Annales de la Société Géologique de Belgique, T. 96, 1973, pp. 347-360 1973

POLYFRAMBOIDAL PYRITE OF THE ROCHELINVAL PYRITE BEDS (BELGIAN ARDENNES) (*)

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(14 fig. dans le texte)

ABSTRACT

A section of the Revinian near Rochelinval (Belgian Ardennes) shows four beds of « quartzite » (greywacke) which contain concentrated bands of polyframboidal pyrite : other beds contain a little dispersed polyframboidal pyrite. The concentrated bands were formed by submarine reworking of superficial, turbidite sediment which, when it was recently deposited, was of a type generally similar to that giving the other quartzites in the succession. The occurrences, and explanation offered are similar to those for a Silurian turbidite succession in North Wales, and indicate the possibility of a fruitful field of research into the sedimentology of the Lower Palaeozoic rocks of Belgium, despite the strongly tectonised and metamorphosed aspect of these rocks.

INTRODUCTION

The polyframboidal texture of pyrite was defined by Love (1971) from Silurian turbidite-greywackes of North Wales. It was shown to be a feature of the early diagenetic pyrite of these rocks and, in particular, concentrations of polyframboidal pyrite were described for the first time and attributed to the reworking and redeposition of pyrite from the newly depositied sediment on the sea-floor before burial. Although rare, such concentrations were found from areas 95 km. apart. Now in the present paper a similar occurrence is reported from the Revinien (Cambrian) of Belgium near Rochelinval. This shows that the phenomenon may be much less rare than formerly supposed, while this evidence of similarity to the Silurian turbidite facies of North Wales could be of value in the study of the Belgian Cambrian stratigraphy and sedimentology.

Also referred to is the work of Corin (1962) and Duchesne (1963) who have recorded from Belgian rocks aspects of the polyframboidal texture.

ROCHELINVAL PYRITE BEDS

The section studied (figs. 1 & 2) lies alongside the railway line from Trois-Ponts to Vielsalm in the valley of the River Salm, between 58.293 km and 58.312 km as

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(*) Communication présentée, et manuscrit déposé le 3 avril 1973.

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indicated by railway distance posts. It is north of the bridge where the road from Rochelinval village crosses the railway and then the river. It exposes black phyllites with green layers and thin beds of grey quartzites of the Revinien, of the Rn1b facies.

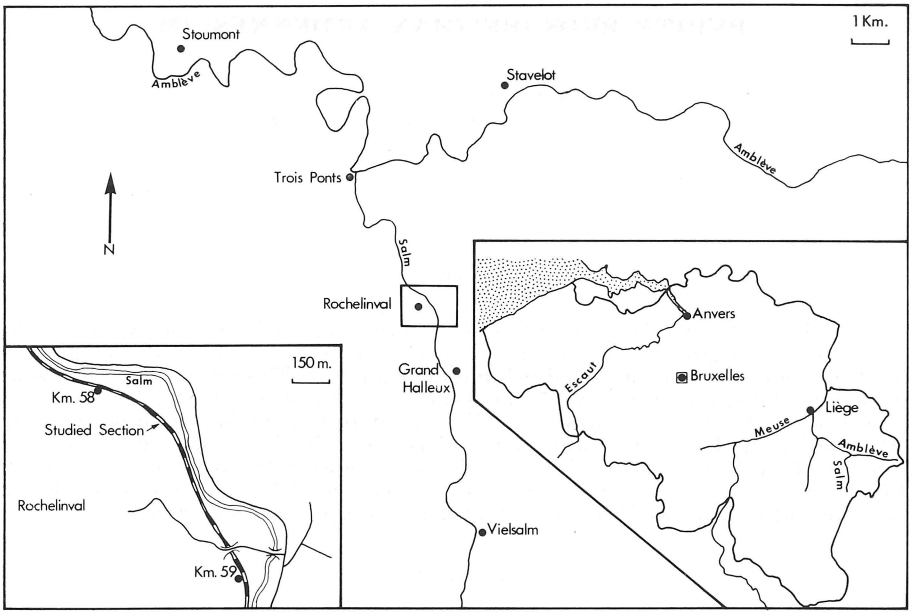


Fig. 1. — Location map, Rochelinval pyrite beds.

This section only in fact represents the middle part of an outcrop 150 m long described in summary by ANTHOINE (1940, p. M6 & fig. 2). He briefly noted one pyrite bed and that it been examined by de Magnée and Legraye. Subsequently Prof. de Magnée made a sample available to Love for preliminary study (LOVE, 1964, unpublished). The Rochelinval section was subsequently reexamined to determine whether this bed might be similar to the Welsh pyrite beds.

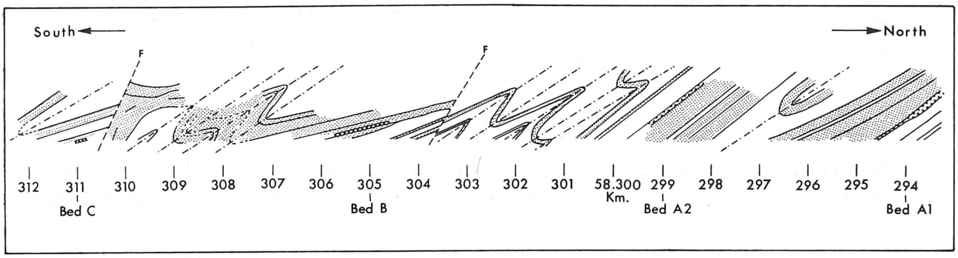


Fig. 2. — Section along cutting of railway from Trois-Ponts to Vielsalm between 58.293 km (as indicated by railway distance-posts). Only the rocks accessible from the level of the railway track are indicated : quartzites — fine stippling; pyrite beds — heavy dots; phyllites and quartzitic phyllites — unshaded.

On the side where the section is upstanding the railway cutting attains several metres in height. Only the part directly ascessible from the railway track, however, has been examined in detail.

From north to south the following successively are distinguished :

1. A syncline whose limbs show 2 m. of quartzite in 10-50 cm. beds, separated by thin phyllites. At the base of these quartzites is a bed rich in pyrite whose thickness attains almost 8 cm. at 59.294 km. (Bed A1) but which is reduced to 3 cm. at 58.299 km. (Bed A2).

2. A more phyllitic series with thin quartzite beds, more strongly folded. It is separated from the succeeding part by a small fault, probably of trivial throw, and emphasized by a thin discontinuous vein of quartz.

3. A different quartzitic group, folded, including a 3 cm. pyritic bed near its base (Bed B, 58.306 km.) directly overlain by a lenticular 13 cm. bed of quartzite. It is possible that this quartzitic group belongs to the same stratigraphical horizon as the quartzites described in part 1 of the section.

4. A synclinal fold in a more phyllitic group separated from that in B by a small fault similar to the previous one. One quartzite shows a ½ cm. of concentrated pyrite (Bed C, 58.311 km.).

MICROSCOPIC DESCRIPTIONS

Framboidal and polyframboidal pyrite

Framboidal pyrite is the microscopic texture of pyrite in which micron or sub-micron sized grains are combined in spherules from 5 μ to 50 μ in diameter. A detailed account with illustrations is given by LOVE & AMSTUTZ (1966). Polyframboids are larger bodies of pyrite, also generally rounded, but composed of a number of framboids in aggregation (figs. 3-6); they range up to perhaps 750 μ or more in diameter, but the smallest can overlap the size range of framboids, especially in section where full diameters are often not shown.

In adopting and defining the term « polyframboidal », LOVE (1971 p. 1038) attempted to combine the special textural relationship to framboidal pyrite with the contrasting emphasis on the higher order of complexity of polyframboids over framboids, and the need clearly to differentiate the textures if their particular significances are to be determined. It was felt by LOVE that this was better attained by incorporating the now well known and established descriptive word «framboidal », admittedly therefore continuing to call on familiarity with the raspberry (French : framboise — introduced by RUST, 1935) rather than with fish-roe (see below) and despite growing need to make more concise the use of « framboidal » itself.

FABRICUS (1961) in using the term Rogenpyrite, was drawing attention to the textural resemblance to roe, fish eggs, in clusters. As early as 1885 VON GÜMBEL used Rogenstein for mineral spherules in rock such as would mainly now be classed as ooliths. Other terms noted by FABRICUS (q.v.) to have been used for clusters of pyrite spheres or related bodies are Pyrithaufchen and Rogenhaufchen. KELLING (1970) has used the term Megaframboid. Other authors however have failed to make any distinction at all from framboidal pyrite or have not seen fit to use a distinguishing name.

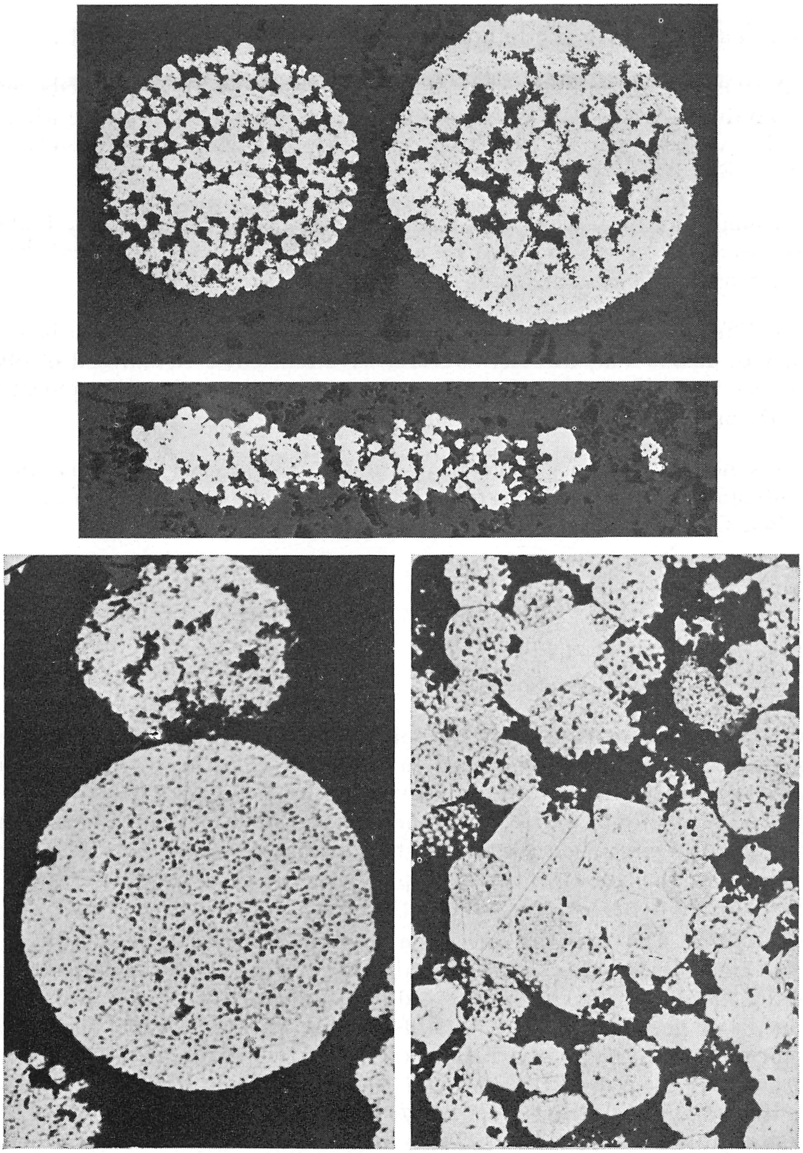


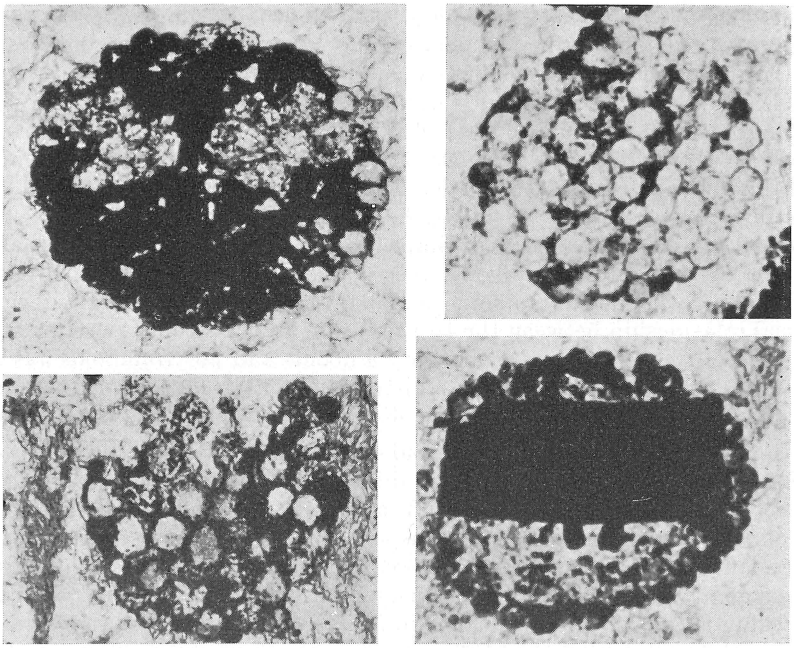
Fig. 3-5. — Polyframboidal groups of pyrite in polished section. That in Fig. 4 has possibly been crushed from an original spherical shape. Magnification about 250 ×. Fig. 6. — Polyframboidal pyrite and secondary pyrite within a concentrated band. Traces of pyrrhotite and chalco-pyrite are present in the secondary pyrite. Magnification about 50 ×.

In the quartzites free of polyframboidal concentrates normal framboidal pyrite and fine scattered grains dominate over polyframboids even to the exclusion of the latter. The combined framboidal and polyframboidal pyrite lie between 0.25 % and 3 % by volume (from point-counting), whereas in the concentrated bands as much as 70 % was recorded, dominantly polyframboidal with single framboids only contributing one unit to this value. In the estimations by point counter the whole of a polyframboid is taken as a single mineral grain, ignoring any cement-filled voids.

The figures 3-6 are chosen to show some of the main characteristics of the polyframboidal pyrite found in the Revinien of Belgium and to allow comparison with that of the Welsh Silurian. In general, however, the Belgian material is much less revealing of detail. The enlarged photographs show, for instance, how much the internal texture, both within and between the component framboids of the polyframboids' has been observed by internal coalescence or intergrowth of pyrite. LOVE & AMSTUTZ found this to be a character of ordinary framboidal pyrite and LOVE (1967) from Recent pyrite confirmed that it can occur in the early crystallisation stage of the pyrite. As a result here even many polyframboids have an almost homogeneous internal texture. It might be, however, that such a body as in figure 5, where even the outer boundary is simple, never actually underwent the process which gave the individual framboids, as is discussed later. In such a case it would be therefore a « very large framboid » and not a polyframboid and so demonstates the special relationship between the two textures. In the size measurements quoted below, the size frequency of recognisable framboids and polyframboids was bimodal around 35 μ in sections and in the size distribution finally quoted this was taken as the lower limit for including bodies of obscure internal texture as polyframboids.

Variations of the simple polyframboid structure may be seen best in the concentrated bands. They are classed under (1) differential internal texture of more or less complete bodies, (2) partial polyframboids appearing as if they are parts of complete ones broken and separated during transport — they are not seen in the normal quartzites, (3) looser aggregates of framboids, both in the main concentrates and in their weaker pyritic bands and in ordinary quartzites; beyond a certain degree of open packing they tend to be irregular in form and in some cases they can be seen clearly as polyframboids crushed between adjacent clastic grains. This is best demonstrated where, for instance, a partly stronger rim has withstood crushing while the remainder did not.

It is in the first variation that polyframboids of two concentrated beds (Beds A1 & A2) at Rochelinval show a difference from those of Conway. This is in the presence of forms where much of the pyrite of the individual framboids is absent, and this may occur (figs. 7-10) progressively until, with the almost total absence of pyrite the body appears, in thin or polished section, like a photographic negative of a normal polyframboid. However, even in the extreme case illustrated most of the component framboids have a peripheral layer of pyrite grains preserved, together with some inter-framboidal pyrite; both of these and the mineral infilling clearly delimit the position of the individual framboids. The exact similarity of structure of the totally and partially negative polyframboids with that of normal polyframboids indicates that negative parts are either incompletely developed or ones that have suffered a secondary removal of pyrite. Normally between framboids of the polyframboids silica and traces of sericitic mica form a cement : in place of the pyrite in the negative forms is a sheet silicate mineral, perhaps phengite, with low 2nd order birefringence colours. Limited by the framboid boundaries, and not providing grains large enough for completely satisfactory identification even under high magnification in thin section, in adjacent framboid positions it appears sometimes to be in optical continuity. It also occurs outside the limits of polyframboids but there subordinate to other micaceous minerals. The textural evidence is that the material is of secondary origin in the framboidal positions and the compositional indications support the extra availability of iron at the time of formation. This is discussed further below, with the origin of polyframboidal pyrite. X-ray diffraction of polyframboids with some secondary sulphide (see below) yielded evidence only of pyrite; no marcasite such as named by ANTHOINE (p. 6) was found.



Figs. 7-10. — Pyrite-deficient (« negative ») polyframboids in thin section in transmitted light. Magnification about 200 ×.

Secondary pyrite

Between and enclosing framboidal and polyframboidal pyrite is a clearer textured pyrite. As much as 25 % by area has been found in a concentrated band (where the total pyrite was as much as 80 %) and up to 6 % in a normal quartzite. In appearance this pyrite is usually euhedral except when in contact with polyframboids, but all gradations of incorporation of the latter are seen until only a relic texture remains. In a few cases the euhedral pyrite has formed a covering of small euhedral grains around part of a polyframboid. Both crushed and intact spheroids are enclosed, showing that the secondary pyrite must be later than compaction of the sediment : it must then have replaced rock matrix and also any material interstitial to the polyframboidal texture. Like the polyframboids the secondary pyrite has resisted tectonic deformation. A small proportion of pyrrhotite and a very little chalcopyrite are found in it (fig. 6).

The quartz veining cuts, and is later than, all pyrite. A few grains and polyframboids appear to have fallen into the vein filling material. The veins contain no evidence of the transport of pyrite but neither is there evidence of the solution of polyframboidal pyrite in the immediate vicinity of the secondary pyrite.

Organic material in the polyframboidal pyrite

When treated by the process described by Love (1956), involving concentrated Nitric Acid, prepared samples of dominantly polyframboidal pyrite from the Rochelinval pyrite beds as well as from the Conway beds yield residues of organic matter. Distinguishable in the Conway beds preparations are forms of the type once named as « *Pyritosphaera barbaria* Love 1956 », singly and in bonded clusters. Undoubtedly they represent organic material interstitial to the pyrite grains in the framboids of the polyframboids. From the Rochelinval beds the organic residues are less well preserved, as appropriate for more highly tectonised rocks but thin films of organic material which had surrounded the individual framboids are most prominent, containing often some form of residue of interstitial material as well (fig. 11). It is emphasised (Love 1964, p. 14) that this organic material is not regarded as representing the remains of any micro-organism essential to the formation of pyrite, but its presence is recorded as a matter of complete description and as an interesting link with simple framboidal pyrite from other rocks in which the phenomenon is well known. Electron microscope study of this organic material and reconsideration of its association with polyframboidal pyrite is being made by Love, C. D. Curtis and associates and will be reported elsewhere.

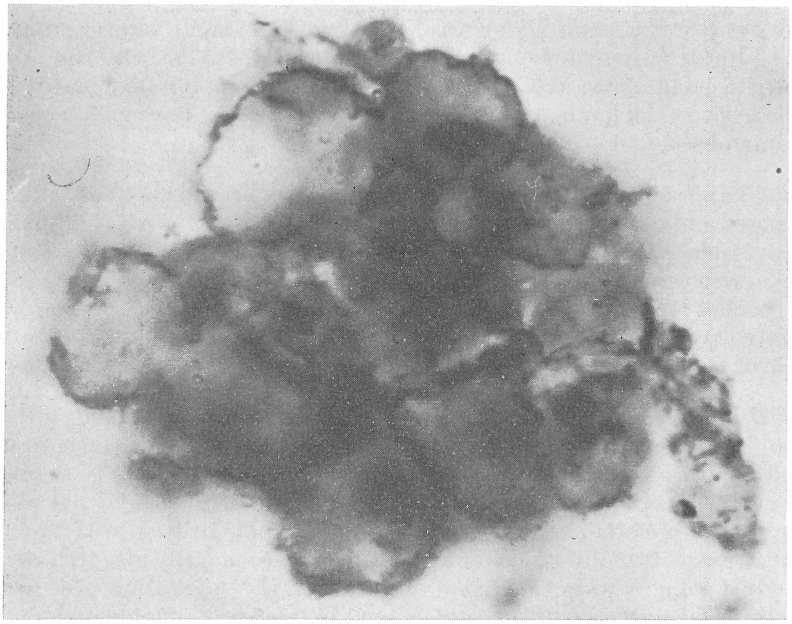


Fig. 11. — Residue of carbonaceous-organic material after solution of pyrite by HNO_0 from part of a polyframboid. The material principally appears as a resistent film of the external dimensions of individual pyrite framboids : more granular organic material lies internally in some. Magnification about × 1600.

Quartzites

Those quartzites associated with the pyrite are similar microscopically to those with little or no pyrite. They are variably sorted arenites sometimes containing, to a visual estimate, enough chloritic material to warrant description as greywacke, but the dominating quartz grains show strong interlocking and regrowth, with loss of much of the original texture to give the hard rock referred to as quartzite by ANTHOINE. They are interpreted to have contained much silt sized material as well as finer and medium sand. Due to the retexturing, clastic grain size analyses have not been made. In the vicinity of some of the pyrite, strain shadowing and radial recrystallisation of the pyrite is prominent in thin section. Stylolitic seams are common, as rather irregular partings in the rock, accompanied by small and irregular concentrations of chloritic material and pyrite of the type generally scattered through adjacent parts.

FIELD DESCRIPTION OF ROCKS

Bed A1 contains the thickest pyrite development and is probably that already known to the authors quoted. The whole quartzite is about 6-8 cm. thick and is exposed for 4 m. up-dip from rail level : possibly it died out above this, but folding occurs and exposure is poor. The bed includes up to 4 cm. of concentrated pyrite but at the upper end of the exposure this is split by a quartzite interlayer and the higher 1.5 cm. part of the pyrite has become much weathered and oxidised. All along, the pyrite is overlain by 2-4 cm. of quartzite.

The pyrite rests upon slatey mudstone : in the strong slickensiding at the contact no linear sedimentary structures have been distinguished. The upper part of the pyrite band shows cross lamination marked by polyframboidal pyrite and above the pyrite weak lamination is shown in the greywacke but much stylolitisation and veining obscures it.

Bed A2 appears as a stratigraphically inverted quartzite about 40 cm. in thickness, exposed only near the level of the railway line. At the stratigraphical base the pyrite is dense for up to 1 cm. thickness, and then for 2.5 cm. follows quatrzite-greywacke with discontinuous and mostly weakly pyrite-bearing laminae indicating mainly parallel-lamination but also some cross-lamination. The variability of the concentrated pyrite beds is emphasised by the differences seen in between beds A1 and A2 over the few metres of along-bed distance separating them.

Bed B is the middle one of three distinct beds together making up a 30 cm. quartzite. It is 12 cm. thick and at the level of the railway line it starts with a concentrated pyrite band up to 2.5 cm. thick, but its impersistence is shown by its absence only 2 m up-dip. Figure 12 shows thct, as in Bed C, the dense pyrite bed passes up into a cross-laminated division, perhaps ripple drifted, with much pyrite along the laminae. Stylolitisation, however, appears secondarily also to have concentrated pyrite along certain prominent surfaces. In the succeeding part of the bed pyrite is rare and slumping becomes prominent in the parallel-laminated and weakly cross laminated greywacke-quartzite. Some laminae marked out by polyframboidal pyrite are included in the slump overturns.

Bed C shows the clearest sedimentary structures associated with the polyframboidal pyrite (fig. 13). In a massive quartzite about 10 cm. thick and about 0.5 cm. above the bottom is a band about 0.5 cm. thick, very rich in polyframboidal pyrite. It passes up into parallel and cross-laminated greywacke clearly marked out by bands and trough-fills of pyrite. Some minor slump-overturn of the upper parts is seen in places. Stylolitisation is less intense than in Bed A1 but probably the cross laminated units have been modified by differential compaction around those richer in pyrite. Both top and bottom of the whole bed are strongly slickensided against slatey mudstone and current markings have not been distinguished.

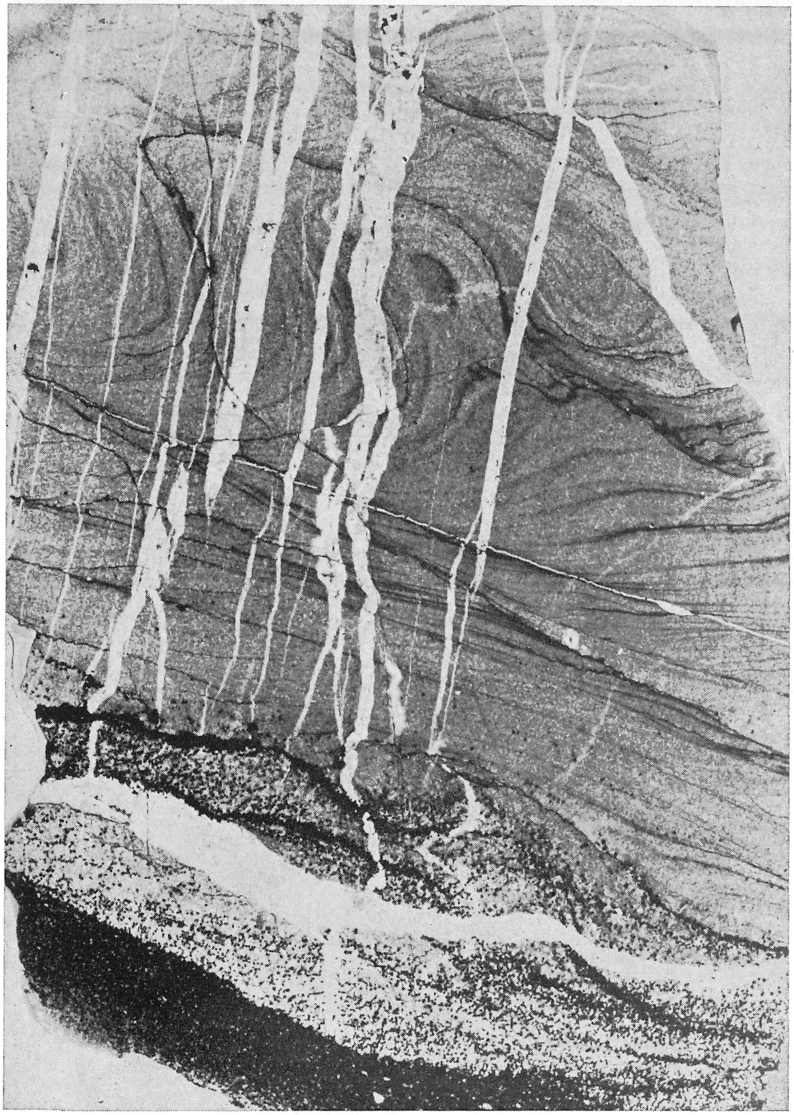


Fig. 12. — Thin section of Bed B to show bedding relationships of the pyrite band (dark) and the overlying quartzite. Magnification about 2 ×.

Other quartzites were collected within this section without finding further pyrite concentrates. Data on ordinary polyframboidal pyrite from some beds are included in figure 14.

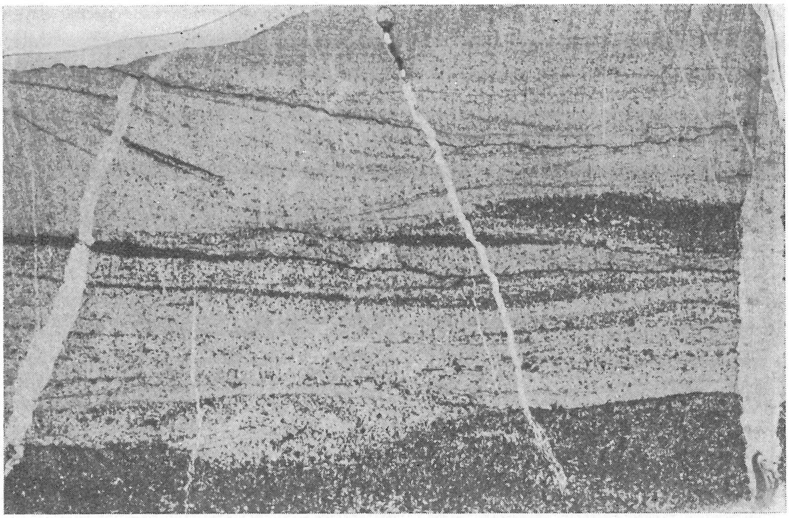


Fig. 13. — Thin section of Bed C to show bedding relationships of the pyrite band (dark) and the overlying quartzite. Magnification about 1.3 ×.

SIZE, ABUNDANCE AND CRUSHING OF POLYFRAMBOIDAL PYRITE

The distribution of median diameter in polished section of polyframboidal pyrite and its abundance and crushing (fig. 14) show a similar trend to that in Love

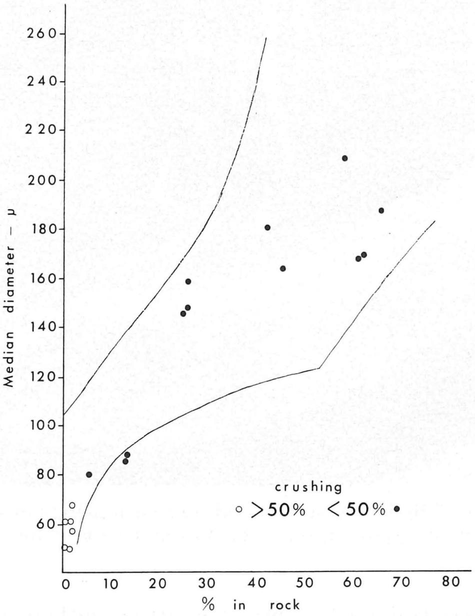


Fig. 14. — The relationship of median diameter of polyframboidal pyrite to its abundance in the rock sample. Sustantial crushing is also indicated in some samples. Lines indicate limits of similar data from Silurian pyrite polyframboids of Conway, N. Wales (Love 1971).

1971, (fig. 10) with the larger median diameters and extended size range being associated with greater concentration and lesser frequency of crushing of the pyrite. In this way the concentrated bands stand out clearly from the scattered occurrences of polyframboids in other quartzites, while between them in the distribution come data from those parts of the rock with the thinner laminae and from lesser concentrations adjacent to the concentrated bands.

DISCUSSION OF OBSERVATIONS

Love (1971) has pointed out that polyframboids appear to represent an extension of the phenomenon of the formation of simple framboidal pyrite and associated minute grains during the very early stages of diagenesis of sediments. While polyframboidal pyrite is not exclusive to turbidite-greywacke sediment, this is a lithology in which it seems regularly to represent at least a fraction of the early diagenetic pyrite. With their unusually rapid deposition, of mixed sediment possibly rich in iron-bearing clay and organic material, and maybe also introduced into stagnant or poorly oxygenated deep sea bottom waters, turbidites might indeed be regarded as a likely environment for the abundant development of early diagenetic iron sulphide. Probably this would initially be a form of FeS, to be followed by crystal· lisation of pyrite. It was Love's suggestion that the polyframboid-forming globules, at an intermediate stage, divided into the smaller globules which became the component framboids.

Authors generally are in agreement now that although the sulphide is of biogenic origin, much of it from suphate reduction, the form of framboidal pyrite owes nothing to the physical form of any micro-organism. Such organic material as may be found within ancient framboidal and polyframboidal pyrite is regarded as being of non-biological entry into the complex, contemporaneously or later.

It is still a point open to discussion whether or not spherulisation of iron sulphide can occur, as often suggested, due to the physical properties of the sulphide-sediment-fluid environment — that is some form of suface tension or immiscibility effect. Rickard (1970) has suggested that some pre-existing form is adopted, possibly of globules of organic substance (not organisms) or gaseous vacuoles. The stability of vacuoles of such a size as to originate polyframboids was not considered by that author, however, and could represent an insuperable problem in the adoption of this particular theory. In either case, however, before compaction the coarse texture of turbidites and the presence of large intergranular volumes of uncompressed clay could ideally accomodate the unusually large individual globules of iron sulphide recorded here together with normal framboids.

The trend combining increasing size and abundance with lower crushing of polyframboids referred to in the previous sections was suggested by Love to indicate that the concentrated bands, and indeed most occurrences above the 3-5 % polyframboid abundance, were caused by reworking of sediment already containing scattered polyframboids and other pyrite, to give these rare concentrated bands in the process. The differing, and decreased, degree of crushing of polyframboid aggregates was taken as a further indication of the energy of the erosive and transporting agency : from an unconsolidated sediment in which framboidal and polyframboidal pyrite was forming, mostly only the stronger, better coherent aggregates could survive the removal and redeposition without breaking up into smaller particles that would be swept on elsewhere with the bulk of the sediment. The thicker concentrates and laminae of polyframboids must represent just those critical conditions of deposition where the larger pyrite alone could be dropped or rolled without so much of the other material accompanying it.

In this the negative polyframboids pose a problem to be resolved, as kindly pointed out by I. de Magnée and G. C. Amstutz (personal communications). It is felt here, however, that their presence does not invalidate the general hypothesis. Among the conditions under which iron sulphide mineral might be differentially removed from polyframboids three appear most worthy of consideration (i) direct solution of pyrite either in the original or in the derived sediment or its subsequent rock (ii) supergene or submarine weathering and solution of pyrite in the original or derived sediment or rock, (iii) submarine oxidation of the unstable primary iron sulphide precursor of fully crystallised pyrite. Against (i) is the selectivity of process having only affected two of the four closely spaced concentrated beds of pyrite and none of the normal quartzites. Both of conditions (ii) and (iii) might initially leave a residue of goethite-type material — $Fe(OH)_3$ — able to influence the iron content of subseqentuly crystallising material, but in (iii), the selective solution of incompletely pyritised polyframboids best fits the environment of formation.

For this case it might therefore be suggested that for Beds A1 and A2 reworking of original sediments occurred before the full processes of framboidal pyrite formation in the polyframboids had been allowed time to pass to completion such as would have happened if they had been left undisturbed. For normal framboidal pyrite, such a state was represented in the Recent sediments of the Wash reported on by Love (1967, Table I & p. 338-9). There the black spherules were respectively completely, incompletely or not at all soluble in dilute acid or were progressively less completely changed to red ferric hydroxide on standing exposed to air, in accordance with the decreasing proportions of FeS to FeS_2 present; this was also inferred to indicate the stage individually attained by the black spherules in the FeS to FeS_2 transformation. In the case of the negative polyframboids of Rochelinval such oxidation most likely would occur during reworking or following redeposition of the concentrates in a chemical environment sufficiently oxidising to affect FeS but not pyrite. This hypothesis requires that the range of undamaged negative polyframboids in the concentrated beds were all in fact physically strong enough to suffer erosion and transportation in the company of completely pyritised polyframboids and also to resist compaction crushing subsequently. Fig. 9 shows an incomplete polyframboid which might have lost part of its structure during transport. Experiments, at present incompleted, on the behaviour of polyframboids in a small water flume indicate that separation from quartz sand can indeed be achieved without violence, at least under conditions conducive to rolling; indeed, the accompanying illustrations of negative polyframboids as preserved in the concentrated beds show always a distinct framework of crystalline pyrite whose intergranular adhesion must have provided enough strength to allow preservation uncrushed. Whether geothite pseudomorphing FeS added some strength, or how early the translucent mineral infilling developed, is not ascertained.

Subsequently to the deposition of reworked material, and subsequently to any temporary oxidation reactions, if the sediment attained a generally reduced condition before, say, burial to 1 metre depth, it might be regarded natural that further framboidal or polyframboidal pyrite could develop. If so this could give some overlapping and blurring between the groupings defined from Figure 14.

It is impossible to point to any individual source bed from which, before its consolidation, sea-floor currents could have taken material for the selective deposition of pyrite to give the concentrates. The latter probably really are localised in occurrence and extent, so a local source is most likely. While it is not so clear, as in the Welsh rocks, that deposition of the concentrated beds immediately followed some erosive and channeling activity of the transporting current, evidence has been given of cross lamination in some of the concentrates. It is likely that somewhere not far away each of these beds was undergoing localised erosion by a late stage of its own depositing current or a subsequent one of a turbid nature.

PREVIOUS BELGIAN RECORDS

The examples illustrated by CORIN (1962) certainly include polyframboidal forms of pyrite. Unfortunately only small amounts of borehole core are now available for reference (personal communication, 1971) and the evidence for the presence of a concentrated band of polyframboids remains only in that author's figures 9-14. From the Revinian of the Bayehon valley, DUCHESNE (1963) has illustrated material from thin sections which show all the appearances of polyframboidal pyrite and yet which are not now pyrite. Possibly with some organic matter present, and certainly mainly siliceous, these bodies might represent cavities from which pyrite was oxidised and removed in solution from pyrite framboids. The polyframboids beds at Conway show at their present weathered surfaces relic forms in limonite preserving much physical detail of the pyrite : and complete replacement could also happen during diagenesis or tectonism and metamorphism.

SIGNIFICANCE OF THE PYRITE BEDS OF ROCHELINVAL

The Cambrian rocks of the Ardennes, being strongly tectonised and partly metamorphosed, have only slowly yielded up finer details of structure and sedimentation. Hence, while GEUKENS (1962) has tentatively suggested a turbidite origin for part of the Revinien (Rv3) it appears useful that for other parts certain sedimentological characteristics are now found comparable with another, better known and more easily studied region of turbidite deposition.

In North Wales LOVE (1971) identified the rare concentrated polyframboidal beds as showing either ripple drifting after erosive channelling by the depositing current, or alternatively, distinct regular lamination. For this, and from the general lithological character of the arenites, it was tentatively suggested that the submarine currents reworking the turbidite sediment to give the concentrates might themselves have been turbidity currents. The current directions determined fitted those already demonstrated regionally for the turbidite deposition. Unfortunately in the case of the Rochelinval examples it is not possible to attribute current directions, nor has this been done for other supposed turbidites in this succession. It appears likely, however, from this first study, that detailed search might reveal further examples of the reworked pyrite beds and other well preserved and recognisable features of tubidite deposition in the Revinien and other Lower Palaeozoic rocks of Belgium.

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