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Polymetamorphism of Ores in Precambrian Stratiform Massive Sulfide Deposits at Ambaji-Deri, Western India

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Massive stratiform zinc-lead-copper sulfide ores, in association with cordierite-anthophyllite rocks, occur in adjacent localities of Ambaji and Deri, in Western India. The metasedimentary country rocks, interlayered with amphibolites and intruded by acidic to intermediate plutonic rocks, belong to the Precambrian Delhi Supergroup. The ore minerals identified by detailed mineragraphic studies include: sphalerite, galena, chalcopyrite, pyrite, pyrrhotite (both monclinic and hexagonal phases), magnetite, ilmenite, rutile, arsenopyrite, molybdenite, cubanite, mackinawite, boulangerite, gudmundite, meneghinite, lautite, tenantite, native bismuth, native silver, chalcocite and covellite. The common sulfide-silicate schistosity in the ores, flowage of sulfide streaks and tails around rotated poikiloblasts and in their pressure shadow region developed during early folding (F_1) and regional metamorphism of the rocks under green schist facies condition. These were superimposed by a pervasive hornfelsic fabric involving sulfides and silicates and including microfabrics due to annealing and grain growth in sulfides, during a subsequent phase of low pressure thermal metamorphism and related tectonism (F_2). Finally certain deformation features and some uncommon fabrics like martensitic lamellae in galena and subgrains in sphalerite developed during a mild deformation episode (F_3) in the waning stages of tectonism in the area. Compositional change in the ores during thermal metamorphism was minimal.

INTRODUCTION

The importance of recognition of the metamorphosed character of ore deposits, particularly in Precambrian terrains, which establishes the time of ore formation with respect to the geological evolution of the enclosing rocks, has been stressed by several workers (e. g. Mc Donald, 1967; Vokes, 1969; Stanton, 1972; Mookherjee, 1976). In many cases this leads to a restriction of the number of possible processes in the genesis of the ores concerned and thus helps in constructing a viable model of ore genesis. Sulfide ores, spatially associated with cordierite-anthophyllite rocks in close proximity to acidic intrusives, can form by any of the many diverse processes suggested for the genesis of these rocks (see e. g. Ramsay and Davidson, 1974; James et al., 1978). However, if the effects of thermal metamorphism are established in such ores, a pre-intrusion

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process rather than metasomatism related to the intrusion, would appear responsible for sulfide ore generation.

The purpose of the present contribution is to record the mineralogy and describe significant metamorphic ore fabrics from two adjacent massive sulfide occurrences showing cordierite-anthophyllite rock association in the late Precambrians of western India. The development of the ore fabrics has been related to the stages of metamorphic evolution of the enclosing rocks. Mineralogical adjustments due to metamorphism and formation of compatible ore minerals in compositionally different metamorphic assemblages have been discussed. This paper is an extension of some earlier preliminary observations by the author (Deb, 1973).

GEOLOGIC SETTING

Massive, stratiform, zinc-lead-copper ores occur at Deri, in the Sirohi district of Rajasthan and at Ambaji, in the Banaskantha district of Gujarat (Fig. 1), at the two ends of a north-south trending 8 km long stretch of folded and polymetamorphosed rocks belonging to the lower part of the Ajabgarh series of late Precambrian Delhi supergroup (Coulson, 1933; Heron & Ghosh, 1938). The ores are concentrated preferentially in lensoid patches of metamorphosed magnesian and calc-magnesian rocks like, chlorite-cordierite-anthophyllite rocks, quartz-chlorite-tremolite schists, forsterite marble etc. enclosed within argillo-arenaceous metasediments or ortho-amphibolites. Acidic to intermediate igneous intrusives occur within or close to the mineralized zones.

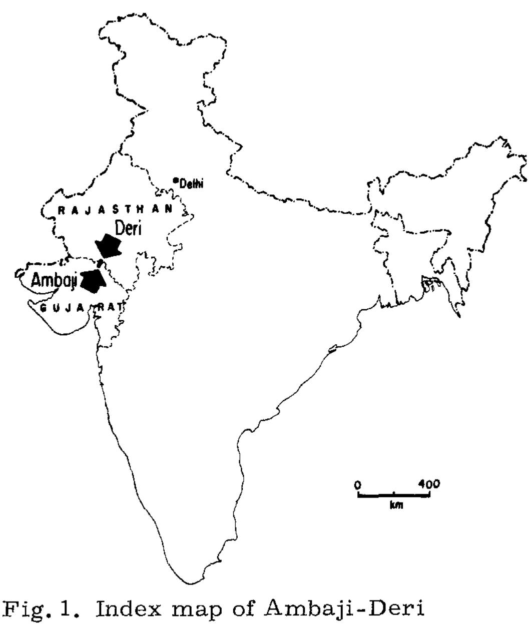


Fig. 1. Index map of Ambaji-Deri

Structural studies (Deb, 1974) have shown that regional metamorphism to upper green schist facies conditions accompanied the first penetrative deformation (F_1) in the area, which produced the pervasive schistosity. Thermal metamorphism under hornblende hornfels facies condition, apparently related to the intrusion of plutonic rocks in the area, extensively superimposed the hornfelsic fabric on the pre-existant schistosity and was broadly coeval with the second phase (F_2) of deformation. The last phase of deformation (F_3) saw the development of steeply plunging down-dip fold in the rocks.

Based on various geological evidences, including the polymetamorphosed nature of the ores, as brought out in the present contribution, together with an analysis of the geological environment and available geochemical data, Deb (1978) has recently suggested a model of syn(-dia) genetic origin of the sulfides within highly magnesian or calc-magnesian sediments.

ORE MINERALIZATION

The sulfide ores at Ambaji and Deri occur as typically "stratiform" (Stanton, 1972) lensoid to tabular bodies, a major part of which on the basis of their metal content and compact nature, can be termed "massive sulfides". Minor discordancy is noted only locally. Zinc, led and copper are the principal metals in that order of general abundance. However, the relative concentration of these major metals varies among orebodies and also within a single ore lense. The average content of these metals, as deduced from bore hole data are: Zn 6.0 %, Pb 5.5 %, Cu 2.0 % and Zn 9.0 %, Pb 6.8 %, Cu 1.1 %, at Ambaji and Deri respectively (G. S. I., 1972). Iron content of the ores varies widely. Minor metals in the ores include: Mo, Ag, As, Bi, Cd, Co and Ni.

Massive sphalerite ores, studded with idiomorphic pyrite crystals are most common. Galena-rich and chalcopyrite-rich ores come next in abundance. Pyrite is an ubiquitous constituent and frequently is sufficiently abundant to form massive or banded pyritic ores. Pyrrhotite-rich ores are found in Deri while locally, magnetite is a major constituent. The ores also show compositional banding, ranging in thickness from a few millimeters to a few centimeters. It is best displayed by alternating concentrations of pyrite and sphalerite or alternating layers rich in quartz, biotite, pyrite and chalcopyrite.

Copper and lead-zinc ores in the area generally tend to form separate bodies, the former being enclosed in more arenaceous units while the latter shows a distinct preference for the more calcareous and magnesian rocks. In composite orebodies no definite stratigraphic zoning of the metals is obvious though in some cases lead apparently is concentrated in stratigraphically lower levels.

MINERALOGY OF THE ORES

About 200 polished and thin-polished sections of ores from 30 drill holes and underground workings were studied under a Carl Zeiss Universal research microscope to decipher the mineralogy of the Ambaju-Deri sulfides.

Sphalerite-rich ores have a simple mineralogy with varying concentration of galena and chalcopyrite and rare flakes of molybdenite. Idiomorphic pyrite grains, 0.03 to 4 mm across, characterize the massive sphalerite ores and occur either as isolated metablasts or in pyritic layers. Where galena is the predominant sulfide, as within typical hornfelsic mass of chlorite and amphibole blades, coarse pyrite porphyroblasts, subrounded grains of sphalerite and a host of minor inclusions are common. Molybdenite flakes, 0.03 to 0.21 mm in length, showing sharp multiple kinks and strong undulose extinction form the most common inclusion. Tenantite is present as irregular to subrounded patches in galena and poikiloblastically contains fine elliptical inclusions of its host. The mineral is also noted in galena-rich ores along galena-chalcopyrite or sphalerite-chalcopyrite interface. Fine, irregular, rectangular or elliptical patches of gudmundite are present within galena matrix. Tiny myrmekitic intergrowth of native bismuth in pyrrhotite is also noted in galena-rich ores. Some fine white specks in galena with very high reflectivity have been tentatively identified as native silver.

The most complex mineral assemblage is observed in chalcopyrite-rich ores. These ores generally comprise: Chalcopyrite + pyrite + magnetite + sphalerite + galena with or without pyrrhotite, cubanite, molybdenite, mackinawite and arsenopyrite. Pyrrhotite occurs as discrete grains. Cubanite (non-isometric) appears as exsolved blades, often deformed in more than one crystallographic direction of chalcopyrite and in close association of pyrite. Mackinawite is commonly present as needles and worm-like aggregates, upto 0.09 mm in length. Arsenopyrite is found to have replaced pyrite along grain boundaries and fractures. Coarse magnetite poikiloblasts in the assemblage include ilmenite lamellae rarely.

Uncommon sulfides and sulfosalts form complex intergrowth within chalcopyrite mass (Fig. 2). They were identified by comparison of their optical properties, including spectral reflectivities, with those described by Ramdohr (1969)

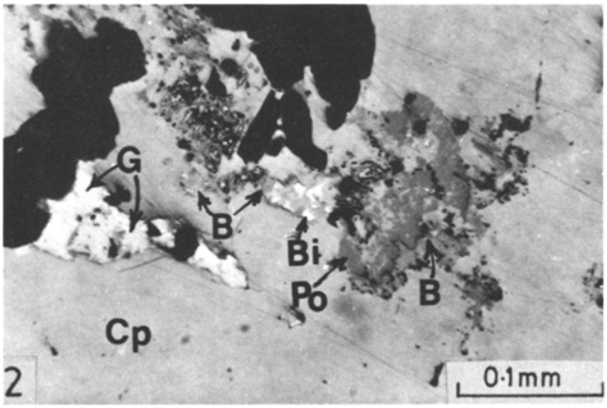


Fig. 2. Complex intergrowth of uncommon sulfides and sulfosalts in Chalcopyrite (C_p). Irregular boulangerite (B) mass include skeletal bodies of native bismuth (Bi), replace irregular patch of gudmundite (G) and associated with pyrrhotite (Po). Reflected ppL, under oil immersion

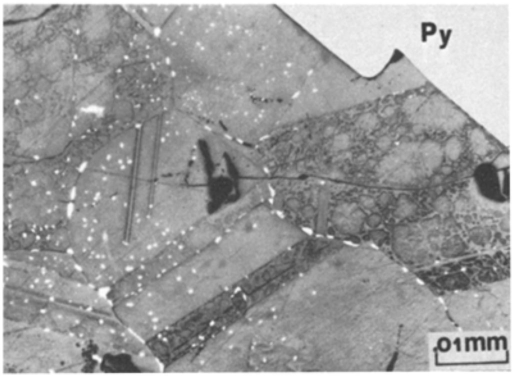
and Uytenbogaardt & Burke (1971). Irregular patches of boulangerite include skeletal masses of native bismuth or replace irregular bodies of gudmundite or are found intergrown with aggregates of lautite. Boulangerite and gudmundite are also found streaked out with the flaky silicates to define a schistose texture. Association of meneghinite, lautite and native bismuth is also not uncommon.

Pyrrhotite-rich ores are characterized by banding of sulfide and silicate layers and a pronounced schistosity defined by chlorite, tremolite and actinolite. Sphalerite, pyrite and chalcopyrite are commonly present in these ores. Electron microprobe analysis of sphalerite and the iron sulfides in these assemblages using a ARL-SEMQ microprobe (15 KV; beam current of 0.1 μ a; 10 sec. counting time; troilite, pure Zn and Mn metal as standards) reveal that FeS mole % of sphalerite in association with pyrite and pyrrhotite varies between 15.3 and 17.6, and in association with pyrite alone, is between 6.0 and 7.5 (see Deb, 1978). Pyrrhotite coexisting with sphalerite and pyrite contains 47.7 - 48.0 atomic percent Fe indicating the phase to be hexagonal pyrrhotite $Fe_{1-x}S$ solid solution. X-ray powder diffraction analysis, using CuK_α radiation provided corroboration when pyrrhotite separates from one of the samples, gave a single sharp 102 peak. In other samples, however, double peaks at the hexagonal 102 position indicated the presence of a two phase, monoclinic + hexagonal assemblage (Arnold, 1966). When treated with magnetic colloid (Scott, 1974) these samples exhibited a lamellar arrangement of magnetic domains of the monoclinic phase within the hexagonal host. Textural relationships such as preferential occurrence of monoclinic pyrrhotite along grain boundaries, fractures and basal partings of hexagonal pyrrhotite suggests that the former is an alteration product, probably representing retrograde variation in pyrrhotite composition (cf. Scott et al., 1977).

METAMORPHIC ORE FABRIC

A detailed study of a series of polished sections of the Ambaji-Deri ores, etched with the appropriate reagents, revealed a host of microfabrics, including a few uncommon ones, which can be ascribed to ore metamorphism. The more significant and interesting features are discussed below.

Granoblastic mosaic of polygonal grains, showing excellent foam structures at places (Figs. 3, 4, 5) is present in massive or banded ores of sphalerite, galena, chalcopyrite and pyrrhotite. The individual grains generally measure 0.1 to 0.3 mm across, although rarely, coarser grains of 0.6 mm width have been noted in chalcopyrite mass. Both chalcopyrite and sphalerite grains show coherent annealing twin lamellae, bent and deformed in places (Fig. 4). Where galena and chalcopyrite are present as minor phases in sphalerite mass they spread out as fine wisps, concave cusps or blebs along sphalerite grain and twin boundaries or at equiangular triple junc-



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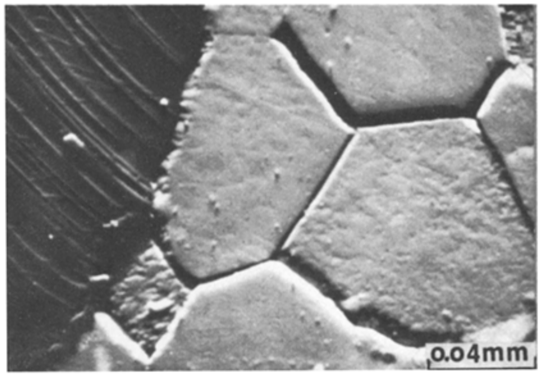

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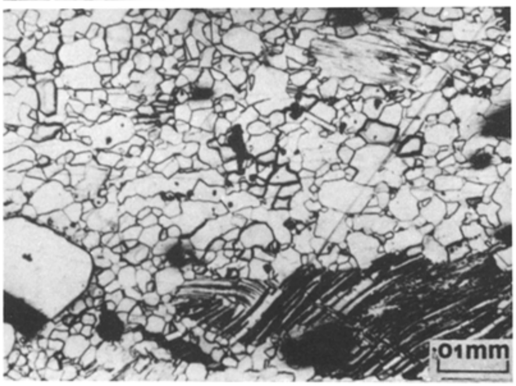

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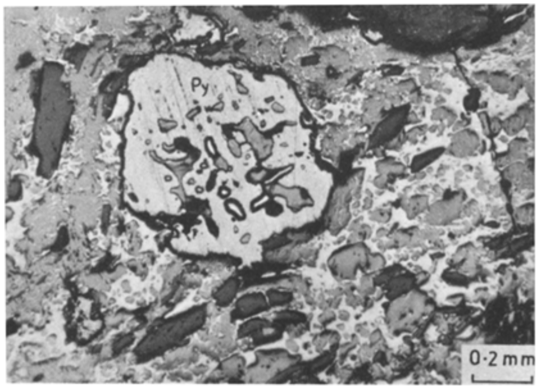

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Fig. 3. Fine blebs of chalcopyrite localized along grain boundaries and annealing twin lamellae of sphalerite. Note the elliptical to rounded patches within sphalerite grain on the right, adjacent to pyrite (Py). Sample etched with HI. Reflected ppL, in air

Fig. 4. SEM photograph (back scatter electron, 20 KV) of galena mosaic showing 'foam structure' with the individual grains meeting three at a point and subtending 120° angle at the junction. Note the slightly curved annealing twin lamellae of sphalerite on the left. Sample etched with thiourea + HCl

Fig. 5. Polycrystalline mosaic of chalcopyrite, with the flattened grains producing a discernible ore-schistosity. Note the translation twin lamellae in a patch of chalcopyrite in the lower right hand corner. Sample etched with sat. chromic acid. Reflected light. Nomarski differential interference contrast, in air

Fig. 6. Rounded poikiloblast of pyrite with inclusions of galena and sphalerite. Granules of sphalerite and laths of tremolite (dark grey) flow on a galena matrix around the poikiloblast. Reflected ppL., in air

tion points (Fig. 3). In certain sections cut normal to the schistosity of the silicates in the ores, chalcopyrite polygons appear flattened with their length: breadth ratio varying between 2:1 and 1.7:1. The preferred orientation of such grains produce a discernible ore schistosity. Interestingly, within this mosaic are also present a few coarse grains with translation twin lamellae, warped and deformed at places (Fig. 5).

Pyritic ores generally show sugary texture, with a normal uni-modal dis-

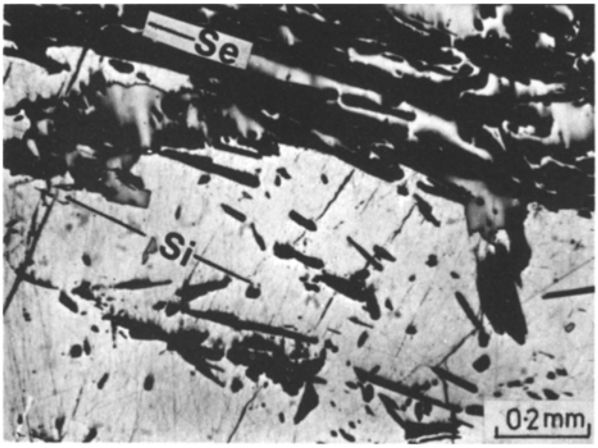

Fig. 7. Subparallel tremolite laths in pyrite poikiloblast describe an internal schistosity (Si) which parallels the external schistosity (Se) described by the same mineral in chalcopyrite mass. Reflected ppL, in air

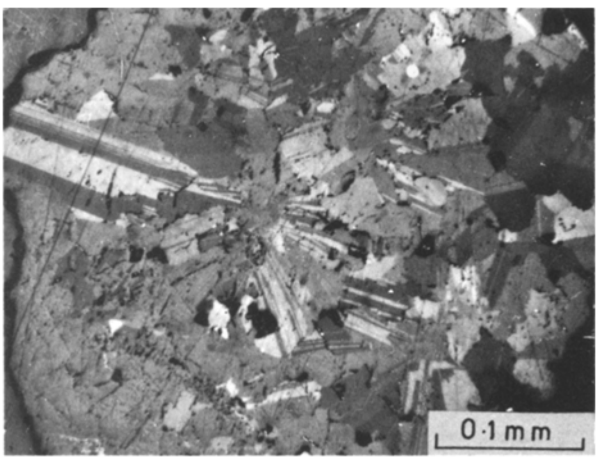

Fig. 8. Rossette-like patch of arsenopyrite within coarse pyrite, individual grains showing lamellar twinning. Reflected light, crossed nicols, under oil immersion

tribution of the triple junction angles around 120°. The individual pyrite grains are mostly poikiloblasts containing rounded to subrounded inclusions of galena, sphalerite, chalcopyrite and less frequently, magnetite grains and tremolite laths. The trains of inclusion are often helicitic or snowballed, with granules of sphalerite, laths of tremolite and smears of chalcopyrite found flowing in a galena matrix around rotated pyrite metacrysts (Fig. 6). Stages of pyrite growth are indicated by the embayment of inclusions, their restriction only along the rim, or incorporation in zones parallel to the grain margins. In sections with a low sulfide: silicate ratio, elongated pyrite grains are oriented parallel to the schistosity, with chalcopyrite, and less commonly galena and sphalerite concentrated in its pressure shadow regions. In certain instances, the schistosity defined by oriented tremolite laths in the rock (Se) passes through elongated pyrite grains to define an internal schistosity (Si), confirming the latter's metablastic growth (Fig. 7). Many of these elongated grains, on etching with dilute HNO_3 turn out to be granular aggregates, their growth having been controlled by a preexistant schistosity. However, certain flattened and S-shaped single pyrite grains in the Ambaji-Deri ores seem incompatible with Graf & Skinner's (1970) observation that pyrite behaves as a brittle substance under all conditions of deformation in the earth's crust. Perhaps such plastic deformation of pyrite is possible under very slow strain rates expected in nature (cf. Sarkar & Deb, 1974) and/or high temperatures and sulfur vapour partial pressures (Atkinson, 1975). Pyrite has also suffered brittle deformation locally, with the softer sulfides healing up the fractures. In rare specimens, etching with HNO_3 brought out zones parallel to the pyrite grain boundary, or irregular to elliptical patches within the grains themselves, which on the whole resembles a relict colloform structure. Coarse magnetite poikoblasts showing sieve structure with included blebs of chalcopyrite, are common in chalcopyrite-rich ores.

Directional fabric is well developed in ores rich in tremolite and chlorite and comprising laminae and streaks of chalcopyrite, sphalerite, elongated grains of magnetite and pyrite. Galena

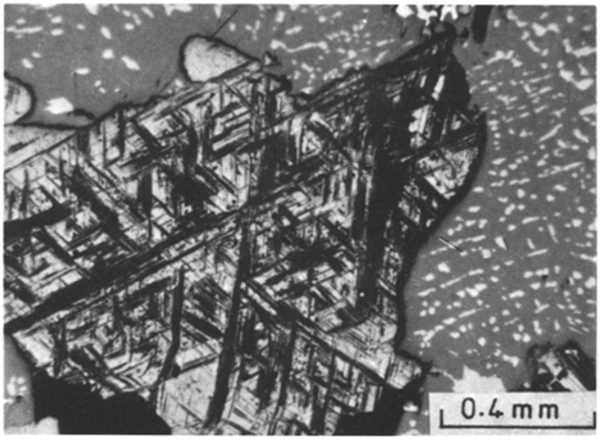

Fig. 9. Slip bands along (hkl) directions of galena. In the adjacent sphalerite, elongated chalcopyrite blebs are oriented along crystallographic directions. Etched with thiourea + HCl. Reflected ppL, in air

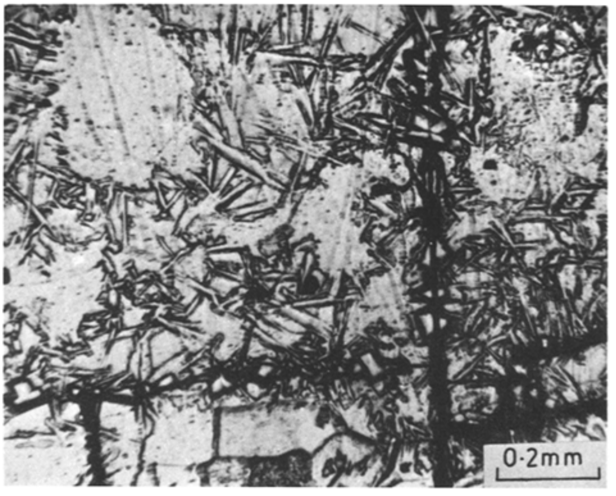

Fig. 10. Irregularly oriented martensitic laths within galena. The squarrish to rectangular platelets are concentrated along grain boundaries. Etched with thiourea + HCl. Reflected ppl, in air

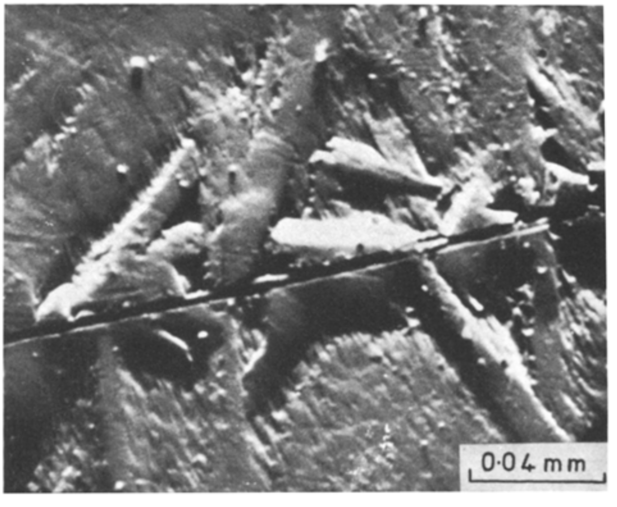

Fig. 11. SEM Photograph (Back scatter electron, 20 KV) of irregularly oriented martensite laths in galena, concentrated along grain boundary. Etched with thiourea + HCl

films and chlorite flakes showing coaxial kinks, are commonly found interlaminated to describe a sulfide foliation, warping around coarse pyrite porphyroblasts. Such sulfide foliations have been extensively imprinted upon and disrupted by a decussate or hornfelsic fabric defined by the flaky and bladed silicates, set within a recrystallized sulfide mosaic. Among the sulfides, arsenopyrite shows simple and lamellar growth twins disposed radially to describe rossette like patches on pyrite grains (Fig. 8).

Deformation in single grains of galena is represented by bent cleavages or the presence of slip bands in more than one crystallographic directions (Fig. 9). In certain coarse chalcopyrite grains parallel sided or tapering, spindleshaped to lensatic twins are revealed on etching.

Two rather unusual textural features are also present in the Ambaji-Deri ores. The HI etch in certain sections of massive sphalerite, revealed the presence of discrete subrounded blebs and irregular to elliptical patches, which range in width between 0.003 to 0.16 mm and are contained mostly within grain boundaries and annealing twin lamellae of sphalerite (Fig. 3). They have a slightly higher relief with respect to their matrix, though optically they are very similar.

Etching of galena with thiourea + dil. HCl, revealed the presence of fine descrete laths, needles and squarish to rectangular platelets (Fig. 10) in almost all polished sections, similar to those described by Sarkar (1974) from the metamorphosed lead-zinc ores of Sargipalli, in eastern India. The laths, which are dominant over the other types, vary in length between 0.09 to 0. 30 mm while the rarer squarish plates are about 0.06 mm across. The laths are totally disoriented and, on the whole, their distribution over the galena matrix is irregular though a strong tendency to cluster near equilibrium grain boundaries is obvious (Fig. 11). Interestingly, the laths and rectangular platelets when studied under the scanning electron microscope, were found to transect grain boundaries.

DISCUSSION

Recognition of pervasive development of metamorphic fabric in the Ambaji-Deri ores, enclosed in polymetamorphosed rocks, raises two possibilities on the temporal relationship of the ores with metamorphism: a) involvement of the ores in both the early phase of regional metamorphism and the later phase of thermal metamorphism or b) involvement of the ores in only the last phase of low pressure thermal metamorphism.

The common sulfide-silicate schistosity, its flowage around rotated pyrite poikiloblasts, streaking of sulfide "tails" in the pressure shadow regions of pyrite augens parallel to the schistosity in the rocks are features believed to have evolved during regional metamorphism of the rocks under green schist facies conditions.

The subsequent phase of thermal metamorphism of the rocks and ores took place under a mean P-T condition of about 3.4 kb/575 °C, as worked out on the basis of the silicate assemblage in the hornfelsic fabric and sphalerite geobarometry (Deb, 1978). The characteristic recrystallized and hornfelsic texture of the Ambaji-Deri sulfides along with features of grain growth and secondary recrystallization developed during this phase and was clearly superimposed on the pre-existant schistose fabric. Evidence of recrystallization of an earlier deformed aggregate of sulfide with a directional fabric is provided by the preferred orientation of flattened polygonal grains within recrystallized chalcopyrite mosaic containing relict coarser grains with warped translational twin lamellae (Fig. 5). Such an interpretation of this fabric is supported by the experimental studies of Stanton and Wiley (1972) with the Coeur d'Alène ores.

The presence of discrete patches in sphalerite matrix and laths and platelets in galena matrix is intrigueing in view of the thoroughly recrystallized nature of the ores. Comparable features in sphalerite, possibly due to higher concentration of iron, have been described from Mississippi valley ores by Barton, Bethke and Toulmin (1963), and from synthetic sphalerite crystals, by Scott and Barnes (1971). The possibility of any compositional difference between the patch and matrix of Ambaji-Deri sphalerites was checked by electron microprobe analyses. Sphalerite grains in a number of sections were found to be homogenous with respect to Fe, Zn, Mn and Cd - the subrounded patches having the same composition as the host. Cd and Mn in some samples were not detectable at 0.1 % level of concentration. In view of this the patches seem to represent later stage brecciation of, or development of subgrains on previously annealed sphalerite. No definite disruption of annealing twin boundaries in sphalerite (Fig. 3) supports the latter possibility.

The laths and platelets in galena, which are obviously not twin lamellae, strongly resemble the bainite and martensite structures, commonly noted in annealed steel. While the formation of bainite involves diffusion of elements (e.g. Carbon in case of steel), martensitic reactions are a diffusion less phase transformation in which a new phase of exactly same composition as the parent phase, but with a new crystal lattice is produced accompanied by shear deformation (Shewmon, 1969; Smith, 1964). Martensites can transect grain boundaries, provided the adjacent grains show a high degree of structural fit.

To check the possibility of anomalous concentration of certain elements in those laths and platelets, about a dozen of various shapes were analyzed with the electron microprobe. They were all found to be pure PbS within detectable limits of analysis. Microprobe scanning for different elements across the laths and matrix failed to detect any difference in chemistry between these apparent phases and their host. The following elements were not detectable at the stated levels of concentration: Ag (0.5 %), Fe (0.1 %), Cu (0.1 %), Bi (0.5 %), As (1.0 %), Sb (0.5 %), Zn (0.1 %).

The above data rules out the possibility of these laths being exsolution lamellae or bainitic structures and strengthens the suggestion of Sarkar (1974, for the Sargipalli ores) that these are analogous to the martensites, though a detailed study of the crystal structure of the plates and matrix would be necessary to confirm this. Since applied stress and plastic deformation of the matrix are known to facilitate martensitic reactions, the laths were probably formed during a deformation phase (F_3) following annealing and recrystallization of the ores. The development of these laths across equilibrium grain boundaries of galena also supports this possibility. Moreover, although martensitic transformations tend to be predominantly athermal, i.e. due to sudden temperature changes, isothermal formation is also known in certain alloys like Fe-Ni (Reed-Hill 1966). In the author's opinion, isothermal formation may be the answer to the problem of rapid quenching of metamorphosed ores under natural conditions required in case of athermal transformation, as pointed out by Sarkar (1974).

The presence of so many other deformational features such as, deformation twin lamellae in chalcopyrite, bent cleavage and slip bands in galena, deformed annealing twin lamellae in chalcopyrite etc. in thoroughly recrystallized ores also seems rather incongruous. In view of the ease and surprisingly low temperatures at which annealing can erase out any trace of earlier deformational features in softer sulfides (Stanton and Gorman, 1968; Gill, 1969; Clark and Kelly, 1973) it seems rather unlikely that deformational features formed during regional metamorphism (F_1) would survive the widespread annealing of the ores during thermal metamorphism (F_2), or that the annealing process itself was incomplete and selectively localized. Therefore, the deformation features mentioned above, as well as the subgrains in sphalerite and martensitic lamellae in galena seem to have formed in all probability during a mild post-recrystallization deformation episode (F_3) - the one which produced downdip folds in the rocks without any marked recrystallization.

Compositional changes in the ores during thermal metamorphism was minimal. In only a few samples, textural relations suggest that pyrrhotite developed from pyrite, probably by desulfurization. In general the process seems to have been rather insignificant during the thermal metamorphism of the Ambaji-Deri ores. However, compositional homogeneity in minerals like sphalerite with respect to Fe, Zn, Cd and Mn (see Deb, 1978), indicating attainment of phase equilibrium in the Zn-Fe-S system under uniform pressure conditions, is believed to have been achieved during this phase of metamorphism. Pyrite and magnetite are the most common iron minerals co-existing with sphalerite in magnesian and calc-magnesian metamorphic assemblages. The buffered assemblage pyrite + pyrrhotite + magnetite and the assemblage pyrrhotite + magnetite, both coexisting with sphalerite are noted occasionally in ferruginous metamorphic assemblages at Deri, and are indicative of successively lower f_{S_2} and f_{O_2} values during metamorphic equilibration (cf. Deb, 1978).

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