Upper Palaeozoic millerite-bearing ironstones of the South Wales coalfield

Introduction

Sedimentary ironstones are widely developed in the Westphalian (Upper Carboniferous) 'Coal Measures') of South Wales, and provided the majority of ore for Welsh iron production. Under this group of iron ores there are two subtypes: blackband ironstone, and brownband or claystone–ironstone. Claystone–ironstone nodules (Figure 5.80) occur throughout the 'Coal Measures', forming bands in the dark-grey mudstones adjacent to coal seams, while blackband ironstones are relatively restricted in occurrence. They are concentrated stratigraphically in the lower Westphalian C strata, particularly in the Margam area, close to the postulated coal basin depocentre (Young, 1993). They were mined for their iron content, but in comparison to some areas, such as the Midlands, where they formed the backbone of the iron industry, production in South Wales was relatively limited in comparison to the brownband or claystone–ironstones.

Between 1855 and 1917, a total of 17.1 million tons of iron ore were produced from the brown-band ironstone beds (Young, 1993). The industry dosed in the 1930s. Since the ironstones often occurred intercalated with coal seams, it was often the case that both coal and iron ore were worked from the same pit. Common throughout the British coalfields, the South Wales claystone–ironstone are of particular importance in mineralogical terms for the well-crystallized sulphide assemblage developed in septarian cracks within the claystone–ironstone nodules. This assemblage, accompanied by siderite, dolomite, calcite, quartz, barite, carbonate-fluorapatite, waxy hydrocarbons and clay minerals, comprises millerite, galena, chalcopyrite, sphalerite, pyrite, marcasite and siegenite. It is, however, the excellent acicular groups of millerite crystals, reaching several centimetres in length on occasion, that have chiefly made the South Wales ironstones internationally famous in mineralogical terms.

Recently, similar, and clearly structurally controlled mineralization, has been observed *in situ* encrusting open joint-surfaces in 'Coal Measures' sandstones in the north-western part of the coalfield (Bevins and Mason, 2000). Together with recent fluid-inclusion and isotopic studies (Alderton and Bevins, 1996; Alderton *et al.,* 2004), the new discoveries throw a considerable amount of light on the genesis of the mineralization, and may have connotations with regard to the overall evolution of the coalfield.

In the South Wales Coalfield the selection of a single representative site for conservation purposes is not practical. The reason for this is that exposures of the productive strata are transient, as they are essentially limited to opencast coal mines, which tend to be short-lived and are covered by rigorous planning conditions, including their complete restoration upon cessation of mining. However, as one opencast closes, another opens, so that opportunities to study this internationally important mineralization *in situ* should continue to be available for the foreseeable future.

The ironstone-hosted sulphide mineralization of South Wales is also present in the once numerous coal tips of the area, albeit to varying degrees. However, due to the grassed over nature of these tips, good opportunities to sample and study the mineralization occur only when the tips are being landscaped, a process which ultimately removes the mineralogical resource. Nevertheless, a comprehensive suite of material is preserved in the Mineral Collection of the National Museum of Wales, where it is available for study.

For the above reasons, the millerite-bearing claystone–ironstone mineralization has not been allocated a specific GCR site, although it is of GCR importance. As such, the paragenesis of the mineralization is described here in detail because of its relevance to the overall metallogenic framework of Wales. In field terms, it is recommended that visits should be arranged to whichever opencast workings are in operation at any given time.

Description

The claystone–ironstones sometimes occur as continuous layers but are more commonly observed as concretionary bands, the concretions varying in size from a few centimetres to over 1 m. The concretions tend to have a flattened appearance, and are often 'bun-shaped', with a convex top and flat base. The ironstone is a hard, splintery micritic siderite-dominated rock, of a dark-grey colour, weathering to brown. The concretions are hardest internally, with relatively soft outer zones.

Most of the concretions contain thin siderite-lined cracks, but a considerable number host networks of open, septarian fissures. These form interconnecting mosaics of principally vertical, often slightly curved, fractures, extending from the centres of concretions to the outer zones, where they die out before reaching the concretion surface. The cracks are lined with crystalline siderite, sometimes in sharp contact with the micritic siderite of the fracture walls, but more often appearing to pass gradationally from micrite through dark-brown, increasingly sparry siderite to the yellowish-white crystalline variety, in a texture suggestive of wall-rock metasomatism. The presence of carbonate-fluorapatite as a late-stage mineral crystallized on siderite and millerite has been described recently by Plant and Evans (2005).

The sulphide minerals occur on the siderite and have clearly grown in an open-space environment. Many fine specimens have been recovered over the years (Bevins, 1994), including some of the world's finest examples of millerite, with sprays of lustrous, sometimes twisted, acicular crystals reaching over 4 cm on occasion, and water-clear quartz crystals 3–4 cm in size, locally referred to as 'Merthyr Diamonds'. Of particular note, and still not uncommon today, are small sprays of millerite upon which are threaded bright cubo-octahedra of galena. Sphalerite also overgrows millerite, while chalcopyrite is so infrequently observed in actual contact with millerite that a strict paragenetic relationship has yet to be determined. Siegenite is relatively minor and forms usually microscopic (< 1 mm) octahedra scattered about on the siderite. Pyrite and marcasite are relatively rare within the septarian cracks, although a few fine specimens have been reported (Bevins, 1994). Barite is also rare and is a late-stage mineral. The various waxy long-chain hydrocarbons, such as hatchettite ($C_{38}H_{78}$), occur as orange to yellowish spheroids (Firth, 1971).

The distribution of the claystone–ironstone sulphide minerals has been debated by various authors (see North and Howarth, 1928; Firth, 1971). It has long been suggested that millerite tends to be confined to the part of the coalfield in which high volatile bituminous coals occur, and is apparently absent from the north-western anthracite zone. However, millerite has recently been identified from the anthracite zone, occurring on joints in sandstones associated with quarrz, ankerite and chalcopyrite (Bevins and Mason, 2000). This sandstone-hosted mineralization is in fact also widespread throughout the South Wales Coalfield, and at the time of writing was well exposed at the Nant Helen Opencast site, Abercraf (Bevins and Mason, 2000).

At Nant Helen, the sandstones carry a strong set of open, mineralized joints orientated approximately north–south. In some sections, conjugate joint-sets striking north-east-south-west and north-west-south-east, are also mineralized in a broadly north–south obtuse zigzag pattern. East-west joints are unmineralized. The joints are most heavily mineralized above the hanging-wall of a significant N–S-trending, E-dipping normal fault plane (with evidence for a separate phase of strike-slip movement). Away from the fault, the amount of mineralization decreases steadily until, at about 50 m distant, only traces are present. The fault appears to post-date the mineralization, as heavily slickensided quartz and ankerite, in both normal and strike-slip orientations, can be seen on the fault plane.

The mineralization is crustiform in nature, consisting generally of simple, fissure-wall-coatings with a well-defined paragenetic sequence. Initial mineralization consisted of the development of thin (< 1 cm) fibre-quartz spanning slowly opening fissures. However, this eventually gave way to more expansive opening of the joints, with the result that the fibre-quartz crystals developed well-formed terminations where detached from the wall-rock. The second generation of quartz, nucleating upon some of the terminated fibre-quartz crystals, occurs as large (up to 4 cm x 3 cm) crystals of the 'Merthyr Diamond' habit, some occurring as sceptres upon the fibre-quartz and others as flat-lying doubly terminated forms.

Both generations of quartz are overgrown, abundantly in places, by rhombic ankerite (typically 10–20 mm), which is yellowish when fresh but a rich tan-brown where weathered. Additionally, further fracturing accompanied the ankerite deposition, so that in places the ankerite coats otherwise unmineralized rock surfaces, or cements cracked quartz. Sulphides occur sporadically on both ankerite and quartz and are most abundant in the widest, most intensively mineralized, sections of the joints. Chalcopyrite is abundant as scattered 1–3 mm sphenoidal crystals. Millerite is more restricted in occurrence, and seems to occur commonly along joints in certain sandstone beds but is absent from others. Millerite forms characteristic sheaves of acicular crystals up to 3.5 cm, the larger crystals completely spanning the open joints.

Finally, unusual mineralization has recently been recorded from bedding-normal micro-fractures ('cleats') in anthracitic coals in the north-western part of the coalfield (Gayer and Rickard, 1993). As well as a variety of sulphides, lead-selenium minerals and microscopic, collomorphic gold occur within this assemblage.

Interpretation

The blackband ironstones have been interpreted as having been formed as the result of the diagenesis of a 'bog iron-ore'-type precursor, although direct siderite precipitation from iron-enriched tropical swamp waters is also thought to be possible (Young, 1993). The genesis of the brown claystone–ironstone beds and nodules has been the subject of considerable debate (summarized in Young, 1993). They may occur close to marine bands, or within totally non-marine parts of the Westphalian sequence. Typically, the largest ironstone beds were generated within fine-grained sediments which were deposited in a lacustrine swamp environment. Within these sediments, concretions nucleated around organic debris and other 'seeding' agents.

The source of the iron (and minor manganese) could have been soil sesquioxides or unstable silicates (Young, 1993). Microbial oxidation of adjacent organic matter, resulting in the reduction of Fe(III) to Fe(II), also released HCO_3, the consequent rise in alkalinity favouring carbonate precipitation. Thus, concretion growth involved the deposition of micritic siderite within the pore spaces of relatively unconsolidated sediments, so that the central part of any concretion has the highest micritic siderite content. These early, and probably rapid, stages of growth may have been initiated under as little as 1 m of sedimentary cover (Curtis *et al.,* 1986). As burial depths increased, concretion growth continued, the final stages, under perhaps a burial depth of several hundred metres (Curtis *et al.,* 1986), producing, in the case of the South Wales ironstones, the characteristic relatively siderite-poor rim.

The formation of the septarian cracks has been the subject of many theories. A popular and long-standing theory, discussed by Astin (1986), involved the existence of an initially soft concretion interior which subsequently dehydrated, leading to formation of shrinkage cracks. However, as Astin (1986) pointed out, the specific nature of the postulated clay-rich centre or precursor gel has not been explained. Furthermore, it is hard to explain the nature of a suitable chemical environment during diagenesis capable of dehydrating clay-rich concretion centres.

Using examples occurring in Jurassic and Eocene strata, Astin (1986) concluded that the septarian cracks developed as stress-induced tensile fractures during progressively deeper concretion burial. Under these conditions, the principal stress involved is the load pressure, related directly to depth of burial. As a consequence of the load pressure, horizontal tensile stresses arise and, for a rock of given tensile strength, tensile fractures will form instantaneously when the effective minimum tensile stress equals the tensile strength.

This theory is attractive, since it explains certain features, in particular the failure of the septarian cracks to reach concretion surfaces. These outer layers, transitional to the mudstone host-rocks, with a much lower micritic siderite content, would have a much lower tensile strength than the intensely cemented inner zones. Under high load pressures, this contrasting rheology would result in progressive plastic deformation of the weak outer zones about the rigid interior, until the effective minimum tensile stress reached the tensile strength of the interior, resulting in the brittle failure of the inner zone.

The mineralization of the septarian cracks within the nodules has yielded important data regarding the P-T conditions under which the assemblages were formed. Alderton and Bevins (1996) examined fluid inclusions in quartz crystals from nodules collected at the landscaped Wyndham Colliery, near the centre of the South Wales Coalfield. Fluids were found to be of low salinity but highly methanoic. The study led to the conclusion that the quartz occurring within the nodules formed at a temperature of around 150° C under a pressure of around 500 bar. More recently, Alderton *et al.* (2004) have compared fluid-inclusion data for samples from the ironstone nodules and from fracture fillings in sandstones (see below), and concluded that the carbonates probably crystallized at relatively low temperatures (< 100° C), whilst the quartz formed at a later stage and at higher temperatures (between 150° C and 200° C). Their evidence of variations in the temperature and composition of the mineralizing fluids which correlate with coal rank variation across the coalfield indicates a probable causal link. Their data also confirmed a geothermal gradient of *c.* 45° C km^{-1} at maximum burial. Such figures suggested a much higher palaeogeothermal gradient in the area than was previously considered. This, combined with other evidence, implies a much higher degree of heat flow than one would expect for a foreland basin setting to the South Wales Coalfield (Kelling, 1988; Frodsham and Gayer, 1997), and the possibilities of other basin development mechanisms, for example lithospheric extension, require further examination (Bevins *et al.,* 1996b).

The recently discovered sandstone joint-hosted mineralization at the Nant Helen Opencast is important because the assemblage in many aspects resembles that of the septarian cracks, suggesting the likelihood that the two styles of mineralization are cogenetic. Additionally, the clearly defined structural trend of the sandstone joint mineralization relates the mineralization to a specific structural regime operating at a specific stage of basin development.

The paragenetic features of the Nant Helen mineralization indicate that it developed over a N–S-trending structural weakness along which tensile stress was focused during regional extension associated with basin deepening. This was relieved initially by the opening of joints, with fibre-quartz development, in the sandstones and by ductile deformation in the mudstones and coals. Increasing tensile stress opened the joints out into small veins and was finally relieved by much larger-scale normal faulting.

Faulting similar to that exposed at Nant Helen is abundant across the whole of the South Wales Coalfield, trending north-west-south-east in the eastern part of the coalfield and NNW–SSE or north–south in the western sector. This intensive fault pattern is consistent with east-west to north-east-south-west extension across the coalfield during basin development. If the fault at Nant Helen is typical, the faulting postdates the mineralization on the sandstone joints, and the common orientations of both fault and joints suggests that they both developed as part of the same overall process U.S. Mason, unpublished interpretation).

Similar quartz-bearing sandstone joint assemblages occur in the western extension of the coalfield, for example in coastal exposures in the Saundersfoot district of Pembrokeshire. However, in this area, which lies to the south on the Variscan Front, tectonism has resulted in the shattering of the mineralization, leaving the open joints crammed with the broken shards of the quartz crystals (Bevins and Mason, 2000), indicating that such mineralization clearly predates the Variscan compressive deformation.

Evidence therefore suggests that the mineralization of the South Wales Coalfield is a pre-Variscan, burial metamorphism-/extension-related event within a rapidly subsiding extensional basin (Bevins *et al.,* 1996b; Bevins and Mason, 2000). The age constraints are demonstrable: the mineralization was clearly developed at some point between sediment deposition in early to mid-Westphalian times, and late Westphalian times, when intense deformation occurred which shattered the mineralization at sites south of the Variscan Front.

Methanoic, low-salinity mineralizing fluids (Alderton and Bevins, 1996) were generated from sedimentary porewaters during burial-related low-grade metamorphism under a high geothermal gradient. The fluids remobilized Ni, Co, Cu, Pb and Zn from the sediments (including the ironstones) and re-deposited them upon accessing nearby low-pressure zones, such as tensile fractures, as small amounts of their sulphides, accompanied by quartz, clay minerals and abundant, metaso-matically recrystallized siderite (in ironstones) and ankerite (in sandstones). Traces of Au and Se were also deposited, particularly on fracture surfaces within anthracitic coals (Gayer and Rickard, 1993).

As subsidence waned, so did the mineralizing activity, so that the Upper Coal Measures (Pennant Sandstone Formation; Westphalian C–D) is, so far as has been observed, unmineralized. At the end of Carboniferous times the area underwent a period of uplift, folding and thrusting along the east–west Variscan trend. To the south of the Variscan Front, in southern Pembrokeshire, the basin sequence was intensely folded, and in this area the majority of the delicate crystal growths developed during the mineralization event were destroyed.

Conclusions

An important form of mineralization occurs in strata of the South Wales Coalfield, principally within septarian cracks in claystone–ironstone nodules but also on open joints in sandstone beds of Lower to Middle Westphalian (A–C) age.

These were deposited in a rapidly subsiding coal swamp basin and rapidly subjected to a steep geothermal gradient and overpressuring as a result of deep burial. Horizontal tensile stresses increased as a consequence of the increasing load pressure and deformed the coals, mudstones and ironstone concretion exteriors plastically, while units of higher tensile strength, such as sandstones and the interior zones of ironstone concretions, underwent brittle failure as the minimum effective tensile stress eventually exceeded their tensile strength. Metals, liberated from the ironstones and associated sediments during porewater migration, were re-deposited as minor amounts of well-crystallized sulphides within the septarian ironstone concretions and on sandstone joints.

References

(Figure 5.80) Photograph of claystone–ironstone nodules exposed in Cwm Gwrelych, Glyn Neath. (Photo: R.E. Bevins.)