirmed this distribution.

Conclusions

Field structures, microscopic textures, mineralogical data and laboratory experiments lead to the conclusion that the Sicilian rocks were formed by liquid immiscibility of a common parent liquid. Synthesizing the foregoing results, the following course of events may be reconstructed.

At a given time a basic magma rose up towards the Earth's surface in the Sicanian Basin (Western Sicily). This magma was not effused, but arrived at very high crustal levels (near the Earth's surface). It entered the liquid immiscibility field and thus started to separate into two immiscible liquids. Liquid-liquid phase separation should have started when the temperature of the magma was still high enough to allow diffusion and the cooling rate was not too fast. The felsic enclaves were certainly formed while the reaction of the titanaugite into kaersutite was taking place (Fig. 4). Although we are not able to tell exactly the P_{tot} and P_{H_2O} conditions, we think that this reaction occurred at low P_{tot} and high P_{H_2O}, in the presence of alkaline fluxes (Na and K-rich fluid phases).

In general, even if typical minerals of the undersaturated rocks are lacking in our rocks, the Sicilian parent magma evolved from slightly undersaturated conditions to saturated ones during liquid-liquid phase separation. This tendency is clearly evidenced by the zoning of the mineralogical phases. In particular, the titanium and aluminium depletion with distance from the core zone to the border zone in the titanaugite crystals (Tab. 4) characteristically indicates transition from undersaturated to saturated conditions (Carmichael et al. 1974). This transition is confirmed by the augite crystals occurring in the fine-grained portions and having the same chemical compositions as the rims of the titanaugite grains. Finally, the presence of pseudomorphs after presumed olivine (Fig. 2) and the feldspar crystals restricted to the groundmass indicate the same evolutive path.

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