# Metamorphism of the Palaeozoic Cinco Villas massif (Basque Pyrenees): illite crystallinity and graphitization degree

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**Abstract**

The degree of metamorphism affecting the Devonian–Carboniferous rocks of the Palaeozoic Cinco Villas massif has been studied by (*a*) petrographic techniques, (*b*) illite crystallinity, and (*c*) degree of graphitization of the carbonaceous material. Some mineralogical differences have been found between the Devonian and Carboniferous rocks; paragonite and mixed-layer paragonite/muscovite, typical of anchimetamorphic areas, appear in the Devonian but are not found in the Carboniferous rocks. These are characterized by the local appearance of chloritoid, garnet, amphibole, epidote, andalusite and biotite, and the generalized presence of muscovite/chlorite. Illite crystallinity shows a metamorphic zonation (anchizone–epizone) towards the granitic Aya massif, and a concentric pattern around the intrusive body.

The optical analysis shows that the first effects of the regional metamorphism began before the $D_2$ deformation, reaching its paroxysm during this phase and continuing afterwards. The degree of graphitization evolves progressively with metamorphism, and this fact is reflected in an exponential relationship between $d_{(002)}$ and crystallinity $Lc_{(002)}$. The similarity in the degree of graphitization between homologous materials within and outside the Aya aureole suggests a similar kinetic factor for both the thermal and regional metamorphisms, or a similar duration time. This fact, as well as the distribution and relationship between the deformation and recrystallization of the minerals, suggests a syn-plutonic regional metamorphism developed at low pressures, in accordance with the value of the $b_0$ parameter for the white micas, and probably a maximum temperature of about 500 °C.

KEYWORDS: metamorphism, illite crystallinity, graphitization, Cinco Villas massif, Spain.

## Introduction

DURING diagenesis and very low-grade metamorphism, different crystallo-chemical changes in clay minerals take place and these produce both polytypical and compositional changes, and a progressive increase in illite crystallinity. Some authors have used this parameter to define the transition between diagenesis and metamorphism (Weaver, 1961; Kubler, 1967, 1968; Dunoyer de Segonzac, 1969; Kisch, 1980; Islam *et al.*, 1982, among others). The $b_0$ parameter in the white potassic micas is related to the celadonitic content (Cipriani *et al.*, 1968) and has been proposed as a relative geobarometric indicator (Sassi and Scolari, 1974).

The pelitic and semipelitic sediments of the region generally contain an important amount of carbonaceous material whose nature evolves with the diagenesis and progressive metamorphism. The 'continuum' between ordered and disordered graphite is known in geological literature as the graphitization process (French, 1964; Griffin, 1967; Izawa, 1968; Landis, 1971). This process determines the chemical activity of carbon and the redox potential during metamorphism. According to Shengelia *et al.* (1977), the graphitization shows an inverse relationship with the *c* parameter of the carbonaceous material which decreases with increasing temperature. By contrast, it is not affected by pressure and, in contrast to silicate materials, retrograde metamorphism has no influence on the *c* parameter, hence its use as a possible geothermometer.

The present work attempts to establish the nature of the regional metamorphism surrounding the metamorphic aureole of the Aya granite in the

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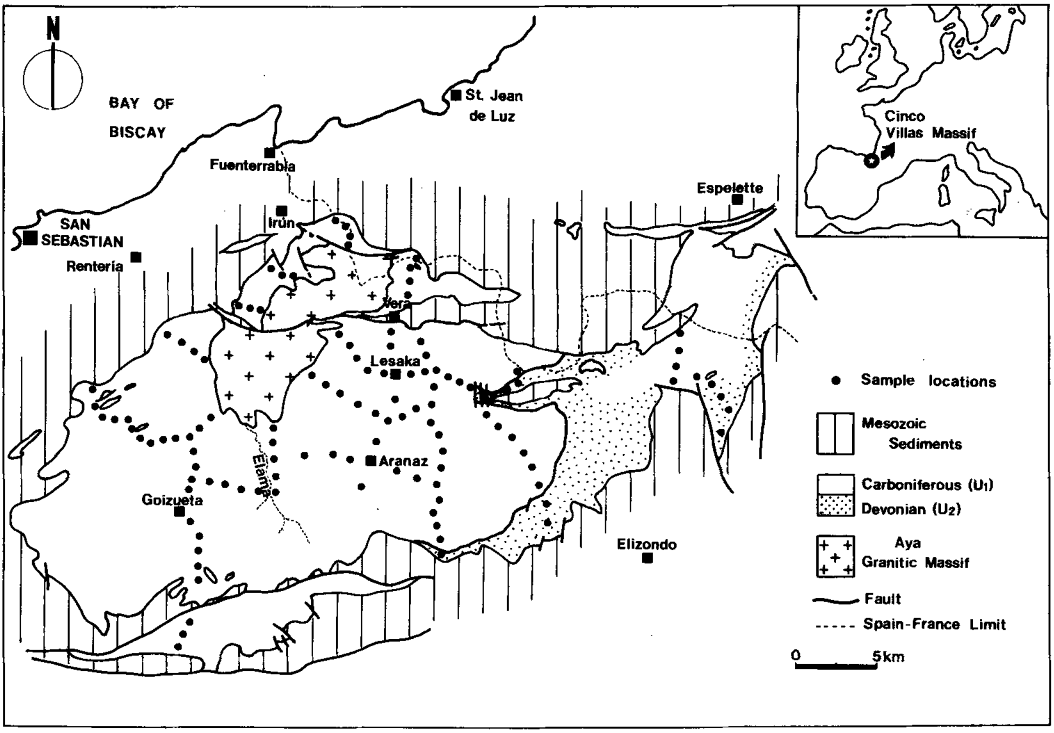


FIG. 1. Simplified geological map of the Cinco Villas massif. Black circles show the location of the analysed samples.

Palaeozoic Cinco Villas massif. Some authors have noted the influence of slight metamorphism on this massif but without mentioning any of its features or distribution (Campos, 1976; Krausse, 1974). The fact that Dunoyer de Segonzac and Heddebaut (1971) recorded an anchimetamorphic event on the Palaeozoic Aldudes massif using illite crystallinity has almost certainly influenced these authors.

## Geological setting

The Cinco Villas massif is an important part of the Hercynian basement of the Pyrenees. It is situated at the western limit of the Basque Palaeozoic, connected to the Labourd massif on the east and limited in the south by the Marble Slip Sheet (Fig. 1). Two main lithological groups can be differentiated: first, the sedimentary series which was deformed and affected by metamorphism during the Hercynian; and second, the igneous rocks related to this orogeny. The sedimentary series contains two lithostratigraphical units, chronologically different, corresponding to the $U_1$ and $U_2$ units of Heddebaut (1973).

The western unit ($U_1$) forms most of the massif and is characterized by a monotonous lithology (Fig. 1) of schistose character with interbedded sandstones, limestones and conglomerates (Cinco Villas Schistose Formation). These rocks lie on some less geographically extended limestones which outcrop in the Lesaka–Aranaz area. This unit still remains chronologically uncertain. The most recent lithostratigraphical studies suggest an age range from possibly Namurian for the Aranaz limestones (Heddebaut, 1973) to middle Westphalian (Requadt *et al.*, 1977). However, Juch and Schafer (1974) have described some rocks in the area east of the massif (Ormateko Formation) and have given them a Visean age.

In contrast, the eastern unit ($U_2$) has a less monotonous and chronologically better known lithology (Fig. 1). It has limited outcrops in the ESE of the massif with schists, sandstones, limestones and dolomites, which extend from possibly Silurian (Anzabal schists) to an upper Devonian age (Heddebaut, 1973; Juch and Schafer, 1974; Requadt, 1974).

All these rocks are affected by the Hercynian regional deformation where the $F_2$ and $F_3$ folding phases are the most evident. The $D_2$ deformation shows the most important structural features and it develops a regional schistosity variably orientated ENE–WSW. This phenomenon is accompanied by the intrusion of the Aya granitoid (Pesquera, 1985) located on the north of Cinco Villas massif and, together with some basic dykes and sills, is representative of the Hercynian magmatism in this region.

## Methods

Bearing in mind the lithological variations, the sample collection of pelitic and semipelitic rocks was carried out along seven principal traverses which are nearly perpendicular to the general strike of the Palaeozoic massif. The locations of the samples are shown in Fig. 1, at each of which several specimens were collected, altogether some 300 samples have been taken.

The mineralogical study has been carried out by combining X-ray diffraction with optical methods. The identification of the micaceous minerals in the anchizone was made primarily by X-ray methods. Orientated aggregates were prepared by separation of the clay fraction ($< 2$ $\mu$m) by sedimentation and, in appropriate cases, they were subject to glycolation and heating to 300–500 °C. In other cases organic matter was digested with $H_2O_2$, and the carbonate fraction removed with HCl (0.2N) or acetic acid (1N). In order to obtain a higher accuracy of the interplanar spacings, quartz and silicon were used as internal standards.

The half-height width of the illite 10 Å diffraction peak was measured using the following conditions: slits 1°–0.2 mm–1°, time constant 1 or 4 at scale $1 \times 10^3$, $2 \times 10^3$, scanning speed 0.5°/min and paper speed 10 mm/min. The peak widths are expressed in degrees $\Delta 2\theta$ rather than in millimetres. The values corresponding to the low and high-grade limit of Kubler (1967) are respectively 0.40° and 0.25° $\Delta 2\theta$.

The $b_0$ parameter of the potassic white micas was obtained following Sassi and Scolari (1974). One hundred and eighty polished slides were prepared from the pelitic rocks cut perpendicular to the foliation. The measurements of the $d_{(060)}$ spacing in the micas were made in the 59 to 63° $2\theta$ region using the quartz (211) peak as internal standard and at a scanning speed of 0.25°/min.

The separation of the carbonaceous matter contained in the pelitic and semipelitic rocks was carried out following a slightly modified method from Tagiri (1981). Altogether 42 samples were analysed. The amount so obtained rarely represented more than 1% of the original sample weight and was generally in the order of 0.2–1% depending on the character of rock. Sometimes purification was reached after two or three treatments.

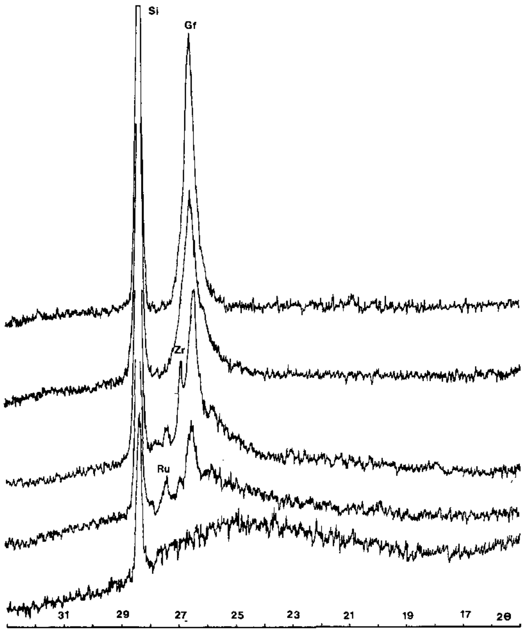


FIG. 2. Representative X-ray diffractograms from 15° to 32° $2\theta$ of the carbonaceous material separated from the pelitic schists of the Cinco Villas massif. Si = silicon; Gf = graphite; Zr = zircon; Ru = rutile.

The resultant material was mixed with Si as internal standard and then analysed by X-ray diffraction. Analytical conditions were; Cu-$K\alpha$ radiation at 40 kV, 20 mA, time constant 1–0.4, at scale $1 \times 10^3$, $2 \times 10^3$, scanning speed 1°/min., paper speed 10 mm/min. Frequently some insoluble minerals such as zircon and rutile were present in the residue (Fig. 2), but their presence did not jeopardize the analysis.

When the half-height width of the (002) reflection for the carbonaceous matter was greater than 0.5°, the correction factor proposed by Otani and Kimura (1972) was used. In this way the crystal size was calculated using the Scherrer formula $Lc_{(002)} = 91/\beta$, where $\beta$ is the real width at half-height.

## Mineralogy and petrography

### *Muscovite–illite*

Following Frey (1978), the presence of micas whose basal reflection remains unchanged at 10 Å following glycolation has been called 'muscovite-illite'. The fact that $d_{(060)} = 1.50 \pm 0.01$ Å indicates

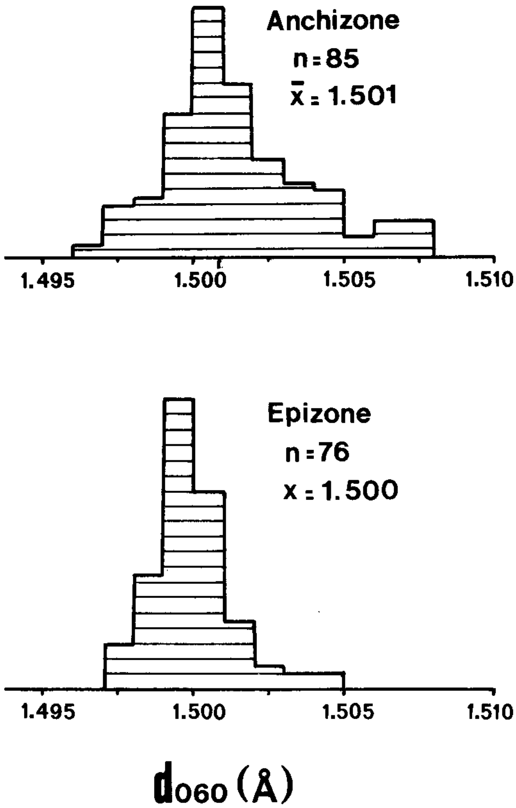


FIG. 3. Histograms of the $d_{(060)}$ spacings of anchimetamorphic and epimetamorphic white K-micas from the pelitic and semipelitic rocks of the Cinco Villas massif. Note the lower mean $d_{(060)}$ value of the epimetamorphic samples compared with anchimetamorphic samples.

its dioctahedral nature and, furthermore, the value

$$Ir = \frac{I_{(001)}/I_{(003)})\,\mathrm{air}}{(I_{(001)}/I_{(003)})\,\mathrm{glycol}} = 1$$

shows that it is not a swelling mineral (Srodon and Eberl, 1984). The polytype corresponds to $2M_1$ using the reflections 3.74 and 2.58 Å (Velde and Hower, 1963). The Si content changes from 6.10 to 6.50 using the relation of Velde (1967). The $d_{(060)}$ values of a number of samples from pelitic and semipelitic rocks were measured and the mean $d_{(060)}$ value is slightly lower for the epizone than for the anchizone samples (Fig. 3). In general, $d_{(060)}$ varies between 1.496 and 1.508 Å with $\Sigma(Mg + Fe_t)$ values between 0.01 and 0.23 according to the expression of Guidotti (1984). This suggests a slight loss of $\Sigma(Mg + Fe_t)$ in white K-mica with progressive metamorphism, which is probably consumed in chlorite formation. The application of the diffractometric methods to establish the $(Mg + Fe_t)$ content in muscovite has been criticized by some authors (Naef and Stern, 1982; Frey *et al.*, 1983), because the interference of the (060) and (331) reflections causes problems. However, the use of polished sections perpendicular to the foliation enhances the intensity of the (060) reflection, with the subsequent diminution of the intensity of the (331) reflection (Guidotti, 1984).

### *Paragonite/muscovite*

This mineral was first described by Frey (1969) in anchimetamorphic shales from the Glarus Alps, and interpreted as a regular interlayered mineral having a main basal reflection at 3.25 Å. In the Cinco Villas massif it appears only in rocks of Devonian and Silurian age and has not been found in the Carboniferous rocks.

Closely linked to this interlayered mineral is paragonite, which is easily recognized by its reflections at 4.82 and 3.21 Å. This phase, together with allevardite, pyrophyllite and chloritoid, has also been noted by Dunoyer de Segonzac and Heddebaut (1971) in Devonian rocks located to the NE of the Palaeozoic Aldudes massif. These phases have not been observed in Siluro-Devonian rocks of the Cinco Villas massif.

### *Chlorite*

Usually chlorite appears in different proportions, together with white mica, and shows a variable grain size (< 500 $\mu$m) depending on the metamorphic grade. The grain size is larger when it is found with biotite or in rocks close to the biotite isograd. It shows a net pleochroism: light green, colourless ($\alpha$) to green ($\gamma \approx \beta$), with typical blue or brown-greenish interference colours. Together with other phyllosilicates it defines the main cleavage of the rocks, but sometimes also defines an earlier cleavage or shows a disorientated growth post-$S_2$. The $b_0$ parameter and $d_{(001)}$ allow the composition of the octahedral and tetrahedral sites to be estimated from the basic formula $(Mg,Fe,Al)_6(Si,Al)_4O_{10}(OH)_8$. The average values of $b_0$ and $d_{(001)}$ from 52 anchimetamorphic chlorites are 9.30 and 14.12 Å respectively, and those from 36 epimetamorphic chlorites are 9.29 and 14.12 Å. Using the data of Bailey (1972, 1975), Brindley (1961) and Nieto (1983), these chlorites can be expressed by the structural formulae:

$$(Mg_{2.4}Fe_{2.5}Al_{1.1})(Si_{2.5}Al_{1.5})O_{10}(OH)_8$$
Anchizone

$$(Mg_{2.7}Fe_{2.1}Al_{1.2})(Si_{2.5}Al_{1.5})O_{10}(OH)_8$$
Epizone

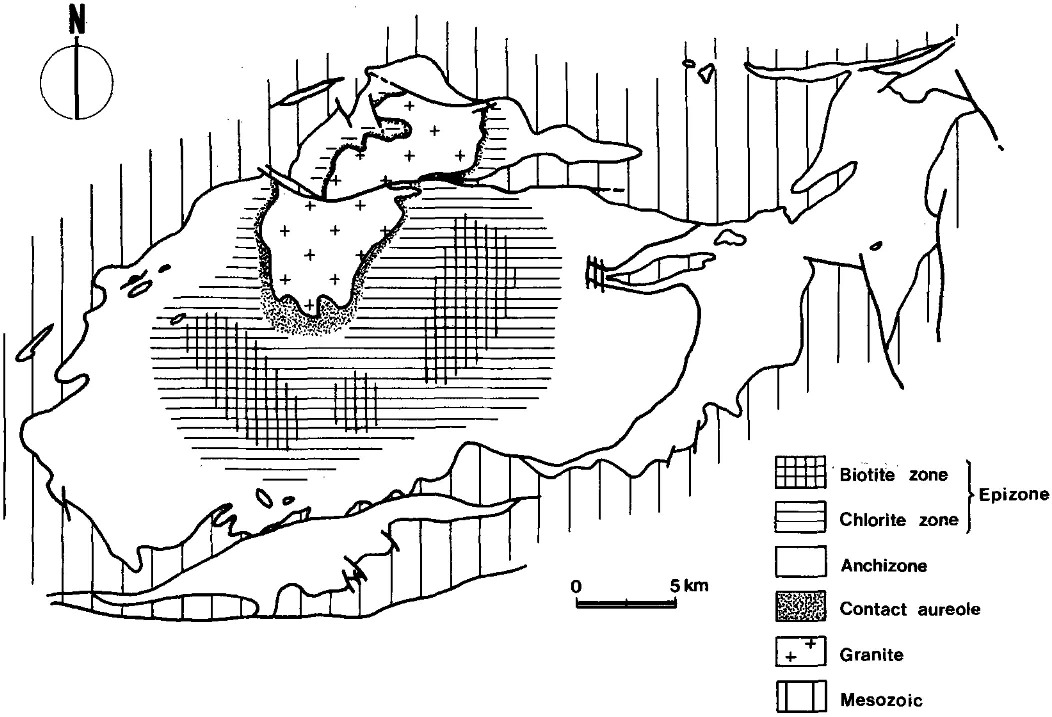


FIG. 4. Outline map of the Cinco Villas massif showing the different metamorphic zones.

According to the schemes proposed by Foster (1962) and Bayliss (1975) these chlorites fall close to the limit of the chamosite and clinochlore fields. The epimetamorphic chlorites are lower in Fe than anchimetamorphic chlorites. According to Nieto (1983), a temperature increase favours the formation of the Mg-chlorites, while its decrease favours the formation of the Fe-chlorites.

### *Biotite*

Biotite is relatively abundant in some pelitic and semipelitic rocks of the biotite zone, but its occurrence is restricted to the areas of Lesaka-Aranaz and Goizueta (Figs. 1 and 4). The crystals are $< 600$ $\mu$m and have a prismatic habit and strong pleochroism: light yellow ($\alpha$) to red brown ($\gamma \approx \beta$). The red colour increases with the grade of metamorphism, indicating a low content in $Fe_2O_3$ which would be compatible with the presence of ilmenite in the rocks rather than hematite or magnetite. Biotite growth is typically syn-$S_2$ although it postdates $S_1$ and $S_2$ (Fig. 5A, B).

### *Feldspar and quartz*

Plagioclase is the main feldspar and its composition varies between albite and sodic oligoclase, as determined by X-ray diffraction (Kroll and Ribbe, 1983). When possible, the plagioclase composition has been determined optically (Suwa *et al.*, 1974), and in the biotite zone it is more calcic, up to $An_{35}$, when associated with carbonate schists. On the other hand, myrmekitic intergrowths and chess-board textures have been observed, indicating transformation processes. K-feldspar is less abundant, appearing occasionally with perthitic lamellae ($< 10$ $\mu$m wide, $< 30$ $\mu$m long).

Quartz occurs in all samples and is a more or less abundant phase. In pelitic and semipelitic rocks its modal proportion varies between 10 and 50% approximately, with a coarse to very fine grain size.

### *Other minerals*

Apart from the above mentioned minerals, other phases whose distribution is more localized appear in the Cinco Villas massif.

*Andalusite* occurs with biotite in schists in the northeast Goizueta area and, particularly, in the Lesaka area. Under the microscope (Fig. 5B) it presents a xenoblastic or idioblastic form, with a

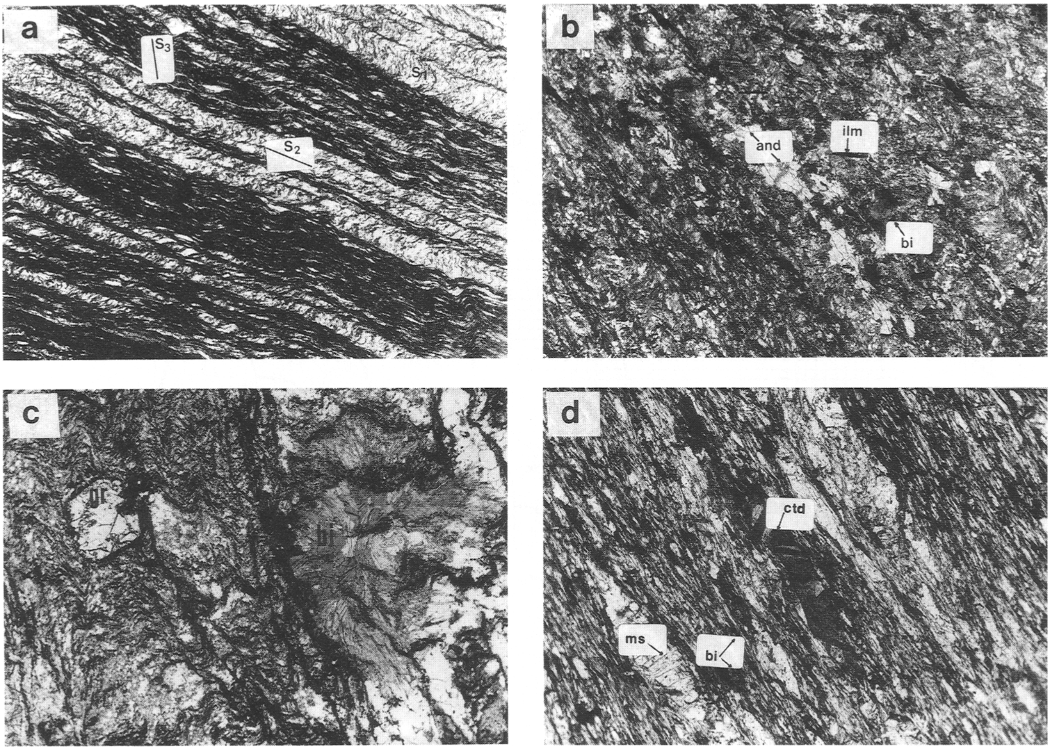


FIG. 5. Photomicrographs of mineral textures. (A) Biotite schist with $S_1$ cut vy planar $S_2$ and a later imposed crenulation ($S_3$) from Goizueta area (4×); (B) Xenoblastic andalusite (and) with biotite (bi) (post-$S_1$, syn-$S_2$) and disorientated ilmenite (ilm) crystals in relation to main foliation from Lesaka area (4×); (C) Schist with idioblastic garnet (gr) (post-$S_1$), poligonized biotite in a fold and amphibole from Goizueta area (4×). (D) Chloritoid (ctd) orientated parallel to $S_2$ foliation marked by lines of mica flakes in biotite-muscovite schist from Lesaka area.

fine to medium grain size, and its growth is syn-$S_2$ to post-$S_2$.

*Almandine* garnet is found only rarely in schists from the biotite zone. The crystals are subautomorphic (Fig. 5C), fine grained and with an ambiguous relation with respect to the regional deformation.

*Amphibole* has been found in schists to the NE of Goizueta, forming fibroid-radial or sheaf-shaped aggregates, less than 5 mm in size. The crystals have an actinolitic character with a weak pleochroism: light green ($\alpha$) to green ($\gamma$), sometimes with blue tones and an extinction angle $\gamma\hat{}c$ of 16° to 18°.

*Clinozoisite/zoisite* is present as either xenoblastic crystals or columnar aggregates of fine size in the biotite zone. Sometimes they can be optically differentiated by their extinction and interference colours.

*Chloritoid* has been observed only in metapelitic rocks near Lesaka. Under the microscope (Fig. 5D) it has the appearance of crystalloblasts which are syn-$S_2$. It is green-brown in colour, size < 500 $\mu$m and has variable extinction: $\gamma\hat{}a$ = 15–18°. It is important to mention its local coexistence with biotite.

The presence of *vermiculite* in semipelitic material represents a greater genetic problem (Velde, 1978). It is a scarce mineral and appears, in some samples, to correspond to the transition from the biotite to the chlorite zones. Using X-ray diffraction it is recognized by a reflection at 14.4 Å which has an intensity higher than a peak at 7.2 Å. Glycol treatment at 80 °C for 24 h does not result in any swelling phenomena, but when heated at 550 °C the basal spacing collapsed to 11.52 Å, indicating that the mineral was partially hydrated. This is diagnostic of the presence of vermiculite.

Optically the material shows certain similarity to biotite, from a light brown colour ($\alpha$) to a golden brown ($\gamma \approx \beta$), and sometimes with reddish hues. The crystal size is less than 100 $\mu$m and, generally post-$S_1$, syn-$S_2$. Chlorite, phengite,

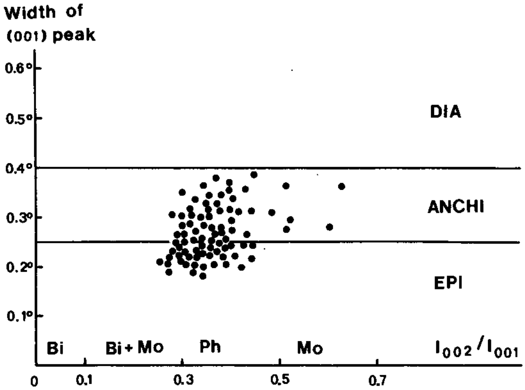


FIG. 6. Relation between the width (in $\Delta 2\theta$) of the 10 Å peak and the ratio of the intensities of the (002) and (001) peaks in the Silurian, Devonian and Carboniferous schists of Cinco Villas massif. Limits proposed by Kubler (1967).

plagioclase and quartz are the main phases present, together with vermiculite.

*Calcite* is the most common carbonate mineral, but sometimes minor ankerite has also been found. *Titanite* appears as characteristic brown xenoblastic or subidioblastic crystals in the carbonate schists of the biotite zone.

Finally, the most frequent accessory minerals are tourmaline, apatite, zircon, pyrite, rutile, ilmenite and graphitic material. Chalcopyrite, pyrrhotite, magnetite and hematite have been found but are rare.

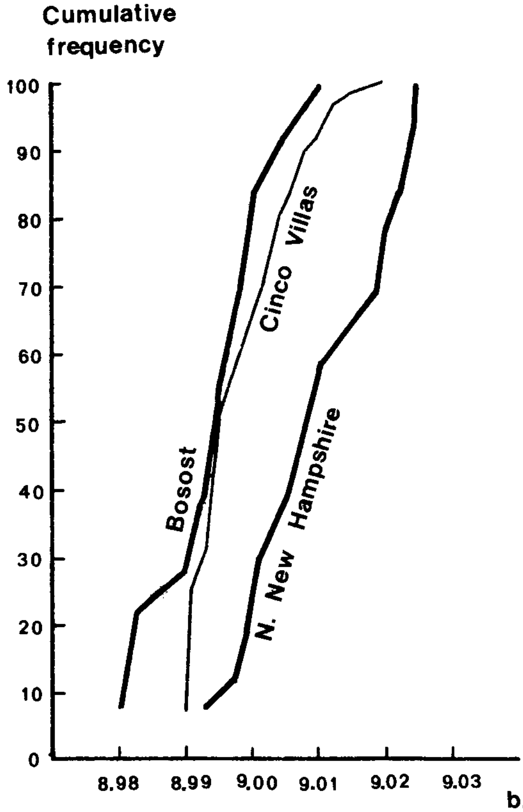


FIG. 7. Cumulative frequency curve of the $b_0$ values of white micas from the Cinco Villas massif in relation to analytical data of Bosost and N. New Hampshire massif.

### Illite crystallinity

Illite crystallinity has been used to indicate relative changes in the metamorphic grade. However, it is necessary to consider several factors that affect the width of the 10 Å peak, e.g. the lithology and the presence of paragonite (Kisch, 1983).

In Fig. 6 the crystallinity values are plotted against $I_{(002)}/I_{(001)}$ showing that most lie in the phengite field. The Silurian, Devonian and some Carboniferous rocks have been recrystallized under anchimetamorphic conditions. Notable differences in the illite crystallinity of these rocks have not been observed. The illite crystallinity increases towards the granitic massif, and the limit between anchizone and epizone appears approximately concentric to this intrusive body (Fig. 4).

### $b_0$ parameter of the white potassic micas

The measured $b_0$ values of white K-micas from pelitic rocks range between 8.990 and 9.020 Å with a mean value of 8.998 Å and standard deviation of 0.007. The cumulative frequency curve is represented in Fig. 7, lying between the Bosost and New Hampshire curves (Sassi and Scolari, 1974). Following the empirical scale proposed by these authors, conditions of low pressure during the Hercynian metamorphism of Cinco Villas can be deduced, which is compatible with the kind of metamorphism affecting the Pyrenees during the Hercynian orogeny. Nevertheless, it must be remembered that the changes in this parameter are due to the celadonite content, which is also related to the chemical composition of the rocks, rather than simply to the baric conditions of the metamorphism (Bertagnini and Franceschelli, 1982).

### Graphitization-degree

The ordering process of the carbonaceous matter during the graphitization can be expressed as a sequence from disordered graphite, graphite $d_3$,

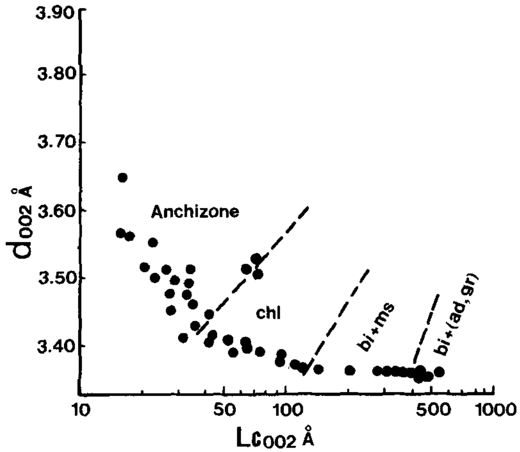


FIG. 8. Plot of $Lc_{(002)}$ *vs.* $d_{(002)}$ of the carbonaceous materials from Cinco Villas metamorphic rocks.

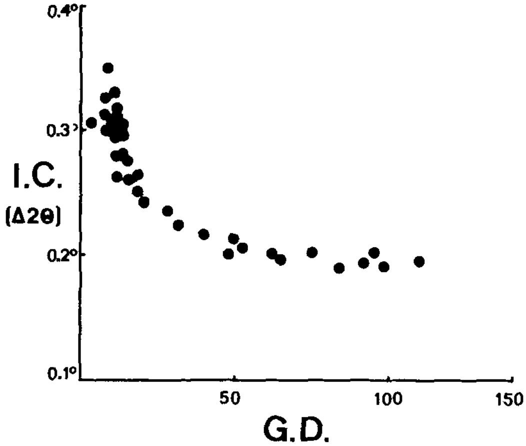


FIG. 9. Plot of illite crystallinity (I.C.) of the white K-micas *vs.* graphitization degree (G.D.) of the carbonaceous matter from Cinco Villas pelitic rocks.

graphite $d_2$, graphite $d_{1A}$, graphite $d_1$ to completely ordered graphite (Landis, 1971). Another way of representing this phenomenon corresponds to the expression contained in Tagiri and Tsuboi (1979), later modified by Tagiri (1981):

$$GD = [(d_{(002)} - 3.70)/\log(Lc_{(002)}/1000)] \times 100$$

where GD = graphitization-degree, and $Lc_{(002)}$ = crystallite size.

According to these authors a hyperbolic relationship exists between $d_{(002)}$ and $Lc_{(002)}$ and when the present values are plotted, a similar relationship is observed (Fig. 8). The anchizone-epizone limit and the different zones are shown superimposed on this diagram (Fig. 8). A hyperbolic relationship also exists between the illite crystallinity and graphitization degree (Fig. 9). However, the graphitization process still continues once the illite crystallinity has reached a maximum value.

In general, the carbonaceous material that characterizes the anchizone is graphite $d_{2-3}$ and $d_3$ (Landis, 1971). Graphite-$d_2$ appears also in the epizone but usually a higher degree of order is indicated. The X-ray diffraction spectra show perfectly the structural change undergone by the carbonaceous material in relation to the grade of metamorphism (Fig. 2). Sometimes ordered graphite coexisting with less ordered material has been found, which is manifested by a well defined (002) peak and another more diffuse one. This represents a clear example of coexistence between detrital graphite and metamorphic graphitic material, comparable to those described by other authors (Landis, 1971; Fujinuki *et al.*, 1974; Tagiri and Tsuboi, 1979; Itaya, 1981).

Variations in the graphitization degree of samples belonging to the same zone are due to different causes—from lithological differences to the influence of shearing stress. The differential behaviour of rocks towards the tectonic movements have been cited to explain these differences (Fischbach, 1971).

Grew (1974) and Itaya (1981) also indicated differences in the degree of completion of the silicate recrystallization and graphitization which correspond to temperature, on the one hand, and the duration of the metamorphism, on the other. Graphitization is a rate-controlled process. Consequently, bearing in mind that the graphitization degree reached by the materials within the contact aureole is similar to that from the analogous zones in the rest of the massif, it would suggest that the plutonism and the regional metamorphism are two closely linked phenomena. In other words, the Hercynian regional metamorphism registered in Cinco Villas would have a synplutonic character.

## Mineral distribution and assemblages

The mineral assemblages observed in the pelitic and semipelitic rocks in each of the metamorphic zones are summarized below. The boundaries of the different zones recorded correspond approximately to those shown in Fig. 4.

*Anchizone.* Muscovite - illite + chlorite + quartz + albite + mixed layer paragonite / muscovite; muscovite - illite + chlorite + quartz + albite; and muscovite–illite + quartz + calcite + chlorite + albite. Accessory minerals include combinations of tourmaline, zircon, apatite, pyrite and graphitic material (graphite $d_3$–$d_2$, Landis, 1971).

*Chlorite zone.* Muscovite + chlorite + quartz + albite; muscovite + chlorite + quartz + calcite (±ankerite) + albite. Accessory minerals include combinations of tourmaline, zircon, apatite, pyrite and graphitic material (graphite $d_{1A}$, Landis, 1971).

*Biotite zone.* Biotite + quartz + plagioclase + muscovite + chloritoid + chlorite; biotite + quartz + plagioclase + calcite (±ankerite); biotite + quartz + amphibole + plagioclase + clinozoisite / zoisite + garnet; and biotite + quartz + andalusite + muscovite. Accessory minerals include several combinations of sphene, tourmaline, zircon, apatite, rutile, ilmenite, pyrite and graphitic material (graphite $d_1$, Landis, 1971).

## Outline of the chronology of deformation and recrystallization

In many metamorphic regions, evolution of the tectonic movements by the superimposition of two or more structures is evident from the observation of the microstructural relations. The relative chronology between deformation and Hercyninian metamorphism in Cinco Villas can be thus established (Fig. 10).

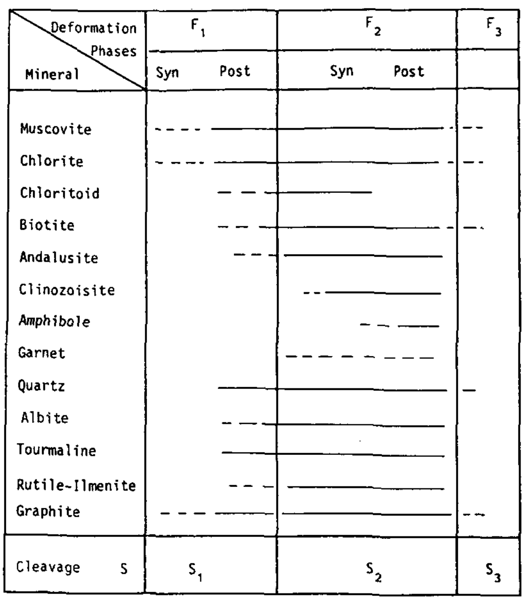


FIG. 10. Outline of the chronology of deformation and recrystallization of some important minerals.

The relationship to $D_1$ is not so clear as to $D_2$. This is due to a less obvious development of $S_1$ than $S_2$. At the same time, the latter is affected by a fracture cleavage ($S_3$) of irregular importance within the massif. The angle formed between $S_2$ and $S_3$ is variable, from a low angle, subparallel, to almost an orthogonal relation (Fig. 5).

$S_1$ is usually defined by phyllosilicates and graphitic material. At the same time it is possible to observe helicitic crystals of chlorite, albite and biotite formed by $S_2$ (Fig. 5A). The formation of biotite continues during and after $D_2$, and sometimes crystals reorientated by $S_3$ have been observed. Amphibole, on the other hand, is clearly post-$S_2$, but in the case of garnet there are not enough criteria to establish a precise correlation.

The highest intensity of the regional metamorphism was reached during $D_2$, although its pre-$D_2$ and post-$D_2$ effects are also relatively important.

## Metamorphic conditions: discussion

It is difficult to establish the precise physical conditions of the regional metamorphism affecting the Cinco Villas massif, as the lack of thermobarometric data limits any kind of discussion.

The potassic white mica polytype corresponds to $2M_1$ and this suggests temperatures higher than 300 °C (Velde, 1965). On the other hand, the general presence of paragonite/muscovite in Devonian rocks would denote a very low grade of metamorphism, below 350 °C. The illite crystallinity increases progressively towards the granitic massif until epimetamorphic conditions are reached, with a biotite zone registered in specific areas within the massif (Fig. 4). The maximum temperatures reached during the regional metamorphism would probably be near 500 °C, considering the local appearance of almandine and the coexistence of andalusite–biotite. The formation of almandine would indicate conditions of the highest temperature zone within the low-grade metamorphism (Winkler, 1979).

Accepting the presence of a high thermal gradient during Hercynian metamorphism of the Pyrenees (Zwart, 1979) and, bearing in mind the $P$–$T$ diagram with variable Si values for the micas of the muscovite–celadonite series (Velde, 1967), the Si contents registered in the biotite zone (6.20±0.10) would reflect pressures below about 1.5 kbar. This would be comparable to both the low-pressure conditions suggested by the $b_0$ parameters for the white micas and the estimated pressure regime within the contact aureole associated with the Aya granite massif (Pesquera, 1985).

In conclusion, the maximum regional metamorphism of Cinco Villas developed at temperatures below 500 °C in a low-pressure regime (presumably at $P_{H_2O} \approx 1 \pm 0.5$ kbar). The fact that the graphitization degree of the carbon matter is similar in analogous materials inside and outside the contact aureole suggests that the length of both the contact metamorphism induced by the Aya pluton and the regional metamorphism has been similar. This fact, together with the distribution and the relation between deformation and recrystallization of the minerals, suggest a syn-plutonic regional metamorphism.

The generation of local thermal domes would be the consequence of the transferred heat due to the influence of igneous material from deep levels, and this could explain the presence of biotite in Carboniferous rocks when it has been established that on the Pyreneean regional scale, the biotite isograd is rarely reached in the Devonian (Kornprobst *et al.*, 1980). This is related to the hypothesis suggested by Zwart (1962) and Fonteilles (1981), where the high thermal gradients reached during Hercynian metamorphism in the Pyrenees would be the consequence of a pluto-metamorphic phenomenon.

The uppermost limit of the Hercynian metamorphism, as evidenced in Cinco Villas, is not posterior to the orogenic paroxysm in contradiction to what Autran *et al.* (1970) and Zwart (1979) established in the Oriental and Central Pyrenees respectively. The first effects of the regional metamorphism in Cinco Villas are manifested before the major Hercynian deformation ($D_2$), being principally developed during $D_2$ and even post-$D_2$, although less important.

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