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METAMORPHIC EVIDENCES FROM THE MONCHIQUE PLUTON (SOUTH PORTUGAL): CONTACT METAMORPHISM VS REGIONAL METAMORPHISM UNDER VERY LOW-GRADE CONDITIONS

Evidencias metamórficas del plutón de Monchique (S de Portugal): Metamorfismo de contacto vs metamorfismo regional en condiciones de metamorfismo incipiente

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Abstract: *During the Cretaceous, the Monchique pluton was emplaced in metapelite and greywacke host rocks, producing a thermal aureole with hornfels containing cordierite, andalusite and biotite. Apparently, thermal effects persist more than 3 km from this syenitic igneous body, evidenced by vitrinite reflectance values higher than 4% and natural coke with a mosaic optical texture. Nevertheless, the host rocks not transformed to hornfels show the same textural, chemical, mineralogical and crystal-chemical characteristics as the equivalent clastic rocks from the distant Brejeira and Mira Formations. This difference between the reaction progress of inorganic and organic materials can be related to the slow reaction rates of the clay minerals and the absence of pervasive tectonic effects during the thermal processes, which are one of the most important driving forces of their reaction progress.*

Key words: *Clay minerals, electron microscopy (SEM and HRTEM), Kübler Index (KI), organic maturation, metapelites, XRD.*

Resumen: *Durante el Cretácico tuvo lugar la intrusión del plutón de Monchique en rocas clásticas (pelitas y areniscas), afectadas por un metamorfismo regional incipiente, en la Zona Sur Portuguesa del Macizo Ibérico. La intrusión produjo una aureola térmica con corneanas en el contacto, en las que se han identificado cordierita, andalucita y biotita. Aparentemente, los efectos térmicos persisten a más de 3 km del plutón sienítico como pone de manifiesto la reflectancia de la vitrinita con valores > 4% y la presencia de coque natural con estructura en mosaico. Sin embargo, las rocas clásticas próximas al contacto, pero no transformadas a corneanas, muestran las mismas características texturales, químicas, mineralógicas y cristalquímicas que los materiales equivalentes, pero más distantes, de las formaciones Brejeira y Mira. Esta diferencia entre el progreso de la reacción de los componentes inorgánicos y orgánicos debe de estar relacionada con la baja velocidad de reacción de los minerales de la arcilla. Además, la ausencia de episodios tectónicos intensos durante los procesos meramente térmicos pudo privar a los minerales de la arcilla de una de las fuerzas motrices fundamentales para el progreso de su reacción.*

Palabras clave: *Minerales de la arcilla, microscopía electrónica (SEM y HRTEM), Índice de Kübler (KI), maduración orgánica, metapelitas, XRD.*

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The patterns of metapelitic zones in very low-grade metamorphic terranes are governed by stratigraphy, structure, depth of burial and geothermal gradient. Merriman and Frey (1999) illustrated some typical patterns that have been well-characterised by a variety of techniques and suggested that specific patterns could be related to geotectonic settings.

Nowadays, the thermal implications of the illitization reaction are well documented from burial-diagenetic sequences. In addition, information on clay mineralogical trends in the vicinity of contact metamorphic environments is becoming more popular because it is a setting relevant to predicting long-term reactions in nuclear waste repository sites (Kemp et al., 2005; Techer et al., 2001; Techer et al., 2006).

Some of the major differences between burial diagenetic and contact metamorphic processes are the heating rate, the duration, the role of fluids and K activity, and the magnitude and origin of rock stress. Terranes that undergo high heat flows are usually characterized by the development of thermal aureoles that are broadly concentric or parallel to the margins of the intrusive body (Kerrick, 1991). Furthermore, burial environments are more subjected to tectonic strains caused by the loading of sediment or the movement of crustal rocks whereas contact metamorphic rocks may also suffer rock stresses due to the intrusion of magma, which usually promote initial dehydration, cracking and hydrofracturing in the host materials. Depending on the timing in relation to orogenic movements, also the regional stresses of tectonic events can affect to the aureole. However, although there has been some research describing the effects of intrusions on clay minerals (e.g. Nadeau and Reynolds, 1981; Smart and Clayton, 1985; Aaron and Lee, 1986; Kisch, 1987; Roberts et al., 1990; Kemp et al., 2005; among others), conclusions about factors controlling thermal evolution are rare and it clearly remains a subject open to debate.

Many model reaction sequences of metapelitic aureoles are assumed to begin with the appearance of biotite, cordierite and/or andalusite (e.g. Pattison and Tracy, 1991). However, when the outer low-temperature zones (150–350º C) of aureoles are characterized by detailed X-ray diffraction and, even, electron microscopy studies, recording phyllosilicate assemblages and their crystallinity, cryptic low-temperature aureoles are detected where regional very low-grade rocks are intruded by late plutons (Merriman and Frey, 1999). According to Bühmann (1992), the effect of igneous intrusions on sedimentary rocks depends primarily on the thickness of the intrusive body. The distance over which changes take place may vary considerably, however; in fact, some sills cause no metamorphism at all due to their relative thinness (Rowsell and De Swardt, 1976). But although being important the thickness of the intrusion, there are other important parameters as the temperature of the melt and host rocks, the fracture network of the country rock, and the fluids.

In the South Portuguese Zone, a very low-grade regional metamorphic evolution occurs from SW to NE along flysch sequences containing an igneous intrusion, the Monchique pluton (Abad et al., 2001). The aim of this research is to determine the thermal effect of the intrusion on the host metapelites. The emplacement in the diagenetic area of the South Portuguese Zone sequences opens the possibility to studying the effect of thermal metamorphism in an area relatively free of other metamorphic effects. Samples collected in the surroundings of Monchique pluton (Fig. 1) have been studied by X-ray diffraction, optical microscopy, scanning electron microscopy and high-resolution transmission electron microscopy. Vitrinite reflectance measurements were also conducted on several samples containing dispersed organic matter to quantify potential temperature-related trends via levels of organic maturity.

The correlation between clay mineral maturity and organic thermal maturity indicators follow a simple trend when the subsidence of a sedimentary basin is gradual and with a normal geothermal gradient. However, these two types of shallow crustal materials mature at different rates probably due to the different nature of their reaction progress (Kisch, 1987; Robert, 1988; Ferreiro Mählmann et al., 2012). This contribution explores the clays behaviour in the surrounding of a pluton to infer aspects related to the thermal evolution in absence of other driving-forces.

Geological overview and materials

The Monchique massif crops out in the western part of the Algarve region, in Southern Portugal (Fig. 1a). The massif is elliptical, about 5 km wide by 15 km long. Its longest axis runs E-W, and it is divided into two topographic sectors, Picota in the east and Foia in the west. The Monchique pluton comprises a mainly syenitic complex of 63 km^2 associated with a further range of dykes, both internal and external to the complex, with approximately 5% igneous breccias. The isotopic study by Abranches and Canilho (1981) indicates an upper Cretaceous age (64.7±13.6 Ma), which is in accordance to more recent isotopic datations by Grange et al. (2010). The emplacement into the upper crustal level was favoured by a set of faults striking in the same direction, according to Valadares and González-Clavijo (2004). Previously, the origin of the Serra de Monchique complex had been related to lithospheric extension and North Atlantic passive margin formation without any evidence of the involvement of a mantle plume (Rock, 1982; Bernard-Griffiths et al., 1997). Recently, Geldmacher et al. (2005) have proposed a genetic link between the Serra de Monchique magmatism and that of the Madeira volcanic province, relating both of them to the activity of a mantle plume anchored in the mantle Transition Zone, whose track is represented by the SW–NE alignment of Madeira, Mt. Ormonde and Serra de Monchique. According to Grange et al. (2010) the resulting ages from north to south of the three major alkaline massifs along the Atlantic coast of Portugal (Monchique, Sines, Sintra) increasing northward are in agreement with the motion of the Iberian plate between 88 and 60 Ma above the mantle plume that has caused magmatism since the Cretaceous. In addition, Grange et al. (2010) suggest

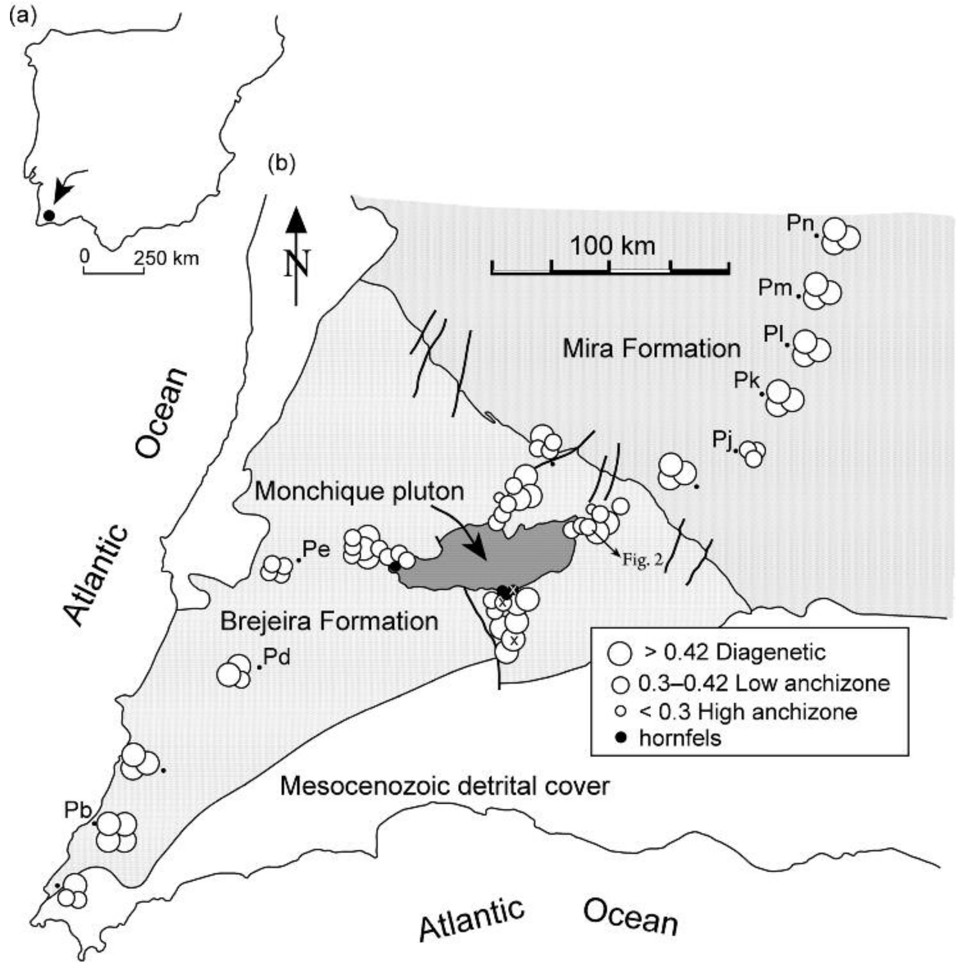

Fig. 1.- (a) Sketch of the Iberian Peninsula showing the location of the Monchique massif, (b) geological map of the southwestern part of the South Portuguese Zone (adapted from Oliveira, 1990; and Quesada, 1991) indicating sample locations and the Kübler Index values. The scale is enlarged for the samples nearest the pluton. x: samples selected for TEM study. Pb, Pd, Pe, Pj, Pk, Pl, Pm, Pn are labels of samples further from the pluton.

that the emplacement of the alkaline massifs occurred along major lithospheric discontinuities, Messejana and Nazare faults, based on the location, close to such structures, of these magmatic complexes.

The syenitic rocks of the Monchique complex consist of feldespars (microcline, albite and/or oligoclase), nepheline, sodalite, augite, aegirine-augite, biotite, sphene, apatite, zircon, muscovite, hornblende and secondary minerals such as cancrinite, analcime and calcite (Valadares and González-Clavijo, 2004). There is a close relationship between the nepheline content and the granularity of the rock, since the coarse-grained syenite has more nepheline than the fine-grained types (Canilho et al., 1978; Rock, 1978).

The host rocks are metapelites and greywackes of marine origin belonging to the Baixo Alentejo Flysch Group, specifically to the Brejeira Formation, which is a turbiditic sequence of heterogeneous lithology, mainly comprising dark sandstones and shales. The sandstones are mostly quartzitic and contain minor quantities of volcanic material and feldspars. The shales are decimeter-thick, greyish to blueish, and contain plant debris. Palinomorphs (miospores) and ammonoid fauna (goniatites) indicate ages ranging from Middle Namurian near the contact with the Mira Formation, to Lower Westphalian (Jorge et al., 2013). Abad et al. (2001, 2002) described the metamorphic evolution of the South Portuguese Zone of the Iberian Variscan Belt, based on the metapelitic and metasammitic rocks. They found diagenetic conditions for the Brejeira and Mira Formations, which form the SW end of the South Portuguese Zone. There is a zone of hornfels at the contact with the igneous massif, where andalusite and cordierite have been reported (Shaw Rock, 1983).

Experimental procedure

Sample collection

This research is based on the study of samples collected along four cross-sections in the surroundings of the Monchique intrusion (Fig. 1b), although there are only a few good exposures of the contact between the Monchique massif and the metapelitic host rocks. The southern cross-section begins close to Caldas de Monchique and extends 3.6 km along the P. Lagos-Monchique road. The last sample point, S-9, was the closest to the contact (<50 m). The western cross-section, close to Marmelete, covers around 2 km. In the northern cross-section, covering more than 5 km, as well as metapelitic rocks, igneous breccias composed of aegirine, albite and andalusite have been identified. Finally, in the eastern cross-section the metapelites collected are more affected by weathering (Fig. 2). Efforts were taken to obtain fresh material from the best exposures, but in general all these samples are dark grey or brown due to the presence of carbonaceous matter and/or Fe oxides.

X-ray diffraction

Seventy-five samples were studied by X-ray diffraction (XRD) to determine the mineral assemblages and the main crystal-chemical parameters of dioctahedral mica. The research was carried out using a Philips PW 1710 powder diffractometer with Cu-Kα radiation, graphite monochromator and automatic divergence slit. The <2 μm fraction was separated by centrifugation. Orientated aggregates were prepared by sedimentation onto glass slides. Dimethyl-sulphoxide treatments were carried out on some samples to corroborate the identification of kaolinite. Preparation of samples and experimental conditions for the Kübler Index of illite "crystallinity" (hereafter KI according to the AIPEA recommendation, in Guggenheim et al., 2002) measurements were carried out according to IGCP 294 IC Working Group recommendations (Kisch, 1991). Our KI measurements (y) were transformed into C.I.S. values (x) according to the equation y= 0.674x + 0.052 (r= 0.999), obtained in our laboratory using the international standards of Warr and Rice (1994). The KI was measured on both 10 Å and 5 Å peaks to check the effects of adjacent peaks (Nieto and Sánchez Navas, 1994). The b cell-parameter of micas was obtained from the (060) peak measured on slices of rock cut normal to the main fabric of the samples. For the spacing measurements, quartz was used as an internal standard.

Scanning electron microscopy and high-resolution transmission electron microscopy

Following XRD and optical microscopy studies, carbon-coated polished samples were examined by scanning electron microscopy (SEM), using back-scattered electron (BSE) imaging and energy-dispersive X-ray (EDX) analysis to obtain textural and chemical information. These observations were performed using a Zeiss DSM 950 SEM, equipped with an X-ray Link Analytical QX-20 energy-dispersive system. The phyllosilicates were analysed by SEM with an accelerating voltage of 20 kV, a beam current of 1-2 nA and counting time of 100 s, using both natural and synthetic standards: albite (Na), periclase (Mg), wollastonite (Si and Ca), orthoclase (K) and synthetic Al_2O_3 (Al), Fe_2O_3 (Fe) and $MnTiO_3$ (Ti and Mn).

The structural formulae of micas were calculated on the basis of 22 negative charges $O_{10}(OH)_2$ and the formulae of chlorites, on the basis of 28 negative charges $O_{10}(OH)_8$. Due to the very fine-grained nature of these samples, the results obtained using EDX on occasion had to be rejected because of contamination. According to Guidotti et al. (1994), it is assumed that 75% of the Fe in the micas is Fe^{3+}.

After BSE examination, three specimens from the southern cross-section were prepared for high resolution transmission electron microscopy (HRTEM) study. TEM observations were obtained with a Philips CM20 (STEM) equipped with an EDAX solid-state EDX detector, operat-

Fig. 2.- Field view of the E cross-section. See its location in Fig. 1b.

ing at 200 kV, with a LaB_6 filament and a spatial resolution of 2.7 Å between points and 1.4 Å between fringes. Both electron microscopes are located at the Centro de Instrumentación Científica (C.I.C.) of the Universidad de Granada.

Vitrinite reflectance

Vitrinite reflectance is the most widely used indicator of thermal maturity in sedimentary rocks containing disseminated organic particles (Ernst and Ferreiro-Mählmann, 2004). Some selected samples from the southern cross-section and some metapelites previously described by Abad et al. (2001) in distant areas of the Brejeira and Mira Formations were studied to check the possible temperature evolution in relation to the distance from the pluton. Samples were crushed to <1 mm, mounted in epoxy resin, cut and polished for petrographic analyses at the Instituto Nacional del Carbón (INCAR-CSIC, Oviedo, Spain). The reflectance analyses require a beam of normal incident white light; the percentage of reflected light from the surface of polished vitrinite is a function of the maturity of the maceral. All organic particles found in each microscopic section of 4x4 cm^2 were measured due to their rarity and dispersion (see Table 3). According to the ISO 7404/5 rule, the final values correspond to the average (mean random vitrinite reflectance = %Rr).

Results

Mineralogy and textural features

XRD data (Table 1) and BSE images (Fig. 3) show that the metapelitic samples comprise a phyllosilicate fine-grained matrix (<5µm) and a coarse fraction, which is assumed to be detrital in origin. This coarse fraction is composed of xenomorphic quartz, alkaline feldspars and phyllosilicates (Fig. 3a-b). No significant differences have been found among the four cross-sections. Quartz and white K-mica are the main phases in all the samples. Illite is the predominant constituent of the <2 µm fraction and significant amounts of kaolinite, intermediate Na-K mica and chlorite were also detected in samples from the different cross-sections. Accessory minerals include iron oxyhydroxides such as goethite, hematite and ilmenite, titanium oxides, ilmenorutile and large euhedral tourmaline. Kaolinite/mica and chlorite/mica stacks <50 µm in length and <20 µm in width are disseminated in the matrix. There is no evidence of a strain-fabric in most of the samples and the orientation of the coarse fraction seems to be in accordance with the bedding (S_0).

The TEM study in two of the very low-grade metapelitic samples located in the S cross-section confirms those textural and mineral observations. In addition, it has been observed that, at the lattice scale, both samples pre-

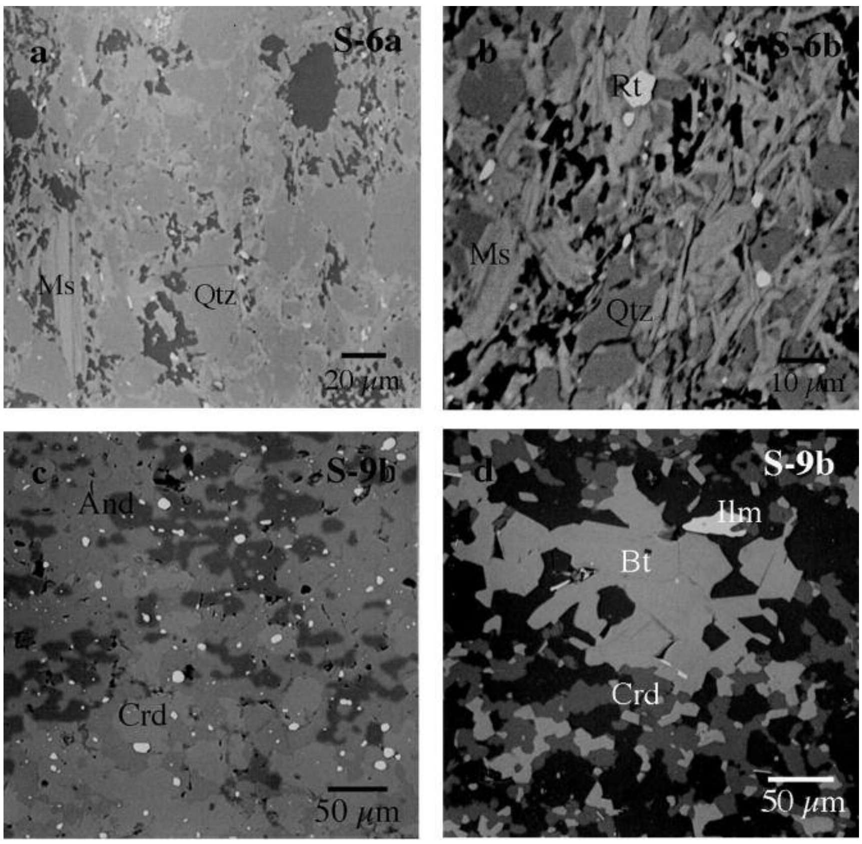

Fig. 3.- (a-b) BSE images showing the textural aspect of the metapelitic rocks near the pluton: a fine-grained matrix and a coarse fraction, with no evidence of strain-fabrics (Qtz: quartz, Ms: muscovite, Rt: rutile); (c-d) BSE images corresponding to the hornfels showing their typical assemblage with cordierite (Crd), andalusite (And) and large biotite crystals (Bt); ilmenite (Ilm).

White mica						Mineral composition
Samples	d(001) Å		**b** (Å)	KI (Δ°2θ)		Qz, K-rich mica, feldspars (all the samples)
	<2μm	bulk		<2μm	bulk	
S-1a	10.007	9.987	8.980	0.49	0,32	Chl, Na-K mica, Fe-oxides
S-2a	10.008	9.991	-	0.50	0.38	Na-K mica, Sme, Kln, Gth, Rt
S-3a	9.987	9.992	-	0.49	0.40	Kln, Tur
S-4a	9.984	9.992	-	0.46	0.34	Kln, Gth, Hem, Na-K mica
S-5a	9.982	9.999	8.987	0.56	0.31	Kln
S-6a	10.003	10.006	8.988	0.31	0.22	Kln, Rt, Ilm-Rt, La, Ce phosphates
S-6b	9.981	9.991 (9.808)	8.988	0.46	0.28	Na-K mica, Kln
S-6c	9.996	-	8.994	0.35	0.22	Fe-oxides
S-8a	9.997	9.993	8.987	0.43	0.38	Chl, Kln, Gth
S-9a	-	10.017	-	0.41	0.26	Bt, Crd, Na-Cafs
S-9c	-	10.028	-	0.41	0.22	Bt, And, Crd, Chl, Ilm, Tur, Na-Cafs, Ilm-Rt, REE phosphates
S-9f	-	10.027	-	-	-	Bt, Crd, And, Pl
Pf-20	-	10.008	8.986	0.40	0.28	Na-K mica, Kln
Pf-21	10.007	10.004	8.985	0.40	0.28	Na-K mica, Kln
Pf-22	10.016	10.003	8.988	0.32	0.25	Na-K mica, Kln
W-1a	10.002	10.017	8.987	0.58	0.37	Na-K mica, Kln
W-2a	10.001	10.000	8.983	0.56	0.37	Na-K mica, Kln, Gth
W-2b	10.003	10.000	-	0.50	0.59	Kln, Gth
W-3a	10.009	10.009	-	0.41	0.25	Na-K mica, Kln, Gth, Hem
W-4z	-	9.976	-	0.26	-	Kln, Gth
W-5a	-	-	-	0.37	-	Bt, Crd, Chl
W-7b	-	9.990	-	0.34	0.28	Na-K mica, Kln, Fe-oxides, Tur, Ap
W-8a	9.977	-	-	-	0.49	Na-K mica, Kln, Gth, Trm
W-9a	10.005	-	8.986	0.61	0.49	Chl, Ilm
N-1b	10.008	10.012	8.987	0.62	0.47	Na-K mica, Kln, Gth
N-2a	10.036	10.019	8.997	0.41	0.31	Kln, Gth
N-3a	10.014	-	9.005	0.46	0.40	Kln, Na-K mica, Gth
N-3b	10.004	9.999	-	0.65	0.41	Na-K mica
N-4a	10.016	10.006	-	0.43	0.35	Kln
N-5z	-	10.065	-	-	0.28	Kln, Gth, Hem
Pg-25	9.993	9.993	-	0.41	0.31	Kln
Pg-26	10.006	10.006	8.987	0.31	0.22	Kln, Na-K mica
Pg-27	9.984	10.005	8.989	0.28	0.25	Kln, Pg
N-7a	-	-	-	0.37	0.31	Kln, Fe-oxides
N-10c	10.000	-	8.995	0.38	0.32	Kln, Fe-oxides
E-1a	10.004	-	9.000	0.41	0.32	Kln, Chl
E-2a	10.003	10.003	8.999	-	0.25	Chl
E-3a	9.996	9.993	9.001	0.29	0.20	Chl
E-3c	9.999	9.997	8.988	0.44	0.23	Kln, Chl, Tur, Gth
E-3e	-	-	-	0.32	0.35	Chl, Grt, Fe-oxides
E-4e	-	-	8.986	0.44	-	Kln
E-4f	-	-	-	0.37	0.29	Chl, Kln, And, Ilm
E-4g	-	9.993	8.990	0.34	0.19	Kln
E-9a	-	-		0.31	0.20	Kln

Table 1.- Crystal-chemical parameters of mica and bulk mineralogy of samples studied by XRD and SEM.
The label capital letter indicates the S, W, N and E cross-sections, except for the Pf and Pg which belong to the previous metapelites sampling (Abad et al., 2001). The label number indicates the proximity to the pluton, high numbers mean closer. The label small letter indicates different rocks in the same sample place. In bold, samples studied by TEM. Mineral abbreviations according to Whitney and Evans (2010).

sent dioctahedral micas characterised by well-defined, crystalline and defect-free packets, 100–1500 Å in thickness and with parallel and sub-parallel grain boundaries, sometimes with an intense mottled texture (Figs. 4, 5). Different polytypes have been found in the two samples, such as 1Md and 2M, and we have recognised superperiodicities of up to 70 Å in the general reflections of the selected area electron diffraction (SAED) pattern, revealing a long-range stacking order (Fig. 4, inset). The presence of 1Md and 2M polytypes in the studied samples is common in these kinds of rocks. The well-defined, and defect-free packets of mica observed at nanometer scale are characteristic of an authigenic origin of the phyllosilicate matrix of clastic rocks from late diagenesis to low epizone conditions. Sample S-6a with kaolinite, which is quickly damaged under the electron beam, shows a parallel alternation of dioctahedral mica and kaolinite packets (Fig. 6). The chlorite packets are frequently damaged and have an altered and poor crystalline aspect.

SEM data of the hornfels show an assemblage based on biotite, cordierite, andalusite, chlorite, quartz and Na-Ca feldspars. The grain size is larger (up to 100 μm) than in

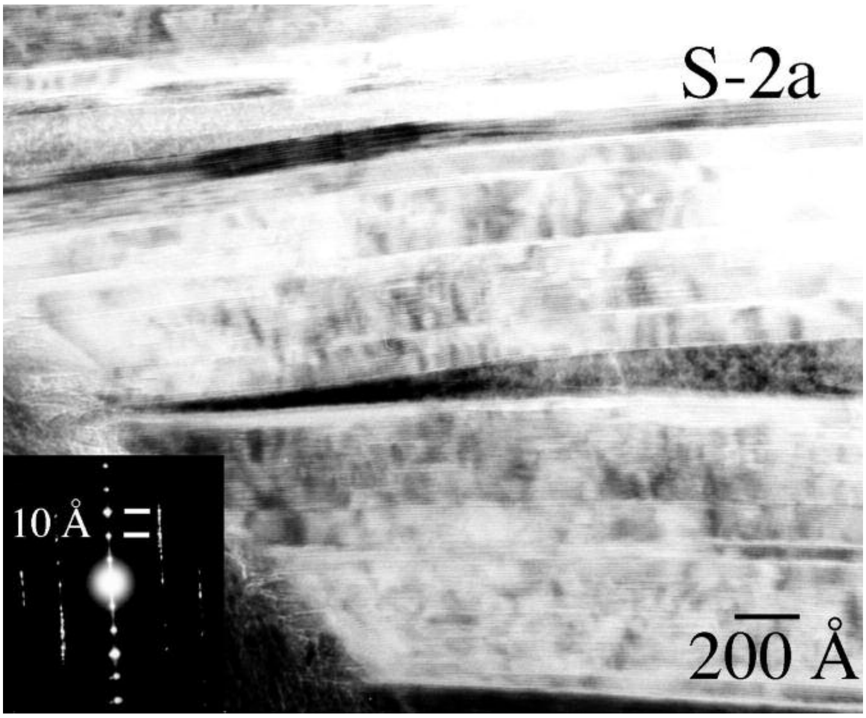

Fig. 4.- Lattice-fringe image of Na-K mica packets, defect-free and parallel. Mica reflections are present in the SAED (inset) with a superperiodicity of 70 Å in the general reflections.

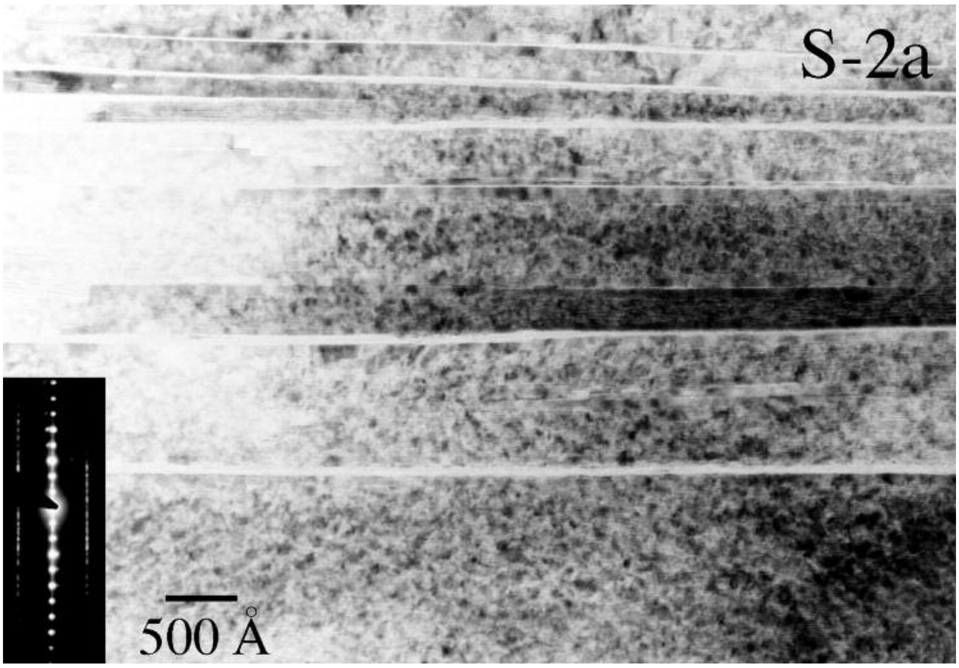

Fig. 5.- Lattice-fringe image showing mica crystals of variable thickness, with mottled texture, and very narrow and elongated fissures. SAED pattern shows very sharp 00l reflections.

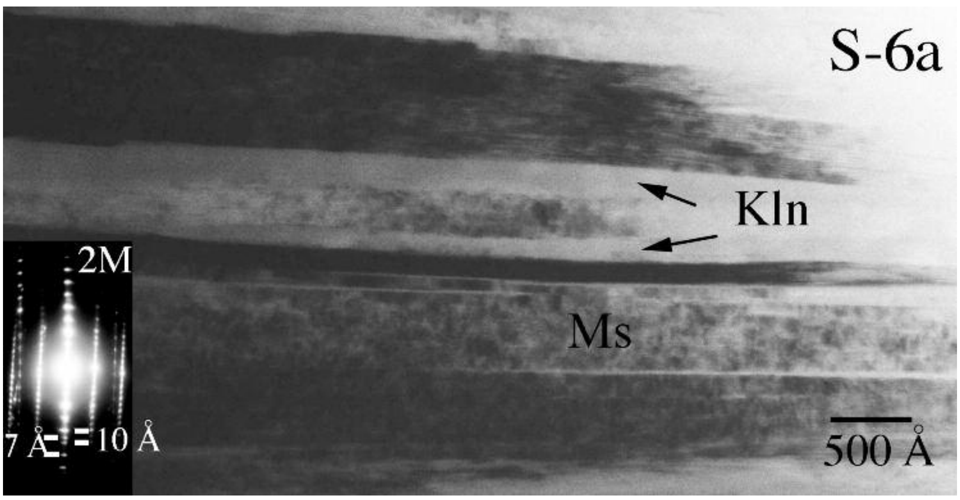

Fig. 6.- Alternation of mica (Ms, dark contrast) and kaolinite packets (Kln, light contrast). Beam damage is obvious in the kaolinite packets. The SAED reveals the reflections of both phases. The 2M polytype for mica have been established in the general reflections (20 Å).

the very low-grade metapelites (Fig. 3c-d). Under the SEM, the hornfels reveal a very different texture from that of the metapelites, more crystalline and with well-defined crystals of phyllosilicates; however, the TEM study of the fine fraction area shows that at the lattice scale these textural differences, in relation to dioctahedral K-mica, are not as evident. Mineral identification by TEM confirms the SEM and XRD data presented in figures 3c-d and Table 1.

Crystal-chemical parameters

Table 1 also shows the white-mica crystal-chemical parameters. The basal spacing of mica is around 10.002 Å (s= 0.01) and an overall average value of 8.990 Å (s= 0.01) was obtained for the b unit-cell parameter. Figure 1b displays illite crystallinity (KI) values in relation to the location of the samples. The map includes not only the data corresponding to the samples from the pluton surroundings, but also some of the results of the metapelites previously described by Abad et al. (2001) in distant areas of the Brejeira and Mira Formations. The limits of the anchizone, the zone of incipient or very low-grade metamorphism, are defined on the KI scale at 0.42 $\Delta^{o}2\theta$ CuKα for the diagenesis/anchizone boundary, and at 0.25 $\Delta^{o}2\theta$ CuKα for the anchizone/epizone boundary (Kübler, 1968). According to these ranges, the irregular distribution of the KI values (Table 1) indicates diagenetic-low anchizone conditions and locally high anchizone, in concordance with previous studies of these flysch sequences (Abad et al., 2001, 2002). The KI values of the bulk fractions are lower than the KI values of the <2 μm fractions, but there is a parallel behaviour between them, suggesting the presence of detrital mica. The highest KI values are coincident with the samples that contain intermediate Na-K mica or paragonite due to the superposition of their (001) peaks on the 10 Å one, which invalidates the use of their KI data.

Chemical compositions of phyllosilicates

The chemical composition of the dioctahedral micas from samples surrounding the pluton were determined by EDX under SEM (Table 2). To determine whether emplacement of the pluton in any way altered the chemical

Samples	Si	Al^{IV}	Al^{VI}	Fe	Mg	Ti	∑ oct.	K	Na	∑ inter.
S-2a/1	3.19	0.81	1.91	0.08	0.06	0.01	2.06	0.61	0.13	0.74
S-3a/1	3.23	0.77	1.62	0.25	0.17	0.05	2.08	0.85	0.06	0.91
S-3a/3	3.36	0.64	1.73	0.10	0.17	0.02	2.02	0.76	0.05	0.81
S-5b/2	3.44	0.56	1.87	0.04	0.02	0.02	1.96	0.66	0.05	0.71
S-6a/5	3.25	0.75	1.81	0.06	0.15	0.01	2.04	0.83	0.00	0.83
S-6a/7	3.11	0.89	1.80	0.08	0.09	0.05	2.02	0.88	0.06	0.94
S-6b/1	3.10	0.90	1.91	0.02	0.00	0.04	1.97	0.89	0.08	0.97
S-8a/10	3.14	0.86	1.72	0.26	0.12	0.01	2.11	0.83	0.07	0.90
S-9a0/3*	3.07	0.93	1.77	0.19	0.12	0.02	2.09	0.88	0.06	0.94
S-9c/13*	3.10	0.90	1.76	0.20	0.10	0.02	2.09	0.86	0.07	0.92
S-9f/7*	3.11	0.89	1.85	0.10	0.08	0.01	2.04	0.89	0.04	0.93
W-1a/3	3.11	0.89	1.94	0.06	0.00	0.02	2.03	0.76	0.09	0.85
W-3b/2	3.30	0.70	1.78	0.13	0.11	0.02	2.04	0.74	0.05	0.80
W-3b/3	3.12	0.88	1.81	0.25	0.03	0.02	2.11	0.65	0.17	0.82
W-7b/4	3.08	0.92	1.77	0.19	0.11	0.02	2.09	0.85	0.06	0.91
W-9a/1	3.15	0.85	1.79	0.15	0.13	0.01	2.09	0.85	0.01	0.86
W-9a/17	3.34	0.66	1.92	0.06	0.05	0.00	2.03	0.66	0.03	0.69
N-1a/1	3.20	0.80	1.72	0.16	0.11	0.04	2.04	0.88	0.04	0.92
N-1b/7	3.11	0.89	1.98	0.10	0.02	0.01	2.10	0.60	0.09	0.69
N-4a/6	3.35	0.65	1.77	0.07	0.09	0.04	1.98	0.72	0.11	0.83
N-8a/2	3.09	0.91	1.86	0.15	0.00	0.03	2.03	0.88	0.04	0.92
N-8a/5	3.23	0.77	1.81	0.16	0.04	0.02	2.03	0.83	0.04	0.87
N-9a/6	3.42	0.58	1.80	0.10	0.07	0.01	1.98	0.71	0.09	0.80
N-9a/14	3.15	0.85	1.81	0.17	0.09	0.01	2.08	0.80	0.05	0.84
E-1a/8	3.34	0.66	1.81	0.11	0.11	0.01	2.04	0.74	0.00	0.74
E-2a/1	3.17	0.83	1.82	0.17	0.09	0.01	2.09	0.71	0.10	0.81
E-2a/4	3.08	0.92	1.87	0.12	0.07	0.01	2.07	0.67	0.20	0.88
E-2b/3	3.22	0.78	1.80	0.11	0.13	0.02	2.05	0.74	0.09	0.83
E-3c/6	3.10	0.90	1.89	0.06	0.05	0.03	2.03	0.82	0.06	0.88
E-4f/6	3.28	0.72	1.91	0.06	0.05	0.00	2.03	0.68	0.07	0.75
E-9a/4	3.08	0.92	1.97	0.04	0.00	0.00	2.01	0.86	0.06	0.92
E-9a/7	3.07	0.93	1.94	0.02	0.01	0.01	1.99	0.90	0.10	1.00
E-9a/8	2.96	1.04	1.97	0.04	0.01	0.00	2.02	0.95	0.07	1.02

Table 2.- Representative chemical compositions for K-rich dioctahedral micas normalized to $O_{10}(OH)_2$.
* indicates mica analyses from the hornfels.

composition of phyllosilicates, these data were compared with the mica analyses of the Brejeira Formation (Abad et al., 2002). The main feature is the high heterogeneity of the data, which combine variable degrees of illitic, phengitic, paragonitic and ferrimuscovitic substitutions in addition to some quite pure muscovite analyses. These findings are equivalent to compositions described by Abad et al. (2002) for samples from the Brejeira Formation. Figure 7 shows that the scatter and the range for the different elements in the dioctahedral mica analyses of these rocks are very similar even quite near to the boundary of the intrusion. Ca and Mn contents are always below 0.01 a.f.u.

It is possible to distinguish several different textural types of dioctahedral mica, that is, in the matrix, with a predominantly illitic character, and as detrital-like grains with more muscovitic composition. Micas in the hornfels include dioctahedral and trioctahedral compositions. The dioctahedral micas are close to ideal muscovite, with a low illitic component (Table 2, analyses marked with *). The trioctahedral micas are Fe-rich biotites.

Chlorites are trioctahedral with a high Fe/(Fe+Mg) ratio (0.41–0.90) corresponding to a chamosite variety.

Vitrinite reflectance

Vitrinite reflectance of dispersed organic particles is particularly useful for estimating maximum temperatures that sedimentary rocks have experienced. Although nineteen samples were prepared, finally organic matter for the measurements was found only in fourteen of them. Table 3 shows the %Rr values obtained from metapelitic rocks from the southern cross-section. With one exception, Rr values are higher than 4.0%. According to Bostick (1979), these Rr values, considering a minimum heating interval of 0.2 My, indicate temperatures higher than 300 ºC. Another value corroborating these temperatures in the surroundings of the intrusion (<3 km) is the presence of natural coke with a mosaic texture. The Rr range for the metapelites located further from the pluton (see their location in Fig. 1b) is larger, with one value lower than 3.0 % and two samples more than 30 km away from the pluton outcrop with a Rr higher than 4.0% (Table 3). In any case, the Rr results indicate meta-anthracite and anthracite conditions for the entire region.

Samples	N	distance from the pluton (km)	%Rr
S-1a	21	3.6	4.22
S-2a	15	2.2	3.60
S-3a	4	0.9	4.14
S-5a	21	0.6	5.41
S-6a	2	0.45	4.92
Pe-19	81	6.5	3.77
Pd-15	35	14	2.70
Pj-37	166	14	4.65
Pk-43	80	19	4.17
Pl-44	38	23	3.57
Pm-48	13	28	3.42
Pn-52	35	33	3.16
Pb-5	55	33	4.52
Po-58	88	37	4.06

Table 3.- Vitrinite reflectance data of selected samples. N is the number of organic particles measured.

Discussion

Patterns of contact metamorphism are generated in the vicinity of igneous intrusions, typically as aureoles. According to Merriman and Frey (1999), the aureoles can be divided, in terms of their metamorphic history, into those that predate and those that postdate the regional metamorphism. In this case, the thermal aureole developed after the Hercynian orogeny, which produced a very low-grade

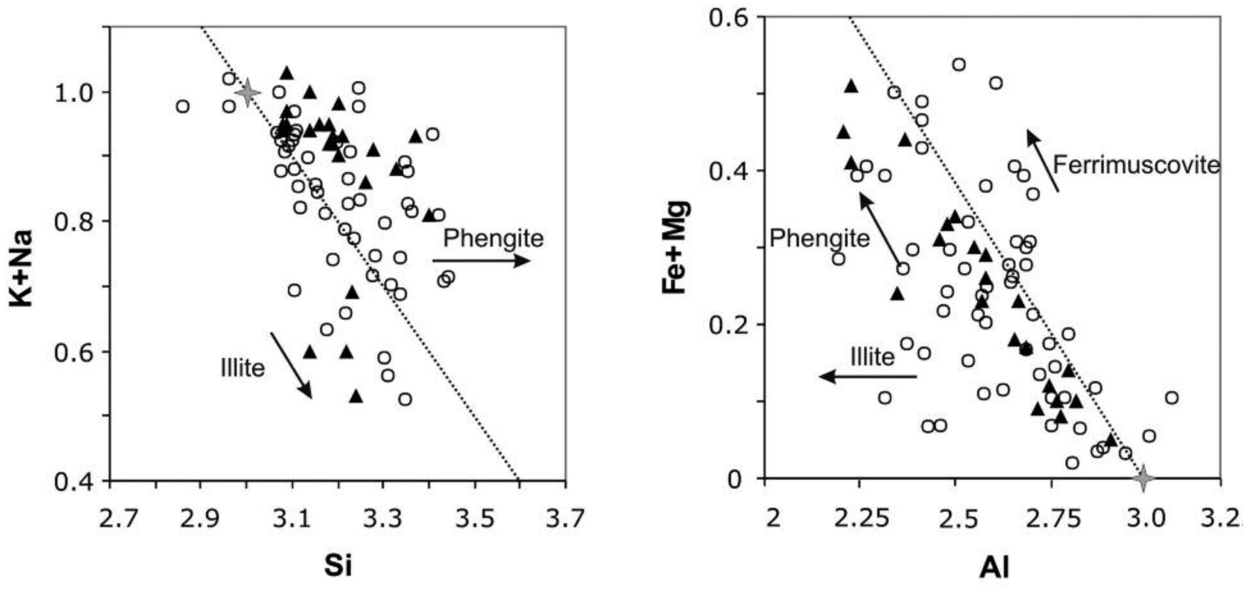

Fig. 7.- Chemical composition diagrams of dioctahedral micas from the metapelitic rocks sampled in the surroundings of the pluton (triangles) compared with dioctahedral micas from the Brejeira Formation, >3 km from the pluton (circles, taken from Abad et al., 2002). Stars and lines respectively indicate the theoretical muscovite position and corresponding exchange vectors.

metamorphism in these terranes. The pattern therefore overprints and should be discordant on earlier regional metamorphism.

Crystal-chemical data for the metapelites close to the pluton coincide with the data reported in Abad et al. (2001) for the metapelites of the Brejeira and Mira Formations. In particular, a) the b cell-parameter of micas indicates, according to Guidotti and Sassi (1986), low-pressure conditions; b) the thermal effect on the KI values, which vary over a range of 0.3–0.6º Δº2θ, is diffuse and does not show a concentric pattern of isocrysts such as those which have been described in other localities (Roberts et al., 1990; British Geological Survey, 1992, 1993a, b, 1997; Hirons et al., 1997). Only sporadic KI values are lower than the general tendency previously described in the Brejeira and Mira Formations (see Fig. 1b). This seems to indicate that these parameters have not been sensitive to the thermal effect of the intrusion.

The mineral assemblage including quartz, white micas (K and intermediate Na-K mica), kaolinite and feldspars, described within 100 m of the contact, is the same as the metapelitic rocks studied in both the Brejeira and Mira Formations in previous researches (Abad et al., 2001 and 2002). Moreover, the chemical compositions of the phyllosilicates show similar ranges and scattering for the different exchange vectors even very near the contact (Fig. 7). This chemical similarity is the reason for the lack of changes in d_{001} and b parameters, which are, in fact, determined by the chemical composition of micas. Focusing on the hornfels, the chemical characterisation of the white micas has shown that the compositions are muscovitic by contrast to the illitic character of the micas in the very low-grade metapelitic rocks.

In relation to the TEM data, at lattice-fringe level the textural aspect of the micas is very similar to the micas described in diagenetic metapelites (e.g. Fig. 5 in Abad et al., 2001) and sandstones (e.g. Fig. 4 in Abad et al., 2002) of the Brejeira Formation. Specifically, crystallite thicknesses of micas measured directly on lattice-fringe images of the selected samples give values in the range to those of the metapelites far from the pluton (Fig. 8 in Abad et al., 2001) which present an average value of 850 Å.

Therefore, only the vitrinite Rr values indicate small differences in relation to the regional rocks (Table 3), with an average value %Rr = 4.5 in a range of 3.6–5.4. The metapelites situated far from the intrusion present for the same parameter a range from 2.7 to 4.7 with an average value equal to 3.8. This is the only data indicating that the low-temperature metapelitic aureole is more extensive than the mappable hornfelsing. This is commonly based on the first appearance of biotite, in this case less than 50 m from the intrusion contact.

The temperature suggested by the KI data and kaolinite persistence (< 200º C) is lower for all the Brejeira and Mira Formations than that indicated by vitrinite reflectance (>300º C), with only minor differences between the cryptic aureole and the regional rocks, affecting the vitrinite reflectance values, as commented in the previous paragraph. Theoretical and laboratory studies demonstrate that vitrinite maturation is attained in a geologically insignificant time interval (Ernst and Ferreiro-Mählmann, 2004); that is to say, the most important variable that determines vitrinite reflectance is host-rock temperature. Therefore the correlation between organic and clay mineral reaction progress depends overall on the thermal history, particularly on basinal heat flow characteristics and geological rates of heating. At high heating rates (contact metamorphism), vitrinite matures more rapidly than clay minerals (e.g. Olsson, 1999), but the maturation of vitrinite is slower where there is a normal geothermal gradient of gradual subsidence (e.g. Francu et al., 1999). When the thermal evolution for a given period of time is not well known, correlating organic maturation with a maximum temperature becomes complicated. To address this question it may be useful to consider research in young basins where thermal evolution and heat flow are well known. According to several authors (Lopatin, 1971; Gretener and Curtis, 1982), the thermal effect on organic maturation is exponential in nature, whereas time is linear. At very low temperatures (<50 ºC), the conversion rate is so slow that the time effect is negligible. Likewise, at temperatures above 130 ºC, the conversion rate is so fast that time is not a dominant factor either. However, the effect of time becomes significant in the interval of 70-100 ºC. Barker (1989) suggested that vitrinite maturation can range from 1 year or less in an intrusive emplacement (contact metamorphism) to 10^4 years in geothermal systems or 10^6-10^7 years in burial diagenetic conditions. Nevertheless, Merriman (2005) concludes that the rate at which clay mineral reactions progress from mature to supermature assemblages can range from 10^4 to $2*10^9$ years (or more). Therefore, the thermal effect of the emplacement of the Monchique intrusion has had very different consequences on these two types of materials due to their different maturation rates. The slow reaction rate of clay minerals does not respond to the short-lived thermal events (Srodon, 1979; Aoyagi and Asakawa, 1984; Kisch, 1987). Interestingly, such difference in the behaviour of inorganic and organic matter is not completely restricted to the cryptic aureola; it is more significant in the vicinity of the intrusion, but persists in all the Brejeira and Mira Formations.

In figure 8 we have compared the KI and vitrinite reflectance values (%Rr) of the rocks surrounding the Monchique pluton, in particular, and those corresponding to all the Brejeira and Mira Formations, in general, with similar characteristics of different geotectonic settings, according to Merriman (2005). The data of the cryptic aureole of the pluton are very similar to the extensional basin, characterized by a high heat flow (> 35ºC/km), whereas those corresponding to more distant areas of the Brejeira and Mira Formations fall in the intermediate field between high and normal heat flows, as defined by Merriman (2005). This seems to be an indication that the intrusion of the Monchique pluton occurred under a general thermal increased flux affecting to the entire region. The punctual area affected by the intrusion would have reached slightly higher values than the rest of the region, but the difference would not have been very significant due

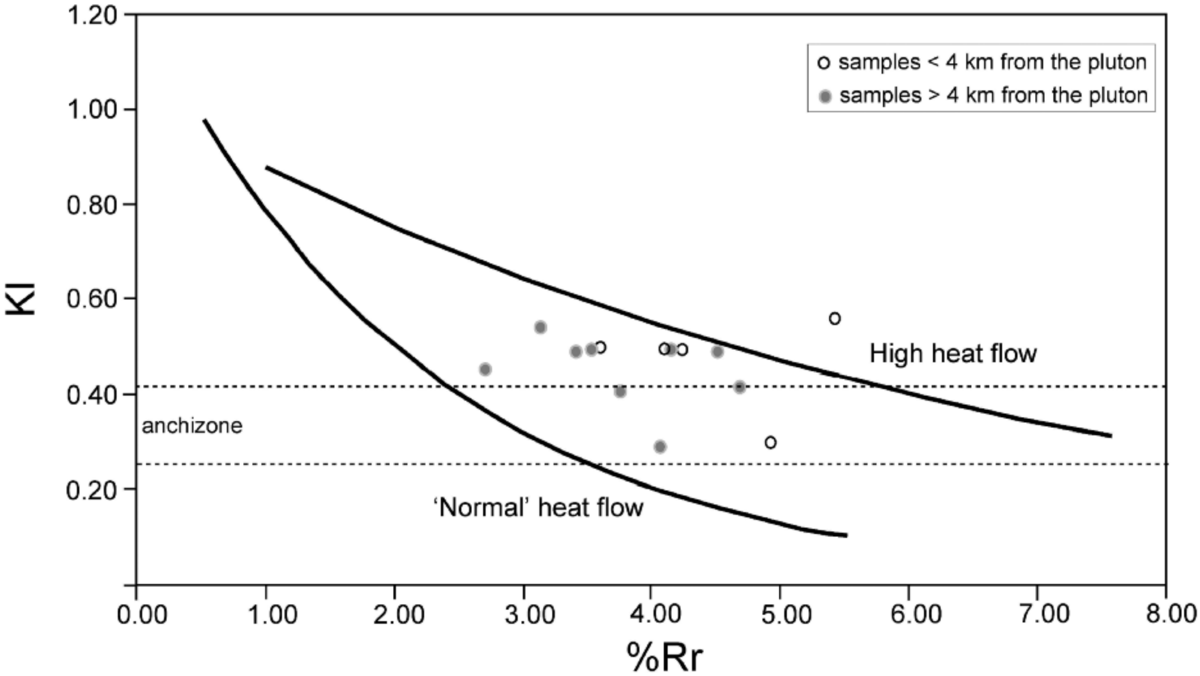

Fig. 8.- Plot of vitrinite reflectance %Rr against the Kübler index of illite "crystallinity" (KI). The curves are reproduced from Merriman (2005, Fig. 9). The right curve is based on data from South Wales and southern Ireland (White, 1992 and Wagner, 2003). The left curve is based on data from a wide variety of basin types (unpublished and from Dalla Torre et al., 1996).

to the regional high thermal plateau occurring during the Cretaceous as a consequence of the presence of a mantle plume below the eastern Central Atlantic region. The magmatism related to the thermal anomaly of the mantle plume is shown in the alkaline igneous rocks of three massifs along the Iberian margin in Portugal, Monchique, Sines and Sintra. The obtained ages for these plutons located along the Atlantic coast of Portugal are in agreement with the northward motion of the Iberian plate between 88 and 60 Ma above the mantle plume responsible of the magmatic activity (Grange et al., 2010).

The thickness of an intrusive body is an important factor in the development of a thermal alteration (Hagelskamp, 1988; Dennis et al., 1982; Bishop and Abbott, 1995) and no metamorphism occurs if the body is not thick enough. In addition, the emplacement depth and temperature of the intrusion also play an important role. If the emplacement of the body is deep enough, the effects on the surroundings may be less marked than if the emplacement is close to the colder terrestrial surface, where differences with the host rocks will probably be notable. Notwithstanding, cooling is faster close to the surface, and the interval for changes is therefore smaller. The intrusion of the Monchique body into the upper crustal level (Valadares and González-Clavijo, 2004; Grange et al., 2010) favored by a set of faults trending in the same direction suggests fast cooling, thereby having little effect on inorganic thermal indicators. Nevertheless, the fast cooling can produce retraction fractures in the host rocks promoting relevant changes in the rock fabrics.

According to several authors (Robert, 1988; Ernst and Ferreiro-Mählmann, 2004) in thermal aureoles, the relationships between inorganic and organic thermal indicators may be influenced not only by different reaction kinetics but also by hydrothermal alteration. The presence of kaolinite in most samples, including the hornfels, is not consistent with the typical prograde reactions and mineral assemblages of higher grade and corroborates a retrograde process previously described throughout the South Portuguese Zone (Abad et al., 2001 and Nieto et al., 2005) that is, a fluid-mediated process occurring at low temperatures and subsequent to regional metamorphism. In this case, it had to be subsequent to the contact metamorphism and probably favoured by post-intrusion hydrothermal fluids. Kaolinite crystallization is, normally, favoured by the circulation of low-temperature waters (< 200 ºC) within the rocks and its presence is usually considered an evidence for low-temperature alteration of other aluminosilicates, especially of feldspars. In samples of Brejeira Formation, Nieto et al. (2005) described some kaolinite crystals transecting the packets of mica and lateral transitions from 10 to 7 Å layers (Fig. 3 in Nieto et al., 2005). These relations imply that kaolinite post-dated muscovite formation subsequent to the peak metamorphism. Kemp et al. (2005) in a study in which they describe the effects of a Tertiary intrusion on mudstones and associated limestones, consider the formation of a late set of contraction joints and fractures when the intrusion and host rocks cooled, providing conduits for cooler fluids. The presence of a fluid phase is critical in driving clay mineral reaction and consequently, kaolinite could be a back-reaction product of higher-temperature minerals. Figure 6 shows parallel and alternating packets of mica and kaolinite. This kind of texture could be interpreted as one phyllosilicate replacing another, being the packets of reaction product parallel or subparallel to reactants, or as the result of the joint crystallization of the two minerals. Both origins are difficult to be distinguished, but, in any case, are compatible with the described processes.

The KI data from the surroundings of the pluton and, in general, from all the Brejeira and Mira Formations support the notion proposed by Merriman and Peacor (1999) that the main factors controlling KI values are kinetics. As in the Narcea Antiform (northern Spain) (Abad et al., 2003), crystallinity cannot be directly indexed with temperature measurements because crystal sizes are governed by the relative rates of crystal growth, pressure solution, and recrystallization processes, which are all controlled in part by fluid activity, heat and tectonism. Whereas strain was a very important factor in the growth of the Narcea micas, for instance, in Monchique the intrusion emplacement and the general passive heat flow did not imply fabric-forming events in the host metasediments, although according to Grange et al. (2010) the alkaline complex emplacement was related to the reactivation of major faults during the extensional rifting phases of the continent in the Jurassic. The absence of tectonic strain during the thermal event of the pluton emplacement is a factor that could have deprived the clay minerals of one of the main driving forces for their reaction progress.

To conclude, we have seen that in the area where the Monchique pluton intruded, whereas the Rr indicates temperatures higher than 300 ºC, that is, anchizone-epizone transition (subgreenschist to greenschist facies) (Bucher and Frey, 1994), the KI values and clay mineralogy point to diagenetic-low anchizone conditions (100-200 ºC). Furthermore, the mineral assemblage is typical of very low-grade metapelitic rocks except for the sample points nearest the contact (<50 m), in which hornfels have been described with mineral composition in coherence with the Rr data. The different nature of the reaction progress in the organic and inorganic materials explains the distinct response to a short-lived thermal event such as the Monchique pluton emplacement. The main reasons are the slow reaction rates of the clay minerals compared to the vitrinite maturation rate and the absence of pervasive tectonic events, which is one of the most important driving forces of the clay minerals reaction progress. In conclusion, the thermal effect of this intrusion had a minimal effect on the clay minerals of very low-grade metamorphism and other tools (vitrinite reflectance, for instance) are essential to identify and characterise the contact metamorphism beyond the hornfels area.

Conclusions

The effect of igneous intrusions on the intruded rocks and its relationships with the regional metamorphism is a complex phenomenon, which cannot be simplified in terms of a simplistic differentiation between contact and regional metamorphism. Igneous activity is usually linked to an enhanced regional thermal flux, which minimizes the differences between the vicinity of the intrusion and the rest of the metamorphic terrain. In the case of SW Iberian Peninsula, the presence of a mantle plume, in addition to the localized magmatic effects, being the Monchique one of the resultant intrusions, was able to produce a regional increase of the thermal flux giving way to similar effects to an extensional very-low grade metamorphism, which, in turn, produced anthracite and meta-anthracite vitrinite Rr values in the diagenetic-anchizone pelites of the Brejeira and Mira Formations. This regional thermal effect and the localized one in the pluton aureole were not able to change the clay-mineralogy of the pelites beyond the hornfels, restricted to 50 m from the pluton, due to a combination of short heating time and absence of tectonic strain.

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