CHAPTER VI

ORIGIN OF GRAPHITE DEPOSITS

In this chapter the author has discussed in detail, the nature of the graphitic rocks in Gujarat and has drawn certain conclusions regarding their probable mode of origin.

Detailed investigations of graphites and graphitic rocks of Gujarat revealed two distinct varieties of graphite - one is the amorphous and the other is flaky. Amorphous graphite is disseminated in the arenaceous and argillaceous metasediments where microflakes or grains of graphite are oriented parallel to the schistosity of the rocks that are exposed around the villages - Ankli, Sewania, Muthai, Chaena, Narukot and Jaloda. Flaky graphite, on the other hand, is found disseminated in arenaceous meta-sedimentary rocks near Virpur.

Interpretations based on field studies and laboratory investigations

The following evidences mainly based on field investigations, indicate that the graphite bands intercalated with graphitic schist form a single stratigraphic unit.

1. The parallelism of the bands/layers or seams of graphite with the bedding of crystalline limestone (Jhab-Redhana, Ankli) or with bedding of quartzite (Nodatod, Narukot) and quartz-mica schist (Virpur).

2. The parallelism of the strike of the graphite bands with that of the bedding foliations of the graphite bands and graphitic schist.

3. Uniformity in the foliations of graphitic rocks for a considerable distance from the intrusive granite contact.

4. Absence of larger concentration near the contact zone and complete absence of crystalline graphite bearing veins in the graphite schist.

The association of limestone with graphite schist leads one to believe that they are genetically related or are the transformed and released products by their contacts with the introducing granites.

Lakshminarayan (1969), on the basis of his preliminary studies, has come to the conclusion that the absence of organic evidences in the rocks of the Sewania and Ankli localities rules out the possibility of the organic origin of the graphite. According to him, the graphite deposits are the result of contact metamorphism of limestone and are of inorganic origin.

It is an established fact that, algal and other primitive plant life were existing during the Pre-Cambrian time. The thin section studies of the graphite, indicate that the grade of metamorphism of these sediments varies between low to moderately high. According to Turner and Verhoogen (1962), the temperature during such metamorphism attains a maximum range of 550° to 700°C. The delicate plant under these conditions will not be able to retain their structures. The transformation to graphite, however, under such conditions may not be impossible.

The present investigations, on the other hand do not indicate any such limestone-granite contacts at Ankli, Sewania, Virpur and Muthai as claimed by Lakshminarayan. On the other hand, it is observed that the limestone formation is separated by a few meter thick formation of graphitic schist. If limestone was expected to be the source rock and the graphite was to be released near the contacts of the granitic intrusions, it would be logical to expect some unassimilated calcite in the matrix of the rocks developed near the contact zone. Thin section studies of graphite-bearing rocks of Ankli, Muthai, Jaloda, and Virpur show complete absence of calcite in the matrix. Calcite, however, occurs as small stringers and small veinlets cutting across the schistosity of the graphitic rocks. The flakes of graphite are developed perpendicular to the vein walls. Hence, the possibility that during the contact metamorphism limestone was dissolved and lime-silicates and carbon dioxide were liberated and further reduced to form graphite in the presence of hydrogen, can be ruled out.

According to Ogilvie (1904), the widespread dissemination of graphite flakes in sedimentary limestone and quartzite can best be explained by the organic hypothesis. Regional metamorphism of the original constituents of the rocks can give rise to the formation of graphite. According to Clark (1921), the bedded deposits of graphite that lie concordant with the beddings in sedimentary rocks, are the result of contact metamorphism of carbonaceous sediments in which heat seems to have been the most important factor, since all of the volatile constituents of the original carbonaceous matter must be dispersed in order to allow the formation of graphite.

At Ankli, Sewania and Muthai, the sharp contacts of the quartz veins, pegmatitic veins and granite with graphite-bearing rocks, the absence of graphite in the contact rocks viz. granite and the foliations almost perpendicular to the instrusive contact, give evidences against the theory of the origin of graphite due to lateral secretion.

Another possibility of the assimilation of graphite through igneous sources is suggested by Bastin (1912). According to his suggestion, the carbon was a primary element of igneous magma. So far, the graphite in Gujarat, are concerned they are associated with the metasedimentary rocks viz. phyllite, schist, gneiss, crystalline limestone and quartzite. The granite and pegmatite occurring across the foliations of the enclosed graphitic rocks show sharp contact and distinctly intrusive character while the bedding of metasedimentaries are parallel to the foliations of the graphitic rocks. The above observations rule out the possibility of the graphite forming carbon being the primary element that was released and deposited during the late stages of consolidation of the pegmatite magma. Winchell (1911), suggested that the carbon, on account of its infusibility and insolubility, does not exist in magmatic solutions. There are, thus no convincing evidences to believe that graphite was a primary element of pegmatitic or granitic magma in Gujarat.

Clark (1921), from his laboratory experimentations has brought out series of chemical equations to show that the oxides of carbon (gases), in the presence of gaseous water, react to form graphite. Much of this water is magmatic while some of it may be derived from the sediments. This may partly be the case in the graphitic rocks investigated and as evidenced from the occurrence of graphite in large amount at the margins of pegmatites. The heat released from the intrusive, possibly acted as some sort of agent to form the graphite. At Muthai, Ankli, Virpur and Sewania the pegmatitic intrusion is very minor in relation to the graphite bearing schists. The foliations in the rocks are almost perpendicular to the intrusive contact. Flakes or grains of graphite are oriented parallel to the foliations. In general, there is an absence of larger concentration of graphite at the margins of pegmatites. All these observations do not support Clark's theory of the origin of graphite.

According to Winchell (1911), the extremely refractory nature of graphite will not allow its existence in the form of liquid or gaseous carbon in the molten magma. The carbon which crystallize~~y~~s to form graphite in pegmatites is drawn from the silicate solutions that contributed the oxides and the reduction of these oxides to reduce the necessary carbon to form graphite. This may be true for the graphite originating in pegmatites and not for the graphite disseminated in phyllite, schist and gneiss. Winchell's suggestions may be partially true in such graphite in Gujarat where the heat emanated by the igneous intrusion could have been responsible for the conversion of indigenous organic carbonaceous matter already present in the original sediments to be converted into graphite.

Another possibility of graphite formations, according to Winchell (1911), is due to sublimation of carbon. In such cases the temperature of liquification and vaporization of graphite are known to be about 3000°C. The rocks containing graphite and the associated minerals in the area under study are characterized by the flakes of graphite that are evenly distributed and closely associated with quartzose matrix and other flaky minerals, forming the schistose structure. At Ankli, Muthai and Virpur, calcite is absent in the matrix of the rocks but occurs as veins, cutting across the schistosity. The flakes of the graphite are almost perpendicular to the vein wall. All these evidences indicate that the rocks in Gujarat were not subjected to any such temperature variations as suggested by Winchell.

The graphitic rocks of the above localities have been considered by Gupta and Mukherjee (1938) to be of magmatic origin, mainly because of the intrusive nature of the associated rocks and the development of pegmatites and aplites representing the residual products of the differentiation of magma (Post-Aravalli). Merh, Jambusaria and Patel (1968) have recognized the intrusive granites as the last phase of the granitic intrusion in Aravalli rocks of Gujarat. The absence of graphite in granite, sharp contact of granite with graphite-bearing formations, distinctly intrusive nature of granite and the fact that graphite is not known to exist in magmatic solution, rules out the possibility of granitic magma as a source of graphite in these areas. ?

The Sewania graphite deposits show calcareous matrix and minor proportions of associated silica. Crystalline limestone bed is not observed anywhere on the surface in contact with granite. The graphite bearing layers are parallel to the yellowish white sandy bands which are devoid of calcite and are of varying thickness ranging from 2" to 1½ feet. Thin section study reveals that graphite flakes are oriented uniformly along with other flaky minerals to form the schistose structure. Calcite occurs in the matrix with unequal grains of quartz in which the grains of calcite are very difficult to identify. Alternate layers of graphite along with mica flakes and calcareous bands are also noticed. From the above evidences, it can be concluded that graphite in the form of indigenous carbonaceous matter was scattered irregularly in impure calcareous to argillaceous sediments. It is, therefore, postulated that due to the low grade regional metamorphism, the schistosity was developed in the rocks, and its heat and pressure were responsible for the formation of graphite and other metamorphic minerals viz. chlorite, muscovite and biotite.

In the graphite deposits of Narukot, amorphous graphite occurs in phyllite. The associated rock is quartzite, which forms a definite member of the meta-sediments. The rocks are affected by regional metamorphism. According to Jambusaria (1970), the metamorphism of the original argillaceous to arenaceous sediments containing carbonaceous matter, has given rise to the formation of graphitic phyllite.

In addition to the known occurrences there are many places where isolated patches or bedded veins or pockety masses of graphite deposits are observed. According to the investigations of Cirkel (1907), most of such occurrences and the resulting graphites are in all or in part developed from the original carbonaceous matter contained in the sediments.

The graphite from all areas except Virpur is amorphous in nature and occurs in the form of minute particles or microflakes (microcrystalline). At Virpur the graphite is of little flaky type, disseminated in schist and gneiss. The percentage of graphite varies from 5 to 22 per cent. There is considerable variation in the flake size, thickness and shape of graphite particles from one deposit to another, depending upon the degree of metamorphism. The schist and gneiss of this region represent the ancient arenaceous and argillaceous sediments that have been transformed under the influence of heat and pressure. The graphite content of such amorphous deposits mainly depends on the amount of carbon originally present in the sediments. Variations in the graphite content of the formation along their strike and dip could be due to depositional irregularities. Quartz is the major constituent in all the graphite bearing rocks.

Three varieties of quartz are observed in the graphite schist. Both strained and unstrained quartz are present in schists, while the milky white quartz occurs in the younger veins that are developed along and across the schistosity of the major rock body. There is no evidence of any enrichment due to the intrusive rocks which have acted as agents of metamorphism. There are no convincing evidences supporting the origin of graphite from limestone. It is, therefore , clear that the graphite has originated from carbonaceous sediments containing some organic floral elements. Newland (1907), has sited the distillation of such organic compounds and the loss of their volatile matter to the effects of metamorphism.

The graphite at Chaena and Jaloda is amorphous and very fine grained. It is even finer than that of Muthai, Ankli, Sewania and Virpur. According to Newland (1907), very fine to medium-sized, flakes of graphite, tending towards amorphous graphite, occurs in metasediments that contained organic remains. According to Winchell (1911), the graphitic layers in metamorphosed rocks viz. quartz-schist and marble, represent the metamorphosed carbonaceous layers resulting in very fine-grained and impure amorphous graphite. He also believes that more intense metamorphism would produce coarser and purer graphite. The same is found true for the graphite deposits of Gujarat that are presently investigated.

In physical appearance, the graphite bearing rocks show foliated nature and well developed schistose structure, which are to be the most important characteristics developed in the regionally metamorphosed rock. On the basis, of graphite flakes and mineral assemblages present in graphite deposits, it can be inferred that those consisting of micro-graphite flakes (amorphous) are the result of low-grade metamorphism (chlorite zone) and those having well-developed flakes of graphite (flaky) are due to medium to high grade metamorphism (biotite-zone).

At Ankli, Virpur and Muthai contact metamorphic effects are observed on regionally metamorphosed graphitic schist. The graphitic schist shows gneissic character near the contact of intrusive granite. The quartz-mica-graphite schist is medium grained near the contact while away from the contact as at Chaena and Kundal, it is fine grained. The presence of tremolite and serpentine in crystalline limestone indicates contact metamorphic effects due to granite e.g. at Ankli and Virpur.

The intimate intergrowth of graphite with quartz and feldspar gives an evidence that graphite has not been added to the rock. Graphite did not exist in the silicate solution in the form of crystal flakes is indicated by the evidence that some of the constituents of the rock are enclosed by graphite and also by the fact that the graphite is uniformly distributed throughout the rock. Graphite did not exist in the silicate solution in the form of liquid or gaseous carbon is indicated by its refractory character and its practical insoluble nature in molten magma at the ordinary temperature attained by magma.

Proximate analysis (Table I) indicate that volatile matter present in the samples containing amorphous graphite varies from 1 to 6 per cent. The amount of volatile matter in the rock also depends on the effect of metamorphism. The low-grade, regionally metamorphosed rocks containing amorphous graphite have varying amounts of volatile matter due to the non-uniform temperature-pressure effects on the carbonaceous sediments. The volatile matter of the samples containing flaky graphite varies from 2 to 2.5 per cent. The deposits containing flaky graphite indicate the effect of medium to high grade metamorphism.

The chemical analysis (Table III) of graphite schist brings out the following points:

1. The percentage of silica is high,
2. MgO does not exceed over CaO,
3. CaO is not abundant in some deposits,
4. The percentage of Al_2O_3 is in excess.

The above results indicate that this graphite schist is of sedimentary origin and is comparable to the one described by Bastin (1909-1910). The original sediments were probably arenaceous to argillaceous with disseminated calcareous and carbonaceous matter.

Thus, in all occurrences of graphite in this area, graphite flakes and grains are associated with meta-sedimentary rocks and are disseminated in them.

The dissemination of graphite in metasediments and the probability of the existence of lower forms of vegetable life in Pre-Cambrian period is expressed by Alling (1918) and Winchell (1910-1911), to suggest the biogenic origin for the graphite. Gelpi et al. (1969). have reported that the most graphitic rocks, ores and commercial samples containing hydrocarbons indicate biogenic origin.

According to Winchell (1910), the hydrocarbons are improbable source of graphite, for they are wholly transformed into hydrogen and carbon monoxide at 700° to 800°C in the presence of water which is generally present during the formation of pegmatites and veins. Carbides of the metals seem to be equally improbable as source of graphite.

Clark (1921), has described disseminated deposits in which graphite is evenly distributed and represents all or in part the original carbonaceous organic matter contained in them. In most cases, it is the result of metamorphism of carbonaceous sediments and not from coal beds. In fact, wherever organic matter has been trapped in strata undergoing metamorphism, graphite is likely to result. This transformation of organic matter to graphite affects siliceous, argillaceous and calcareous sediments in the same way.

The present investigations thus indicate the following sequence of events and the role of the regional metamorphism that brought the conversion of the organogenic matter to be transformed into graphite.

As already mentioned, the foliations in the country rocks that bear graphite are uniform and well developed.

They are found obliterated near the granitic contact. In the vicinity of granite, the graphite bearing rocks are medium to coarse grained. Near the granite contact, the phyllitic rocks have been transformed to schistose or even gneissose rocks.

The regional metamorphism gives rise to a number of mineral assemblages and is mainly dependent upon its parent rock material. The graphite bearing rocks of Ankli, Sewania, Muthai, Chaena and Jaloda which were originally pelitic sediments, now show the presence of quartz, chlorite, muscovite, biotite and calcite. The crystalline limestone consisting of calcite, tremolite forsterite, quartz, chlorite, epidote and mica, suggest that the original rock must have been an impure argillaceous to siliceous limestone. The mineral assemblages of the graphitic rocks indicate low-grade metamorphism of chlorite zone. The mineral assemblages present in crystalline limestone on the other hand suggest contact metamorphism. In general, the mineral assemblages of the metasediments point out that the metamorphic changes on argillaceous, arenaceous and calcareous sediments must have taken place at low temperature and moderate pressure conditions. Shearing stress was also quite effective (Graphite occurrences of Ankli, Sewania, Muthai, Narukot, and Jaloda etc.). The appearance of abundant biotite and occasional sillimanite in Virpur graphite deposits suggest the effects of moderate to high grade metamorphism. But here the absence of garnet is noticed.

The original pelitic rocks are transformed to slates and phyllites due to higher grade of metamorphism. The detrital quartz has recrystallized and has formed small lenticles or streaks. Mica and chlorite have also undergone regeneration and appear as coarser flakes. The schistosity has become more pronounced and the rock is transformed to quartz-mica schist which contains quartz, chlorite, muscovite and sericite as important constituents. Thus it can be concluded that low to moderate regional metamorphism of carbonaceous sediments has given rise to graphite-bearing schist and gneiss.

It is evident that the regional metamorphism in the Champaner rocks is generally of chlorite zone and the mineral assemblages belong to Green-schist facies of Turner and Verhoogen (1962) who have estimated temperatures of low-grade regional metamorphism between 300° to 500°C and pressure pH_2O = 3000 to 8000 bars.

From field characters, mineral assemblages and chemical compositions it seems most probable that the sediments giving rise to graphite deposits were of two types:

1. arenaceous-argillaceous-carbonaceous sediments,
2. calcareous-argillaceous-carbonaceous sediments.

These sediments were interbedded with clay, limestone and quartzite bands. Low to moderately high grade regional metamorphism of these sediments have given rise to schistose structure and have transformed carbonaceous matter into graphite. Granitic intrusion was responsible for the contact metamorphism of both types of sediments.