Mineralogy of the Silvermines district, County Tipperary, Eire.

(With Plates IV to VI.)

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Summary. The paragenesis of a suite of hydrothermal minerals (sulphides, baryte, rhombohedral carbonates, quartz, sericite, and chlorite) at four mines in this district is discussed. Of particular interest are the conversion of pyrrhotine to marcasite, which is shown to take place by loss of iron, the replacement of blende by chlorite, and the possible formation of primary goethite. The occurrence of gudmundite is recorded for the first time in the British Isles.

JUST over fifty years ago, Russell (1907) published in this journal 'Notes on the Mines and Minerals of the Silvermines District, County Tipperary'. A recent survey of various eighteenth- and nineteenth-century workings and one modern productive lead mine near Silvermines, coupled with microscopic examination of the ores, has revealed the presence of additional minerals and some unusual textural features.

A fuller description of the mines, local geology, and economic mineralization has been published elsewhere (Rhoden, 1958a, 1958b). Briefly, the ore deposits occur in Devonian and Carboniferous rocks bordering the Silvermines Fault, which is a complex system of major and tributary fractures that can be traced for over 25 miles across northern Tipperary and southern Clare.

Shallee and Gortnadyne mines.

At Shallee mine, ramifying veinlets of galena, baryte, and other minerals, listed in order of deposition in table I, are concentrated beneath gently dipping shale horizons in strongly fractured Devonian sandstones. At Gortnadyne mine the copper lode consists mainly of brecciated Devonian sandstone, re-cemented by quartz, sulphides, baryte, and carbonates.

Although dislocation of the beds preceded the onset of mineralization, structural dilation of the fractures continued throughout the sulphide phases, and finally ceased during the deposition of baryte, which filled TABLE I. Epigenetic minerals at Shallee and Gortnadyne mines.



the remaining space in the veins and breccias. During mineralization, the red and buff sandstones and shales were bleached to light grey and olive-green colours respectively, with redeposition of sedimentary hematite and goethite as pyrite; at the same time, ilmenite grains were replaced by clusters of rutile crystals set in translucent leucoxene and quartz.

The first epigenetic mineral of which traces still remain was pyrrhotine. Platy crystals, up to 1 inch square, grew parallel to the sandstone wallrocks, giving the veins a streaky appearance. After slight fracturing, the pyrrhotine was often surrounded by euhedral crystals of arsenopyrite, and of a rhombohedral carbonate that was afterwards converted to ankerite and goethite (see p. 133), before being finally set in a matrix of quartz and later sulphides. During the subsequent deposition of copper, antimony, zinc, and lead minerals, all the pyrrhotine changed to marcasite and pyrite (see p. 131).

Of the principal minerals, pyrite was deposited earliest. Small pyrito-

hedra line the veinlets at Shallee mine, and are scattered throughout the sandstone and shale wall-rocks, having formed at the expense of sedimentary iron oxides. A few thin beds of dolomitic and sandy shale, however, contain abundant needles of arsenopyrite, to the exclusion of pyrite, indicating that arsenic may have been an original constituent of the rock (Goldschmidt and Peters, 1934).

Two forms of *blende* occur: one is dark brown, iron-rich, and often contains tiny inclusions of exsolved chalcopyrite; the other type is light yellow and free from inclusions. Deposition of the dark variety, which is more common at Gortnadyne than at Shallee, overlapped the formation of pyrite, producing alternate colloform banding. The purer blende was the last sulphide to crystallize, and is often associated with veinlets of ankerite. *Chalcopyrite*, followed closely and partly replaced by *tetrahedrite*, succeeded pyrite at both mines, but attained economic grade only at Gortnadyne.

The most abundant sulphide in the veins at Shallee mine, and probably also in the breccia at Gortnadyne, is galena; but if the 'ore' is assumed to include the inter-vein sandstone, galena may be exceeded by pyrite. Although galena slightly replaced all the earlier minerals, most of it was deposited in open fissures; but in places where the vein-walls had not been lined by pyrite or chalcopyrite, galena also replaced quartz and claymica minerals to give the marginal sandstone a distinct blue-grey coloration. Galena at Shallee mine contains 44 to 53 oz. of silver per long ton of lead metal (production data for 1949 to 1958), and at Gortnadyne, 20 to 25 oz. per long ton (Wynne, 1861).

A remarkable feature of the galena at Shallee mine is the multitude of inclusions of bournonite, boulangerite, and tetrahedrite. Some tetrahedrite grains are plainly relics left during the replacement of surrounding chalcopyrite; part of the bournonite may be a reaction 'front' between partly replaced copper minerals and galena; but most inclusions are remote from such replacement contacts, and the microscopic texture indicates that they have exsolved from the galena during cooling. Boulangerite occurs as a 'rash' of tiny untwinned laths up to 40 μ long, oriented along crystallographic planes within the galena. Bournonite inclusions, showing excellent parquet-like twinning, are larger and more rounded in shape, and tend to segregate at grain boundaries, leaving peripheral areas of the galena free from inclusions. Tetrahedrite is less common away from contacts with copper minerals, but occurs in the galena like bournonite. Inclusions of different minerals are never intermingled, but may occur in neighbouring grains of galena. Although

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direct evidence is lacking, exsolution probably commenced with tetrahedrite, continued with bournonite, and ended with boulangerite; the type of inclusion produced in each grain of galena would depend on the concentration of copper and antimony, and on the original temperature of deposition. In direct contrast, galena at Gortnadyne mine rarely contains any exsolved minerals.

The growth of fibrous mats of *chlorite* and *sericite* in open veins at this stage of mineralization is described below (p. 134). Deposition of creamy-coloured *ankerite* (ω 1.712 to 1.720), containing a trace of manganese, probably started shortly after the pyritic stage, but reached a modest peak as sulphide mineralization ended with the crystallization of light yellow blende (see table I). The carbonate at Gortnadyne is contaminated by the presence of dolomite in parts of the breccia. Any remaining space in the veins or breccias was filled by pure white *baryte*, the last primary mineral deposited. The usual range of secondary minerals, listed in table I, developed during erosion.

Conversion of pyrrhotine to marcasite. The alteration of pyrrhotine to marcasite and pyrite is not a rare phenomenon in moderate to low temperature sulphide deposits,¹ having been first reported over fifty years ago (referred to by Allen, Crenshaw, Johnston, and Larsen, 1912). Some writers (summarized by Edwards, 1954) consider that the change occurred during the closing stages of mineralization, while others (Foslie, 1950) regard it as a supergene process. Although similar conversions of pyrrhotine to marcasite have been observed in ores from Abbeytown, Co. Sligo, Eire, and from Stari Trg, Trepča, Yugoslavia, the unique suite² of minerals from the Silvermines district establishes the exact point during the primary mineralization at which the alteration occurred.

As the first epigenetic mineral, pyrrhotine crystallized only in the 'strongest' veins at Shallee mine, and in the breccia at Gortnadyne. Single platy crystals with sharp edges, illustrated in pl. V, fig. 3, grew parallel to the vein wall, giving an appearance of 'flow texture' to the veins; pyrrhotine never occurs in the granular or rosette form commonly found at other deposits,¹ including Abbeytown and Trepča. After being lightly fractured, the pyrrhotine plates were partly covered by arseno-pyrite (shown in pl. IV, fig. 1) or carbonate (possibly chalybite, see p. 133) crystals, before being finally set in quartz and later sulphides.

¹ Described by O'Malley and McGhie, 1939; Stillwell and Edwards, 1943, 1945, 1946; Stillwell, 1944; Carter, 1948.

² Type samples from Shallee and Gortnadyne mines are included in the reference collections of Economic Mineralogy at the Imperial College of Science and Technology, London.

Pyrrhotine remained stable throughout the pyritic stage of mineralization, but changed entirely to marcasite and pyrite during the subsequent deposition of copper, antimony, lead, and zinc minerals. Despite the simultaneous crystallization of iron-rich minerals, such as blende, chalcopyrite, and tetrahedrite, nearby and even within the pseudomorph, the pyrrhotine altered to disulphide by expelling roughly half its original iron content, and without consuming additional sulphur (see below). As the conversion progressed, tiny shrinkage cracks opened parallel to the basal parting of the pyrrhotine crystals, which otherwise retained their shape. The reduction in volume from pyrrhotine to disulphide can be measured microscopically, and is remarkably constant at 32 to 38 %.

Theoretically, the conversion of pyrrhotine to marcasite can take place in three ways (for convenience pyrrhotine is assumed to be FeS (troilite)): constant volume, in which sulphur must be added to the system and iron removed in the proportion expressed in the equation 4FeS+2S = $3\text{FeS}_2+\text{Fe}$; constant iron, in which sulphur is added (FeS+S = FeS₂), and the resulting marcasite occupies over 33 % more space than the original pyrrhotine; and constant sulphur, in which almost half the iron is expelled (2FeS = FeS₂+Fe), leaving marcasite or pyrite to occupy only 66.8 % or 65.2 % of the pseudomorph respectively. Although natural pyrrhotine, which has a defect structure (Hägg and Sucksdorff, 1933), varies in composition between Fe_{0.991}S and Fe_{0.893}S with consequent change in density, its original composition should not change the ratio of atomic sulphur to molecular volume, nor affect the theoretical shrinkage to marcasite or pyrite in a constant sulphur system.

By comparing the theoretical reduction in volume by $33 \cdot 2$ % for marcasite or $34 \cdot 8$ % for pyrite with the measured 32 to 38 %, there can be no doubt that the pyrrhotine at Shallee and Gortnadyne mines changed to disulphide solely by the loss of iron. The same applies to the conversion of granular pyrrhotine at Abbeytown mine and Trepča, and to the typical 'bird's eye' textures observed in other ore deposits.

As the pyrrhotine decomposed, the shrinkage cracks were filled by the mineral being deposited in the vicinity. Most cracks contain chalcopyrite, tetrahedrite, iron-rich blende, galena, or quartz (see pl. IV, figs. 1 and 2), any single crack usually being filled with more than one mineral; less commonly, bournonite, boulangerite, gudmundite, a rhombohedral carbonate, and even later minerals, like iron-free blende and baryte, may be included. Although most of the pyrrhotine changed to marcasite, some idiomorphic pyrite developed near the edges of the pseudomorphs, breaking the continuity of the tiny shrinkage cracks (see pl. IV, fig. 1). The marcasite occurs as minute lath-shaped crystals, oriented at right angles to the cracks and often separated from each other by almost sub-microscopic partings containing other minerals.

Gudmundite. Liberation of iron from the decomposing pyrrhotine produced some unusual results in the narrow shrinkage cracks and along the edges of the pseudomorphs. During an influx of tetrahedrite, a momentary excess of iron over copper would cause the deposition of gudmundite (FeSbS), and the remarkable angular texture illustrated in pl. IV, fig. 2; the tetrahedrite crystals shown in pl. V, fig. 4, were partly replaced by chalcopyrite, then covered by a thin, even layer of gudmundite, before remaining space in the centre of the crack was filled by quartz. Rarely, intergrowths of gudmundite and tetrahedrite occur separately from the pyrrhotine pseudomorphs.

Although the properties of gudmundite closely resemble those of arsenopyrite (Buerger, 1939), it can be readily identified in polished sections by its strong rose-coloured pleochroism. Ridland (1941) reported a Talmage hardness of F for gudmundite, similar to arsenopyrite; but most observers, including the writer, prefer a rating of D, closer to tetrahedrite. The apparent rarity of gudmundite may be due solely to the failure of many geologists to distinguish it from arsenopyrite.

Gudmundite, which is usually found in hypothermal deposits like the complex sulphide masses at Boliden (summarized by Sampson, 1941), or the gold-stibnite veins of Southern Rhodesia (Morgan, 1957), has not been previously reported from the British Isles, and may occur in other leptothermal deposits similar to those at Silvermines.

Replacement of carbonate mineral. The crystallization of a rhombohedral carbonate around pyrrhotine and on vein-walls before the deposition of pyrite was mentioned above (p. 129). Despite their characteristic outline, the rhombs are invariably filled with a granular aggregate of ankerite and goethite, which vary widely in relative proportion. Interlocking grains of ankerite are rarely in definite orientation either to one another or to the pseudomorph margins. In many pseudomorphs, like that illustrated in pl. V, fig. 5, galena appears to vein and partly to replace the aggregate along grain boundaries, or along the ankerite cleavage. In most samples, the galena seems to be related to the existing aggregate rather than to any 'ghost' structure of an original single carbonate crystal.

Theoretically, three interpretations of the texture are possible. Galena

may have veined and partly replaced either the original carbonate, the remainder of which changed to ankerite and goethite during supergene alteration; or an aggregate of ankerite or chalybite, only the goethite being formed later; or the present aggregate of ankerite and goethite, this being a primary replacement of an original chalybite crystal.

The third hypothesis accounts for the texture best, for goethite is always present in the pseudomorphs, while surrounding sulphides show no sign of supergene alteration. If the original carbonate was chalybite that had crystallized shortly after pyrrhotine, but before the complete sulphidation of iron oxides in the surrounding sandstones, it may have become unstable during the later phases of mineralization, when true ankerites were being deposited; as the chalybite changed to ankerite, some of the excess iron would remain in the pseudomorph as primary goethite. In a low-temperature deposit, like that at Shallee mine, such residual goethite might survive through the final stages of galena-baryte mineralization, for the decomposition temperature of goethite is about 150° C. in neutral or weakly alkaline hydrothermal solutions (Smith and Kidd, 1949). The formation of crystalline goethite in similar ores was reported by Edwards (1955).

Redeposition of pyrite. Crystallization of pyrite and blende overlapped at Gortnadyne mine, producing alternate colloform banding and mutual replacement. The dark brown iron-rich blende illustrated in pl. V, fig. 6, was lightly brecciated, then partly replaced by pyrite; subsequently, both minerals were replaced by galena. But the texture shown in this plate is unusual, for pyrite is moulded round typical galena crystals, and forms long, thin, broken veinlets along the cubic cleavage. This moulded form of pyrite is usually devoid of relics of blende.

The best explanation is that as galena replaced both minerals, some of the pyritic material remained almost *in situ*, and was redeposited around the growing crystals of galena, or along the cleavage; slight pressure would break these narrow veinlets shortly after formation. Zinc, however, would be removed from the site of replacement.

Chlorite and sericite in veins. Although extensive zones of alteration are not discernible in the wall-rocks at Shallee mine, the sericitic groundmass of sandstones and shales is often replaced by chlorite near some of the 'stronger' veins. During the later phases of mineralization (see table I), platy mats of chlorite and sericite lined the edges of freshly opened fissures, which were subsequently filled with galena, ankerite, or baryte. The feathery plates of sericite illustrated in pl. VI, fig. 9, curve and branch within galena without regard to the cubic cleavage; similarly, in other samples, chlorite plates finger into baryte crystals without disturbing their optical continuity or utilizing their excellent cleavage.

In each instance, the microscopic texture indicates crystallization of galena and baryte around the chlorite or sericite plates, rather than development of the latter in fractures in, or replacing, the galena and baryte. Sometimes, rows of these plates lining a vein-wall are bent the same way, suggesting the flow direction of the solution depositing the encasing mineral. Growth of chlorite and sericite in veins, probably from material derived mainly from the wall-rocks, must be regarded as a distinct phase of mineralization, occurring just before the main deposition of galena and baryte.

Replacement of blende by chlorite. An unusual replacement phenomenon occurs at Shallee mine, in which isolated crystals of dark brown blende containing exsolved inclusions of chalcopyrite are marginally replaced by bright green chlorite. The original outline of a euhedral blende crystal, the margin of chlorite, and the unreplaced core, are illustrated in pl. VI, figs. 7 and 8. Irregular masses of light yellow blende, shown in pl. VI, figs. 8, have replaced clay-mica minerals in the rock groundmass surrounding the pseudomorph. Since the quantity of the light yellow form is comparable with the amount of replaced copperrich blende, the inescapable conclusion is that blende replaced by chlorite was immediately redeposited nearby purged of its inclusions. Much of the uncommon, pure blende found at Shallee may have been derived from an earlier copper-rich variety (see paragenesis in table I).

According to Hey's (1954) optical data, both chlorites ($\omega = 1.64$, $\epsilon - \omega = -0.004$) mentioned above can be classed as brunsvigite.

Knockanroe and Ballygown South mines.

Along the northern side of the Silvermines fault at Knockanroe and Ballygown South mines, fractured dolomite near the base of the Carboniferous Limestone series contains disseminated pyrite, marcasite, blende, galena, and baryte. During prolonged pre-glacial erosion of the orezones, sulphides were oxidized and the products, with baryte and silica, became progressively concentrated with the removal of the host-rock. Less soluble minerals, such as anglesite, cerussite, unaltered galena, and baryte, accumulated on the surface with gossan. More soluble material, like zinc and some iron, migrated downwards in sulphate solution to replace fresh dolomite, forming smithsonite and chalybite respectively; these, in turn, changed to hemimorphite and goethite as the level of oxidation gradually deepened. The high-grade residual lead-silver 136

deposit, which gave rise to the name 'Silvermines', was exhausted during the seventeenth and eighteenth centuries; below it, the enriched zinciron ore-zone still remains, known as the 'Calamine Bed'. Primary minerals identified at these mines are listed in order of deposition in table II, and secondary minerals in order of depth zones in table III.

TABLE II. Primary minerals at Knockanroe and Ballygown South mines.



* Pyrrhotine exsolved from the earliest variety of blende, during cooling.

The first epigenetic minerals in the dolomite, or in the sandstone faultbreccia known as the 'Sulphur Lode', were *pyrite* and *marcasite*. Spherulites, up to 2 inches in diameter and composed mainly of radiating pyrite crystals, often contain wedges or segments of marcasite; the relic texture illustrated in pl. VI, fig. 10, indicates that clumps of marcasite crystals in the centres of spherulites may subsequently change to pyrite. Slight variations in pH caused by the dissolution of dolomite, rather than small changes in temperature, pressure, or concentration, would determine the mineral formed at the point of precipitation at each moment (Allen, Crenshaw, Johnston, and Larsen, 1912). As Tarr (1927) suggested, marcasite would crystallize during an influx of slightly acid solutions, but pyrite would be deposited as the quantity of dissolved lime increased and the temperature of the solutions decreased.

TABLE III. Secondary minerals at Knockanroe and Ballygown South mines.



As in similar rocks at Shallee, abundant needles of arsenopyrite

developed in certain thin shaly beds within the dolomite, to the exclusion of pyrite or marcasite.

Deposition of *blende* overlapped that of pyrite and marcasite, causing alternate colloform banding round spherulites. The earliest blende was a dark brown variety, containing up to 4 % Fe, from which pyrrhotine subsequently exsolved; thereafter the blende became progressively purer, until clear, light yellow crystals were formed at the end of mineralization. The exsolution of pyrrhotine from blende at Knockanroe and Ballygown South mines, rather than chalcopyrite as at all other deposits in the Silvermines district, is noteworthy; it indicates that the total absence of copper minerals in the present superficial workings will probably continue in depth.

Deposition of galena started shortly after the earliest blende, but finished just before the final and purest variety of the latter. The high silver content of the lead ores made the locality famous in olden days; the figure of 80 oz. of silver per long ton of smelted lead quoted by Rutty (1772) and Wynne (1861) referred mainly to the oxidized ores, but Apjohn (1860) confirmed the same quantity in fresh galena. The last truly epigenetic mineral was *baryte*.

The hydrothermal minerals are closely associated with irregular veinlets of a light pink *dolomite* (ω 1.69) containing traces of iron and manganese, which has the same composition as the dark blue-grey host-rock. Redeposition of dolomite started during the later phases of sulphide mineralization, and continued afterwards until most fissure and solution cavities were sealed. At the western end of Knockanroe mine most veinlets were filled with baryte, which effectively restricted the recrystallization of dolomite.

The secondary ores at these mines resemble 'calamine' deposits elsewhere. In the residual lead-silver zone, cerussite and anglesite replace galena, often leaving a core of sulphide protected from oxidation. In the deeper 'calamine' zone, clear crystals of hemimorphite (first discovered by Capt. King (Wynne, 1860), and identified by Apjohn (1860)) occur in an ochreous mass of goethite, chalybite, smithsonite, unaltered primary minerals, and brown-stained dolomite.

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EXPLANATION OF PLATES IV TO VI.

PLATE IV.

- FIG. 1. Edge view of pyrrhotine pseudomorph, composed chiefly of marcasite, and surrounded by euhedral arsenopyrite crystals, quartz, chalcopyrite, and tetrahedrite. In the pseudomorph, shrinkage cracks contain quartz (black), blende (dark grey), tetrahedrite (medium grey), chalcopyrite (light grey without relief), and galena (light grey with strong relief); areas of plain relief near margins are pyrite, which break the continuity of the cracks. Shallee mine, 320-ft. level, 9.6 W. stope; polished section $\times 170$.
- FIG. 2. Pyrrhotine pseudomorph containing marcasite (light grey) and quartz. (black spots), with tetrahedrite (dark grey) in the shrinkage cracks. Angular lines are gudmundite, which covered the tetrahedrite crystals during a momentary pause in growth. Gortnadyne mine, breccia; polished section $\times 930$.

PLATE V.

FIG. 3. Side view of platy pyrrhotine crystals, showing sharp edges, surrounded by quartz (dark grey) and later sulphides (light grey shades). Pseudomorph filled

Plate IV



H. N. RHODEN: MINERALOGY OF THE SILVERMINES DISTRICT

Plate V



H. N. RHODEN: MINERALOGY OF THE SILVERMINES DISTRICT

Plate VI



H. N. RHODEN: MINERALOGY OF THE SILVERMINES DISTRICT

with marcasite, with other sulphides in shrinkage cracks parallel to basal parting of original pyrrhotine. Shallee mine, 320-ft. level, 9.6 W. stope; polished section, $\times 41$.

- F1G. 4. Shrinkage cracks within pyrrhotine pseudomorph. Triangular-ended tetrahedrite crystals, which grew out from the marcasite edges, were partly replaced by chalcopyrite, then covered by a thin layer of gudmundite; remaining space in the crack was filled by quartz. Gortnadyne mine, breccia; polished section, $\times 1230$.
- FIG. 5. Pseudomorphs of early carbonate (chalybite?) crystals, filled with ankerite (darker grey shades) and goethite (lighter grey). Galena (white) embayed and veined the pseudomorph, partly replacing both minerals. Shallee mine, 320-ft. level; polished section, $\times 148$.
- FIG. 6. Brecciated iron-rich blende (dark grey), partly replaced by pyrite (white); both minerals were subsequently replaced by galena (light grey, deep relief). Some of the pyrite was redeposited around the developing crystals of galena, or in long, thin, broken veinlets along the cubic cleavage. Gortnadyne mine, breccia; polished section, ×90.

PLATE VI.

- FIG. 7. Pseudomorph of euhedral blende crystal (outlined in white), marginally replaced by chlorite; unreplaced core of blende (black) contains exsolved chalcopyrite. The surrounding rock is a shaly, dolomitic sandstone. Shallee mine, 260-ft. level, borehole 39 at 45 ft.; thin section with X-nicols, ×47.
- FIG. 8. As for fig. 7. Unreplaced core of blende (white) surrounded by chlorite (medium grey). Irregular masses of light yellow blende (white) have replaced rock groundmass around pseudomorph. Copper-rich blende replaced by chlorite was redeposited nearby purged of inclusions. Shallee mine, 260-ft. level, borehole 39 at 45 ft.; polished section, ×47.
- FIG. 9. Feathery plates of sericite (black) surrounded by galena (white) in a narrow vein, between sandstone wall-rocks. Shallee mine, borehole 2E at 12.5 ft.; polished section, $\times 67$.
- FIG. 10. Relic texture in centre of pyritic spherulite indicating former presence of clump of marcasite crystals. Knockanroe mine, 'Sulphur Lode'; polished section, ×51.