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MORPHOLOGICAL AND CRYSTALLOGRAPHIC CHARACTERISTICS OF THE GRAPHITE OF OSUMITA, VENEZUELA, AND ITS USE AS A GEOTHERMOMETER

M. Martínez*, M. Vivas*, S. Marrero*, W. Meléndez*, G. Garbán**, A. Benavides***

RESUMEN

Se realizó un estudio morfológico, petrográfico y cristalográfico en muestras representativas de grafito diseminado del Cerro Osumita, estado Cojedes, Venezuela, con el fin de establecer el grado de cristalinidad mineral y la temperatura máxima alcanzada por la roca. A partir de la roca original (Gneis cuarzo-feldespático-grafítico La Aguadita), se obtuvieron fracciones enriquecidas en grafito mediante la combinación de métodos químicos (HCl-HF) y físicos (flotación y acumulación en interfaces líquidas). El grafito obtenido por este procedimiento fue analizado por difracción de rayos X. Adicionalmente, se estudió la morfología del grafito aislado mediante Microscopía Electrónica de Barrido (MEB).

La asociación mineralógica del gneis (cuarzo – oligoclasa – hornblenda (verde) – microclino – clorita – biotita – epidota) ubica el metamorfismo alcanzado por la roca en la zona superior de la facies de los esquistos verdes, en la zona de la biotita. El grafito presenta un aspecto de hojuelas con una concentración que oscila entre 7 y 10% en peso. El proceso de enriquecimiento permitió obtener fracciones con más de 92% en grafito. Bajo el microscopio electrónico de barrido, los granos de grafito presentan una morfología definida por láminas muy bien definidas y ordenadas. El índice de cristalinidad alcanzado oscila entre 68–70, con un grado de grafitización promedio de 60, y un valor del parámetro cristalino d_{002} de 3,35 Å, lo que sugiere un alto grado de ordenamiento de la red cristalina. La temperatura máxima alcanzada por la roca, de acuerdo con el grado de grafitización, es de 472 ± 27 °C.

Palabras clave: grafito, grafitización, cristalinidad, difracción de rayos X, Venezuela.

ABSTRACT

A morphologic, petrographic and crystallographic study was performed in some representative samples of disseminated graphite of the *Cerro Osumita*, Cojedes State, Venezuela with the aim of establishing the degree of mineral crystallinity (graphitization) and the highest temperature reached by the rock.

From the original rock (*La Aguadita* quartz-feldespatic-graphitic Gneiss), fractions enriched in graphite were obtained by means of leaching with HCl-HF and physical procedures (flotation and accumulation in liquid interfaces). The isolated graphite was analyzed by X-Ray Diffraction, whereas morphology was studied by Scanning Electronic Microscopy (SEM).

The mineralogical association exhibited by the gneiss (quartz – oligoclase – hornblende (green) – microcline – chlorite – biotite – epidote) allows establishing the metamorphism of the rock in the upper zone of the green schist facies, in the biotite zone. Graphite grains present aspect of flakes, with a concentration oscillating between 7 and 10% in weight. The used enrichment process allowed obtaining fractions with more than 92% in graphite. Under Scanning Electronic Microscopy, graphite samples showed morphology dominantly as sheets, very well defined and ordered. The crystallinity index (measured by XRD) is around 58 and 60, with a mean graphitization degree of 60, suggesting a high order in the crystalline net. The higher temperature reached by the rock, according to the graphitization degree, is of 472 ± 27 °C.

Key words: graphite, graphitization, crystallinity, X-ray diffraction, Venezuela.

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* Centro de Geoquímica, Instituto de Ciencias de la Tierra, Facultad de Ciencias, Universidad Central de Venezuela. Apartado 3895, Caracas 1010 A, Venezuela. E-mail: manmarti@gea.ciens.ucv.ve

** Centro de Geología, Instituto de Ciencias de la Tierra, Facultad de Ciencias, Universidad Central de Venezuela. Apartado 3895, Caracas 1010 A, Venezuela.

*** Centro de Geofísica, Instituto de Ciencias de la Tierra, Facultad de Ciencias, Universidad Central de Venezuela. Apartado 3895, Caracas 1010 A, Venezuela.

Introduction

Graphite is in high global demand due to its numerous applications. However, relatively few countries produce this resource (Korea, Mexico and Madagascar together account for nearly 90% of world graphite base reserves, Mendoza, 1996). World reserves of this mineral are on the order of 4 × 10^{11} MT.

Venezuela ranks among the top 20 importers of pure graphite worldwide. More than 1,700 MT of the mineral are imported annually, even though the country possesses several mineralizations of economic interest, such as the deposits of Cerro Osumita in Cojedes State and Montecano in Falcón State. Due to different natural graphite varieties — both crystallographically and morphologically — adequate characterization of the different deposits present in the country is necessary.

The objective of this study is to classify the graphite of the La Aguadita Gneiss, establish its degree of crystallinity and the level of graphitization of the carbonaceous material contained in the gneiss. This information is fundamental for establishing the commercial value of this deposit and its potential as a future graphite resource. The study also explores the use of graphite as a geothermometer, comparing temperature values obtained by this method with those of the metamorphism reached by the associated rocks, established through mineralogical associations.

Origin and Evolution of Graphite in Metamorphic Rocks

Amorphous carbonaceous material is present as an accessory in numerous sedimentary and metamorphic rocks. As the level of metamorphism increases, the structural, chemical and textural characteristics of this material undergo numerous transformations, largely due to the high chemical reactivity of organic matter relative to other materials present in the rock, such as silica and carbonates.

The variety of changes undergone by carbonaceous material in the sedimentary domain can be tracked relatively easily by conventional petrographic or geochemical methods (e.g., vitrinite reflectance, bitumen yield, biomarker detection, Rock-Eval pyrolysis, among others). However, upon entering the metamorphic environment, the transition from kerogen in metagenesis or anthracite to graphite *s.s.* can be tracked through X-ray diffraction. Other techniques include: Raman spectroscopy (Leech and Ernst, 1998), solid-state ^{13}C NMR spectroscopy, mass spectrometry (stable isotopes in graphite, Hoefs and Frey, 1976) and differential thermal analysis (Luque et al., 1998).

Graphite can be used as an indicator of metamorphic grade in rocks whose chemical composition does not allow the development of index minerals, and in rocks that have undergone polymetamorphism or retrograde metamorphism (Tagiri et al., 2000). Since graphitization is irreversible, any indicator based on graphite crystallinity will only record the highest metamorphic conditions reached, making it more significant than other geothermometric methods.

Varieties and Classification of Graphite

Several varieties of graphite are known, depending on morphology and mode of occurrence. Amorphous graphite, the most common in nature, occurs in nebular form in thin section, sometimes with microcrystalline structure (French, 1964); flake graphite results from a higher degree of crystallization. Spheroidal, crystalline, and diamond-pseudomorph graphite are also recognized. Flake graphite derives from high-grade metamorphism under elevated pressure and temperature; amorphous graphite from low-grade regional metamorphism (phyllites) and contact metamorphism in coal sequences.

French (1964) proposed one of the earliest classifications of carbonaceous material:

1. Amorphous carbonaceous material.
2. Asphaltic carbonaceous material.
3. Disordered graphite (graphite-d).
4. Graphite.

This classification was refined by Landis (1971) in terms of degree of disorder:

1. Fully ordered graphite, with a very intense 002 peak at 3.35–3.36 Å, and presence of other reflections.
2. Graphite-d1, with a relatively intense 002 peak at 3.35–3.36 Å, but noticeably broadened toward lower angles. Other reflections absent.
3. Graphite-d2, with a broad 002 peak and larger spacing (~3.5 Å).
4. Graphite-d3, with a very broad 002 band, low-intensity peaks, and mean spacing 3.50–3.75 Å. Asphalts and poorly evolved organic materials.

Factors responsible for graphite crystallinity include: temperature (Winkler, 1976); pressure (Oberlin, 1980); directed stress (Wilks et al., 1993); duration of the metamorphic event; type of initial kerogen (Ross and Bustin, 1990); silicate catalysts (Hoefs and Frey, 1976); and fluid phase composition (Rumble and Hoering, 1986). The interpretation of metamorphic history may be complicated by the simultaneous presence of graphitizable and non-graphitizable organic matter, producing complex XRD spectra (Buseck and Bo-Jun, 1985).

Tagiri et al. (2000) used the Graphitization Degree (GD) and other crystallinity parameters in the Sanbagawa metamorphic belt (Japan) to establish a shuffled-cards structure model.

Graphite is therefore a mineral sensitive to the degree of metamorphism reached; its characterization allows temperature or pressure values to be estimated with reasonable precision. Its use as a geothermometer should be restricted to discrete regions.

X-Ray Diffraction

Several parameters extracted from X-ray diffraction (XRD) analysis allow the degree of crystallinity achieved by graphite to be determined. The Crystallinity Index (I_c) originally proposed by Landis (1971):

$$I_c = 2 \times h_{002} / w_{002} \qquad (1)$$

where h_{002} and w_{002} are the height and base of the 002 reflection in the graphite diffractogram. Crystal thickness (L_c) is obtained from the Scherrer equation:

$$L_c = [k\lambda / \beta_{002}] \cos\theta \qquad (2)$$

where λ is the radiation wavelength; $k = 0.9$ for graphite; b_{002} is the half-width of the d_{002} peak in radians; and θ is the diffraction angle. L_c measures the number of graphene layers stacked in the crystal.

A third parameter is the Graphitization Degree (GD), obtained from the Tagiri (1981) equation:

$$GD = [d_{002} - 3.70] \times 10 / \log(L_c / 1000) \qquad (3)$$

GD relates to crystallization temperature through equations derived empirically by Wada et al. (1994) from the Ryoke metamorphic belt, central Japan:

$$T\ (°C) = 3.8\ GD + 200 \qquad (4)$$

$$T\ (°C) = 3.2\ GD + 280 \qquad (5)$$

Equation 4 applies to rocks with a calcareous sedimentary protolith; Equation 5 for pelitic protolith. These were obtained from 86 rock samples affected by regional and contact metamorphism, using temperatures from conventional geothermometers and mineral paragenesis.

Geological Setting

From a geological standpoint, the north-central region of Venezuela is dominated by the Caribbean Mountain System, a complex igneous-metamorphic province. Menéndez (1965) subdivided the central massif into four tectonic belts, later expanded to eight by Bell (1968). More recently, Ostos et al. (2001) defined the following tectonostratigraphic belts (approximately E-W trend, from north to south):

1. *Dutch and Venezuelan Islands:* Early Cretaceous oceanic igneous rocks coexisting with Late Cretaceous island arc rocks.
2. *Cordillera de la Costa – Serranía del Litoral:* Precambrian granitic basement and Mesozoic metasedimentary cover; oldest rocks affected by high P/T metamorphism, overprinted by intermediate P/T metamorphism.
3. *Caucagua-El Tinaco belt:* Paleozoic basement, Permian metasedimentary cover, and Cretaceous metavolcanic/metasedimentary association. Basement affected by low P/T amphibolite facies; Paleozoic and Mesozoic sequences show green schist facies.
4. *Paracotos belt:* flysch sequences with serpentinite mega-lenses and dismembered ophiolites of Late Cretaceous–Paleocene age; locally very low-grade metamorphism.
5. *Villa de Cura belt:* metasediments, meta-volcaniclastics, island arc volcanics and mafic to ultramafic plutons; high P/T metamorphism overprinted by intermediate P/T.
6. *Guárico Mountain Front:* unmetamorphosed sequences Late Cretaceous to Oligocene; deep marine to continental environments.

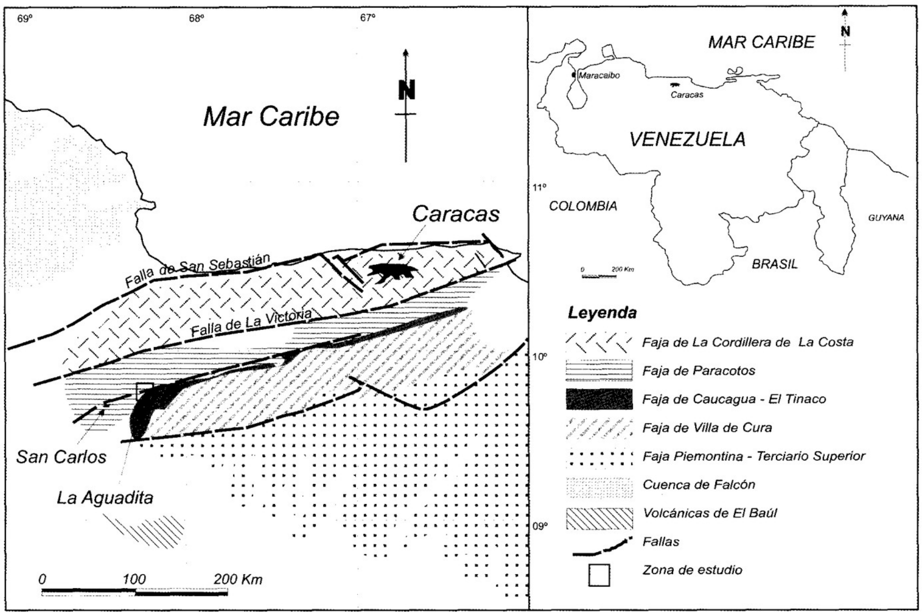

Fig. 1.—Simplified geological map of the north-central zone of Venezuela, showing the main tectonic features and the location of the study area.

The graphite-bearing rocks in Cojedes State fall within the Caucagua-El Tinaco tectonic belt. This complex, located between the Cordillera de la Costa belt to the north and the Guárico Mountain Front to the south, presents two metasedimentary units: the La Aguadita Gneiss and the Tinapú Schists (Fig. 1, González de Juana et al., 1980).

The La Aguadita Gneiss constitutes the oldest part of the complex, of probable volcano-sedimentary origin. It is composed mainly of interbedded hornblendic gneisses, biotitic gneisses, amphibolites, and granitic rocks. The unit is an alternation of mafic and felsic layers averaging 5–30 cm in thickness, rarely exceeding 1 m; this alternation produces the banded appearance of the gneiss. Felsic layers (quartz-plagioclase rocks) tend to be thinner than the mafic ones (hornblende-quartz-oligoclase gneiss and amphibolites).

Transitions between the two types are common, particularly between the hornblendic and biotitic gneisses. In the upper part, the unit contains quartz-plagioclase-chlorite schist layers and schistose conglomerates with quartz and granitic rock clasts.

The mineralogical composition is governed by plagioclases (25–70%), mainly oligoclase (average 18–27% An). Feldspars occur as porphyroblasts and recrystallized neoblasts. Predominant mineralogical associations (Rojas, 1989):

Quartz-feldspathic gneisses:

- Quartz + oligoclase + hornblende (green) ± microcline ± chlorite ± biotite ± epidote ± actinolite.
- Quartz + oligoclase + biotite + epidote ± hornblende (green) ± garnet.
- Quartz + oligoclase ± microcline ± chlorite ± epidote ± muscovite.
- Quartz + oligoclase ± hornblende (green) ± epidote.

Amphibolites:

- Oligoclase + hornblende (green to brown) ± quartz + chlorite ± epidote ± biotite.
- Oligoclase + hornblende (green to brown) ± chlorite ± epidote.

Graphite is usually found as an accessory mineral, though some samples reach concentration levels from 2 to 13%.

The La Aguadita Gneiss apparently represents a sedimentary sequence with minor volcanic additions. According to González de Juana et al. (1980), its mineralogical associations correspond to the epidotic amphibolite facies.

Materials and Methods

A total of 5 gneiss samples were collected from different outcrops of the La Aguadita Gneiss at Cerro Osumita. The first 5–10 cm of exposed rock were discarded; only fresh, unweathered samples were collected. Rock samples were pulverized in a Shatterbox with a tungsten carbide vial. The graphite enrichment process combined physical and chemical methods, which proved more efficient than either alone. The pulverized sample is initially treated with concentrated HCl and HF to eliminate the silicate mineral matrix. The residue is washed several times and, if necessary, re-subjected to acid treatment. Subsequently, the residue is placed in a separating funnel with equal volumes of water and cyclohexane. Graphite preferentially concentrates at the water-cyclohexane interface, separating from remaining mineral impurities. This procedure is essentially a modification of Bonijoly et al. (1982). Figure 2 shows a summary of the experimental method.

This combined process yielded graphitic fractions enriched up to 12 times the original mineral content — a

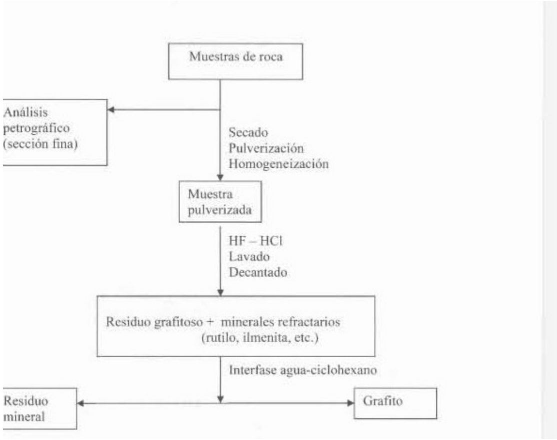

Fig. 2.—Experimental scheme used in the present work.

graphitic fraction with 92% graphite from rock with an original content not exceeding 8.5%.

The graphite concentrate was subjected to: %C (UIC Coulometrics), XRD (Siemens D8 Advanced), and Scanning Electron Microscopy (Hitachi S-500).

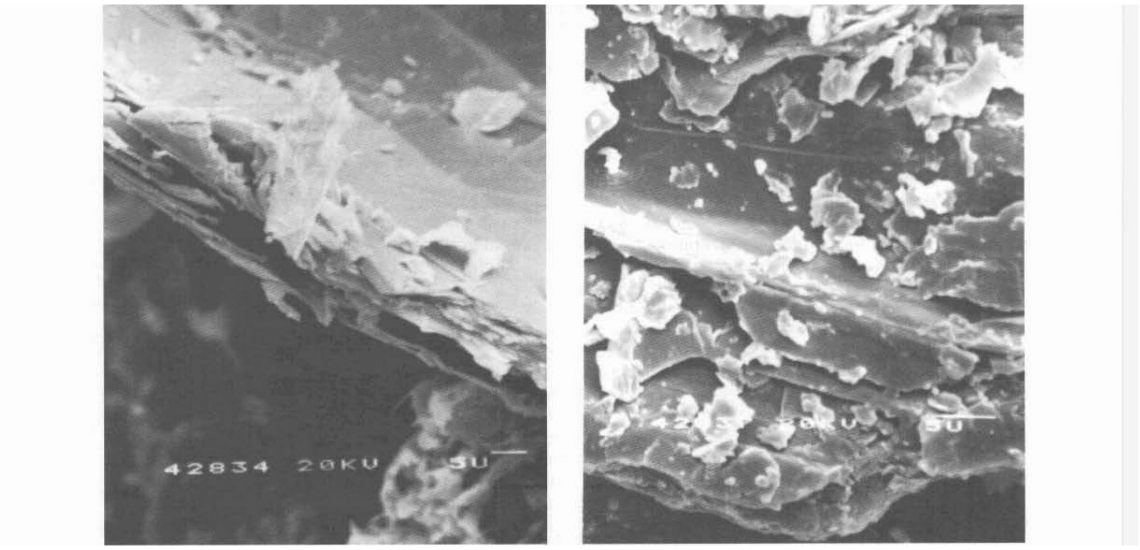

Fig. 3.—Micrograph of the Osumita graphite.

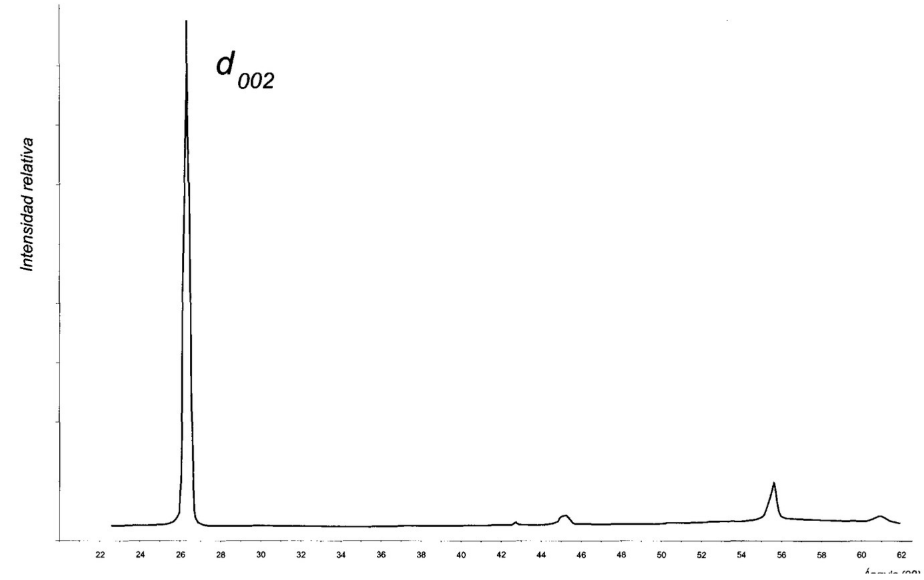

Fig. 4.—Diffractogram of the Osumita graphite.

Results and Discussion

Petrography

The La Aguadita Gneiss can be formally classified as a quartz-feldspathic graphitic gneiss. Petrographic analysis of the samples allowed the identification of quartz (mono- and polycrystalline), microcline, oligoclase, chlorite, muscovite, epidote, graphite, and biotite. The metamorphic grade, determined from mineralogical associations, reaches the green schist facies, biotite zone. In thin sections, graphite appears as well-differentiated flakes, laminae or scales of up to 3 mm in length.

Graphite Analysis by Electron Microscopy and XRD

The morphological analysis of the studied graphite (Fig. 3) reveals well-differentiated and structured laminae, providing a preliminary indication of the degree of crystallinity achieved. This is confirmed by the diffractogram of the graphitic concentrate (Fig. 4). Several observations point to a highly structured and organized graphitic material: the perfect definition of the peaks and the high resolution of the 002 diffraction peak at a 2θ angle of 26.3° (Cu Kα), corresponding to an interatomic distance (d_{002}) of 3.36 Å. The Osumita graphitic material is therefore well-crystallized graphite according to French (1964) and Landis (1971).

Table 1 presents the results of calculations established from the diffractogram. All parameters support the concept of a well-structured, thick, well-crystallized graphite with a high degree of graphitization. The plot of d_{002} spacing vs. the ratio of the 002 peak height to its half-width places the Osumita graphite in the well-ordered structure field (Fig. 5a). The d_{002} vs. L_c plot (Tagiri et al., 2000) places it in the biotite zone of the green schist facies (Fig. 5b).

Table 1.—Parameters obtained from the diffractogram of the graphite of Cerro Osumita

Parameter	Graphite of Osumita					Carbonaceous Material*
d_{002} (Å)	3.36	3.35	3.36	3.35	3.36	3.46
Crystallinity Index (I_c)	68	70	68	69	68	5
Crystal thickness (L_c)	265 Å	290 Å	265 Å	305 Å	255 Å	35 Å
Graphitization Degree (GD)	57	63	59	66	56	20
Temperature (°C) — pelitic rocks	462	481	470	491	460	344

* Carbonaceous sample from Kempis (Miranda State, Venezuela), very low evolutionary grade, included as reference.

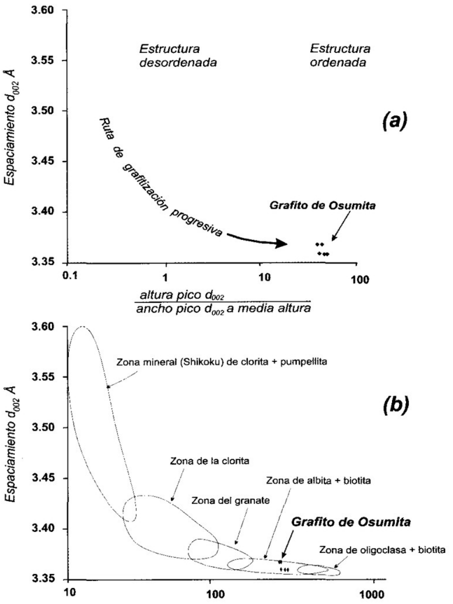

Fig. 5.—(a) d_{002} spacing vs. the ratio of the height of the 002 peak to its half-width (after Diessel et al., 1978). (b) d_{002} spacing vs. L_c, showing the different mineralogical zones (after Tagiri et al., 2000).

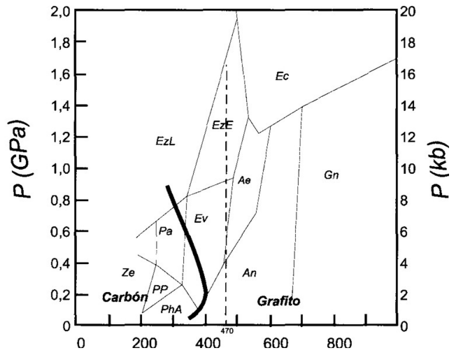

Fig. 6.—P-T diagram with fields of the different metamorphic facies. Ze = Zeolite; PP = Prehnite-pumpellite; Pa = Pumpellite-actinolite; EzL = Blueschist-lawsonite; PhA = Prehnite-actinolite; Ev = Green schist; EzE = Epidotic blueschist; An = Amphibolite; Ae = Epidotic amphibolite; Ec = Eclogite; Gn = Granulite. (Yardley, 1989). Dashed line = temperature found for Osumita graphite. Shaded area = P-T field where GD varies 55–65. GD of Osumita graphite = 59.

Relationship Between the Obtained Temperature and the Metamorphic Grade

Due to the pelitic origin of the La Aguadita Gneiss, Equation 5 was applied to calculate the maximum temperature. The maximum crystallization temperature is 472 ± 27 °C, corresponding to the upper zone of the green schist facies field in the P-T diagram, bordering the epidotic amphibolite facies (Fig. 6). The petrographic information is consistent with the temperature found: green hornblende predominant in the La Aguadita Gneiss lithologies corresponds to "medium temperatures" (Miyashiro, 1973). The predominant oligoclase with 20–30% An indicates low-pressure terrains. The lithologies have been affected by metamorphism corresponding to the epidotic amphibolite facies, with intermediate to low P/T ratio (Miyashiro, 1973), in the almandine zone (Turner, 1981), or low to medium metamorphic grade (Winkler, 1978). Maximum pressure is estimated at 0.4–0.5 GPa (4–5 Kb).

The temperature values obtained are significantly lower than those required in industrial crystalline graphite production. However, in the natural metamorphic environment, graphitization processes occur at lower temperatures: Landis (1971) notes that graphitization begins at at least 300 °C, with perfectly crystalline graphite at 450 °C. Diessel et al. (1978) specify that graphitization begins within the chlorite zone of the green schist facies and is completed before the amphibolite facies, at 380–450 °C for pressures of 0.2–0.3 GPa.

The temperature value is consistent with the metamorphic grade from mineral paragenesis. Limitations of the method:

a. The temperature value corresponds to the metamorphic peak or climax.
b. No reliable pressure estimator is available, introducing imprecision in P-T field placement.

One obvious limitation of the method is the requirement for graphite presence in the lithology. However, given its character as an accessory mineral in metasedimentary rocks that included organic matter, its presence is quite frequent. At least 0.5 wt% graphite, measured as percentage of total carbon, is required to apply the flotation and acid technique. Pure fractions are not needed: fractions with more than 30 wt% graphite yield satisfactory diffractograms.

Conclusions

The disseminated graphite present in the La Aguadita Gneiss consists of well-defined scales or laminae. Morphological analysis and XRD parameters characterize it as *well-crystallized graphite* with a high degree of molecular ordering.

The maximum crystallization temperature reached by the graphite is 470 ± 30 °C. This value, in association with general barometric indicators (% An of plagioclases and hornblende color), suggests that the metamorphism experienced by the La Aguadita Gneiss corresponds to the biotite zone, in the central to upper part of the Green Schist Facies.

The X-ray diffraction technique proved adequate for rapidly and non-destructively characterizing graphite. The information obtained permitted using graphite as a geothermometer in estimating the maximum temperatures reached by the rock containing this mineral.

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