

## Graphitic Hornfels Dikes in the Ronda High-Temperature Peridotite Massif

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### Abstract

The Ronda high-temperature peridotite body of southern Spain contains hornfels dikes with up to 16 wt percent graphite. These dikes, which occur in many places along the peridotite contact, are most conspicuous along the eastern edge of the Ronda massif. They contain abundant cordierite, garnet, and biotite and resemble hornfelses of the metamorphic aureole around the peridotite massif. Fe:Mg distribution between coexisting cordierite, garnet, and biotite in two dike specimens suggests equilibration at approximately 800°C. Stable carbon isotope ratios ( $\delta^{13}\text{C} = -20.8$  to  $-24.1$  per mil relative to the "PDB" belemnite from the Peedee formation of South Carolina) are compatible with a sedimentary hydrocarbon source for the graphite. The hornfels dikes are believed to be metamorphosed, organic-rich muds which entered fractures in the hot, solid peridotite body as it was emplaced into or onto crustal sediments. It is suggested that carbon was deposited by pyrolysis of hydrocarbons in the mud during metamorphism.

### Introduction

The Serrania de la Ronda peridotite intrusion is located on the south coast of Spain within the deformed internal zone of the Betic-Rif orocline (Fig. 1). Recent papers have described the petrology and geochemistry of the intrusion (Dickey, 1970); the petrology, structural geology, and K/Ar "age" of the contact metamorphic aureole (Loomis, 1972a,b,c); and the gravity field across the intrusion and southwards to Morocco (Bonini, Loomis, and Robertson, 1973). From these studies the following interpretation emerges:

(1) The Ronda peridotite massif is near the crest of one of two narrow ridges of high density material ( $\sim 2.9$  g/cm<sup>3</sup>) which lie along the western margins of the Alboran Sea. The southern ridge surfaces as the Beni Bouchera high-temperature peridotite intrusion in Morocco. Probably during early Miocene times both ridges intruded thick ( $>10$  km) sections of crustal rocks. The Ronda intrusion, which is contacted by marbles, hornfelses, and high-grade gneisses, is estimated to have intruded the crust at more than 800°C.

(2) The Ronda intrusion is 85 to 90 percent peridotite. In addition, the intrusion contains discontinuous sheets of mafic rocks (some of which

may have formed by partial fusion of the ascending mass) and a variety of discordant dikes.

The origins of the various dikes within the Ronda intrusion are not known. Some dikes may be indigenous to the intrusion, some may be related to exotic magma systems, and some may represent anatectic magmas formed by partial fusion of the country rocks during emplacement of the massif. This report describes a group of graphite- and cordierite-rich dikes which apparently originated by intrusion and metamorphism of hydrocarbon-charged mud during the final emplacement of the peridotite massif.

### Field Occurrence

Dark gray, graphitic dikes (5 cm to 2 m thick) occur in peridotite at the eastern end of the Ronda massif, most abundantly along the lower valley of the Rio del Hoyo del Bote near the village of Istán. The dikes occur as far as 2 km within the boundaries of the peridotite body; they have not been found in the country rocks. Typically arrayed along parallel joints, the dikes cross-cut layering and foliations in the peridotite. The dike contacts are sharp and, in places, coated with graphite. There is little or no evidence of marginal chilling.

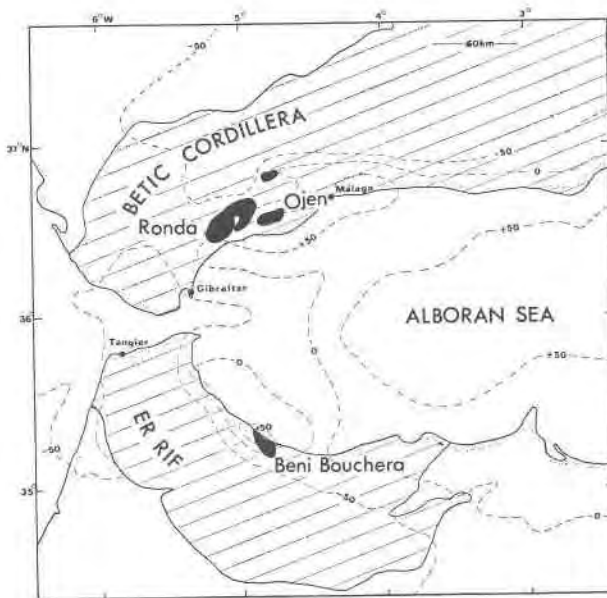


FIG. 1. Locations of the major peridotite massifs in the Betic-Rif orocline. Bouguer gravity anomalies (in mgal), shown as dashed lines, are from Bonini *et al* (1973). Positive anomalies as high as +110 mgal occur within the +50 mgal closure SE of the Ronda massif. Petrological descriptions of the Ronda, Ojen, and Beni Bouchera massifs can be found in Dickey (1970), Hernández-Pacheco (1967), and Kornprobst (1969) respectively.

The dikes vary in aspect from chaotic to ordered. The chaotic dikes form irregular, branching intrusions which bear xenoliths, angular fragments of

quartz (to 3 cm) and veinlets of quartz, sodic plagioclase, and pectolite. The xenoliths are gneisses from beyond the massif and mafic and ultramafic rocks which may originate within the massif. The ordered dikes contain few xenoliths, quartz fragments, or subsidiary veinlets; they have more regular, simple contacts with the peridotite.

### Petrography

The dikes closely resemble the high-grade hornfels which locally contacts the peridotite intrusion (Loomis, 1972a). Cordierite and quartz are the predominant minerals. Garnet, biotite, and ilmenite (up to 0.8 mm) lie in a groundmass of cordierite, micaceous material, quartz, and subordinate plagioclase. Accessory phases are K-feldspar, zircon, hercynite, rutile, pyrrhotite, and other minor sulfides.

Graphite is an essential constituent of all the dikes. The chaotic and the ordered dikes (each represented by one specimen) differ in graphite concentration and graphite distribution. The chaotic specimen (Fig. 2), collected from a dike 400 m inside the peridotite body, contains a jumble of small (5 mm) polycrystalline aggregates rich in garnet and cordierite. In this specimen, graphite constitutes 16 wt percent of the rock and is concentrated around the margins of the polycrystalline aggregates. This graphite is xenomorphous but produces sharp X-ray diffraction peaks. The more homogeneous specimen (Fig. 3), collected from a dike 1.5 km inside the

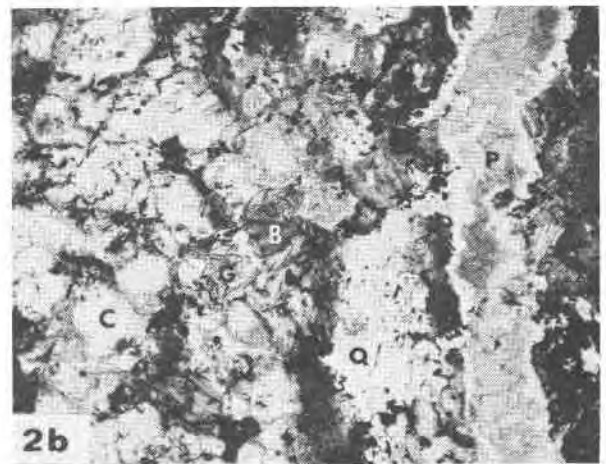
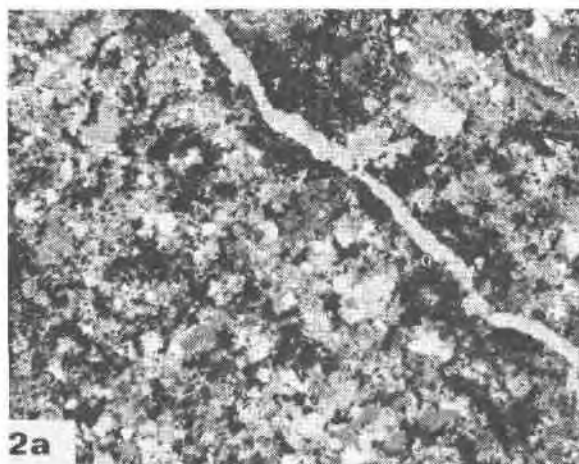


FIG. 2. Photomicrographs of the chaotic dike (R267). a. Wide field view with oblique nicols. Note the veinlet of pectolite, plagioclase, and quartz. Much of the opaque material is graphite. b. Detailed view in plane polarized light. Prominent phases are garnet (G), cordierite (C), and biotite (B). Pectolite (P), plagioclase, and quartz form a veinlet that traverses the field. The murky to opaque material is a mixture of micaceous material, graphite, ilmenite, and sulfides.

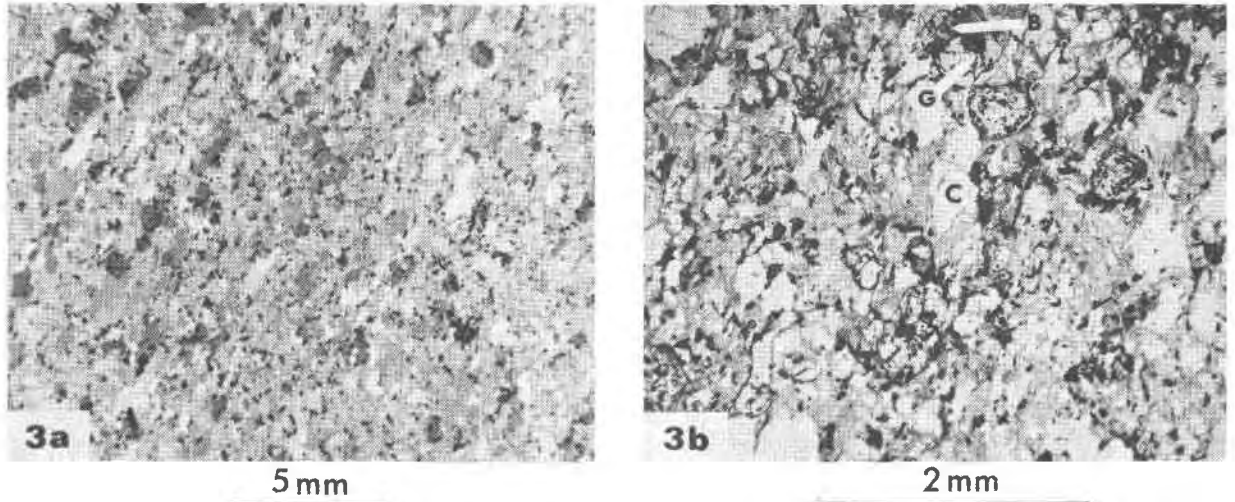


FIG. 3. Photomicrographs of the ordered dike (R269). a. Wide field view with oblique nicols. Note the dimensional and optical preferred orientation. b. Detailed view in plane polarized light. Prominent phases are garnet (G), cordierite (C), and biotite. Most of the opaque material is ilmenite. Graphite occurs as sharp flakes.

peridotite body, has an even hornfelsic texture with mafic and opaque phases lying in a cordierite-rich groundmass. Sharp flakes of graphite (2 wt percent of the rock) are distributed uniformly throughout. No xenoliths are present. The rock shows dimensional and optical preferred orientation of grains parallel to the dike margins.

**Mineral Compositions**

*Analytical Method*

Mineral analyses were performed on an automated MAC-5 electron microprobe programmed with the Geolab System (Finger and Hadidiacos, 1972) running at 15 kV accelerating voltage, 0.02 to 0.05  $\mu$ A specimen current, and counting times of 20 to 30 sec. Silicate and oxide analyses were corrected by the method of Albee and Ray (1970). Sulfide analyses were corrected by the method of Boyd, Finger, and Chayes (1968). Synthetic silicate glasses, synthetic sulfides, and natural chalcopyrite were used as standards. Data are reported to three significant figures for concentrations of 10.0 wt percent or more, two figures for concentrations of 1.0 to 9.9 percent, and 1 figure for concentrations below 1.0 percent. Observed concentrations below 0.5 percent are reported as "tr." All iron is reported as divalent. The tabulated data are averages of 1 to 36 analyses.

*Silicates (Table 1)*

The garnets are almandine-rich and generally similar in both the chaotic and the ordered speci-

mens, except that the chaotic specimen (R267) shows higher average MnO and a wider range of MnO concentrations (1.1 to 7.0% vs 1.1 to 1.3%). In R267, Mn varies inversely with Fe and is concentrated in the interiors of some crystals. The garnets are otherwise uniform in both specimens.

Biotites in both rocks are virtually identical. The mineral is characterized by rather high concentrations of Fe, Al, and Ti.

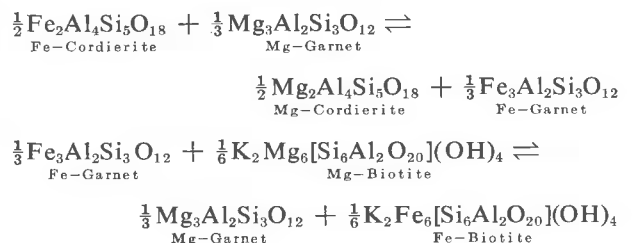
Cordierites in the chaotic and the ordered specimens are also very similar. The high summations indicate low water contents.

Plagioclase compositions vary from calcic labradorite to sodic bytownite. The greatest variation occurs in the chaotic specimen (R267) which also contains nearly pure albite in veinlets.

Traces of K-rich feldspar were found by chance with the microprobe. The phase occurs as small grains in the groundmass. The analysis from R269 may be contaminated with extraneous material.

*Fe-Mg Partitioning*

Distribution coefficients for the reactions:



based upon the average mineral compositions (Table 1) are given in Table 2. According to the interpretation of data of Hensen and Green (1971, 1972) and Currie (1971) by Wood (1973), the observed distribution of Fe<sup>2+</sup> and Mg between cordierite and garnet indicates equilibration at 625°C to 775°C depending upon  $P_{H_2O}$ . According to Saxena's (1969) interpretation of Fe-Mg distribution between garnet and biotite, the equilibration temperatures are near 800°C. The temperature estimates are closest for  $P_{H_2O}$  between 1 and 3 kbar. These temperature estimates are compatible with Loomis's (1972b) data for the contact aureole. The slightly higher temperature indicated for R269 than for R267 is qualitatively reasonable, because R269 crystallized deeper within the peridotite body; such fine distinctions, of course, may be meaningless.

#### Oxides (Table 3)

Ilmenite is the most abundant oxide in both specimens. It is more abundant in the ordered specimen (R269) than in the chaotic specimen (R267). The mineral generally contains rather high concentrations of MnO which increase toward the margins of grains. Small amounts of TiO<sub>2</sub> are commonly intergrown with the ilmenite. Hercynite was identified in R267 as a discrete grain and probably as small inclusions in cordierite of both specimens.

#### Sulfides (Table 4)

Pyrrhotite dominates with subordinate amounts of chalcopyrite and pentlandite. Traces of cubanite were also found. Although the sulfides are similar in both rocks, all varieties are more abundant in the chaotic specimen (R267).

TABLE 1. Silicate Compositions

	Garnet		Biotite		Cordierite		Plagioclase		K-Feldspar	
	R269	R267	R269	R267	R269	R267	R269	R267	R269	R267
SiO <sub>2</sub>	38.1	37.8	33.0	33.2	49.3	48.7	50.8	48.0	65.4	66.5
TiO <sub>2</sub>	0.1	tr	4.7	4.8	tr	tr	0.0	0.0	0.0	0.0
Al <sub>2</sub> O <sub>3</sub>	20.5	21.0	17.4	17.5	32.4	32.6	30.6	33.5	16.8	18.3
Cr <sub>2</sub> O <sub>3</sub>	0.3	0.2	0.3	0.7	tr	tr	tr	0.0	0.2	0.0
FeO*	35.1	34.1	23.0	22.8	11.2	11.5	0.1	0.3	0.2	tr
MnO	1.2	2.2	0.1	0.1	0.1	0.2	tr	0.0	tr	0.0
MgO	3.4	3.0	6.7	6.6	6.2	6.4	tr	0.0	0.2	0.0
CaO	1.6	1.6	0.0	0.1	0.05	0.1	14.4	16.7	1.5	tr
Na <sub>2</sub> O	0.0	0.1	0.3	0.3	0.0	0.2	3.5	2.0	1.6	0.1
K <sub>2</sub> O	0.0	tr	8.2	8.6	0.0	tr	0.3	0.1	13.9	16.5
Total	100.3	100.0	93.7	94.8	99.25	99.7	99.7	100.6	99.8	101.4
Cations Per Formula Unit										
Si	3.04	3.03	5.46	5.44	5.08	5.01	2.32	2.19	3.02	3.02
Ti	0.005	0.002	0.58	0.59	tr	tr	0.00	0.00	0.00	0.00
Al	1.93	1.98	3.39	3.38	3.93	3.96	1.65	1.80	0.914	0.981
Cr	0.02	0.01	0.04	0.09	tr	tr	0.00	0.00	0.007	0.00
Fe*	2.35	2.29	3.18	3.13	0.963	0.995	0.004	0.01	0.007	tr
Mn	0.082	0.15	0.01	0.01	0.007	0.01	0.00	0.00	tr	0.00
Mg	0.41	0.36	1.6	1.6	0.95	0.98	0.00	0.00	0.01	0.00
Ca	0.14	0.14	0.00	0.02	0.005	0.007	0.706	0.815	0.074	tr
Na	0.00	0.01	0.1	0.09	0.00	0.04	0.31	0.17	0.14	0.009
K	0.00	tr	1.7	1.8	0.00	tr	0.02	0.005	0.819	0.957
Total	7.977	7.972	16.06	16.15	10.935	11.002	5.010	4.990	4.990	4.967
Oxygens	12	12	23	23	18	18	8	8	8	8
Analyses	10	36	6	15	3	36	10	5	1	1

\* Total Fe as FeO.

TABLE 2. Fe/Mg Distribution Coefficients

	R267	R269
$K_D$ (Gn/Cd)	6.3	5.7
$K_D$ (Gn/Bi)	0.30	0.33
Trans. $K_D$ (Gn/Bi)*	-0.093	-0.061

\* The Transformed  $K_D$  of Saxena (1969) is an empirically derived function which incorporates the concentrations of Ca and Mn in garnet and Al and Ti in biotite as well as Fe/(Fe+Mg).

Niccolite, other arsenides, and chromite were described by Oen (1973) in cordierite-bearing veins and lenses in the Ojen ultramafic massif near Málaga (Figure 1). Similar phases have not been found in the Ronda dikes (except for chromite in ultramafic xenoliths). The maximum observed concentration of As is 0.1 wt percent in ilmenite.

Carbon Isotopes

Stable carbon isotopic ratios of graphite from the dikes were determined by the Geochron Laboratories, Cambridge, Massachusetts. Their results, expressed as  $\delta^{13}C$  relative to PDB, are: center of chaotic dike (R267), -24.1 ‰; graphite coated contact surface of chaotic dike (R267), -20.8 ‰; ordered dike (R269), -21.3 ‰. The precision of these data is  $\pm 0.5$  ‰ (R. Reesman, personal communication). Compared with other carbon isotope data (Fig. 4), the data for the Ronda dikes fall within the overlapping ranges for reduced carbon in many terrestrial rocks. They are similar to ratios found in petroleum and some natural gases.

TABLE 3. Oxide Compositions

	Ilmenite Hercynite			Cations Per Formula Unit			
	R269	R267	R267	R269	R267	R267	
SiO <sub>2</sub>	1.8	1.3	0.7	Si	0.045	0.03	0.02
TiO <sub>2</sub>	51.1	51.4	0.1	Ti	0.961	0.973	0.002
Al <sub>2</sub> O <sub>3</sub>	0.3	0.3	59.3	Al	0.008	0.008	1.98
Cr <sub>2</sub> O <sub>3</sub>	0.2	0.0	0.2	Cr	0.004	0.00	0.004
FeO*	44.9	44.4	35.5	Fe*	0.939	0.935	0.840
MnO	1.3	1.3	0.2	Mn	0.027	0.026	0.005
MgO	0.1	0.2	3.2	Mg	0.003	0.008	0.13
CaO	tr	0.1	0.1	Ca	tr	0.003	0.003
Total	99.7	99.7	99.3	Total	1.987	1.983	2.984
				Oxygens	3	3	4
				Analyses	2	13	1

\* Total iron as FeO.

TABLE 4. Sulphide Compositions

	Pyrrhotite		Chalcopyrite		Pentlandite	
	R267	R269	R267	R269	R267	R269
S	39.5	38.6	36.2	36.1	34.9	34.0
Fe	60.5	61.0	29.6	28.2	35.6	35.6
Co	0.2	0.2	0.1	0.1	3.8	0.3
Ni	0.2	0.1	0.3	0.2	22.3	24.6
Cu	0.1	tr	32.1	33.5	1.7	0.3
Total	100.5	99.9	98.3	98.1	99.0	98.4

Discussion

The concentration of graphitic hornfels dikes near the eastern edge of the peridotite massif, the similarity of these dike rocks to the contact hornfels, and the carbon isotope ratios indicate that the dikes intruded the peridotite from a crustal source, as suggested by Loomis (1972a). The source material must have contained substantial quantities of carbon and have been mobile at temperatures below 800°C. Partial fusion of argillaceous sediments of pelitic

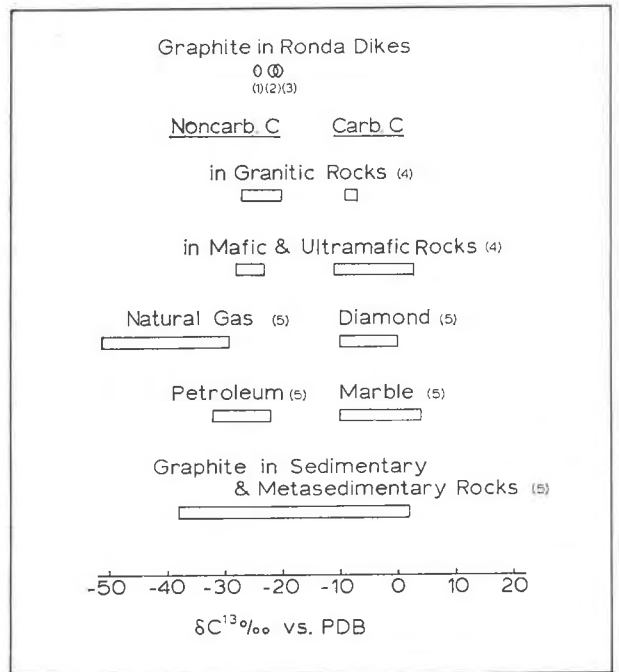


FIG. 4. Stable carbon isotope ratios, expressed as  $\delta^{13}C$  relative to PDB, of the graphite-bearing dikes from Ronda and a variety of terrestrial materials. Key: 1. graphite from center of chaotic dike (R267); 2. graphite from ordered dike (R269); 3. graphite from graphite-coated surface of chaotic dike (R267); 4. data from Fuex and Baker (1973); 5. data from Schwartz et al. (1969). Carb. C = carbon in carbonate. Noncarb. C = reduced carbon.

schists is possible at such temperatures in the presence of water; however, the liquids so produced would be granitic, and the unmelted residue would be rich in garnet, cordierite, and calcic plagioclase (Grant, 1973). Garnet and cordierite are prominent constituents of these dikes; alkali feldspars are not. Furthermore, the composition of the ordered dike is somewhat more refractory than the composition of an average shale (Table 5). The dikes, therefore, were probably not formed by anatexis of pelitic rocks.

Perhaps the graphitic dikes formed as intrusions of unmetamorphosed, unconsolidated argillaceous sediments (muds) which were metamorphosed to hornfels after emplacement in the peridotite. During this thermal metamorphism, small amounts of calcium, aluminum, alkali metals and silica were mobilized and formed veinlets of quartz, plagioclase, and pectolite.

The source of the carbon might have been sedimentary carbonates or hydrocarbons. The low calcium concentrations and light carbon isotope ratios, however, favor sedimentary hydrocarbons as the principal source. Sedimentary hydrocarbons may have contributed force as well as substance to the intrusions: many deeply buried argillaceous sediments contain large quantities of natural gas, chiefly methane. Not uncommonly, fluid pressures in such sediments exceed the hydrostatic pressure and, in some instances, even approach the lithostatic pres-

sure (Rubey and Hubert, 1959). If the rising Ronda peridotite intrusion intruded such a hydrocarbon-rich, uncompacted and overpressurized sediment (a so-called "heaving shale"), hydrocarbons and mud could have been injected deep into fractures in the hot, solid peridotite. The forcible intrusion of hydrocarbon-charged sediments may have occurred at substantial depth: Hedberg (1974) states that undercompacted (overpressurized) shales are known to occur at depths of 5 km.

Among the first reactions to occur as the mud entered the peridotite would be pyrolysis or cracking of the hydrocarbons to produce carbon and hydrogen. If the hydrocarbon source was natural gas the principal reaction would be:



Pyrolysis of methane or natural gas to produce carbon is a well-established commercial process (Smith and Leeds, 1970). Commercial pyrolytic graphite is deposited at temperatures above 1750°C upon substrates of refractory metals, graphite, or metal compounds such as carbides. Such high temperatures are not always required, however; pyrolytic graphite has been deposited upon transition metals below 1000°C (Robertson, 1969). To be certain that natural gas would crack when passed over peridotite at relevant temperatures, we passed natural gas over crushed harzburgite (<100 mesh) at 800°C, 1 atm. After 4 hours the grains of olivine, pyroxene, and spinel were completely coated with carbon. This experiment did not produce graphite, however. The carbon deposited is amorphous to X-rays. Perhaps annealing at high temperatures (600° to 800°C) for long periods of time would produce crystalline graphite like that in the Ronda dikes.

A positive correlation between graphite and sulfide concentrations is predictable if the sulfides formed by reactions between sulfur compounds in the natural gas, principally H<sub>2</sub>S, and the mud. Such a relationship was observed in the two specimens studied.

A negative correlation between graphite concentration and distance of dike injection would result if pyrolysis of hydrocarbons caused deposition of carbon immediately after the mud entered the hot peridotite. Such a relationship is also apparent in the two specimens studied.

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TABLE 5. Bulk Composition of a Hornfels Dike Compared to an Average Shale

	a	b
SiO <sub>2</sub>	57	58.38
TiO <sub>2</sub>	1	0.65
Al <sub>2</sub> O <sub>3</sub>	20	15.47
FeO*	10	6.06
MgO	4	2.45
CaO	1	3.12
Na <sub>2</sub> O	0.6	1.31
K <sub>2</sub> O	0.6	3.25
C	2.1	0.81

\*Total Fe as FeO.

a) R269. Semi-quantitative analysis based on 120 defocussed-beam microprobe analyses, x-ray fluorescence data, and differential leaching.

b) Average shale (Clark, 1924).

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