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The relationship between euhedral and framboidal pyrite in base-metal sulphide ores

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SUMMARY. Specimens of layered baryte containing a range of pyrite forms, including framboids, framboidal aggregates, spherulites, polyhedra, and zoned euhedral crystals from Kempfield, New South Wales, Australia, have been examined by mineragraphy, X-ray diffraction (XRD), X-ray fluorescence (XRF), and electron-probe microanalysis (EPMA). These investigations showed that the pyrite occurred in two forms—an arsenical variety containing approximately 5% arsenic in solid solution within the pyrite and a non-arsenical variety. EPMA studies indicate that the framboids and the cores of the zoned euhedral crystals have similar levels of arsenic and a genetic relationship is deduced to exist between these forms.

THE common occurrence of euhedral pyrite in sulphide ores and in their country rocks is well known to geologists involved in the study of economic mineral deposits, and a variety of models is available to explain its origin, including: magmatic segregation, hydrothermal solutions, sulphurization of iron-bearing minerals, volcanic exhalations, sedimentation, and diagenesis.

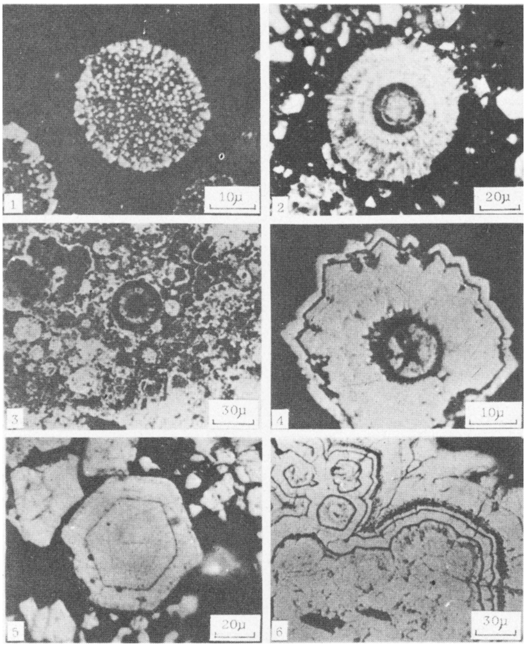
Framboidal pyrite is another important form of pyrite found in similar sedimentary and metasedimentary rocks and associated sulphide ores and also in recent sedimentary environments. It is this latter mode of occurrence that precludes anything except a low-temperature sedimentary/diagenetic origin for this particular form of pyrite. In particular the presence of abundant pyrite framboids in the Kupferschiefer suggested to Schneiderhohn (1923) that these pyrite forms were pyritized bacterial colonies, a concept that initiated a long and not fully resolved controversy into the origin of these minute pyrite arrangements. The discovery of similar framboids in a wide range of sulphide ores has made the interpretation of these structures a matter of difficulty. Here we are faced with a fundamental problem related to the principle of geological uniformitarianism—the framboidal forms present in sulphide orebodies, often apparently of high-temperature paragenesis, are found at the present time in low-temperature deposits such as peat, freshwater and marine muds, and coals.

The present paper is directed partly at this general problem and more specifically to an additional complication noted first by Love and Amstutz (1966). From their examination of pyrite mineralization in the Devonian Chattonooga Shale and the Rammelsberg Banderz they deduced that the euhedral pyrite of the Banderz was the 'equivalent' of the framboids of the Chattanooga Shale, and they concluded 'there is some suggestion not yet adequately supported that the outcome is conversion of framboids into homogeneous pyrite either in single grains or larger masses'. They mentioned the possibility of complete alteration of framboids to euhedral pyrite in certain deposits, with all trace of framboidal forms being obliterated—in other words, that the euhedral pyrite grains observed in certain sulphide ores may have passed through a framboidal stage in their development.

This paper represents an attempt to validate the tentative assumption of Love and Amstutz (that the euhedral pyrite of some ores may be a modification of earlier framboidal pyrite) and is based on a study of a small pyrite/baryte occurrence in New South Wales, Australia.

Occurrence. The Kempfield deposits (Stevens, 1974), which occur south-west of Bathurst, NSW, are essentially stratiform, and consist of lenses of baryte, chert, and disseminated sulphide enclosed in siltstones, arenites, and acid volcanic agglomerates. The rocks have been deformed by two periods of folding and have been metamorphosed to greenschist level. Stevens (1974) noted the presence of pyrite (partly framboidal aggregates) sphalerite, galena, tetrahedrite, and rare chalcopyrite in the deposits.

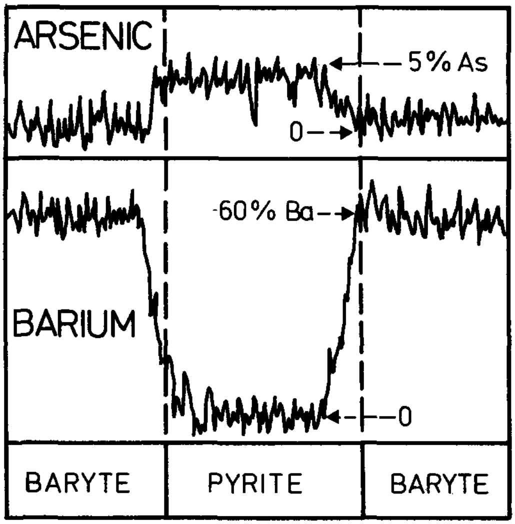
The consistently close parallelism of the baryte lenses and layers with the lithological bedding of the enclosing sediments suggests that the baryte

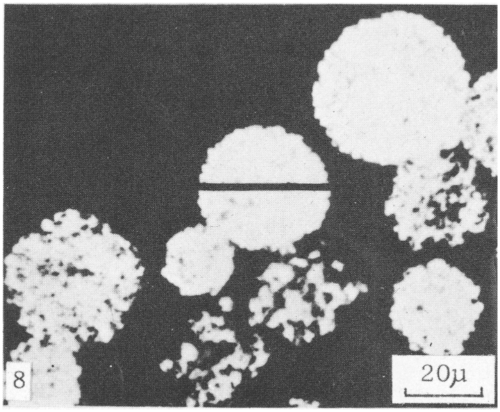


FIGS. 1-6: FIG. 1. Individual pyrite framboids in baryte matrix. FIG. 2. Pyrite spherulite, with internal cavity, in baryte matrix containing pyrite anhedra. FIG. 3. Cellular area of polyframboidal pyrite. FIG. 4. Pyrite polyhedron in baryte matrix. FIG. 5. Zoned pyrite euhedral crystal, and anhedral grains, in baryte matrix. FIG. 6. Pyrite showing fortification-type zoning in baryte matrix.

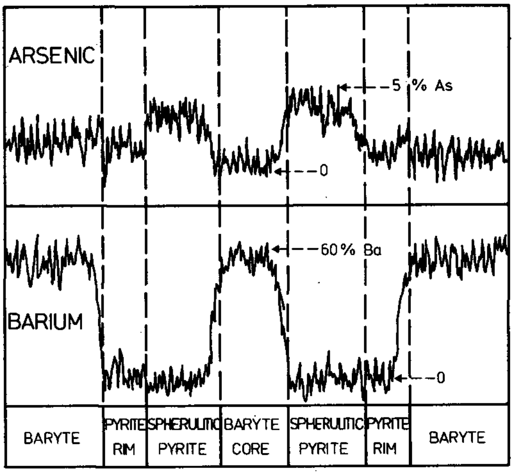
concentrations were either deposited at the same time as the sediments (syngenetic origin) or represent metasomatic replacements of selected beds in the sediments by $BaSO_4$ solutions (epigenetic origin). A syngenetic origin with the baryte and sulphides being derived from volcanic exhalations derived from submarine acid vulcanism is suggested by Markham (1974). This mode of origin for the baryte and sulphides appears likely to the writers on the basis of the parallelism noted above, of the absence of cross-cutting sulphide-gangue mineral veins of hydrothermal nature, and of the lack of obvious replacement structures within the baryte lenses.

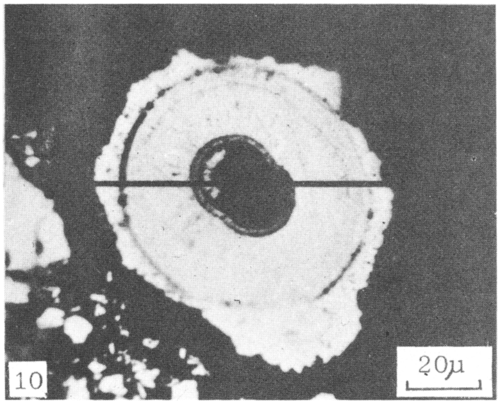
The specimens examined mineragraphically and petrographically by the authors were mineralogically simple, consisting of layered baryte containing disseminated pyrite within a fine-grained metasedimentary host rock. The pyrite occurs as masses of grains showing a range in grain size and morphology set in crystalline baryte. In particular the pyrite occurs: As individual framboids ranging in size from 2 μm to 20 μm (fig. 1). As radiating pyrite spherulites, from 30 μm to 80 μm in diameter, often with an enclosing crust of pyrite with crystallized margins (fig. 2); these spherulites sometimes enclose a framboidal core, and sometimes an internal cavity filled with baryte. As framboidal aggregates, consisting of compacted masses of framboids, from 0.1 mm to over 3 mm in dimension (fig. 3). As pyrite polyhedra, up to 0.1 mm in dimensions (fig. 4). And as pyrite euhedra, often with internal zoning, up to 0.5 mm in dimensions (fig. 5), and larger areas, some centimetres across, of pyrite showing fortification-type zoning (fig. 6).





FIGS. 7 and 8: FIG. 7. EPMA line scans across isolated pyrite framboid (pyrite 1) in baryte matrix. (Traced from original chart.) FIG. 8. Isolated pyrite framboids in baryte matrix. Black bar indicates direction of EPMA line scans shown in fig. 7.





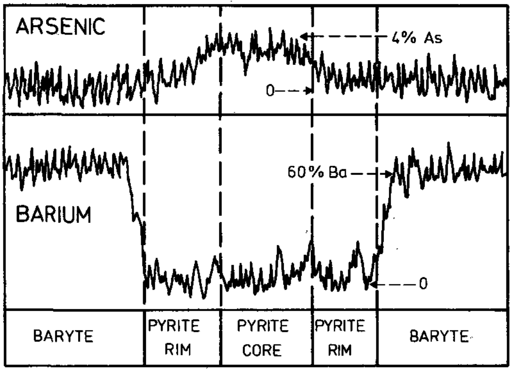
FIGS. 9 and 10: FIG. 9. EPMA line scans across a pyrite spherulite (pyrite 1) with hollow core and secondary pyrite crust (pyrite 2) in baryte matrix. (Traced from original chart.) FIG. 10. Pyrite spherulite with hollow core and secondary pyrite crust set in baryte matrix. Black bar indicates direction of EPMA line scans shown in fig. 9.

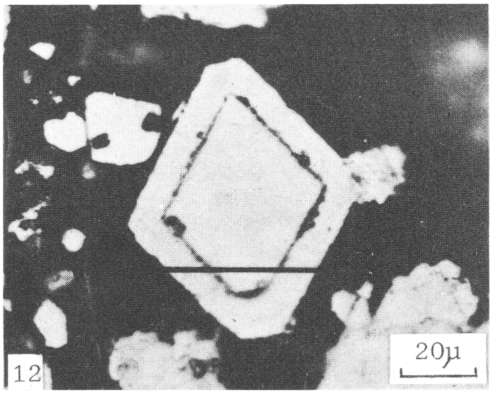
Pyrite mineralogy. The pyrite-bearing baryte samples were further examined by X-ray diffraction (XRD), X-ray fluorescence (XRF), and electron-probe microanalysis (EPMA). The XRD analysis indicated that the sample consisted of pyrite and baryte only, with no unidentified peaks. XRF scans indicated the presence of Fe, Ba, As, and S (major) and Sb, Cu (trace). Examination of the specimens by EPMA confirmed the XRF results. Arsenic was found to be present in most areas of the pyrite (at 4–5 %) and practically absent from other areas (less than 0.1 %). These investigations suggest that the arsenic occurs in solid solution in the pyrite and not as a discrete, arsenic-containing mineral. The mode of occurrence of the arsenic appears to follow particular patterns, which are illustrated by the EPMA line scans in figs. 7, 9, and 11:

Fig. 7 shows that the framboidal crystallites in baryte (fig. 8) contain up to about 5 % As, which is generally evenly distributed through the framboidal grouping. About 90 % of individual framboids examined by EPMA contained arsenic in the range 3–7 %.

Fig. 9 is from a scan across a pyrite spherulite body with a hollow, baryte-filled core and a crust of pyrite with crystallized margins (fig. 10). (Scans for sulphur and ion (not reproduced) show a similar pattern in both figs. 7 and 9; 55 % and 45 % respectively). Arsenic shows an even development (up to 5 %) in the spherulitic pyrite but is essentially absent from the pyrite crust.

Fig. 11 shows that the core of a zoned euhedral crystal in baryte (fig. 12) is arsenic-containing while the rim is essentially free of arsenic.





FIGS. 11 and 12: FIG. 11. EPMA line scans across a zoned euhedral pyrite grain in baryte matrix. (Traced from original charts.) Core is pyrite 1 and rim is pyrite 2. FIG. 12. Zoned euhedral pyrite crystal in baryte matrix. Black bar indicates direction of EPMA line scans shown in fig. 11.

It is thus possible to define two types of pyrite in the Kempfield specimens: Pyrite 1, with 5% arsenic, which makes up the free framboids, the pyrite spherulite bodies, and the cores of euhedral pyrite grains; and pyrite 2, without appreciable arsenic, which makes up the crusts of the spherulitic bodies and the rims of the pyrite euhedra.

This classification of Kempfield pyrite into arsenical and non-arsenical is paralleled by a variation in physical properties. Microhardness and reflectance values could only be carried out on the zoned euhedral pyrite but the results indicate that the arsenical pyrite is lower in microhardness and in reflectance at 589 nm than non-arsenical pyrite.

	Pyrite 1	Pyrite 2
As content	4-5%	less than 0.1%
R% (589 nm)	52.5	53.5
VHN (100 gm)	1340	1580

Discussion. These data may be interpreted in terms of a basic two-stage process: Recrystallization of primary arsenical pyrite (pyrite 1) framboids through an intermediate stage to euhedral pyrite, followed by precipitation of a later generation of pyrite (pyrite 2).

The intermediate-stage material is considered to be intermediate in morphology between the extremes of polycrystalline framboidal pyrite and single-crystal euhedral pyrite, and it may be represented here by the spherulitic bodies (figs. 2, 10).

In the pyrite occurrence at Allandale, New South Wales (Ostwald and England, 1977), recrystallized pyrite polyhedra appeared to be intermediate between framboids and euhedra in morphology.

Evidence for this genetic relationship between the framboids and euhedral crystals is the occurrence of similar levels of a replacement element (arsenic) in both framboids and euhedral crystals, and the absence of evidence for the formation of the euhedral crystals by any other mechanism, e.g. hydrothermal activity.

The mechanism by which the framboids are converted into euhedral crystals is not well understood. The possibility that euhedral pyrite grains are simply the product of crystal lattice oriented overgrowths (epitaxial deposits) on dispersed framboid microcrysts was mentioned by Chauhan (1974) in his study of the lead-zinc deposits of Zawar, India. While this growth mechanism has the virtue of simplicity the present studies indicate that whole framboids pass through a more compact intermediate form to form euhedra. This concept is supported by the discovery of framboidal cores in euhedral pyrite grains in certain base metal ores (Raybould, 1973) and by the framboidal pyrite cores in euhedral cobaltite described by Croxford (1974).

Although the genetic relationship between euhedral and framboidal pyrite has been investigated in a relatively simple environment, the Kempfield deposits as a whole contain disseminations and occasional bands of galena, sphalerite, chalcopyrite, tetrahedrite, and pyrite and Stevens (1974) considers they have a similar origin to the stratiform base metal sulphide deposits of the near-by Hill End Synclinorial Zone.

In general terms the transition from a stratiform disseminated sulphide deposit of the Kempfield type to a stratiform sulphide-rich 'ore' deposit of the Mt. Isa or Zambian Copperbelt type is not really difficult to envisage. Further modification under the influence of such earth processes as folding, shearing, and regional metamorphism with associated recrystallization, deformation, and mobilization of sulphide zones could well produce pyrite-containing, polycrystalline sulphide ores of 'magmatic' appearance. Remembering the well-known resistance of pyrite crystals to differential stress it is interesting to consider that the euhedral pyrite grains of certain base metal sulphide ores may be the end product of crystallization of pyrite framboids developed in saline muddy environments.

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