

Secondary clay in Rajmahal basalts of India and its relation to palagonite-chlorophaeite

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Summary. The mineralogy of the green, yellow, and brown secondary constituents occurring as amygdalae, irregular infillings, and replacements in the Rajmahal basalts of eastern India and commonly described under the ill-defined names palagonite and chlorophaeite, has been revised on the basis of optical, X-ray, and thermal studies. The results indicate that these coloured constituents, which have been chiefly derived by the alteration of pyroxenes, iron ores, and primary glass, belong to the clay minerals, particularly smectite and vermiculite. This brings in question the validity of the terms palagonite and chlorophaeite which, though widely current in basaltic literature, have no definite mineralogical identity. It is suggested here that they are but forms of clay.

SECONDARY minerals in basalts comprise the material deposited in vesicular and interstitial voids by late magmatic fluids and the products of their reaction on primary constituents. In the Rajmahal basalts they fall into 3 main groups: zeolites, silica, and a widespread series of green, yellow, and brown minerals, which have commonly been described as palagonite and chlorophaeite. These constituents have frequently been described from the Deccan traps as well as from other basalts, but their mineralogical nature is highly variable and their identity uncertain. Hence re-investigation of their mineralogy was called for. The material supporting this study was collected from different parts of the Rajmahal hills in the district of Santal Parganas in Bihar, eastern India, and supplemented by collections in the museum of the Department of Geology, Presidency College, Calcutta. About 100 selected specimens were studied in thin sections; optical measurements were made on 25 of them, out of which 6 were chosen for X-ray and thermal examinations. The X-ray investigation was undertaken in the National Professor's Laboratory, University College of Science

and Technology, Calcutta, while the optical and thermal studies were carried out in the Geological Laboratory, Presidency College, Calcutta.

Previous work. Of the two terms chlorophaeite and palagonite, the former is the older. It was first described by Macculloch in 1819 from Rum (Scotland) and in greater detail by Heddle in 1880 as secondary infillings in doleritic rocks (cited in Fermor and Fox, 1916). Peacock and Fuller (1928) and Peacock (1930) subsequently investigated it and concluded that it was a potential chlorite produced by deuteritic alteration of ferromagnesian minerals and glass in doleritic rocks. In Dana's (1949) text-book chlorophaeite is described as a chlorite, while Winchell (1951) does not recognize it at all.

Palagonite was first proposed in 1846 by S. von Waltershausen (cited in Peacock, 1930) to denote an impure substance produced by hydration of glassy basaltic tuff. It has since been described by many others (Middlemiss, 1889; Peacock, 1930; Fermor, 1925, 1927, 1931; Wadia, 1925) and has been regarded as an impure mixture of potential chlorite and zeolite or silica (Peacock, 1930). Palagonite is however more current as a rock name and is not recognized in the mineralogical text-books of Dana (1949) and Winchell (1951).

References to this class of constituents in the Rajmahal traps were made first by MacMohan (1887) and Middlemiss (1889) and subsequently by many others.

Since the home of both chlorophaeite and palagonite is basaltic rocks and since they are both regarded as products of secondary alteration, considerable confusion has arisen on the distinction (if any) between them. Peacock and Fermor respectively have contributed much to this controversy. The main conclusions of Fermor were that palagonite and chlorophaeite were chemically substantially similar, a hydrous substance derived by the hydration of primary glass and alteration of pyroxenes, iron ores, etc. However, he restricted the term chlorophaeite to the light-coloured, low-refracting members of this group while the dark-coloured, high-refracting members were named palagonite. He also stated that chlorophaeite was always accompanied by zeolite (chabazite) and palagonite by chalcedony. Peacock objected to some of Fermor's views and emphasized the compositional difference between chlorophaeite (potential chlorite) and palagonite (mixture of potential chlorite and zeolite or silica). He considered them genetically distinct, the former resulting from deuteritic alteration of doleritic rocks and the latter from hydration of basaltic tuff by external water. Wadia (1925) attempted to draw a distinction in their mode of occurrence,

assigning the rounded amygdales to chlorophaeite and the irregular replacement products to palagonite.

Recent contributions on this subject come from: Allen and Scheid (1946), who have cited examples of basaltic constituents changing to palagonite, which finally gave place to nontronite; Prider and Cole (nontronite), Earley and Milne (interstratified montmorillonite-chlorite), both cited in Wilshire (1958); Wilshire (1958), who has identified this type of material as smectite-chlorite; Sen (1954) who has optically determined the green earths of the Deccan traps as nontronite-montmorillonite; coloured amygdales from the Rajmahal traps were identified as clay by Sarbadhikari (1958).

Mode of occurrence and structure. The green, yellow, and brown secondary minerals in the Rajmahal traps closely resemble the so-called chlorophaeite-palagonite series as described by Fermor (1925) from the Deccan traps. Here these minerals are confined to basalts and basaltic tuff, being very rare in the more acidic lava. One possible reason may be that the latter is very poor in clinopyroxene, which is the principal parent of these minerals. They have formed mostly by the deuteric alteration of clinopyroxenes, partly by replacement of iron ores, less commonly after primary glass and olivine, but never from plagioclase. In hand specimens they occur as spherical amygdales of dark brown or black colour (ranging in diameter from 1 cm downward) or as irregular streaks. Under the microscope they are found to occur as sub-rounded amygdales, as linings to cavities infilled by zeolite, chalcedony, etc., as cores of cavities lined by the above minerals, as irregular amoeboid patches, as partial or complete pseudomorphs of crystals of clinopyroxene, iron ore, or olivine, as inclusions in plagioclase, and as an alteration product of tuff and ash. There is no selective association of any particular variety of constituent with any particular mode of occurrence.

In thin sections, these secondary minerals are seen to be bundles of thin fibres or aggregates of minute granules, crystalline, cryptocrystalline, or nearly amorphous. When in amygdales, the mineral may show zonary structure. Commonly the cores show a granular structure and lesser birefringence while the margins show fibrous structure normal to the cavity walls and higher birefringence. This structural sequence may however be reversed.

Many earlier workers have sought to establish differences between the green and the yellow or brown products in their mode of occurrence and corresponding differences in their derivation. However, examina-

tion of a large number of thin sections by the present writers shows that the green and yellow or brown constituents do not differ substantially in any other microscopic character except colour. No systematic distinction can be made between them in sequence of formation or mode of derivation. Crystals of clinopyroxene in the same slide show alteration now to the yellow substance and now to the green substance within a span of a few millimetres. Primary glass has been replaced sometimes by the brown product and sometimes by the green product in the same slide. Moreover, the green and the yellow or brown varieties occasionally occur in structural continuity with each other. Thus they show in all respects a common mode of origin.

Optical characters. The material used for optical, thermal, and X-ray examinations was separated by hand picking from the amygdalae and cavities in rock samples. The amygdular fraction is closely similar in thin section to the interstitial and replacement fractions and was therefore considered as representative of the whole mineral group.

These minerals exhibit a wide range of colour, refringence, and birefringence and the variation is gradual with no conspicuous break. The colour varies from deep and dark brown to orange, yellow, and pale yellow on the one hand and impure greenish brown, yellowish green, and pale green on the other. The general presumption is that the variation in colour is related (Fermor, 1925; Peacock, 1930) to the oxidation state of the constituent iron, being mainly ferric in the brown and ferrous in the green product. Pleochroism is absent.

The birefringence ranges from 0 to 0.022; commonly it is maximum with the fibrous variety and minimum with the granular variety. Some authors (Wilshire, 1958; Peacock and Fuller, 1928) have described chlorophaeite as isotropic while others (Fermor, 1925; Sen, 1954) have described it as anisotropic. Palagonite is also described as isotropic to anisotropic (Wadia, 1925; Fermor, 1925). Optical isotropy is therefore not an essential feature of these constituents. The Rajmahal samples grade from isotropic (rare) to anisotropic and there is no substantial difference between the types, which have the same derivation and often a structural continuity with each other. The apparently isotropic patches might prove feebly birefringent under high resolution.

Measurement of the refractive index was made on 25 samples powdered to -120 mesh (B.S.S.) in monochromatic light by the immersion method; it ranges from 1.53 to 1.61, being low in pale yellow and pale green varieties and increasing with depth of colour to a maximum in the dark brown and bright orange forms. Elongation is always positive.

X-ray data and identification. Six carefully selected samples of these secondary minerals were analysed by powder methods, using Co- $K\alpha$ radiation in a 114.59 mm-diameter camera, the samples being packed into Lindemann glass capillaries of 0.3 mm diameter. Photographs were taken before and after glycerol treatment.

In four of the samples, the first strong reflection between 17.7 and 17.8 Å from glycerol-treated material, together with higher orders, indicates the mineral to be essentially a montmorillonoid (smectite), mixed in some cases with a little kaolinite and quartz. Samples 5 and 6 gave the characteristic diffraction patterns of vermiculite or chlorite. In order to distinguish between these possibilities, samples were heated at 700° C for 12 hours; this caused a complete replacement of the 14 Å basal reflection by one at 9.3 Å, showing that these samples are essentially vermiculites. These results are in conformity with the optical and thermal properties of the minerals.

Differential thermal analysis. In order to confirm the results of the X-ray examination, all the 6 samples were examined with the help of a differential thermal apparatus. The thermograms of all the specimens show sharp endothermic peaks between 140° and 180° C, which is very typical of montmorillonoids (smectite) and vermiculite. While the low temperature parts of these curves are quite indicative, the high temperature parts are not very characteristic. High temperature peaks are not conspicuous (this might be due to low sensitivity of the apparatus or presence of impurities).

Discussion. In the Rajmahal basalts clinopyroxene, iron ore, and primary glass, stated in order of decreasing susceptibility, have been commonly transformed in varying degrees into a series of yellow, brown, and green secondary products. They exhibit a wide range of gradual variation in refringence, birefringence, mode of occurrence, and structure. Optically they resemble the material described by Fermor (1925) as chlorophaeite and palagonite from the Deccan basalts and are described by the same names in the literature. The present study suggests from optical, X-ray, and thermal data that the Rajmahal chlorophaeite and palagonite essentially consist of common clay minerals, particularly montmorillonoids and vermiculites.

The above observations cast doubt in general on the validity of the minerals chlorophaeite and palagonite, which are very common names in the basaltic literature. They tend to raise the question if these products are indeed anything distinct from clay.

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