DEVELOPMENT OF RECENT STROMATOLITIC STRUCTURES AND PHOSPHATIC ENRICHMENT IN PRECAMBRIAN MARBLE OF SRI LANKA

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Introduction

An apatite ore commonly referred to as the Eppawala phosphate deposit has been discovered in the township called Eppawala situated in the Anuradhapura district of Sri Lanka—lying about 200 km from the capital city of Colombo (Fig. 1). It extends over an area of about 6 km^2 . The ore was formed principally by leaching of an apatite marble formation and an apatite-scapolite-diopside rock. Under tropical weathering conditions, these rocks have given rise to an enriched orebody consisting of both primary and secondary apatite. The primary minerals are chlor-fluor-apatite (Jayawardena, 1976) and hydroxylapatite (Tazaki et al., 1987). The secondary products are carbonate-fluorapatite (collophane), carbonate-hydroxylapatite (dahllite), and crandallite.

The phosphate deposit occurs in a deeply weathered terrain formed of several north-south-trending turtleback hillocks rising to about 170 m above sea level. The apatite bodies are found in the vicinity of migmatitic gneisses which together with crystalline limestone (marble), charnockite, and quartzite form metasedimentary successions characteristic of the Precambrian of the central highlands of Sri Lanka. The P_2O_5 content of the phosphate deposit varies from \sim 25 to 43 percent, values above 35 percent being for the primary apatite crystals. A magmatic origin has been suggested for the Eppawala apatite bodies by Jayawardena (1976). This conclusion was based on the presence of euhedral crystals of apatite associated with magnetite, and the occurrence of crosscutting dikes where apatite is associated with scapolite and diopside. Mineralogies similar to the latter were noted in the cores of complex folds found in the adjacent migmatitic gneisses.

A metasedimentary origin can also be supported by: the existence of plastic flow and deformation structures associated with apatite-bearing rocks and adjacent metasedimentary apatite-free rocks, the presence of spinel in the Eppawala apatite marble, and the presence of appreciable quantities of Cl (nearly 2%; see also Jayawardena, 1976), which are all suggestive of early sedimentary conditions. Furthermore, the apatite bodies are found within metasedimentary successions and their general trend parallels the strike of the metasedimentary rocks of the region. Sharp contacts between apatite bodies and the surrounding basement rocks have not been noticed and no fenitization has been reported. However, only detailed geologic, petrological, and geochemical studies can help to establish the origin of this important phosphate deposit of the Sri Lankan Precambrian.

Currently, the Eppawala phosphate is crushed to fine silt size and used as a fertilizer for long-term crops such as rubber, coconut, and tea. At the present rate of exploitation, the deposit can be utilized for more than 100 years and has been estimated to contain 40 million tons of rock phosphate. At present, efforts are underway to produce more soluble superphosphates from the Eppawala material for short-term crops.

Apart from the general geologic investigations by Jayawardena (1976), geochemical studies have been done on the Eppawala deposit by Tazaki et al. (1986, 1987). Various agronomic experiments have been done (Sivasubramaniam et al., 1981) and studies have been undertaken with a view to beneficiating the deposit to produce a soluble fertilizer (Gunawardane and Glasser, 1979; Weeraratne, 1983). The principal apatite mineral in the Eppawala deposit has been identified variously as hydroxylapatite, chlor-fluorapatite, or fluorapatite and only a systematic study can reveal the exact mineralogical distribution of the ores (Jayawardena, 1976; Amerasekera et al., 1981; Tazaki et al., 1986). The senior author is presently in charge of a research project with a view to carrying out a systematic geologic, mineralogical, and geochemical investigation of the deposit.

The present study is intended to shed light on the geologic features that are responsible for the deposition of the apatite ore at Eppawala.

Geology and Petrology of the Eppawala Phosphate Deposit

In Sri Lanka, more than 90 percent of the area is underlain by Precambrian igneous and metamorphic rocks. These are broadly classified into at least three units: the Highland Group and the Eastern and Western Vijayan series (Fig. 1). The oldest of the units is the Highland Group which occurs in the central highlands of Sri Lanka and is dated to be 2 to 3.2 b.y. old. The Highland Group is characterized by metasedi-



FIG. 1. Map of the main geologic units of Sri Lanka and the location of the Eppawala area.

mentary rock successions of quartzite, marble, charnockite, and migmatitic and other gneisses. Both the Eastern and Western Vijayan Series consist mainly of granites, granitic gneisses, and migmatites with occasional calc-silicate rocks and quartzites. The Vijayan rocks are 1,100 m.y. old (German-Sri Lanka Consortium, 1987). The Eppawala phosphate deposit lies within the Highland Group rocks. It is surrounded by migmatitic gneisses (Figs. 1 and 2).

The phosphate deposit occurs in weathering profiles which can be as thick as 100 m and which developed on apatite marble formations interbanded with migmatitic gneisses-mostly of the hornblende biotite variety. The apatite-bearing marble formation occurs in the core of a north-south-trending synform whose axis is in conformity with the regional strike. The contact between the apatite marble and the gneisses is characterized by complex geologic structures such as kink folds, tight folds, plastic deformation phenomena, and dikes. Within the folds and crosscutting dikes, the apatite is concentrated in the cores. In these cores yellowish apatite mineralizations are surrounded by dark, finer scapolite- and diopside-rich groundmasses. In the apatite marble, apatite occurs as large crystals which have long diameters greater than 25 cm. These crystals are associated with calcite, dolomite, and accessory spinel, magnetite, and phlogopite (Figs. 3 and 4).



FIG. 2. Geologic map of the Eppawala apatite deposit with respect to other formations. Modified after Jayawardena (1976).



FIG. 3. Photomicrograph of apatite (clear) crystals (A) in the dolomite marble rock (dark), Eppawala phosphate deposit. Under plane-polarized light, bar = $300 \ \mu m$.

The deeply weathered regions of the apatite marble formation and the associated rocks can be divided into two distinct zones: weathered and leached zones (Fig. 5). The weathered zone consists of loose sandy clay associated with laterites rich in iron and aluminum. Isolated patches of phosphate occur in cavities. Loose individual apatite crystals also are found disseminated in the weathered zone. Discontinuous outcrops of apatite marble are rarely observed. Chemical analyses indicate enrichments of Al_2O_3 and Fe_2O_3 in this zone. The leached zone is conglomeratic in appearance caused by the disposition of rounded primary apatite grains in finer groundmasses. The primary apatite crystals have corroded margins displaying finegrained outer envelopes. The groundmass displays faint fine-grained laminated sequences which have developed as cavity fillings. Both the weathered and leached zones are distinct chemical entities. The weathered zone is rich in Fe and Al whereas the leached zone shows P enrichment. Microunconfirmities are widespread in both zones.

Development of Stromatolitic Structures and Apatite Enrichment

Microscopic observations of the rock samples from the leached zones of the phosphate deposit show the development of the following structures and textures: (1) laminated matrices with pellets, oncoids, and microoncoids; (2) apatite grains within finer groundmasses; (3) apatite grains with rounded edges (coated grains), both simple and composite oncoids; and (4) intraclasts.

1. The laminated matrices have fine-grained clear and dark laminations. The clear laminations are formed of carbonate-hydroxylapatite (dahllite; microcrystalline apatite) and the darker ones are formed of carbonate-fluorapatite (collophane; cryptocrystalline apatite). In thicker laminations, dark pellets of carbonate-fluorapatite on the order of a few microns or millimeters are found. In still thicker laminations, cauliflower-shaped oncoid structures have formed of clear and dark laminations (carbonate-hydroxylapatite and carbonate-fluorapatite). These oncoids are either single or composite in character (Figs. 6 and 7). The laminated matrix is extensively enriched by secondary phosphate mineralization. Scanning electron microscopic (SEM) studies show filamentous networks (of microorganisms?) in the fine-grained matrix. Euhedral microcrystals of secondary apatite are commonly observed. These crystals show filamentlike protrusions (Fig. 8). P_2O_5 values of the matrix are slightly less than those in the apatite crystals or in the coated region (envelope) (Table 1).

2. The apatite crystals appear to be crystals that are characteristic of the parent apatite marble. The apatite grains from a few millimeters to several centimeters show a sharp contact with a microcrystalline carbonate-hydroxylapatite matrix and associated pellets. The matrix at other points shows a laminated character (Figs. 6, 8, and 9).

3. The size of coated grains can vary from a few microns to several centimeters. They are primary apatite grains around which one or more envelopes made of dark fine grains or clear coarse apatite crystals can be observed. The finer cryptocrystalline coating gives a corroded appearance to the apatite grains which show a faint impression of the initial crystal outlines. The microcrystalline carbonate-hydroxylapatite envelope produces a rimlike texture, and chemical analysis shows high phosphate enrichment in such margins (Fig. 6 and Table 1). The coated grains also occur as aggregate bodies bound by common coatings of cryptocrystalline and microcrystalline apatite. These bodies are reminiscent of compound oncoids described by Dahanayake (1977).

4. The intraclasts are composite bodies, on the or-



FIG. 4. Photomicrograph of hexagonal crystals of apatite (A), mostly associated with scapolite (S) in a dark matrix dominated by diopside, Eppawala phosphate deposit. Under plane-polarized light, bar = $600 \ \mu m$.



FIG. 5. A section through the weathering profile of the Eppawala apatite marble formation with the corresponding vertical distribution of P_2O_5 , Fe_2O_3 , and Al_2O_3 in the various zones. a = apatite marble, b = leached zone, c = weathered zone. Based on field and laboratory observations by the authors and data from Jayawardena (1976).

der of a few microns to several centimeters, which engulf primary apatite grains, coated apatite grains, pellets, oncoids, or even smaller interclasts in a common matrix (Fig. 9).

Summary and Conclusions

Field and laboratory observations suggest that under tropical conditions of subaerial weathering, the



FIG. 6. Photomicrograph of apatite grains (A) with corroded margins showing clear (carbonate-hydroxylapatite) and dark (carbonate-fluorapatite) laminations. Note also a substantially corroded apatite grain (c) and the ovoid-shaped dark collophane-rich pellets in the matrix which shows a laminated character (see left upper area), Eppawala phosphate deposit. Under plane-polarized light, bar = $300 \ \mu m$.



FIG. 7. Photomicrograph of the cauliflower-shaped oncoid bodies formed of clear and dark laminations within a thick stomatolitic lamination. The darker areas are formed of cryptocrystalline carbonate-fluorapatite and the white regions of microcrystalline carbonate-hydroxylapatite. Note the occurrence of pellets (middle left and lower right areas), Eppawala phosphate deposit. Under plane-polarized light, bar = 500 μ m.



FIG. 8. SEM photograph of the microcrystalline matrix showing the occurrences of branching filamentous networks (of microorganisms?) suggestive of stromatolites (top left and bottom, not clear). Note the growth of microcrystals of apatite in a cavity and the protrusion of filaments visible in one crystal (arrow, bottom right). These features are reminiscent of mineralization in stromatolites, Eppawala phosphate deposit. Bar = 20 μ m.

original apatite-bearing lithologies (the dominant being apatite marble) have undergone leaching and subsequent removal of carbonate minerals such as calcite and dolomite. With the continuation of the leaching processes, a residual lateritic cap rock with a network of dissolution cavities was left behind in place of the parent apatite-rich rock(s). The resistant metamorphic apatite grains derived from the parent rocks formed part of a residuum in the dissolution cavities consisting of laminated fine-grained clavey material produced during weathering. In such cavities, microbial activity was prevalent as shown by laminated structures typical of stromatolites (Figs. 6 and 7). The laminated structures on diagenesis were mineralized to give secondary phosphate minerals such as carbonate-fluorapatite and carbonate-hy-

TABLE 1. Electron Microprobe Analysis (in wt %) of the Principal Components of Enrichment in the Eppawala Phosphate Deposit

	Primary apatite crystal		Enrichment zone on the envelope	
	Core	Margin	ot the apatite crystal	Matrix
P ₂ O ₅	42.53	42.64	43.01	39.84
CaO	54.44	54.25	54.51	53.77
K₂O	0.05	0.05	0.05	0.02
Na ₂ O	0.01	0.01	0.01	
SiO2	0.11	0.11	0.11	0.06
Al ₂ O ₃				0.17
FeO	0.01	0.01	0.01	1.88
ZnO	0.09	0.09	0.09	0.26
MnO				0.15
Total	97.24	97.16	97.79	96.15



FIG. 9. Photomicrograph of primary apatite crystals (P), coated grains (C), and intraclasts (I) in a microcrystalline carbonate-hydroxylapatite-carbonate-fluorapatite matrix (see Fig. 8). Note the initial stages of formation of clear and dark laminations around the elongate apatite grain in the lower right area, Eppawala phosphate deposit. Under plane-polarized light, bar = $300 \ \mu m$.

droxylapatite (Fig. 8). These neoformations account for an appreciable fraction of the phosphate enrichment of the whole deposit. The available analysis though not statistically representative confirms such an enrichment in the matrix as shown by the P_2O_5 composition (Table 1). Further evidence for this enrichment is the occurrence of corroded envelopes around the apatite grains. These envelopes, which are formed of secondary microcrystalline apatite, are comparable in texture to the peloids or micritized envelopes which form by a process of grain diminution-a degrading neomorphism phenomenon-in carbonate environments (Folk, 1965; Wolf, 1965; Bathurst, 1973). The presence of phosphatic corroded (coated) grains, intraclasts, and oncoids within the dissolution cavities implies the existence of a sedimentary environment which was nourished by downward-percolating meteoric waters carrying carbonatephosphate nutrients derived from leaching of the apatite marble. The secondary mineralization which produced apatite minerals such as carbonate-hydroxylapatite and carbonate-fluorapatite in both coated grains and the groundmass, occurred perhaps during weathering and diagenetic processes active in the microenvironments within the dissolution cavities. The EDS microanalyses for the secondary phosphates (Tazaki et al., 1987) are comparable in the coated parts of the grains and in the matrix suggesting a common mineralizing pathway. Furthermore, evidence of anaerobic conditions in such an environment is suggested by the presence of sulfur in the so-called "strings" of Tazaki et al. (1987) which occur in the matrix. The cauliflower-shaped phosphatic oncoids can only form during such diagenetic processes (Gerdes and Krumbein, 1987).

It is interesting to note that carbonate oncoids form

during diagenesis of stromatolitic carbonate sediments. The phenomenon of apatite oncoid formation in a carbonate-phosphate sediment can be best understood as a product of mineralization in a phosphatic microbial mat undergoing diagenesis in dissolution cavities (see also Dahanayake et al., 1985; Gerdes and Krumbein, 1987). The association of phosphatic allochems, such as intraclasts, pellets, and coated grains (which are characteristic of the carbonate stromatolitic deposits) in laminated matrices formed of filamentous networks in this modern carbonate phosphate environment, suggests the participation of microorganisms in their development (Fig. 8). Thus the modern carbonate-phosphate environment at Eppawala is comparable to a carbonate stromatolitic environment. Similar analogies can also be drawn with ancient phosphorite-forming stromatolitic environments spread out at various stratigraphic levels (Baturin, 1971; Rao and Rao, 1977, 1980; Riggs, 1979; Soudry and Champetier, 1983; Dahanayake and Krumbein, 1985).

The Eppawala phosphate deposit is characterized by microenvironments where neoformations of apatite developed under conditions similar to those of phoscrete-type phosphorites, e.g., as in Georgina basin rocks of Australia (Southgate, 1986a). The phoscretetype phosphorites are characterized by phosphatic peloids, pellets, and intraclasts which occur within laminated crusts where algal filaments have been found. Recent studies by Tazaki et al. (1986, 1987) on Eppawala have also shown that in the clayey matrix where secondary phosphate enrichment is common. branching networks of strings (filaments?) and diatoms are common. A vague allusion to mineralization by microorganisms was also made by the same authors. The branching strings resemble fungal filaments that are generally associated with sedimentary stromatolitic phosphorite or ironstone deposits (Dahanayake and Krumbein, 1985, 1986; see also Fig. 8).

The following sequence of events can be envisaged for the phosphate enrichments in the weathering profiles which developed on the Precambrian apatite marble formation at Eppawala:

1. Crystallization of primary apatite (chlor-fluoror hydroxylapatite) occurred during Precambrian metamorphic event(s) when a carbonate-rich phosphate deposit in a sedimentary sequence metamorphosed and gave rise to the apatite marble formation which is now located within the Highland Group rocks.

2. Uplift, weathering, and erosion of Precambrian metasedimentary rocks took place under equatorial tropical conditions, perhaps since Late Cretaceous times when the Sri Lankan landmass reached its present position near the equator (Cantwell et al., 1978). Subaerial weathering and erosion of the Highland

Group rocks continued to the present and the development of a soil profile with a significant phosphate enrichment developed on the apatite marble (parent rock) at Eppawala.

3. At least three phases of enrichment are apparent. The first is an accumulation of primary metamorphic apatite grains in the dissolution cavities of the profile which were nourished by downward-percolating meteoric waters. In the tropical fresh-water vadose environment at Eppawala, solar heating increases the concentration and temperature of the percolating carbonate-phosphate nutrient-rich waters. Under such conditions, the second phase of mineral enrichment will occur, namely, phosphate precipitation, perhaps facilitated by microbial participation (Fig. 8; Read, 1976; Southgate, 1986b). Repetitive precipitation and sedimentation generates a laminated stromatolitic groundmass in the dissolution cavities. Subsequently, diagenesis can occur in the sedimentary microenvironments of the dissolution cavities. The third phase of mineral enrichment occurs during diagenetic alteration processes by way of mineralization within the laminated stromatolitic groundmasses, and the development of phosphatic allochems.

4. With continued weathering and erosion phenomena characteristic of the tropical environment at Eppawala, the above-mentioned phases of apatite enrichment occurred during a considerable time period which was punctuated by constructive and destructive episodes as manifested by the network of microunconformities that exists in the weathering profile.

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