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GEOLOGIC IMPLICATIONS OF JAROSITE, PSEUDOMORPHIC AFTER PYRITE

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Jarosite, $KFe_3(SO_4)_2(OH)_6$, occurs in the Boy Scout-Jones molybdenite prospect, Halifax County, North Carolina, where it exists sparingly as thin encrustations on fracture surfaces or as an alteration product of pyrite or mica. It is finely crystalline, but pure material gives a sharp *x*-ray pattern. *X*-ray line and peak values may vary slightly but are well within the limits of a relatively pure Jarosite end member as proposed by Warshaw (1956).

The sulfide deposit in which the jarosite occurs is hosted by a granite body enclosed by gneiss and cut by quartz veins and irregular areas of siliceous replacement (*U.S.B.M., Rept. Inves.* **4156,** 1947). All the units may contain zones of abundant to sparse pyrite with molybdenite and minor amounts of accessories. Jarosite occurs in the outer portion of the oxidation zone, which limits its formation to, or very near to, normal atmospheric temperature and pressure conditions. It is further limited in extent to the immediate presence of pyrite and mica or potassium-bearing feldspar.

All components of the jarosite were derived locally, at or very near the site of crystallization. During oxidation of pyrite in this zone, iron sulfates and sulfuric acid were released. This process is discussed by Merwin and Posnjak (1937). The ferric sulfate furnished iron while excess sulfuric acid released potassium from the mica and feldspar. A similar process for the derivation of potassium from volcanic rocks is discussed by Srebrodolsky (1959). In reference to his own work and that of other Russian workers, he states that during oxidation pyrite gives rise to cold sulfuric acid solutions which mix with potassium-bearing andesites and tuffs to give rise to a strong potassium sulfate solution. This later forms jarosite.

Deposition from such solutions along fracture systems accounts for most of the jarosite volume in the Halifax County deposit. Rapid dilution of the transporting solutions through the addition of surface water took place within a few inches to a few feet from the source of the jarosite components causing a rapid rise in pH and drop in ion concentration of the transporting solution with a resultant gradation from jarosite to goethite away from the jarosite component source. This pattern of gradation through the stability range of jarosite to goethite is consistent with the conclusions of Merwin and Posnjak (1937), though they consider it to be in the dilute, low acid section shown by their phase equilibrium diagram for the basic iron sulfates, in which they consider jarosite to be analogous to the basic salt $3Fe_2O_3 \cdot 4SO_3 \cdot 9H_2O$.

Jarosite also occurs to a very limited extent in the Halifax County deposit as an alteration of mica. Evidence to indicate the type process which occurred in this alteration is lacking and no conclusion was reached as to whether the process involved alteration by replacement or simple filling of a solution void. The universal closeness and relationships of altered mica to pyrite indicates that replacement was involved. Briggs (1951), noted a parallel occurrence in which he had little doubt that jarosite replaced glauconite. Similar conditions of pH and ion concentration would exist in the filling of a mica solution void as occurred in fracture filling. If, however, jarosite replaced mica, a different set of transport conditions would exist. Under these circumstances the necessary iron and potassium would be derived from the mica at the site of jarosite crystallization and sulfur from solution. Not only would the pH of the transporting solution be raised by dilution en route from the oxidizing pyrite source, but a further rise in pH would take place by reaction with alkalies along the route as well as at the site of deposition.

Of special interest in the Halifax County occurrence is the pseudomorphic relationship of jarosite after pyrite in which a complete sequence of steps in the replacement process was observed. Many instances occur where only incipient replacement of the pyrite has taken place along fracture surfaces within the pyrite crystal or along the surface junction of the pyrite crystal with its matrix. There are instances in which a remnant of pyrite was observed quasi-suspended in the interior of the pseudomorphic form. Many times the pyrite crystals are completely replaced by jarosite. Optical properties are continuous.

Where pyrite and jarosite occur together in the pyrite negative crystal form, neither microscopic nor *x*-ray diffraction study revealed the existence of a transition zone consisting of other, more soluble, basic ferric sulfates. This must mean either that the pyrite altered directly to jarosite or that, if a transition zone of basic ferric salts did form, it was subsequently altered to jarosite or was removed in solution. The latter possibility does not seem likely in view of the fact that pyrite was in direct contact with jarosite in all instances in which they were found together in the pyrite negative crystal form. Pough (1914) found jarosite both in contact with, and " . . . separated by a hair's breadth from the unaltered sulphide" in the subsurface weathering of pyrite ornaments.

Where pyrite is replaced by jarosite as it was in the Halifax County occurrence, potassium ions must be transported and introduced to the isolated pyrite-jarosite interface by solution. The potassium ions of the transporting solution were derived through the reaction of excess sulfuric acid with feldspar and mica of the deposit. A rise in pH of the transport solution would have taken place during this reaction with alkalies; therefore, the only further rise in pH which could have taken place, either to this solution or to the more concentrated solution at the immediate jarosite-pyrite interface, where oxidation was occurring, would have been through dilution by the addition of ground water. Continued oxidation of pyrite during the replacement process would not only furnish iron and sulfur but would institute and maintain a low pH value at the surface of jarosite crystallization on the pyrite-jarosite interface.

Warshaw (1956), who concurs with the work of Merwin and Posnjak (1937), concluded that intermittent dilution and the lowering of acidity by reaction with bases are the two processes leading to the formation of jarosite from the highly acid solution which first develops from the oxidation of pyrite. In contrast are the results obtained by Brophy *et al.* (1961), in the synthesis of jarosite at 105° C. under normal atmospheric pressure in a solution of 0.2 N sulfuric acid. The authors list an initial pH value of 0.5 for run 1–10 which contained 4.4 g K_2SO_4 and 39 g $Fe_2(SO_4)_3$ in 100 ml of 0.2 N sulfuric acid.

After dissociation a 0.2 N concentration of sulfuric acid would yield a pH of 0.958 $\cong$1.0. During dissolution of the jarosite-forming components in this solution, simultaneous reactions would occur, ~~with~~ the ferric ion hydrolyzing to increase acidity.

A comparable low pH value on the pyrite-jarosite interface of jarosite crystallization is indicated by the present study and would extend the stability range for jarosite into a much higher acid area than has previously been considered in natural occurrences.

Warshaw (1956), also concluded that the formation of jarosite in pyritic sediments results from the intermingling of solutions carrying the jarosite components. Oxidation of pyrite is considered the source of the sulfur and iron, and the alkalies are derived from other minerals. Tyler (1936), discussing the St. Peter sandstone in Wisconsin, suggests that jarosite may be an alteration product of glauconite attacked by sulphuric acid which was produced by oxidation of pyrite and marcasite. Mitchell

(1961), indicates a similar origin for jarosite in the Eocene Nanjemoy formation of Virginia. Neither of these authors has stated that alteration took place by replacement of glauconite or mica. Briggs (1951), who described jarosite formation in the Eocene of the San Joaquin Valley, California has indicated that jarosite can form as a near-surface diagenetic process involving the reaction of sulfate-bearing interstitial solution, derived from pyrite oxidation, with glauconite. Continuous optical relationships which exist leave little doubt but that jarosite has replaced glauconite. He further states that well cores from the buried sediments of the valley contain only glauconite and pyrite whereas the outcrop areas of the same horizons contain only glauconite and jarosite.

The present study has shown that jarosite can form naturally in the oxidation zone of a sulfide deposit under low pH values as an *in-situ* replacement of pyrite or from solution at a short distance from the derivation of the jarosite components. It is proposed that these conditions would also hold in the formation of jarosite from pyrite-bearing sediments. Not only could jarosite form as a replacement of glauconite and from the intermingling of solutions, as presently described in the literature, but as a replacement of the pyrite in pyrite-bearing sediments where potassium is available. High acid conditions would exist at the site of jarosite formation, and circulation of water would be important to transport but not in terms of increased dilution.

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