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Decarbonation Reactions and the Origin of Vein-Graphite in Sri Lanka

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Abstract : The central question in this problem of the origin of vein-graphite is the source of the carbon. This question can only be viewed against the metamorphic background of the rocks of the island. The CO_2 derived from decarbonation reactions in the calcareous environment provided an adequate source of the required carbon and is postulated to have entered into rock-structures such as joint-planes, etc., and reacted with the country rocks to produce carbon. The practical significance of this theory is that we can now recognise the geological environment in which vein-graphite can occur.

1. Introduction

The two main modes of occurrence of graphite in Sri Lanka are (1) as disseminated flakes in the country rocks such as granulites, charnockites, crystalline limestones, graphite schists, etc. forming the Highland Series, and (2) as veins occupying fractures in the rock. The disseminated flakes evidently represent metamorphosed organic carbonaceous matter in the original rocks. Vein-graphite is not of the same origin because it is not an original rock-forming mineral as the disseminated graphite but occurs in the rock structures such as joints and faults, mostly joints, oblique, transverse or longitudinal in orientation and is therefore of later origin.

It will be of interest to compare the C12/C13 ratios for vein-graphite of inorganic origin and for disseminated graphite in the country rocks (charnockite-metasedimentary series) evidently of organic origin.

Some of the earlier views regarding the origin of graphite are contained in a paper by Wadia[9] and more recently by Erdosh.[2] Wadia postulated the theory of absorption of limestone by charnockitic intrusions resulting in the production of various lime-silicates and pyroxenes in the magma and the elimination of carbon in gaseous or volatile form. Wadia, in keeping with the geological thinking of the times, regarded all charnockitic rocks as being intrusive into what was then known as rocks of the Khondalite system. Regional geological mapping carried out by the Geological Survey Department during the last 20 years has shown that the charnokites, with the exception of a few minor local occurrences, bear no intrusive relationships to the quartites, granulites and marbles of the Khondalite Series and occur interbanded and conformable with these rocks to form what was at first known as the charnockite-metasedimentary series and subsequently termed the Highland Series.

Charnockites have thus been shown to form an inseparable part of one and the same succession. Therefore, the very premise from which Wadia attempted to build up his theory is shown to be erroneous and his views cannot therefore be taken into consideration to explain the origin of vein-graphite.

2. Decarbonation reactions

The central question in this problem of the origin of vein-graphite is the source of the carbon, a question which can only be dealt with in a background of elucidation of the metamorphic processes to which the rocks of the island have been subjected. These processes are marked by metamorphic reactions in different petro-chemical environments such as pelitic, calcareous and basic, only the decarbonation reactions in the calcareous environment bearing relevance to this question.[4,5,6,7,1,8]

In addition to calcite and dolomite, the crystalline limestones (marbles) of the Highland Series consist of a variety of minerals such as forsterite, diopside, phlogopite, spinel, apatite and graphite and some of the mineral assemblages recognised are as follows :—

1. Calcite ± dolomite — forsterite.
2. Calcite ± dolomite — forsterite — diopside.
3. Calcite ± dolomite — forsterite — phlogopite.
4. Calcite ± dolomite — forsterite — diopside — phlogopite.

Spinel, apatite or graphite may be additional minerals in any of these assemblages.

The calc-granulites and gneisses consist of different combinations of minerals such as diopside, scapolite, calcite, wollastonite, sphene and perthite. The relevant assemblages are only those which bear wollastonite and these assemblages are as follows :—

1. Diopside — scapolite — wollastonite ± sphene.
2. Diopside — scapolite — wollastonite — perthite ± sphene.

Considering both the 1st and 2nd periods of granulite facies metamorphism to which the rocks of the island have been subjected, the type of decarbonation reactions involved in the production of the above assemblages are as follows :—

$$\underset{\text{dolomite}}{2CaMg(CO_3)_2} + \underset{\text{quartz}}{SiO_2} \rightleftharpoons \underset{\text{calcite}}{2CaCO_3} + \underset{\text{forsterite}}{Mg_2SiO_4} + 2CO_2 \quad \text{......... A.}$$

$$\underset{\text{forsterite}}{Mg_2SiO_4} + \underset{\text{calcite}}{2CaCO_3} + \underset{\text{quartz}}{3SiO_2} \leftrightharpoons \underset{\text{diopside}}{2CaMgSi_2O_6} + 2CO_2 \quad \text{......... B.}$$

$3CaMg(CO_3)_2 + KAlSi_3O_8 + H_2O \rightleftharpoons (OH)_2KMg_3AlSi_3O_{10} + 3CaCO_3 + 3CO_2$
dolomite K'feldspar phlogopite calcite .... C.

$CaCO_3 + SiO_2 \rightleftharpoons CaSiO_3 + CO_2$ .... D.
calcite quartz wollastonite

3. Discussion of the CO_2 — producing reactions and source of the carbon.

The complete elimination of dolomite and quartz in reaction A resulted in calcite-forsterite with release of CO_2. The elimination of quartz in A resulted in calcite-dolomite-forsterite with release of CO_2. The elimination of quartz in reaction B resulted in the assemblage calcite-forsterite-diopside with CO_2 released. The elimination of dolomite in reaction A resulted in the association calcite-forsterite-quartz with release of CO_2. This assemblage is unstable in the pyroxene granulite subfacies, forsterite reacting with calcite in the presence of quartz to yield diopside and CO_2 according to reaction B ; this reaction proceeded until quartz was completely eliminated resulting in the assemblage calcite-forsterite-diopside with CO_2 released. Thus, calcite-forsterite-quartz apparently stable in the garnet-diopside-quartz subfacies was converted to calcite-forsterite-diopside (with CO_2 released) through reaction B under conditions of the pyroxene-granulite subfacies. Any potash feldspar present in forsterite-bearing marbles (pyroxene-granulite subfacies) would have reacted with dolomite in the presence of water to yield phlogopite with release of CO_2 according to reaction C under conditions of the hornblende-granulite subfacies. This reaction ceased when all the potash feldspar was consumed resulting in the association calcite-dolomite-forsterite-phlogopite. The elimination of calcite and quartz in reaction D resulted in the production of wollastonite with CO_2 released.

From the above mentioned reactions it is important to note : (1) that the CO_2 released provided an entirely adequate source of the carbon required for the production of vein-graphite, (2) that these reactions produced minerals such as forsterite, diopside, phlogopite and also wollastonite, the significance of which is explained below.

The CO_2 derived from the reactions found its way into joint-planes, fault-planes, etc. and reacted with the country rock with which it came into contact to produce carbon by means of some reaction, at present not known ; however, the type of carbon-producing reaction suggested, is as follows :

$CO_2 + 4FeO = 2Fe_2O_3 + C$.[3] Such a mechanism is postulated to explain the origin of vein-graphite.

Impure marbles bearing forsterite, forsterite-diopside, forsterite-phlogopite or forsterite-diopside-phlogopite and wollastonite-bearing calc-granulities are most significant because they have been shown to be the source of the CO_2 which in turn provided the source of the carbon required for the formation of vein-graphite.

That is, a genetic relationship is clearly seen between calcareous rocks such as forsterite, forsterite-diopside, forsterite-phlogopite or forsterite-diopside-phlogopite bearing marbles and wollastonite-bearing calc-granulites on the one hand and vein-graphite on the other, thus implying that the environment in which vein-graphite occurs is marked by the presence of such calcareous rocks, although such rocks may not always be seen in the vicinity of graphite veins, in which case, they may well be found to occur at depth, as for example at the Bogala Mines at Kotiyakkumbara and also, as recent investigations showed, at Katuwana, near Morawaka. Wadia[9] too, had observed a close association of calcareous strata with graphite bodies.

4. Practical significance

The geological environment in which vein-graphite can occur must satisfy either or both of the following requirements : (1) the environment is marked by the presence of impure marbles bearing minerals such as forsterite, forsterite-diopside, forsterite-phlogopite or forsterite-diopside-phlogopite, especially when these minerals are present in larger amounts. It is noted that pure marbles or impure marbles bearing minerals other than those above mentioned (i.e. spinel, apatite or sphene) are of no significance from the point of view of vein-graphite; (2) the environment is marked by the presence of wollastonite-bearing calc-granulites.

The practical significance of this theory is that we can now recognise the geological environment, as defined above, in which vein-graphite can occur and that any search for commercial graphite bodies either on the surface or in depth should be confined to this particular environment, especially when marbles show an abundance of the above mentioned minerals.

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