PHOSPHATE MINERALS OF THE BORBOREMA PEGMATITES: I—PATRIMONIO

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ABSTRACT

The beryl-tantalite pegmatites of northeastern Brazil carry a considerable number of phosphates, both primary and secondary, which are to be considered in a projected series of papers covering individual localities, individual minerals, or groups of localities. The mineral arrojadite, reported as occurring in most of these pegmatites, is known with certainty only from the original locality, and minerals locally so called constitute a wide range of species. The first report is on the "alto" Patrimonio, where amblygonite and lithium phosphite are the primary phosphate minerals. Hydrothermal and other alteration has formed brazilianite (a second Brazilian occurrence for this mineral), crandallite, wardite and apatite, from the amblygonite. Eosphorite, hureaulite, metastrengite, stewartite, ferrisicklerite, heterosite, rockbridgeite(?), and a number of unidentified minerals, have been derived, mainly by oxidizing solutions, from the lithium phosphite. Lepidolite with minor microcline, beryl and tantalite, are primary accessory minerals. Brazilianite and wardite occur sometimes in measurable, although small crystals. Details of observations on various unknowns are included, with the hope that some may be identified by the writer or other observers at some future date.

INTRODUCTION

This paper is the first of a projected series of reports on the phosphate minerals of the beryl-tantalite pegmatites of northeastern Brazil. These pegmatites have been described in some detail by several authors: W. D. Johnston, Jr., P. M. A. Rolff, S. C. de Almeida, L. J. De Moraes, D. Guimaraes, H. C. A. De Souza, F. H. Pough and others. Johnston (1945), gives an extensive bibliography on these occurrences. These writers have paid particular attention to the minerals of economic value, (Pough, 1945, has described simpsonite), and the following brief outline of the geologic picture has been taken from their writings. For a more detailed description of general paragenesis and distribution, the reader is referred to the original papers.

The region in general is one of Precambrian gneisses and mica schists, intruded by granites and cut by pegmatites and quartz veins. Pre-cretaceous (?) erosion reduced the area to a peneplane—the Borborema "planalto," later overlain by Cretaceous and younger sandstones. The present erosion cycle has stripped off most of these cover-rocks, and in the Paraiba-Rio Grande do Norte area has cut into the peneplane surface, forming relatively steep-walled valleys, exposing batholithic granite masses as monadnocks, and leaving many pegmatites standing as walls or eminences above the surrounding terrane, because of their greater resistance to erosion. From these projecting masses has come the local name "alto" given to the pegmatite outcrops in general, even when they
do not rise above the surrounding surface. In the northern area, notably in the State of Ceará, the peneplane surface is less dissected, and the pegmatites show no relief, merely appearing as patches or strips of white quartz on the essentially flat surface.

The pegmatites may be classified as "homogeneous" or "heterogeneous," depending on their mineralogy and texture. The former are simple granite pegmatites, of relatively uniform texture and mineral distribution, with quartz, muscovite, and microcline which has been more or less albitized. They are in general rather regular dikes, and are extremely numerous in the area. The latter (heterogeneous) are usually lenticular in horizontal section, and show a high degree of mineralogic differentiation, which produces a more or less well-developed zonal pattern. They are also numerous, but far less so than the homogeneous type. Field evidence does not indicate a different source for the two types, but the accessory minerals in the heterogeneous dikes are more abundant, often in large crystals, and afford an economic source of beryllium, tantalum and sometimes tin.

The beryl-tantalite pegmatites may be grouped roughly into three

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**Fig. 1**
areas: the Picui-Parelhas, Quixeramobin-Cachoeira and Cascavel-Cristais regions, the first two of which are shown on the accompanying key map (Fig. 1).

According to Johnston, (1945) p. 1040 these pegmatites have been formed by an orderly sequence of crystallization, with little or no evidence of replacement, except for some late stage albitization. This sequence is normally divided into four zones, beginning at the margins with abundant, usually coarse-grained muscovite. Next comes a mixture of normal quartz-feldspar-muscovite with a texture like that of ordinary granite pegmatites. Inside this is a zone characterized by enormous feldspar crystals, up to several meters in dimensions and tons in weight. Most of the beryl, tantalite and spodumene occur here, as do also the primary phosphates, which appear as patches and nodules in the quartz and feldspar. The final, innermost zone is essentially massive quartz, usually milky, but sometimes a curious bluish gray, or a beautiful rose-pink. Many large crystals of beryl appear here also, although large portions of the core may be absolutely barren. Occasional vugs in this quartz are lined with quartz crystals, which may be accompanied by apatite.

Beryl and tantalite have apparently been essentially unaffected by later solutions or by weathering processes, but spodumene may show extensive kaolinization, and in some cases is completely altered. The associated phosphates, however, have been affected by extensive alteration, both hydrothermal and superficial, with the resulting development of a considerable number of later minerals, many of which occur intimately intergrown with each other, in fine-grained aggregates. Many of the pegmatites are lithium-bearing, with lepidolite or spodumene, or both, appearing in considerable amount. Here the primary phosphates are lithiophilite, amblygonite, or graftonite, any one of which may be the source of a series of alteration products. In the absence of lithium, phosphates such as triplite do not appear to have produced such a variety of secondary minerals.

The author was privileged, when on sabbatical leave from the University of California at Los Angeles in 1948, to visit approximately twenty of these pegmatites, and to collect specimens, mainly of the phosphate minerals, for laboratory study. I wish here to express my thanks to the following: the University of California, for a research grant which made the trip possible; the Divisão de Fomento da Produção Mineral, and its Director Dr. A. I. Erichsen, for facilitating arrangements for transportation and guidance in the field; Dr. Onofre Chavez, engineer of the D.N.P.M. at Campina Grande, who was assigned to me as guide and mentor, since without him I should not have been able to accomplish my task; Dr. W. D. Johnston, Jr. for invaluable aid in infor-
mation and introductions; and finally, Señor Silveira Danton, of Silveira Brasil & Cia, who presented me with a magnificent large crystal of tantalite for the University Museum.

Patrimonio

The “alto” Patrimonio is a few hundred yards south of the little town of Pedra Lavrada, in the State of Paraíba (see index map, Fig. 2). It forms a gentle eminence on the landscape, rather than a wall-like outcrop, since it is lenticular rather than tabular in form. It is one of the smaller heterogeneous pegmatites which have been mined, and is not described in detail in the literature. It was originally (1914–1918) opened for mica, which was a rather poor quality of ruby muscovite, and reported production was only a few kilograms. A production of 45 tons of beryl and 2.9 tons of tantalite was reported for 1940–1942, and in 1943 the records show about 200 kilograms of bismutite and native bismuth, with small amounts of mangano-tantalite, tantalite and betafite (?). Lazulite and “arrojadite,” some uranium ochres, aquamarine beryl, and smoky and bluish quartz, were also noted from this dike.

The general pattern of mineral distribution apparently follows roughly Johnston’s zoning, with a quartz-rich central core. Little detail of this pattern was observable at the time of the writer’s visit, especially since the pegmatite had not been worked actively for some years, and the outcrops were obscured by vegetation and by rubble of the dumps. The
principal primary minerals are quartz and microcline, with lesser muscovite and albite (cleavelandite). The minor minerals include beryl, tantalite, lepidolite, amblygonite and lithiophilite. These last two are the source of a considerable number of alteration productions which will be discussed in some detail. The ordinary early minerals were not collected, but their appearance was normal, and they do not need further consideration here.

The beryl and tantalite had been quite thoroughly mined out before my visit, so their characteristic occurrence could not be checked by observation. Specimens of lepidolite, amblygonite, lithiophilite and others, were collected only from fragments on the dumps, so that their exact relationship to the zonal distribution of the dike could not be directly determined. However, judging by their relationship to quartz and feldspar in the dump fragments, there seems to be no reason to assume other than the usual space-relationships. Accordingly, it may be assumed that here the principal phosphates were deposited only moderately late in the sequence of dike crystallization, and occurred along with beryl and tantalite in irregular masses in the quartz and feldspar of Johnston's third zone. In the following sections the various minerals of the deposit will be considered individually. Arrojadite was not observed in the Patrimonio deposit, but should be discussed here because of its supposed widespread occurrence. One of the early phosphates has been named arrojadite, and was described from the “alto” Serra Branca, by Guimaraes (1942). It has been reported by casual observers from many of the pegmatites of this region, and Rolff (1946A) p. 31 says that it is frequently altered to a varied assortment of minerals ranging in color through yellow, blue, olive green, etc., to black, the most common product being vivianite, followed by purpurite. The present writer has found, however, that true arrojadite is rare or absent from any but the original locality, that vivianite is almost never formed, and that purpurite is relatively uncommon. Nevertheless, the varicolored series noted by Rolff is frequently present in the lithia-bearing pegmatites, and forms an interesting group. The alteration sequence ranges from huréaulite, through sicklerite (or ferri-sicklerite) to purpurite (or heterosite), with several other varieties, to purple or blue meta-strengite (phosphosiderite), and black manganese oxides. Most of the alterations appear to be due to descending oxidizing solutions. Descriptions of the individual minerals, with discussion of their relationships, are given in the following sections.

**Lepidolite and Microlite**

Pale lilac lepidolite occurs in massive aggregates of small (2–3 mm.) platy grains intergrown with clear quartz, some cleavelandite and an
occasional grain or poorly developed crystal of colorless to faintly yellowish microlite. This microlite was identified by its isotropic character, high index of refraction, and spectroscopic determination of major calcium and tantalum with small sodium, and very small niobium, aluminum, etc. The x-ray powder pattern checks closely with that of type material from Topsham, Maine, in the Harvard collection, as shown in the accompanying Table 1.

<table>
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<th>Patrimonio</th>
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**Amblygonite**

Massive cleavable amblygonite was found in irregular fragments on the dump. In the specimens collected it is milky white in color and carries clear-cut veins and irregularly bounded patches of later minerals which appear to be hydrothermal in origin. Some of these minerals are in good or poor crystals, some in fibrous aggregates, and some in powdery form as very fine-grained patches. In this group appear apatite, brazilianite, wardite, crandallite, carbonatian apatite and some species so far unidentified. Some specimens show fracture-coating of dehrnite (?) along with carbonatian apatite. A few well-formed crystals of manganotantalite, with a little massive orangeite (?) were found on one amblygonite specimen. Clusters and nests of muscovite occur here and there in
the massive amblygonite. Analysis of the amblygonite (Table 2) shows it to be fairly high in fluorine, and low in hydroxyl. A second generation of amblygonite, very fine grained and associated with eosphorite, was noted in veinlets cutting across lithiophilite.

**Table 2**

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(1) E. L. Martin, analyst.
(2) L. C. Peck, analyst.

**Brazilianite**

Narrow (5–10 mm.) veins in massive amblygonite show numerous small glassy grains, and occasional poor to fairly good crystals of a colorless mineral which goniometric measurements and x-ray powder photographs prove to be brazilianite. This is the fourth known occurrence for this mineral, and the second for Brazil. The other localities are: the original find in the Conselheiro Pena district, Minas Geraes; the Palermo pegmatite near North Groton, N. H.; and the Smith mine near Newport, N. H. The Patrimonio crystals are relatively simple, somewhat elongated on [001], and measured crystals show the following forms: [110] usually well-developed and always present; [101] usually present and prominent; [010] always present though invariably narrow; [320], [130], [670?], [111], [121], [301?], [311], [321], [112], sometimes present but never prominent. Some of the crystals are to be found in a fine granular matrix of crandallite, along with lavender apatite crystals. As observed in thin section, fine-grained brazilianite was deposited along the walls of a fissure in the amblygonite, and followed by deposition of larger grains and
terminated crystals embedded in, or projecting from the walls into the fine-grained crandallite, which filled the remaining space. In one instance, brazilianite is somewhat coarser textured, appearing as sub-parallel or radiating prismatic groups, none of which show crystal faces. Here it is associated with glassy wardite, a yellowish green isotropic mineral which may be orangeite and an unidentified pink phosphate.

**Wardite**

Glassy, granular wardite, sometimes in imperfect crystals, occurs associated with brazilianite in several of the veins and patches of later minerals in the amblygonite. One or two crystals were measurable, and showed {001}, {012}, and {011}. It is somewhat difficult to distinguish from brazilianite by inspection, but is usually in larger grains, and often shows its perfect basal cleavage. X-ray powder photographs confirm its identity, and cleavage flakes show anomalously biaxial segments as reported in crystals from Montebras. Dana (1951) p. 940.

**Crandallite**

This mineral occurs only as fine powdery aggregates filling in the space between lavender apatite, brazilianite and wardite, in veins in amblygonite. It was identified by micro-chemical tests, confirmed by x-ray powder pattern. Associated with it are at least two other minerals in fine prismatic or fibrous form. This matrix is so fine grained and intergrown that the optical properties are confused, but they may be members of the apatite group, since carbonatian apatite has been found in some of the specimens.

**Apatite**

Apatite occurs in various fashions in the deposit. In massive amblygonite, there are large patches of imperfect crystals of dark green manganapatite, essentially contemporaneous with its host. In the veins or replacement patches in the ambygonite, carbonatian apatite appears as pale lavender or bluish prisms up to 2–3 mm. across, heavily striated vertically, and usually with a colorless center. A basal section shows biaxial segments in the lavender portion, with a sensibly uniaxial core. These crystals are somewhat later than most of the brazilianite, and occur principally in regions which were originally cavities, but which have since been filled in with powdery crandallite. An area of massive greenish material adjoining the crandallite filling is shown in thin section to be a feathery aggregate of carbonatian apatite prisms intergrown with numerous prismatic grains of an unidentified mineral.

Hydroxyl-manganapatite also occurs as fine-textured veins cutting
massive altered lithiophilite, ferri-sicklerite, etc. Here the mineral is very fine grained, waxy in luster and pale orange on the borders of the veins, white and sugary in the midportion.

Carbonatian apatite occurs as a drusy to botryoidal crust coating lithiophilite and its alteration products, and also as a fine-grained, almost waxy coating, also on lithiophilite, and partly altered to a very soft unknown mineral.

A late stage of apatite appears as minute, water-clear grains and crystals in narrow veins cutting across spongy aggregates of late alteration products or lining the surfaces of cavities in such minerals as meta-strengite, pyrolusite (?), etc. Here it seems to be almost the last mineral to be deposited.

**Lithiophilite**

Lithiophilite occurs in fair-sized masses of irregular shape, essentially contemporaneous with the primary quartz and feldspar of the pegmatite. When perfectly fresh it is practically colorless, but it is often brownish due to partial oxidation, or greenish yellow from abundant inclusions of later minerals. Chemical analysis of selected pure mineral shows a moderate excess of MnO over FeO, so that it is lithiophilite rather than triphilitc. The analysis is given in Table 2.

**Huréaulite**

Huréaulite occurs in crystalline crusts, occasionally with measurable crystals showing \{100\}, \{110\}, \{201\}, \{111\}, and \{311\}. These crusts are cavity or fracture fillings in lithiophilite. In color the mineral varies from salmon pink to colorless. Chemical tests show only a very small amount of FeO, which apparently varies with the depth of color of individual samples. Huréaulite is one of the fairly early oxidation products of lithiophilite, and is apparently confined to this mineral, not being observed in ferri-sicklerite. Huréaulite is essentially contemporaneous with a yellowish-green mineral (mineral A, which colors much of the lithiophilite), underlying it in part and covering it in part.

**Ferri-sicklerite and Heterosite**

Ferri-sicklerite where present forms an alteration zone around fresh cores of lithiophilite, and as is usually the case, is in crystallographic continuity with it, so that cleavage surfaces on one mineral are continued uninterruptedly on the other. Locally, further oxidation has formed purple patches of heterosite, still with the same orientation crystallographically. The last stage of oxidation is the formation of porous or powdery black manganese oxides of indeterminate character, which sometimes carry small amounts of phosphorus. Little or no iron is left in these
oxides, but the element does occasionally appear in the form of cellular or powdery limonite. Frequently other secondary phosphates are formed directly or indirectly by the oxidation and hydration of the lithiophilite, usually later than huréaulite or mineral \( A \), and earlier than the oxide coatings.

**Meta-strengite**

This is one of the late-formed minerals, and occurs as fine botryoidal crusts, or porous, sometimes crumbly, masses of minute crystals. Less commonly it appears as fine-grained compact patches in ferri-sicklerite, and in one or two cases as pseudomorphs after imperfect, nearly square prisms of an earlier unknown mineral, possibly lithiophilite. In color meta-strengite is quite variable, ranging from a rather bright light blue, which is commonest, to blue-violet, purple, or even pink. In some cases it appears to have faded to almost colorless. Chemical tests show a small amount of manganese, but not enough material is available from this locality to make a quantitative determination. The X-ray powder pattern matches exactly that of analyzed material from another locality. In general it is earlier than the manganese oxides, but sometimes it seems to be later, forming crusts or cavity linings superimposed on the latter.

**Stewartite**

Occasional acicular or bladed crystals or irregular grains, of canary yellow stewartite occur in the meta-strengite, and are in general somewhat later than the latter.

**Variscite**

Pale pink to colorless variscite was observed as a finely drusy coating on one of the fracture surfaces of a highly oxidized specimen. In part this mineral coats even the manganese oxides. Identification was made on the basis of optical and chemical properties, and confirmed by X-ray powder pattern.

**Eosphorite**

One specimen of massive lithiophilite shows a vein of massive flesh-colored or colorless coarsely crystalline material with one good cleavage, associated with fine sugary white amblygonite. Occasional crystal faces show striations on the somewhat elongated grains. This mineral has a hardness of about \( 4 - 5 \), and is optically biaxial negative, with \( 2V \) near \( 10^\circ \); moderate dispersion \( v > r \); index of refraction close to 1.64 for \( \beta \). The mineral is insoluble in \( \text{HCl} \), but on moderate heating turns dark brown and becomes soluble. Chemical tests show \( P, Fe \) small, \( Mn \) moderate, \( Al \) large, no \( Ca \). The X-ray powder pattern matches very closely that of
the childrenite-eosphorite group. The index of refraction and low iron content indicate eosphorite, which seems probable in spite of lack of solubility, and the very small value for 2 V.

**Unidentified Minerals**

A considerable number of minerals in the secondary groups occur in such small quantity or in such intimate intergrowth with others, and in such small particles, that optical determinations are difficult, and only qualitative chemical tests are possible. In none of these cases has identification been possible, but all available information will be given in their descriptions, in the hope that some other worker may encounter better material, and be able to complete an identification. It has been possible to get X-ray powder patterns for most of these, but none has been found to match anything in the A.S.T.M. or Harvard files. In many cases, too, optical determinations have been only approximate. The unknowns will be designated by letters.

**Mineral A = Tavorite***

This mineral occurs as a minor alteration product of lithiophilite, along cleavages, fractures, and in solution cavities. It is quite soft, yellowish green in color, and usually in microscopic spherules. Rarely it forms separate crystals, or crystal aggregates, but in no case in individuals large enough to make any determination of the symmetry. Optically, it is biaxial positive, with 2 V about 70°–80°. Pleochroism is moderate, pale yellow to colorless. Dispersion strong, with r > v. The index of refraction is variable, from 1.77 to 1.81, and extinction is oblique to elongation of the grains. Chemically, it is easily soluble in acid, giving micro-tests for Fe and P; Ca and Mn are absent; no water given off in the closed tube, but the mineral quickly turns bright red-brown at moderate heat. The X-ray powder pattern shows the following spacing and intensities for the strongest lines: 4.96 Å—3; 4.68—5; 3.93—3; 3.41—3; 3.29—10; 3.03—7. The complete pattern rather closely matches that of amblygonite.

**Mineral B**

This mineral appears as dark brown, nearly square prismatic crystals, crudely developed, and showing two perfect cleavages at approximately 90°, parallel to the prism direction. Hardness 3–4; streak pale yellow-brown; optically biaxial positive, with 2 V very small; beta is close to 1.75; pleochroism slight, pale to darker brown; extinction parallel to

*This mineral is apparently identical with the mineral tavorite, identified and described by Lindberg and Pecora (1954) after the submission of this manuscript to the editor.
prism edges. It is readily soluble in acid, and gives tests for Fe, Mn and P. Gives off water readily in a closed tube. Its age relationship to lithiophilite could not be determined, as the two do not occur in contact. Where found, it was in some cases replaced by compact meta-strengite, which forms imperfect pseudomorphs. X-ray powder photographs show spacings which do not match any recorded pattern. Spacings and intensities of the stronger lines are as follows: 8.6 Å—3; 6.6—3; 4.76—3; 4.36—4; 4.11—3; 3.52—4; 3.38—3; 2.83—10; 2.43—3; 2.37—3; 1.52—3.

**MINERAL C**

In the crumbly crystalline coating of late alteration products, one of the minerals is canary yellow in color, occurring as minute square flat crystals, tetragonal in aspect, showing a flat pyramid and base. Optically it appears to be uniaxial negative, with omega about 1.78. Some sections which are square in cross section, and also some which are diamond shaped, show abnormal blue interference colors with crossed nics. The mineral is soluble in acid, giving tests for ferric iron and phosphorus, but not for calcium or manganese. It may be hydrous, but not enough pure material could be isolated to check this. Spacings and intensities of the stronger x-ray powder spacings are as follows: 4.48 Å—10; 3.20—3; 3.10—3.

**MINERAL D**

Another finely-crystalline mineral occurring in similar powdery crusts, is dark red-brown, in extremely small platy or tabular crystals, with index well above 1.83. It appears to be biaxial negative, with 2 V very small. Extinction is symmetrical, and pleochroism is strong, pale yellow to dark red-brown. The obtuse bisectrix is not far from normal to the plates. It is soluble in acid, and gives tests for ferric iron, Mn, P, and probably H₂O, though this is not sure, because of possible admixture of impurities. The x-ray powder pattern is practically identical with those of two other minerals from this locality (Minerals E and F), although their optical and physical properties are different. Prominent lines with spacing and intensity for all three are as follows: 8.65 Å—10; 5.60—5; 2.74—6.

**MINERAL E**

This mineral is compact, fine grained, olive green in color with yellow streak. Index near 1.75. Soluble in acid, and gave tests for ferric iron, ferrous iron, a little Mn, some Ca, and little or no water. A variant of this has a somewhat higher index, and no Ca. The mineral in general in the hand specimen resembles alluaudite from Varuträsk, Sweden, but the x-ray powder patterns do not match.
MINERAL F

Mineral F is yellow-brown in color, apparently pseudomorphous after some earlier mineral, as it shows large cleavage surfaces, but is actually fine grained. Optically it is biaxial negative, with index much above 1.76, and with no pleochroism. It is soluble in acid, and gave tests for ferric iron, Ca and Mn. Physically it somewhat resembles xanthonenite, but the x-ray powder patterns do not match.

MINERAL G

This mineral occurs as radiating clusters of blades intergrown with a mass of other secondary minerals. It is deep red-brown in color, with one perfect cleavage. Optically it is biaxial negative, with 2 V about 5°, and β well over 1.76. The obtuse bisectrix is nearly normal to the platy surface. Pleochroism is strong, from nearly colorless to dark brown. The mineral is readily soluble in acid and gave tests showing much Mn, little Fe, and some water in the closed tube. An identical mineral is found in two other pegmatites of the region, Boqueirao and Taboa. Spacings and intensities of the stronger lines of the x-ray powder pattern are as follows: 8.6 Å—10; 2.90—4; 2.75—4.

MINERAL H

A deep green colored mineral occurring in patches in lithiophilite, and as compact fine-grained masses, is close to rockbridgeite, and may be this species.

Various other minerals of unknown character also occur here in minor amounts, mostly in the porous, friable crusts, and include some manganese oxides; patches of reddish material suggesting salmonsite (index near 1.68, biaxial negative? 2 V small); minute black crystals, green by transmitted light, which have index about 1.76, biaxial (?) negative (?) with large 2 V, and giving tests for Fe, Mn, and P; also a powdery yellow-brown fibrous mineral, possibly an oxidized vivianite.

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