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Pyrrhotite Isograd in Southeastern Tennessee and Southwestern North Carolina

ABSTRACT

The first appearance of pyrrhotite in the Barrovian-type metamorphic succession of the Blue Ridge province of southeastern Tennessee and southwestern North Carolina is interpreted to represent a metamorphic isograd located in the upper chlorite zone slightly west of the biotite isograd. At lower grades of metamorphism, pyrite is the only recognizable iron sulfide, and it is characterized by textures indicating an arrested state of growth. Concentrations of pyrite are commonly aligned parallel to bedding planes. Pyrrhotite appears as streaks along slaty cleavage and is preferentially developed in pyrite-bearing zones. Hexagonal pyrrhotite is dominant, but monoclinic and monoclinic + hexagonal intergrowths also occur. Where pyrrhotite is present, coexisting pyrite is typically replaced by quartz, chlorite, and biotite. The sulfur and some of the iron contained in pyrrhotite is interpreted to have been derived locally from the replaced pyrite simultaneously with the development of metamorphic minerals.

INTRODUCTION

This paper describes certain aspects of the occurrence of iron sulfides in low-grade metamorphic rocks of the Ocoee Series in southeastern Tennessee and southwestern North Carolina. In a previous study (Carpenter, 1970), the distribution of metamorphic isograds was determined for the area. The primary objective of the present study was to determine what, if any, relations exist between the occurrence of iron sulfides and metamorphic intensity. The following aspects of iron sulfide occurrence are described: (1) the first appearance of pyrrhotite, (2) the structural-compositional types of pyrrhotite present and their distributions, and (3) textural features of pyrite and pyrrhotite.

GENERAL GEOLOGY

The study area lies within the Blue Ridge province of the southern Appalachians. Classical prograde metamorphism of the Barrovian-type facies series is developed in the Ocoee Series, a late Precambrian sequence of clastic sedimentary rocks. Radioactive age determinations indicate a mid-Paleozoic age for the metamorphic event (Hadley, 1964).

The Ocoee Series is subdivided into three groups (King and others, 1958). The lowermost Snowbird Group consists mainly of arkose, feldspathic sandstone, and argillaceous siltstone. It is overlain by the Great Smoky Group which is comprised of graywacke with lesser quantities of interbedded siltstone, shale (commonly carbonaceous), and arkosic conglomerate. The Walden Creek Group is the youngest group of formations and the most variable lithologically. Quartz pebble conglomerates, calcareous chloritic siltstones, and carbonaceous slates predominate in the study area. The combined thickness of the Ocoee Series is estimated to be at least 12,000 m in the vicinity of the Great Smoky Mountains (King and others, 1958). The distribution of these groups of formations is shown in Figure 1.

Structurally, the Blue Ridge is an allochthonous thrust sheet that has overridden Paleozoic rocks of the Valley and Ridge province to the west along the Great Smoky fault (Bryant and Reed, 1970). Northeast of the study area, relations in the vicinity of the Grandfather Mountain window reveal that the Blue Ridge province has been tectonically transported to the northwest a distance of at least 65 km. The movement occurred mainly during the Appalachian orogeny at the end of the Paleozoic (Bryant and Reed, 1970; Carpenter, 1970).

In addition to being metamorphosed, the rocks within the Blue Ridge thrust plate are intensely folded and locally faulted. At least two generations of folds are widespread (King, 1964; Hadley and Goldsmith, 1963), and as many as four generations are recognized in some areas (Magee, 1968; Power and Forrest, 1971). Bryant and Reed (1970) have concluded that in most areas metamorphism began during the earliest stage of folding and reached a climax during the second stage. A prominent premetamorphic fault termed the "Greenbriar fault" has been mapped by Hadley and Goldsmith, (1963) and King (1964). Most faults in the Blue Ridge, however, appear to postdate. regional metamorphism (Bryant and Reed, 1970).

FIRST APPEARANCE OF PYRRHOTITE

In the Valley and Ridge province of Tennessee, pyrite and, rarely, marcasite are the predominant iron sulfides in the unmetamorphosed sedimentary rocks. Pyr-

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Figure 1. Map showing the occurrence of iron sulfides and the structural types of pyrrhotite present along traverses A through E in southeastern Tennessee and southwestern North Carolina. Geology adapted mainly from Hardeman (1966), and the positions of metamorphic isograds are taken from Carpenter (1970.

----- Metamorphic Isograd

rhotite is known to occur locally in quartz pebbles, but the origin of the pyrrhotite predates its incorporation into the sedimentary environment (Hill, 1957). Similarly, FeS₂ is the only recognizable iron sulfide in the lowest zones of regional metamorphism in the northwestern portion of the Blue Ridge thrust plate. Pyrrhotite (Fe_{1-x}S), however, is common throughout the more intensely metamorphosed portions of the Ocoee Series in the Blue Ridge

The first appearance of pyrrhotite in the area was investigated to determine whether or not the distribution pattern of pyrrhotite occurrence could be related to a known metamorphic parameter. Such a relation could conceivably constitute a new metamorphic isograd.

Traverses were established along five principal highways (Fig. 1) that trend approximately perpendicular to the northeast-trending isograds. Traverse designations are summarized as follows: traverse A, U.S. 64 along Ocoee River from Parksville to Ducktown; traverse B, Tellico River Road from Tellico Beach to North Carolina line; traverse C, U.S. 129 and North Carolina 28 from Chilhowee to Fontana Village; traverse D, U.S. 441 from McCookville to Newfound Gap; and traverse E, I-40 from Hartford, Tennessee, to Walters Dam. Samples were collected at intervals of 0.4 km or less from traverses A, B, and C; and at about 0.8 km from traverses D and E.

Samples were slabbed with a diamond saw and examined under a binocular. microscope to identify pyrite and pyrrhotite. Polished sections were prepared from sulfide-bearing specimens collected from traverses A, B, and C.

As shown in Figure 1, pyrrhotite appears in traverses B through E at, or slightly below, the biotite isograd. In traverse A the first appearance is well below the biotite isograd. Elongate blebs of pyrrhotite along cleavage planes in slates and metasiltstones mark the first appearance. In most cases, pyrite is also present in the samples containing pyrrhotite. The close spatial relation with the biotite isograd is interpreted to indicate that the first appearance of pyrrhotite is controlled by regional metamorphism.

One explanation for the anomalous occurrence of pyrrhotite in traverse A is a postmetamorphic anticline bringing the biotite isograd close to the surface several kilometers west of the position of this isograd shown in Figure 1. Evidence for this is presented by Hurst and Schlee (1962) who noted an exposure of the Great Smoky Group in the core of an anticline several kilometers west of the contact shown in Figure 1. Thus, a cross section might show a close association with the biotite isograd

even though this is not evident from the geologic map.

COMPOSITION AND STRUCTURE OF PYRRHOTITE

Naturally occurring pyrrhotite can be subdivided into three types - troilite, hexagonal pyrrhotite, and monoclinic pyrrhotite. All possess a NiAs-type substructure but differ in composition and superlattice dimensions. Variation in composition is due to omission of iron atoms in the structure (Hagg and Sucksdorf, 1933). Troilite, the iron-rich member, is stoichiometric FeS and has a superlattice of the 3A, 2C type (Hagg and Sucksdorf, 1933). Monoclinic pyrrhotite is the most iron-deficient type with a composition of approximately Fe₇S₈ (46.67 atomic percent iron) and a 2A, 4C cell (Bertaut, 1953; Wuensch, 1963; Carpenter and Desborough, 1964). Hexagonal pyrrhotite is intermediate in composition between troilite and monoclinic pyrrhotite. According to recent studies by Morimoto and others (1970), the composition ranges between Fe_9S_{10} and $Fe_{11}S_{12}$ (47.37 and 47.83 atomic percent iron). Superlattice types thus far recognized for hexagonal pyrrhotite include 5C (Carpenter and Desborough, 1964), 4C (Fleet and MaCrae, 1969), 11C (actually orthorhombic), and 6C (Morimoto and others, 1970). Intergrowths between troilite-hexagonal pyrrhotite and hexagonal-monoclinic pyrrhotite have been recognized in nature and produced experimentally by several investigators. A reliable technique for determining composition from d-spacing measurements has been described by Arnold (1962).

In 20 samples the type of pyrrhotite present was identified from x-ray powder photographs and diffractograms. Hexagonal, monoclinic, and hexagonal + monoclinic intergrowths were found. Troilite and troilite + hexagonal intergrowths were not identified. Based on the apparent absence of troilite, identification in the remaining samples where only traces of pyrrhotite were present was made utilizing a magnetic coating technique. A slurry mixture of hematite-rouge-chromic acid water, previously described by Cameron and Van Rensburg (1965) for hand buffing of polished sections to remove scratches and surface imperfections, was found to produce a coating of ferric oxide particles on monoclinic pyrrhotite, which appears bright red under crossed nicols. Reference specimens of troilite and hexagonal pyrrhotite were not coated by these particles. This technique is basically the same as that developed by Bitter (1931) who used fine-grained magnetite precipitated from a KOH solution as a coating agent to outline domains in magnetic materials. Papunen

(1970) previously used the Bitter method for identifying monoclinic pyrrhotites.

The structural types of pyrrhotite identified in samples collected from traverses A, B, and C are shown in Figure 1. In general, monoclinic and monoclinic + hexagonal (flame-type) intergrowths are more abundant at the lowest grades of regional metamorphism. Occurrences of a monoclinic rim (delineated by the iron oxide coating) around hexagonal grains were identified in several samples. This association is clearly different from the well-known flame-type exsolution intergrowths characteristic of hexagonal-monoclinic mixtures and may represent incipient weathering.

Although the first appearance of pyrrhotite shows a close correlation with the biotite isograd, the distribution of hexagonal and monoclinic pyrrhotites in traverses A, B, and C does not correlate with established silicate isograds. Williamson and Myer (1969) have proposed an isograd between monoclinic and hexagonal pyrrhotite in a Buchan-type metamorphic succession in Maine. There is no basis for concluding that such an isograd exists in the area shown in Figure 1.

TEXTURES OF IRON SULFIDES

Pyrite and pyrrhotite masses display a wide variety of forms, sizes, intergrowths, inclusions, and grain-boundary relations. Only the most common or distinctive relations are summarized here, but it is believed that they provide new insights into the behavior of these minerals during low-grade metamorphism.

Pyrite shows a variety of grain characteristics, but most of these can be classified into two categories - prograde and retrograde. Prograde features are indicative of an arrested state of growth. Crystals are mainly euhedral with straight crystal edges lacking serrations or embayments by adjacent minerals. Inclusions, if present, show little or no tendency toward elongation along the crystallographic directions of pyrite and are considered to be relict. Retrograde features, on the other hand, consist of irregular serrated grain boundaries, replacement veinlets of later minerals, and distinct control of the distribution of included phases by the crystallographic directions of pyrite. The latter suggests interior replacement of pyrite by later phases. This type of pyrite is indicative of an arrested state of decay. Pyrite commonly shows both prograde and retrograde features in the same grain. Because retrograde features are later, their presence indicates that sulfur has been removed from the volume of pyrite under observation. In the absence of other iron-bearing sulfides, oxides, or silicates as replacement products, migration of iron is also indicated.



Figure 2. Sketches of pyrite aggregates observed in sample TR-3 (Traverse B) 6.5 km northwest of the first appearance of pyrrhotite. A, planar concentration of pyrite grains; B, equidimensional (spheroidal) concentration; C through E, partially coalesced pyrite grains in spheroidal concentration; F, partially recrystallized subhedral pyrite grain.

Figure 2 illustrates prograde features of pyrite from the middle chlorite zone approximately 6.5 km west of the first appearance of pyrrhotite. Small grains of pyrite are shown to occur in varying degrees of "coalescence." A progression from isolated grains to coalesced grains is illustrated. Subgrain boundaries (not shown) can be observed microscopically in the coalesced aggregates which represent the boundaries of the grains prior to coalescence. Recrystallization of the coalesced pyrite grains is interpreted to yield euhedral cubes that lack subgrain boundaries. Inclusions present in Figure 2 represent entrapment of matrix during coalescence.

Figures 3A and 3B contrast the appearance of inclusions in prograde versus retrograde pyrite. Figure 3A shows angular inclusions (quartz) which are interpreted to be relict. Some replacement by pyrite is indicated. In Figure 3B the inclusions (quartz and chlorite) show a distinct orientation along the (100) crystallographic directions of pyrite. Interior replacement of pyrite by the included phases is indicated. Small blebs of pyrrhotite occur throughout the pyrite.

Figure 4 is likewise interpreted to show retrograde features. The distinct zonation along the margin of the cube is due to alignment of very small elongate silicate inclusions, and it probably represents primary growth zoning. The truncation of these bands by metamorphic minerals in the matrix clearly attests to the retrograde status of the pyrite.

The retrograde features described above are typical of rocks containing pyrrhotite. Thus the source of sulfur, and probably some of the iron in pyrrhotite, is almost certainly pyrite. The loss of sulfur from pyrite must be synchronous with the growth

of metamorphic minerals (chlorite, quartz, biotite, sericite) in the matrix.

A relation which appears to portray the development of pyrrhotite in low-grade metamorphic rocks is summarized in Figure 5. The rock consists of alternating dark-gray to black shaly beds and gray silty beds. Pyrrhotite is confined to the intersections of cleavage planes with pyrite-bearing silty



Figure 3. A, photomicrograph showing angular relict quartz inclusions (black) in pyrite (white). Some vein replacement of quartz by pyrite is indicated. Prograde pyrite texture. B, photomicrograph showing interior replacement of pyrite (white) by quartz (black) and chlorite (black). Scattered blebs of pyrrhotite(light gray). Distinct orientation of inclusions along cubic directions of pyrite.

beds. Under microscopic observation, the pyrite shows retrograde features. These relations may be explained by corrosion of pyrite, migration of sulfur in more permeable beds to cleavage planes, and deposition of pyrrhotite by replacement of silicates adjacent to the cleavage surfaces at these sites. The indicated scale of movement of the mobilized sulfur is on the order of a few centimeters or less. Figure 6 illustrates an elongate mass of pyrrhotite developed on a cleavage plane. Irregular grain boundaries and inclusions indicate that pyrrhotite formed by replacement outward from the cleavage planes. Locally, pyrrhotite streaks are enclosed in a sheath of quartz suggesting that silica, as well as sulfur, has migrated.

Near Murphy, North Carolina, pyrrhotite occurs along cleavage developed during the second period of folding (F_2) recognized by Power and Forrest (1971). It was not observed to be associated with cleavages developed during subsequent F_3 and F_4 fold episodes. Power and Forrest demonstrate that staurolite is post- F_2 but pre- F_3 folding. Thus, available evidence indicates that in the. Murphy area, pyrrhotite is emplaced along cleavage during the same time interval in which diagnostic silicate metamorphic minerals developed.

CONCLUSIONS

Prior to regional metamorphism, pyrite was the dominant iron sulfide in the Ocoee Series, occurring mainly as small disseminated grains. Evidence has been cited for coalescence of these particles as a mechanism for pyrite growth. The coalescence could represent movement along either chemical or pressure gradients to points of nucleation and growth. If dominantly a chemical process, small pyrite



Figure 4. Photomicrograph of retrograde pyrite (white) showing banding of inclusions (black) along the margin of the grain. Quartz (black) and chlorite (black) in the matrix have replaced pyrite, truncating the bands of inclusions.

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A showing the occurrence of pyrite along coarser silty beds and pyrrhotite along the intersections of these beds with the slaty cleavage.

grains would dissolve and reprecipitate at points of nucleation. This mechanism may be applicable to large cubes of pyrite that show compositional growth zoning. The nature, however, of the subgrain boundaries observed for certain coalesced aggregates. suggests an alternate mechanism. This type of occurrence is more readily explained by stress-induced migration during tight folding where pyrite grains subjected to differential stress would tend to move physically as discrete particles toward sites of minimum differential stress. This could be accomplished by continued recrystallization with growth on the surfaces facing the direction of minimum stress and by continued loss of volume on the opposite surfaces. Coalescence of grains at sites of



Figure 6. Photomicrograph showing typical occurrence of a "streak" of pyrrhotite (light) along slaty cleavage. Inclusions and irregular grain boundaries indicate replacement of silicates (black) by pyrrhotite.

minimum differential stress would yield aggregates of grains with subgrain-boundaries. With subsequent recrystallization of the aggregate, subgrain boundaries would be obliterated and euhedral outlines would develop. Silicate inclusions would develop by entrapment during coalescence.

As regional metamorphism progressed into the upper chlorite zone, pyrite became chemically unstable with respect to fluids generated within the recrystallizing pelitic sedimentary rocks. As pyrite was replaced by silicates, the mobilized sulfur and iron migrated to cleavage planes where pyrrhotite was deposited. The degree of pyrite stability and the extent of sulfur migration are possibly related to the carbon contents of the various rock units present. Empirical observations suggest that at a given grade of metamorphism, the pyrite/pyrrhotite ratio is higher for carbonaceous units. In addition, pyrrhotite commonly replaces pyrite directly in carbonaceous units, whereas pyrrhotite masses are typically separated from pyrite in rocks lacking graphite or other carbonaceous phases.

Quantitative estimates of the physicochemical conditions, which simultaneously promote the breakdown of pyrite and the growth of pyrrhotite, cannot be reliably determined with the limited data available and the complexity of the systems involved. The universal occurrence of hexagonal pyrrhotite in high-temperature environments (such as hypothermal ore deposits, igneous intrusives and contact zones, and high-grade metamorphic rocks) and the absence of reported occurrences of this phase in sedimentary rocks or lowtemperature environments, in general, suggest that temperature is a major factor that determines the formation of pyrrhotite. Experimental work by Toulmin and Barton (1964) indicates that both the fugacity of sulfur and temperature may determine the stability and composition of pyrrhotite. Interaction of iron sulfides with other volatile constituents such as CO2, H2, F, CL, O₂, CO, and CH₄ may also affect stability relations.

The parallelism between the first appearance of pyrrhotite and established silicate isograds and intimate textural relations between sulfides and silicates demonstrate that sulfide and silicate reactions during metamorphism are, to some degree, interdependent. Kullerud and Yoder (1963, 1964) and Guidotti (1970) have previously demonstrated the importance of the fugacity of sulfur on silicate equilibria.

Another aspect of this study (which is potentially very significant) is the demonstrated migration of sulfur during metamorphism. Sulfurization as an oreforming process has received considerable

attention in recent years. The migration of syngenetic sulfur from sulfide minerals in the rocks during metamorphism, and then the reaction of the sulfur with metal-bearing silicates to form base-metal sulfide deposits, has been advanced as a theory of ore genesis. The largest massive sulfide deposits in the United States are located in the Ducktown-Copperhill district, a short distance southeast of the study area. Fullagar and others (1967) have concluded from wallrock alteration studies that massive sulfide deposits in the Blue Ridge formed by sulfurization of wall rocks during regional metamorphism. They do not speculate on the ultimate source of the introduced sulfur. This study demonstrates a limited mobility of sulfur in the lowest grades of regionalmetamorphism effected by the breakdown of pyrite during metamorphism. Currently, the degree of sulfur mobility possible at higher grades of metamorphism where the massive sulfide deposits occur is conjectural but clearly warrants investigation.

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