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Biotites from biotite-rich crusts of enclaves and clots in the Monopigadon pluton (Macedonia, northern Greece)

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Abstract. — Biotite flakes from the biotite-rich crust of three enclaves and a clot, from the Monopigadon pluton (Macedonia, northern Greece), are examined from the textural and chemical point of view, and compared with those of host-rocks and enclaves. Biotite crystals, forming the biotite-rich crusts and the clot, are not oriented, but they are longer than those of the host-rocks. In particular, biotites of the clot are more than 2.5 times longer than those of the host rock, and biotites of biotite-rich crusts of enclaves are 10 to 50% longer relative to those of the host rock. Their colour is reddish to reddish-brown differing from that of the host-rocks where it is reddish-brown to brown and yellow-brown to brown. Generally, contents of Si, Fe and Mg in biotite increase in the host-rock towards the enclave (or clot), whereas contents of Al and Ti contents decrease in the same direction. Biotites of crusts and the clot are Ti-richer relative to the host-rock biotites. The main substitutional mechanism for Ti^{4+} can be described from the relationship $(R^{2+})^{VI}+2(Si^{4+})^{IV}=(Ti^{4+})^{VI}+2(Al^{3+})^{IV}$. The euhedral lath-like biotite crystals of biotite-rich crusts and the clots were formed simultaneously with the crystallization of the host-rock as the result of the surrounding granitic magma temperature decreasing in a narrow space at contact with colder enclave. It is suggested that the biotite composition of biotite-rich crusts and the clot depends on that of the host-rock biotites, whereas it is independent from the enclave composition.

Riassunto. — Sono state esaminate lamine di biotite sia della crosta ricca di biotite intorno a tre inclusi, due surmicacei ed uno cumulitico, sia di un *clot* – incluso costituito essenzialmente da biotite – del plutone di Monopigadon (Macedonia, Grecia settentrionale). L'esame è stato sviluppato sia sotto il profilo strutturale che chimico ed i dati sono stati confrontati anche con quelli delle rocce incassanti. I cristalli di biotite che compongono le croste ricche di biotite ed il *clot* non sono orientati, ma sono più lunghi di quelli delle rocce incassanti. In particolare, le biotiti del *clot* sono lunghe più di 2,5 volte rispetto a quelle della roccia incassante, a differenza delle biotiti appartenenti alle croste che hanno una lunghezza che varia da 1,10 a 1,50, sempre rispetto alle corrispondenti della roccia incassante. Il colore delle biotiti delle croste e del *clot* varia dal rossiccio al rosso-marrone e differisce da quello delle biotiti della roccia incassante che mostrano una gamma cromatica che varia dal rosso-marrone al marrone e dal giallo-marrone al marrone. In genere, i contenuti di Si, Fe e Mg delle biotiti delle rocce incassanti aumenta verso l'incluso (o *clot*), in parallelo con la diminuzione dei contenuti di Al e Ti. Al contrario, le biotiti delle

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croste e del *clot* sono più ricche di Ti rispetto a quelle delle rocce incassanti. Il principale meccanismo di sostituzione del Ti^{4+} può essere descritto tramite la relazione $(R2+)^{IV}+2\ (Si4+)^{IV} = (Ti4+)^{IV} + 2(Al3+)^{IV}$.

Le lamine euedrali dei cristalli appartenenti alle croste ed al *clot* si sarebbero formati simultaneamente con lo sviluppo della roccia incassante, in seguito alla diminuzione della temperatura del magma granitico inglobante, nello spazio ristretto che circonda l'incluso. I dati strutturali e chimici indicano inoltre che la composizione delle biotiti appartenenti alle croste ed al *clot* dipende da quella delle biotiti della roccia incassante, mentre tale composizione è indipendente da quella dell'incluso.

Key words: *Mineralogy, biotite composition, biotite-rich crust, surmicaceous enclaves, Monopigadon granite*

Introduction

A common characteristic of granitoid rocks is the presence of xenoliths and enclaves whereas surmicaceous enclaves and crystal clusters of biotite, known as biotite clots, are not so common. Micaceous crusts, biotite-rich crusts, and shells of biotite are usually develop around xenoliths and surmicaceous enclaves, although rarely, could be found around cumulate enclaves.

Some information will be given here about the not so common surmicaceous enclaves and clots. Surmicaceous enclaves are present in many intrusions and they are particularly common in anatectic granites associated with migmatites. Didier (1973) suggested that the term "surmicaceous enclave" must be restricted to those where the mica content forms more than 50% by volume of the enclave. Latter, Didier and Barbarin (1991) reported that a distinguishing feature of the surmicaceous enclaves, in the field, is their biotitic crust. Such enclaves are reported by many authors for different granitic rocks e.g. in the granite intrusion on the Velence Mts. (Hungary; Buda, 1993), in the Deddick granodiorite (Lachlan Fold Belt, SE Australia; Maas *et al.,* 1997), in the peraluminous granite of the Nelas area (Central Portugal; Silva *et al.,* 2000) and within the Monte Capanne pluton (Elba Island, Italy; Gagnevin *et al.,* 2004).

Didier (1973) accepted that the surmicaceous enclaves are quite clearly restites whereas their presence was interpreted as a proof that continental crust participated in the genesis of granites (Didier, 1987). Many authors (e.g. Montel *et al.,* 1991; Schödlbaner *et al.,* 1997; Williamson *et al.,* 1997; Nemchin and Pidgeon, 1999; Barnes *et al.,* 2001, Waight *et al.,* 2001; Meli and Sassi, 2000 and 2003; Clarke *et al.,* 2005) argued that surmicaceous enclaves represent either peculiar lithologies from the source area that resisted anatexis or country-rock xenoliths picked up by the pluton at or close to the emplacement level, or partially digested xenoliths of country rocks. Didier (1973) accepted that clots are originated from enclaves or xenolithic rocks due to a combination of processes causing disintegration movements within the magma, aiding for the final disruption and dispersion of the clots from their place of origin (the enclave) to their place of rest (the surrounding granitic magma). The outer biotite rich-crust occurring around xenoliths and enclaves is interpreted as the result of reaction between the ferromagnesian minerals of the enclave and the surrounding granite magma (Didier, 1973). A different interpretation was proposed by Perugini *et al.* (2003) on the basis of large biotite crystals within the host magma, disposed mainly along the periphery of enclaves found in the Sithonia pluton (Greece). The tabular habit characterizing biotites allowed them to align along path lines and hence remain in the host rock.

Although the significance of xenoliths, enclaves, surmicaceous enclaves and clots and their possible origins have been discussed in detail by many authors, little attention has been paid to the texture and chemistry of biotites forming biotite-rich crust and their relations with biotites of host-rocks and enclaves. A characteristic feature of the Monopigadon pluton is the presence of numerous enclaves, xenoliths of different rock types, surmicaceous enclaves, as well as biotite clots. Among enclaves and xenoliths there are many offering the possibility to study their biotite-rich crust and to decipher their peculiar characteristics. A detailed study can give, in fact, information about the factors controlling the composition of the biotites and the role of the enclaves, with the goal of clarify the genesis of the biotites, at least from a qualitative point of view.

The Monopigadon Pluton: Regional setting, field relations, Age

The Monopigadon pluton is an intrusion occupying an area of about 10 km^2 situated between the villages Ag. Antonios, Monopigadon and Krini of Chalkidiki (Fig. 1) and composed of three bodies elongated NW-SE with the most northwestern being the larger. According to Michard *et al.* (1988a and b) the Monopigadon pluton intrudes both the Chalkidiki ophiolites as well as a supra-ophiolitic formation (ophicalcites, volcanoclastic flysch, reefal limestones and conglomerates with acidic rock pebbles). The Chalkidiki ophiolites (Gauthier, 1984) belong to the eastern part of the Vardar ophiolitic belt (Peonias Zone; Mercier, 1968) and in the area show autochthonous cover formations (reefal limestones of Kimmeridian-Tithonian age, and clastic deposits). Ophiolites and supra-ophiolitic formations thrust over the Circum Rhodope and part of Rodopian units (Michard *et al.,* 1998a, b). The Monopigadon pluton consists

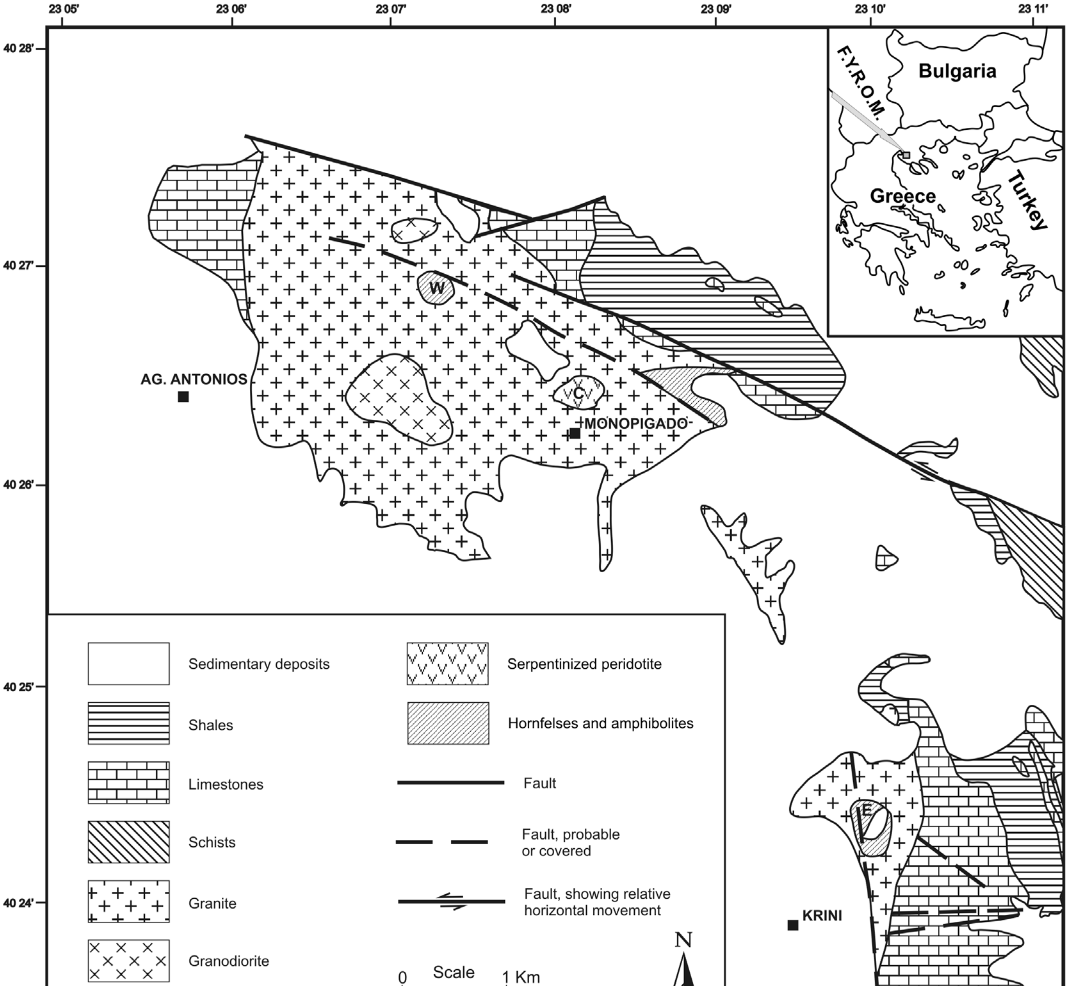

Fig. 1 – Geological sketch map of Monopigadon pluton. W, C and E: western central and eastern bodies enclosed in the plutonic rocks. Arrow in the inset map indicates the area of investigation.

of three different rock-types: biotite granodiorite (BGrd), biotite granite (BGr) and leucogranite (LGr). Aplitic dykes (Apl) occur intruding the BGrd, BGr and LGr. Enclosed rocks are very frequent in BGrd and BGr while they are very rare in LGr. The pluton, exhibits mainly I-type characteristics with ASI mostly ≤ 1.1 although the initial Sr isotopic ratio ranges from 0.7093 to 0.7291 (Koroneos in preparation).

Three rounded bodies of country rocks, covering an area of about 0.25 km^2 each one, are enclosed in the pluton. The central (C, Fig. 1) body is composed mainly of serpentinized peridotite whereas the western and the eastern (W and E, Fig. 1) are composed of hornfelses and amphibolites. Except the tertiary and neogene sediments, covering the broader area, the Monopigadon pluton is unconformably overlain by the Kimmeridian-Tithonian Petralona peri-reefal limestones (Ricou, 1965; Kockel *et al.*, 1977; Gauthier, 1984; Michard *et al.,* 1998b). The latter are followed by the Prinochori beds (calcschists, calcarenites, and conglomerates with slate and quartz pebbles).

The age of the Monopigadon pluton is constrained by the above mentioned stratigraphic evidence and radiometric measurements as well. The pluton was dated by K/Ar on biotite separates at 180 Ma (Ricou, 1965), 149 Ma (Mussallam & Jung, 1986), and 141.5±3.1 Ma (Michard *et al.*, 1998a, b). Kostopoulos *et al.* (2001) reported recently a single zircon evaporation Pb/Pb age at 192.5±3.8 Ma. Preliminary zircon U/Pb dating gives a minimum emplacement age of 184.3±0.8 Ma (Christofides *et al.*, unpubl. data). The last two ages confirm an early Jurassic intrusion for the Monopigadon pluton.

Petrography

The host-rocks

BGrd: Biotite granodiorite is a grey to dark-gray, fine to medium-grained rock with quartz (25.1 to 31.4 vol%), slightly zoned plagioclase (35.0 to 42.0 vol%), often altered to sericite, orthoclase (1.5 to 4.9 vol%), sometimes kaolinized, and biotite existing in higher amounts relative to the other rock-types (28.2 to 34.6 vol%). In thin sections, biotite is reddish-brown to brown and very rarely is discoloured or altered to chlorite. Zircon with pleochroic haloes is a common inclusion in biotite, and apatite, titanite and opaque minerals are present as accessories.

BGr: Biotite granite is a grey-whitish coarse to medium-grained rock intruding the BGrd. The main mineral constituting granites are: quartz (20.2 to 28.9 vol%), zoned plagioclase (32.2 to 40.7 vol%), orthoclase (23.8 to 25.1 vol%), and biotite (12.0 to 14.6 vol%). Orthoclase is very rarely kaolinized, occurring both as phenocrysts and groundmass constituents. It is perthitic and encloses small crystals of plagioclase and biotite in poikilitic texture. Biotite is yellow-brown to brown and often discoloured or altered to muscovite and chlorite. Rarely, rag-shaped amphibole crystals are found. Zircons with pleochroic haloes, apatite, titanite and opaque minerals are present as accessories.

LGr: Leucogranite is a fine to medium-grained leucocratic rock intruding BGr. In some places it is altered in coarse-grained granitic sand. It is richer in quartz (34.8 to 38.1 vol%) and K-feldspar (29.3 to 49.3 vol%), relatively to BGrd and BGr, and poorer in biotite (1.7 to 6.3 vol%). Perthitic K-feldspars (orthoclase and microcline) are kaolinized and slightly zoned plagioclases are altered to sericite. Biotite is yellow-brown to brown in colour and is very often discoloured or altered to chlorite. Muscovite is also present in minor amounts. Zircons, opaque minerals and rarely apatite are present as accessories.

Apl: Aplitic veins rich in quartz (55.2 vol%) and perthitic K-feldspars (orthoclase and microcline; 42.4 vol%), with minor amounts of plagioclase (0.9 vol%) and biotite (0.8 vol%). Zircons and opaque minerals are present as accessories.

The enclosed rocks

Different kinds of rocks are found enclosed in the BGrd and BGr. The majority of them have lenticular to ovoid shape although some angular pieces are also found (Fig. 2a-c). These rocks can be found all over the pluton. They can be grouped in the following six (a to f) groups.

a. Serpentinized peridotite. These are 10-20 cm in size and occur mainly close to the contact of the pluton with the enclosed central (C, Fig. 1) body.

b. Amphibolites. They are rich in amphibole and plagioclase, whereas quartz occurs in very low quantities. Their size ranges from 3 to 10 cm. It is

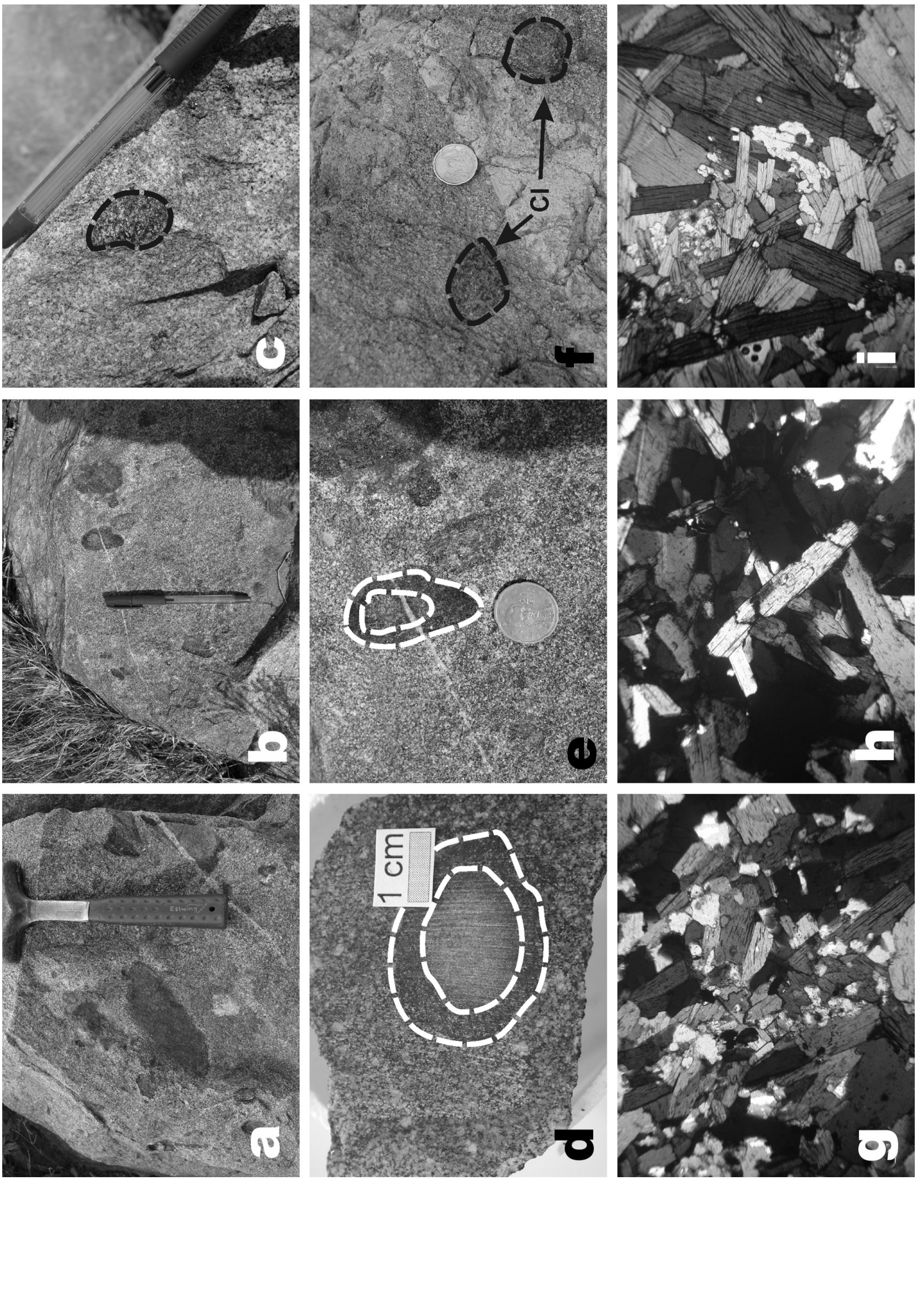

Fig. 2 – Angular and ovoid enclaves of different size in sample MO-23E (a); ovoid enclaves in samples MO-89A (b) and MO-67A (c). Biotite-rich crust (between the two dashed white lines) in sample MO-23E (d) and sample MO-89A (e), and biotite clots (Cl) in sample MO-101 (f). Microphotographs of biotite-rich crusts in sample MO-23E (g) and sample MO-89A (h), and the biotite clot in sample MO-101 (i). The larger side of the last three photos measure 3 mm.

to note here that no micaceous crusts are developed around them.

c. Rocks rich in sericitized plagioclase, clinopyroxene, orthopyroxene, hornblende, and some quartz. They probably represent mafic granulites. Their size ranges from 3 to 10 cm.

d. Rocks rich in biotite (more than 50%), chlorite, some quartz, apatite and epidote. Their size ranges from 3 to 8 cm (Figs. 2a-c). In some of these rocks more than 40% of the biotite is discoloured or altered to muscovite and chlorite. It must be stressed that these altered biotite crystals show a uniform distribution and are not concentrated close to the contact with the host rock. These rocks represent surmicaceous enclaves. They show sharp contacts with the host-rocks, and are very often surrounded by biotite-rich crusts (Figs. 2d-e).

e. Small (2-4 cm) coarse-grained enclaves, rich in biotite, sericitized plagioclase and magnetite, with small amount of quartz and K-feldspar. Their mineralogical assemblage is similar to that of host-rocks, although they differ from the latter for the crystal size being bigger in the enclaves. They also differ for the size and amount of apatite and zircon which are more abundant and form bigger euhedral crystals in the enclaves, relatively to the host-rock. These enclaves are surrounded by biotite-rich crusts similar to enclaves d) and entirely made of not oriented biotite flakes, with widths ranging between 2 and 5 mm. Based on their texture and mineralogy, these, very rare enclaves, are probably of igneous origin, and they could be considered as cogenetic with the pluton, representing, hence, cumulate enclaves. Moreover, they cannot represent mafic microgranular enclaves since their crystals are bigger of those of the host-rocks and needle-like crystals are lacking. Similar type of enclaves is reported by Waight *et al.* (2001) in the Cowra Granodiorite (Lachlan Fold Belt, SE Australia), among five different varieties of enclaves, ranging in size from a few centimetres to 50 cm in diameter, and having a biotite-rich crust.

f. Rocks made chiefly of biotites with diameter ranging from 0.7 to 2 cm (Fig. 2f), and very small quartz and plagioclase crystals occurring often between biotite flakes. They represent biotite clots.

Biotites of four representative samples belonging to groups d), e) and f) were chosen (Table 1). In the following the term enclave (Enc) includes both the surmicaceous enclaves and the cumulate enclave.

Analytical Methods

Biotite analyses were carried out at the Scanning Microscope Laboratory, Thessaloniki University, using a Jeol JSM-840A Scanning

Table 1
Dimensions of the investigated biotites

Sample	Enclave Group	Host rock	Area	Length (mm)				Width (mm)			
				mean	std	min	max	mean	std	min	max
MO-23E	d	BGrd	Host	0.50	0.09	0.31	0.71	0.39	0.14	0.17	0.71
			Crust	0.68	0.07	0.57	0.86	0.34	0.13	0.17	0.71
			Enc	0.16	0.06	0.06	0.23	0.07	0.02	0.03	0.09
MO-67A	d with discoloured biotite	BGr	Host	0.39	0.12	0.23	0.77	0.23	0.11	0.11	0.43
			Crust	0.64	0.13	0.29	0.80	0.20	0.05	0.11	0.29
			Enc	0.26	0.09	0.17	0.34	0.14	0.09	0.06	0.23
MO-89A	e	BGrd	Host	0.70	0.13	0.51	1.00	0.30	0.11	0.14	0.51
			Crust	1.03	0.23	0.57	1.43	0.27	0.10	0.17	0.51
			Enc	1.65	0.80	0.43	3.00	0.56	0.33	0.14	1.29
MO-101	f	BGr	Host	0.68	0.33	0.29	1.43	0.29	0.07	0.14	0.40
			Clot	1.78	0.54	0.91	2.86	0.40	0.17	0.14	0.71

Electron Microscope (SEM) equipped with an Energy Dispersive Spectrometer (EDS) with 20 kV accelerating voltage and 0.4 mA probe current. A set of simple oxides and silicates was used as primary standards. Based on repetitive analyses of the standards, the precision of the analyses was estimated to be better than 2% (relative) for elements with concentration higher than 1% (SiO_2, TiO_2, Al_2O_3, FeO, MgO and K_2O) and better than 5% (relative) for other elements (MnO, CaO and Na_2O).

The whole rock samples were analysed by X-ray fluorescence for major elements on a Phillips PW2400 sequential X-ray fluorescence spectrometer using an Rh-anode X-ray tube (Saint Mary's University Regional Geochemical Centre, Canada). Loss on ignition (LOI) was determined by treating the sample for 1.5 h at 1050 °C in an electric furnace. Major elements were carried out on fused glass discs. International standards were used for calibration. Analytical precision, as determined from replicate analyses, is generally better than 1% (MgO, Al_2O_3, SiO_2, K_2O, CaO, TiO_2, MnO), 2% (Na_2O, P_2O_5) and 3% (FeO).

Results

Biotite morphology

Biotite crystals of biotite-rich crusts and the clot show some differences, concerning size and colour, compared with both biotites of enclaves and host-rocks. For each sample, dimensions (length and width) were measured, in thin section, for fifteen lath-like biotite crystals from each one of host-rocks, biotite-rich crusts enclaves, and the clot; results are presented in the form of minimum, maximum, mean value and standard deviation in Table 1, and compared in Fig. 3. The biotite-rich crusts and the biotite clot contain biotite crystals ranging in length from 0.29 mm to 1.43 mm (Figs. 2g-h) and 0.91 mm to 2.86 mm respectively (Fig. 2i). Among the biotite-rich crusts the sample MO-89A contains the longest crystals relative to the other two enclaves which is also the case when the host-rock biotites are compared. The biotite crystals of the host-rocks range between 0.23 mm and 1.43 mm, while the enclave-biotites from 0.06 mm to 0.34 mm. However, longer biotites were found (up to 3.00 mm) in the MO-89A enclave, and these are longer than the host-rock biotites. It should be noted that in the case of sample MO-101 (biotite clot) there is no difference in crystal-size between the outer and the inner part of the clot. Moreover, sample MO-101 shows the highest difference (1.1±0.24 mm) between the biotites in host-rock and biotite in clot in length.

The width of the biotite crystals ranges from 0.11 mm to 0.71 mm in the crusts, from 0.14 mm to 0.71 mm in the clot and from 0.11 mm to 0.71 mm in the host-rock biotites. The width of the crusts and clot biotites can be considered comparable (taking into account the deviations), which is also the case when the host-rock biotites are compared. Biotite crystals are smaller in the surmicaceous enclaves and bigger in the cumulate enclave MO-89A where crystals up to 1.3 mm wide were found. Concerning colour of biotites in thin sections, it is reddish-brown to brown in BGrd, yellow-brown to brown in BGr, whereas it is reddish to reddish-brown in the biotite-rich crusts and clot.

Biotite chemistry

Polished thin sections were prepared in order to contain at least part of the host- rock, the biotite-rich crust and part of the enclave or the whole clot. In each sample analyses were performed approximately on a transect, from the host-rock to the enclave through the biotite-rich crust or from the host-rock to the center of the clot. In the host-rock five positions containing biotites were selected; the distance between them was 3 to 4 mm. These positions were numbered from H1 to H5 with the latter close to the biotite-rich crust. In the biotite-rich crust, three positions namely C1, C2 and C3 were chosen. The first is close to the border with host-rocks, the third is close to the border with enclaves, and the second is in the middle. Lastly, in each enclave, two positions were selected, E1 and E2, one close to the border with the biotite-rich crust and the other 3 to 4 mm far from the center of the enclave. E1 and E2 positions are lacking for sample MO-101 (biotite clot). In each position, at least 3 biotite crystals (one to two spots in each crystal) were analysed and the results, in the form of mean values, are presented in the Table 2. The number of atoms per formula unit (apfu) was calculated on the basis of 22 oxygens. All the analysed samples are classified as biotites

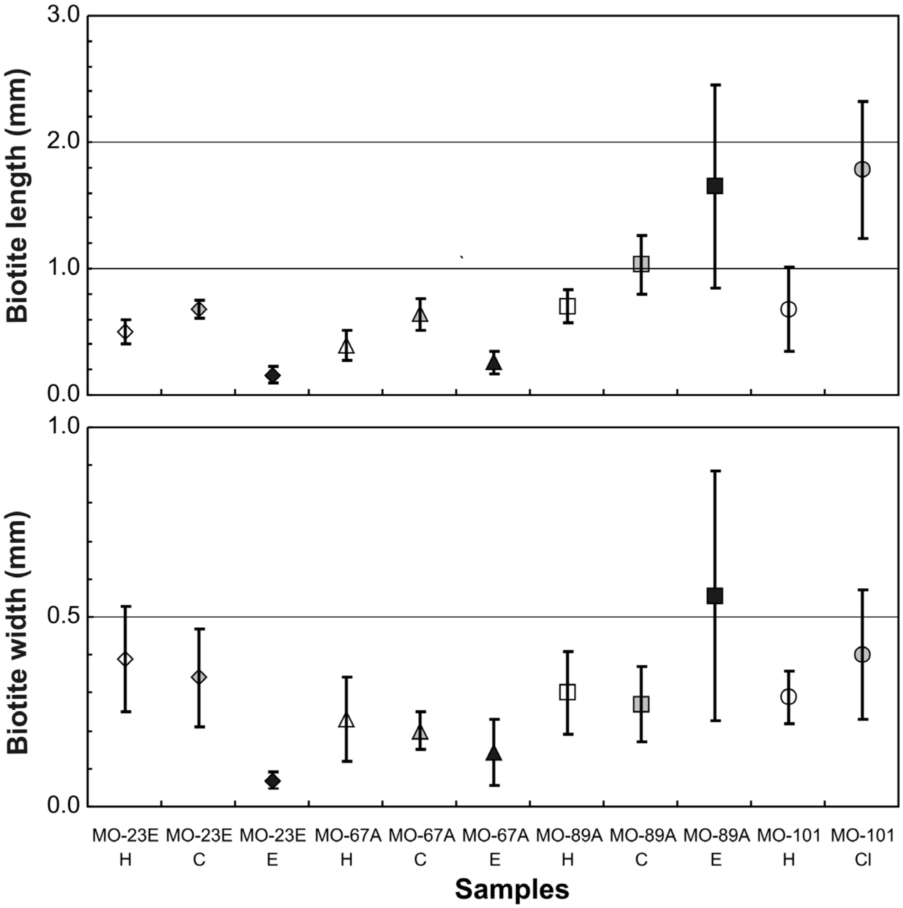

Fig. 3 – Biotite's dimension (length and width) in mm of host-rocks (H), biotite-rich crust (C), enclave (E), and clot (Cl). Diamonds: MO-23E, triangles: MO-67A, squares: MO-89A, and circles: MO-101. Open symbols: host-rock; shaded symbols: biotite-rich crust or clot; solid symbols: enclave.

on the basis of Fe/(Fe+Mg) ratio *vs.* Al_{tot} (Deer *et al.*, 1966; Fig. 4), inasmuch as they are not very variable in composition.

Compositions of the analysed biotites are compared in the diagrams of Fig. 5. For the sake of simplicity the distance between analysed positions H1 to E2 is reported equal, even if this is not the case in the biotite-rich crust (C1 to C3), where positions are closer each others. Using regular spacing from C1 to C3 some details will be, in fact, out of sight. The Si content increases in the host-rock towards the enclave (or clot) in all samples while the Fe content increases also in the host-rock towards the biotite crust, in all samples except MO-23E. The Mg content increases slightly. On the contrary the Al and Ti contents decrease in the host-rock towards the biotite crust (or clot). Al^{IV} decreases while Al^{VI} increases. Although the biotites were analysed also for Mn, Ca and Na, and some differences between host-rock and biotite-rich crust could be noted, these elements are not used due to their low concentrations and possibly low analytical precision.

However, taking into account the errors calculated for the above elements (3.4%) the only statistically robust variations are those of Ti content; the Ti contents plot with error bars in figure 5 while such a plot of the rest elements would

TABLE 2

Biotite compositions from host-rock (H), biotite-rich crust and clot (C), and enclave (E) of the Monopigadon samples

Sample	MO-23E										MO-67A									
Position	Host-rock					Biotite-rich crust			Enc		Host-rock					Biotite-rich crust			Enc	
Area	H1	H2	H3	H4	H5	C1	C2	C3	E1	E2	H1	H2	H3	H4	H5	C1	C2	C3	E1	E2
SiO_2	36.03	36.11	36.37	36.73	37.44	36.89	36.23	36.60	36.77	37.17	37.05	36.72	37.8	38.21	36.77	36.44	35.75	35.94	37.00	37.91
TiO_2	3.48	3.49	3.18	3.09	3.12	3.33	4.00	3.45	4.29	4.06	3.34	3.37	3.28	3.25	2.86	3.17	3.36	3.07	3.32	3.21
Al_2O_3	17.57	17.52	17.14	17.48	17.24	17.56	16.52	17.81	16.38	16.59	18.61	18.56	18.61	18.47	17.03	17.52	17.03	17.94	16.63	16.79
FeOt	18.64	18.03	17.67	17.73	18.56	17.94	18.36	18.05	18.24	18.99	17.76	18.11	17.95	18.32	19.40	18.91	19.32	19.41	18.13	17.36
MnO	0.24	0.28	0.31	0.31	0.24	0.46	0.31	0.31	0.34	0.25	0.19	0.35	0.17	0.47	0.25	0.45	0.37	0.41	0.58	0.42
MgO	10.50	10.52	10.54	11.06	11.14	11.12	10.85	10.54	10.71	11.12	9.51	9.47	9.73	9.68	9.47	10.23	9.83	9.61	10.22	11.68
CaO	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.01	0.00	0.00	0.01
Na_2O	0.42	0.40	0.59	0.20	0.49	0.37	0.29	0.38	0.21	0.37	0.76	0.84	0.72	0.47	0.38	0.43	0.14	0.49	0.55	0.46
K_2O	8.63	8.30	8.35	8.64	8.81	8.68	8.39	8.60	8.65	8.62	9.38	8.44	8.44	8.5	8.49	7.96	7.88	7.99	8.06	8.35
Total	95.52	94.67	94.16	95.23	97.03	96.34	94.94	95.74	95.57	97.16	96.60	95.86	96.70	97.37	94.70	95.10	93.70	94.86	94.49	96.18
Site allocations (22 O)																				
Si	5.440	5.473	5.538	5.524	5.548	5.495	5.493	5.484	5.534	5.514	5.510	5.493	5.575	5.605	5.604	5.508	5.501	5.465	5.616	5.620
Al^{IV}	2.560	2.527	2.462	2.476	2.452	2.505	2.507	2.516	2.466	2.486	2.490	2.507	2.425	2.395	2.396	2.492	2.499	2.535	2.384	2.380
Z	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al^{VI}	0.566	0.604	0.614	0.622	0.558	0.578	0.444	0.630	0.439	0.416	0.772	0.765	0.809	0.798	0.664	0.629	0.590	0.680	0.591	0.554
Ti	0.395	0.397	0.364	0.349	0.347	0.373	0.456	0.389	0.485	0.453	0.374	0.379	0.364	0.358	0.328	0.360	0.389	0.351	0.378	0.357
Fe^2	2.353	2.286	2.250	2.229	2.301	2.235	2.328	2.262	2.297	2.356	2.209	2.265	2.214	2.247	2.472	2.390	2.487	2.468	2.301	2.153
Mn	0.031	0.036	0.040	0.039	0.030	0.058	0.040	0.039	0.043	0.031	0.024	0.044	0.021	0.058	0.033	0.058	0.048	0.053	0.075	0.052
Mg	2.364	2.378	2.393	2.480	2.462	2.469	2.452	2.355	2.403	2.458	2.108	2.112	2.139	2.117	2.151	2.305	2.255	2.178	2.312	2.582
Y	5.709	5.700	5.661	5.720	5.697	5.713	5.721	5.675	5.667	5.714	5.486	5.565	5.547	5.578	5.647	5.741	5.769	5.729	5.657	5.699
Ca	0.000	0.004	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.009	0.000	0.002	0.001	0.000	0.001
Na	0.123	0.116	0.175	0.058	0.139	0.105	0.086	0.110	0.060	0.105	0.219	0.244	0.206	0.134	0.113	0.125	0.043	0.144	0.162	0.133
K	1.663	1.605	1.622	1.657	1.666	1.649	1.622	1.643	1.661	1.631	1.780	1.611	1.588	1.591	1.651	1.535	1.547	1.550	1.561	1.579
X	1.786	1.725	1.798	1.715	1.805	1.754	1.708	1.756	1.721	1.736	1.999	1.854	1.794	1.724	1.773	1.661	1.592	1.695	1.722	1.713
Mg/(Mg+Fe)	0.501	0.510	0.515	0.527	0.517	0.525	0.513	0.510	0.511	0.511	0.488	0.482	0.491	0.485	0.465	0.491	0.476	0.469	0.501	0.545

FeOt=total Fe as FeO

TABLE 2 — *Continued...*

Sample	MO-89A										MO-101							
Position	Host-rock					Biotite-rich crust			Enc		Host-rock					Biotite clot		
Area	H1	H2	H3	H4	H5	C1	C2	C3	E1	E2	H1	H2	H3	H4	H5	C1	C2	C3
SiO_2	36.61	36.26	36.45	36.18	35.55	36.36	36.58	36.76	36.84	36.80	37.02	36.77	36.30	36.57	37.35	36.62	36.30	36.30
TiO_2	3.67	3.78	3.40	3.16	3.03	3.98	4.54	4.13	4.69	4.65	3.29	3.08	2.81	2.76	2.74	3.45	3.96	3.75
Al_2O_3	17.25	17.32	17.40	16.99	16.24	16.95	16.50	17.06	16.97	16.52	18.89	19.22	18.83	19.00	18.62	18.55	17.71	18.71
FeOt	18.82	19.23	18.89	19.20	19.36	19.05	18.89	19.46	19.33	19.66	16.75	16.51	16.85	16.10	16.98	17.40	18.28	18.22
MnO	0.39	0.34	0.13	0.45	0.45	0.34	0.34	0.39	0.21	0.30	0.24	0.32	0.26	0.16	0.13	0.23	0.38	0.38
MgO	9.98	9.85	9.84	9.91	9.70	9.90	9.80	10.01	9.91	9.70	9.79	9.86	9.97	9.90	10.03	9.89	9.89	9.83
CaO	0.00	0.01	0.00	0.00	0.11	0.08	0.00	0.00	0.01	0.00	0.09	0.05	0.00	0.02	0.01	0.00	0.00	0.00
Na_2O	0.48	0.52	0.28	0.46	0.28	0.64	0.68	0.57	0.35	0.55	0.44	0.68	0.69	0.63	0.55	0.62	0.47	0.47
K_2O	8.41	8.41	8.42	8.06	8.22	8.44	8.50	8.51	8.36	8.39	7.53	7.81	8.07	7.90	8.06	7.87	7.96	7.90
Total	95.62	95.72	94.80	94.41	92.94	95.75	95.83	96.88	96.68	96.58	94.04	94.29	93.78	93.05	94.47	94.64	94.94	95.55
Site allocations (22 O)																		
Si	5.516	5.473	5.530	5.529	5.546	5.489	5.514	5.486	5.493	5.513	5.558	5.518	5.504	5.552	5.602	5.505	5.480	5.432
Al^{IV}	2.484	2.527	2.470	2.471	2.454	2.511	2.486	2.514	2.507	2.487	2.442	2.482	2.496	2.448	2.398	2.495	2.520	2.568
Z	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al^{VI}	0.579	0.555	0.642	0.590	0.531	0.505	0.446	0.488	0.475	0.431	0.902	0.917	0.869	0.952	0.893	0.792	0.631	0.732
Ti	0.416	0.429	0.388	0.363	0.356	0.452	0.514	0.464	0.526	0.524	0.372	0.347	0.320	0.315	0.309	0.390	0.449	0.422
Fe^2	2.372	2.427	2.397	2.454	2.526	2.405	2.382	2.429	2.411	2.463	2.103	2.072	2.136	2.044	2.129	2.188	2.308	2.280
Mn	0.050	0.044	0.016	0.058	0.059	0.043	0.043	0.050	0.027	0.039	0.031	0.041	0.033	0.021	0.017	0.030	0.048	0.048
Mg	2.242	2.216	2.225	2.258	2.256	2.228	2.204	2.227	2.202	2.166	2.192	2.205	2.253	2.240	2.243	2.217	2.225	2.193
Y	5.658	5.671	5.669	5.723	5.727	5.633	5.589	5.657	5.642	5.622	5.599	5.581	5.612	5.572	5.591	5.617	5.661	5.675
Ca	0.000	0.001	0.000	0.000	0.018	0.013	0.000	0.000	0.002	0.000	0.014	0.007	0.000	0.004	0.002	0.000	0.000	0.000
Na	0.139	0.151	0.083	0.136	0.085	0.186	0.200	0.164	0.100	0.160	0.127	0.199	0.203	0.186	0.160	0.179	0.136	0.135
K	1.617	1.618	1.630	1.571	1.636	1.625	1.634	1.620	1.591	1.603	1.443	1.495	1.560	1.530	1.542	1.509	1.533	1.508
X	1.756	1.771	1.713	1.708	1.740	1.824	1.834	1.784	1.693	1.763	1.584	1.701	1.763	1.720	1.704	1.689	1.670	1.643
Mg/(Mg+Fe)	0.486	0.477	0.481	0.479	0.472	0.481	0.481	0.478	0.477	0.468	0.510	0.516	0.513	0.523	0.513	0.503	0.491	0.490

FeOt=total Fe as FeO

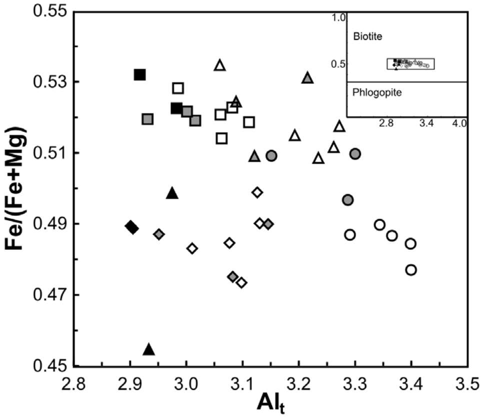

Fig. 4 – Chemical compositions for analysed biotites plotted on classification diagram by Deer *et al.* (1966); see inset for zoomed region. Symbols as in figure 3.

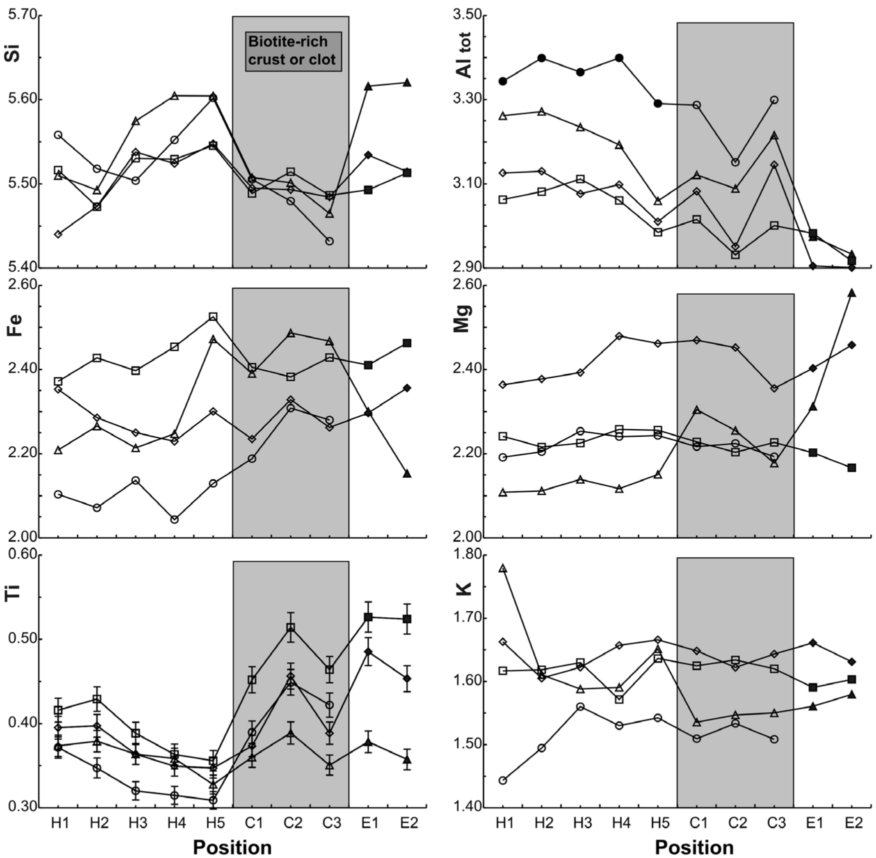

Fig. 5 – Plot of selected elements of the analysed biotites from different positions. H1 to H5: host-rock; C1 to C3 (shaded area): biotite-rich crust or clot; E1 to E2: enclave. Symbols as in figure 3.

make the trends rather indistinguishable. The Si increase and Al decrease in all host-rock biotites towards the enclaves or clot can be also considered remarkable, since these trends are observed in all samples, even if this is not well constrained from the statistical point of view.

From Fig. 5 it is noticeable that in all samples, biotites from the biotite-rich crusts and clot are Ti-richer relative to the host-rock biotites. The Si-, Al-, in the same page Fe-, Mg- and K-contents of the biotite-rich crusts and clot do not differ significantly from that of the host-rock (except K and Fe of MO-67A) tacking into account the errors. It must be emphasized that in all samples, the center (C2) of the biotite-rich crust (or clot) is Ti-richer relative to the borders both to host-rock or to the enclave (C1 and C3 respectively), whereas other elements either do not follow such a symmetry or they have relatively similar concentrations.

Several substitution mechanisms have been suggested for Ti^{4+} in biotites (see Dymek (1983) for a review). These can be described from the following relationships: $(R^{2+})^{VI}+2(Si^{4+})^{IV}=(Ti^{4+})^{VI}+2(Al^{3+})^{IV}$ (1), $(Al^{3+})^{VI}+(Si^{4+})^{IV}=(Ti^{4+})^{VI}+(Al^{3+})^{IV}$ (2), $2(Al^{3+})^{VI}=(Ti^{4+})^{VI}+(R^{2+})^{VI}$ (3), $2(R^{2+})^{VI}=(Ti^{4+})^{VI}+(\square)^{VI}$ (4) and a dehydrogenation reaction $(R^{2+})^{VI}+2(OH)^{-}=(Ti^{4+})^{VI}+2(O^{2-})+H_2$ (5). The fairly good negative correlation between Mg+2Si and $Ti+2Al^{IV}$, indicated by the high correlation coefficients (R^2=0.961, 0.992, 0.965 and 0.991 for MO-23E, MO-67A, MO-89A and MO-101 biotites of the host-rocks respectively; Fig. 6a), confirms that the

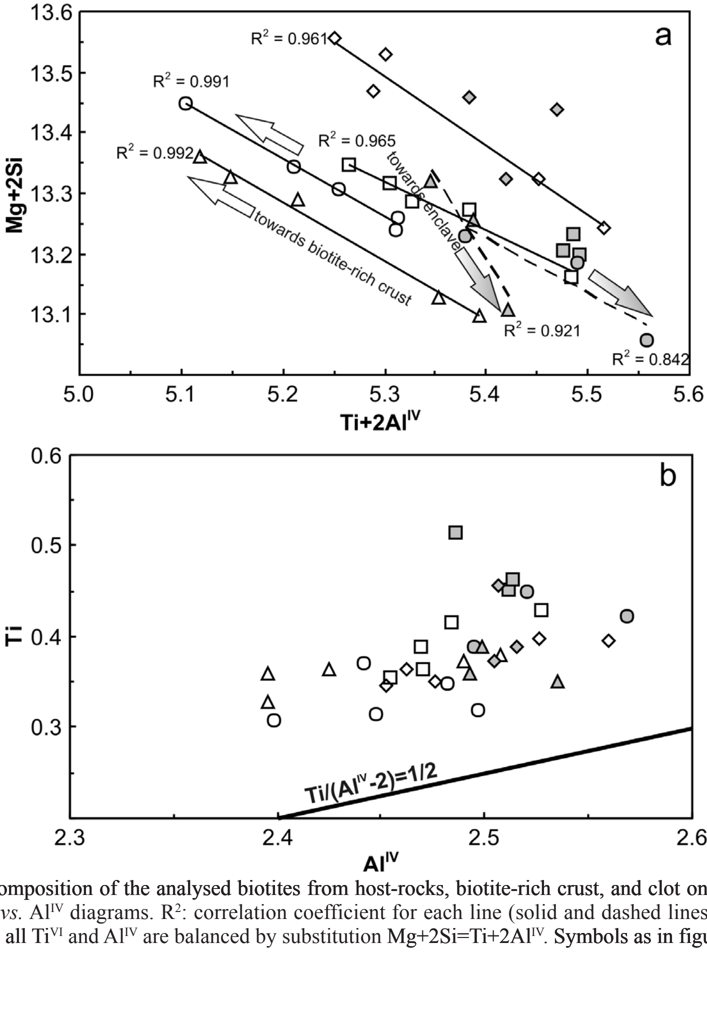

Fig. 6 – Chemical composition of the analysed biotites from host-rocks, biotite-rich crust, and clot on the: (a) Mg+2Si *vs.* $Ti+2Al^{IV}$ and (b) Ti *vs.* Al^{IV} diagrams. R^2: correlation coefficient for each line (solid and dashed lines). The line $Ti/(Al^{IV}-2)=1/2$ indicates that all Ti^{VI} and Al^{IV} are balanced by substitution $Mg+2Si=Ti+2Al^{IV}$. Symbols as in figure 3

substitution of Ti for Mg is compensated for by substitution of Al^{IV} for Si (type 1). This substitution decreases in the biotites of the host-rocks towards the biotite-rich crust or clot. The same mechanism can explain the substitution of Ti in the biotites of the biotite-rich crust of the samples MO-67A and MO-101 (R^2=0.921 and 0.842) but fails to explain the MO-23E biotite-rich crusts and the clot MO-89. The latter are better explained by the type (2) substitution (R^2=0.746 and 0.926 respectively, not shown).

However, if all Ti^{VI} and Al^{IV} were balanced by substitution (1), then the data points (Fig. 6b) should lie along the line labeled Ti/(Al^{IV}-2)=1/2, when the structural formula of biotite has been calculated on the basis of 22O (Dymek, 1983). All analysed biotites plot above the Ti/(Al^{IV}-2)=1/2 line indicating that they contain Ti impossible to balance by Al^{IV}, and thereby requiring an additional Ti-substitution mechanism. Potentially, this substitution can be the Ti for 2Mg (type 4) as indicated by the relatively high correlation coefficients (R^2) ranging from 0.695 to 0.884 (not shown).

The relation of Al-content among the host-rock-samples is (taking into account the errors) MO-101>MO-89A~MO-23E or MO-101>MO-67>MO-89A~MO-23E, if the mean values are compared, and remains the same when the biotite-rich crust is examined (Fig. 5). The MO-23E biotites are Mg-richer relative to the others both in the host-rock and in the biotite-rich crust. These observations imply that the biotite composition of the biotite-rich crust depends mainly on that of the host-rock biotites. It is to note that the biotites of the clot do not differ from that of the biotite-rich crusts.

Rock chemistry

In order to examine the possible influence of the enclaves chemical composition to that of the biotites of the biotite-rich crust, major element chemical analyses of two pairs, host-rock (MO-23A and MO-67) and enclave (MO-23E and MO-67A respectively), are listed in Table 3 and compared in Fig. 7b. At the same time, the composition, in terms of mean values, of biotites of the biotite-rich crust is compared with that of the host-rock biotites (Fig. 7a). All comparisons have been made on water free basis. Since biotites have not been analysed for P_2O_5 the host-rocks are not compared in figure 7A for P_2O_5. For sample MO-23, the biotites of the biotite-rich crust are richer in TiO_2 (1.1 or 10%), MnO and CaO (1.3 for both elements) and poorer in Na_2O (0.82) relative to biotites of the host-rock. However, the enclave (MO-23E) is richer in TiO_2 (2 times), MnO and CaO (1.8 for both elements) and poorer in Na_2O (0.56) relative to the host-rock (MO-23A; Fig. 7b). For sample MO-67, the biotites of the biotite-rich crust are richer in FeOt (1.07), MgO (1.05) and MnO (1.46), poorer in K_2O (0.94), CaO (0.57) and Na_2O (0.10), while SiO_2, Al_2O_3 and TiO_2 are the same, relative to biotites of the host-rock (Fig. 7a). However, the enclave (MO-67A) is much more enriched in FeOt (4.40), MgO (5.16), MnO (3.69), as well as in Al_2O_3 (1.27), CaO (1.95), TiO_2 (4.35), and CaO (1.8 for both elements) and poorer in Na_2O (0.54) and SiO_2 (0.74) relative to the host-rock (MO-67).

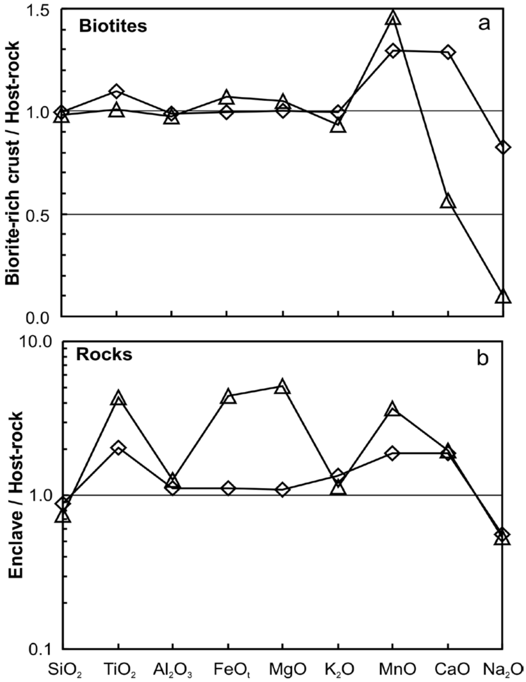

Fig. 7 – Biotite composition of biotite-rich crust over that of host-rock ratio (a); whole-rock major element composition of enclave over that of host-rock ratio (b). Diamonds: sample MO-23; triangles: sample MO-67.

Table 3
Whole-rock major element composition of host-rocks and enclaves

Sample	MO23E	MO23A	Enrichment-depletion factor	MO67A	MO67	Enrichment-depletion factor
Rock-type	Enclave	Host-rock	Enclave/ Host-rock	Enclave	Host-rock	Enclave/ Host-rock
SiO_2	57.46	64.75	0.89	53.12	71.53	0.74
TiO_2	1.67	0.82	2.03	1.25	0.29	4.35
Al_2O_3	16.76	15.25	1.10	18.77	14.84	1.27
FeOt	6.61	5.91	1.12	8.92	2.03	4.40
MgO	2.74	2.52	1.09	4.24	0.82	5.16
MnO	4.82	3.63	1.33	4.83	4.26	1.13
CaO	0.19	0.10	1.89	0.16	0.04	3.69
K_2O	4.63	2.45	1.89	3.16	1.62	1.95
Na_2O	1.79	3.21	0.56	1.80	3.33	0.54
P_2O_5	0.29	0.11		0.34	0.09	
LOI	3.03	1.25		3.40	1.16	
Total	100.00	100.00		100.00	100.00	

Discussion and conclusions

Regarding formation of biotite-rich crusts around xenoliths of different kind of rocks e.g. amphobolites, gneisses shists and basic rocks, Didier (1973) reports that crusts separate xenoliths from the host, points out that they are petrographically very similar to the surmicaceous enclaves, and suggests that these crusts are the result of reaction between ferromagnesian minerals of xenoliths and surrounding granitic magma. A reaction between the enclaves and the granitic melt was suggested also by Montel *et al.* (1991), for the development of such the biotite-rich crusts. A reaction between the granite melt and the mafic material of enclaves was accepted also by Nemchin and Pidgeon (1999) and Long *et al.* (2005). Biotite-crusts surrounding xenoliths have been reported for other rocks also, except granitoids (e.g. Johnson *et al.,* 2003 and Lefebvre *et al.,* 2005), and they were interpreted as the result of reaction with the melt. Based on the above interpretations, the biotite-rich crusts surrounding enclaves can be considered, according to the literature, as the result of reaction between the former and the melt. Concerning the clots, Didier (1973) suggested that during the final stage in the assimilation of xenolithic rocks they disintegrate in the magma. As a result they crumble into little crystal clots which are disseminated in the assimilating magma. Clots are interpreted as restite unmixing products or partially assimilated fragments (xenoliths) of the surrounding country rocks (Drummond *et al.*, 1988; Speer *et al.*, 1989; Fornelli, 1994; Stephens, 2001). They can be also a consequence of the extensive fragmentation of microgranular enclaves (MME), but in this case the clots should be composed of ferromagnesian phases and An-rich plagioclases as well (e.g. Poli and Tommasini, 1991).

Biotite-rich crusts and biotite clots from Monopigadon pluton contain biotite crystals showing not strong differences, when they are compared with those of the host rocks and enclaves. However, some differences do exist concerning their chemical composition, size and colour as well.

The Ti and Al decrease and Si increase in all host-rock biotites towards the crusts (or clot) can be considered remarkable. Comparing (in terms of mean values) the biotites from the biotite-rich crusts with those of the enclaves, it is noticeable that the former are Si-poorer, and Al- and Ti-richer relative to the latter.

The biotite crystals of the biotite-rich crusts and biotite clots are longer than those of the host-rocks and those of the enclaves (except the MO-89A sample). The longest crystals exist in the clot where the biotites are 2.6±1.5 times longer than those of the host rock, while among the biotite-rich crusts of the enclaves the biotites are longer relative to those of the host rock (4 to 12% min. up to 64 to 100% max.). The longest biotite crystals occur in the biotite-rich crust of the cumulate enclave. The length of the biotites from the biotite-rich crusts is also 2 to 4 times longer than those of the enclaves (except the MO-89A sample). The width of the biotite crystals is slightly bigger (10 to 20%, in terms of mean values) in the host-rock biotites compared with those from crusts but not from biotites of the clot where the biotites are 20% wider than those of the host-rock. However, the differences in width seem to be insignificant when the range is taken into account. Both dimensions of biotites from the crusts and clot are interconnected with the corresponding of host-rock biotites. The bigger the biotite in the host-rock the bigger is in the biotite-rich crust. Concerning the colour of biotites, it is reddish-brown to brown and yellow-brown to brown in the host-rocks (BGrd and BGr respectively), differing from that of the biotites consisting the crusts where it is reddish to reddish-brown.

Taking into account the differences, in size and chemistry, existing between the biotites of the biotite-rich crusts and those of the enclaves, an origin from the latter must be excluded. Moreover, the biotite-rich crusts do not grow at the expense of the enclaves but they represent overgrowths. In addition, the differences in chemistry indicate that the formation of these large crystals in solid state due to heating of the biotites of the enclaves is rather impossible. On the other hand, since enclaves were completely solid, we cannot invoke diffusion or matter exchange because these processes are too slow for dimensions of the crusts (a few centimeters). It must be noted also that enclaves are enriched in some elements relative to the host-rocks (e.g. Ti, Fe, Mg, Mn, Ca) but biotites from crusts are not enriched in those elements or they are slightly enriched (Ti).

The euhedral lath-like biotite crystals of the biotite-rich crust and of the clot along with their higher, in respect to the host-rock, length and width (except clot), imply that these crusts and clots were crystallized from the magma simultaneously with the crystallization of the host-rock, when enough space for their growth was available. The mineralogical assemblage of the crusts (or clot) made almost completely by biotites, and given that the biotites were not yet crystallized, implies that magma at the contact with enclaves has a peculiar composition. The part of magma at contact with cold enclave decreases its temperature in a narrow space (some millimetres), hence, at constant lithostatic pressure, solubility of water (fluid in general) increases (Yamashita, 1999, and references therein); therefore water is drew toward the part of magma at contact with cold enclave. This local high content of water stabilizes biotite crystallization.

However, the biotites from crusts and clot are richer in Ti relative to the host-rock biotites. Probably this local Ti-enrichment is the result of the Ti-depletion of host-rock biotites up to a minimum near the crust. Ti was concentrated by diffusion in the magma considering that diffusion has reasonable time extent in magmas, in order to crystallize Ti-richer biotites. K and Mg are not different because they are present already in the magma in enough amounts. Si, even if not statistically robust but with the same behaviour for all samples, is higher where Ti is lower (close to the crust) and it is lower where Ti is higher (in the crust), whereas Al^{IV} decreases from host to crust. These are the results, in all the host-rock biotites, of the substitution of Ti for Mg which is compensated for by substitution of Al^{IV} for Si and this substitution decreases in the biotites of the host-rocks towards the biotite-rich crust or clot. These findings are in agreement with the two proposed substitution mechanisms (1) and (2). It must be noted that the two substitutions act reversely in the host close to crust and in the crust: in the host close to crust Ti and Al^{IV} are depleted, Si and Al^{VI} are enriched whereas in the crust Ti and Al^{IV} are enriched, Si and Al^{VI} are depleted (e.g. samples MO-67A and MO-101; Fig 6A). Accordingly cations R^{2+} should increase, but concomitant substitutions could decrease such cations like dehydrogenation reaction.

Concerning biotites of the clot, they do not differ from that of the biotite-rich crusts. The similar behaviour of biotites from clots with the biotites from crusts around enclaves, implies that clots are clearly pieces of crusts already formed and disrupted. Based on their euhedral crystal shape a restite origin for the clots must be ruled out. Moreover these biotites are longer and wider from the biotites of the host- rock, having the highest difference in length and width. This makes unlikely to be the result of extensive fractionation of microgranular enclaves.

Taking into account that for most of the cases the higher or lower concentration of an element in the biotite of the host-rock results in higher or lower concentration respectively of the element in the biotite-rich crust, it can be concluded that the biotite composition of the biotite-rich crust depends on that of the host-rock biotites and it is independed from the enclave composition. This relation between the compositional characteristics of host-rock and biotite-rich crust biotites is also analogous with their size relation corroborating the findings.

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