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Ankerites and chalybites from the northern Pennine ore-field and the north-east coalfield.

(With Plate III.)

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INTRODUCTION

ANKERITES from the Northumberland coalfield were first described, with chemical analyses, some ten years ago (Hawkes and Smythe, 1935), when it was found that examples from veins exposed on the coast near Hartley and from sludge from nearby coal-washeries showed a remarkable uniformity in composition. The operations of the collieries concerned cover an area roughly 6×6 miles, and range from the Beaumont to the High Main Seam. In order to ascertain whether this uniformity is maintained over a wider area, five additional samples of ankerite from veinlets in rocks of Coal Measure age, and one from partings in the Lower Carboniferous Scremerston coal have been examined. The Coal Measure rocks cut by the veinlets have been proved to contain substantial proportions of ferriferous carbonates, while for comparative purposes two examples of rocks of similar age with carbonate but unaccompanied by ankerite veinlets have been investigated. The range in composition of ankerites in the coalfield has proved to be small.

Meanwhile, during the Geological Survey reinvestigation of the northern Pennine orefield, it was found that many of the carbonates formerly described as dolomite are really ankerite, large concentrations of which accompany the galena-blende-fluorite-baryte veins in the form of replacements of limestone or quartz-dolerite. The presence and

¹ By permission of the Director, Geological Survey and Museum.

general range in composition of the northern Pennine ankerites was established by means of refractive index determinations and confirmed by commercial analyses of iron ores of the 'coarse grey' type (Dunham, 1941), but the present account contains the results of the first analyses and determinations on purified material. Here the range in composition is much greater than in the coalfield and the ankerites are accompanied by chalybites of epigenetic origin, samples of which have also been examined.

In the present work, the purification of the samples, the chemical analyses, and the determinations of specific gravity were carried out by Smythe. Optical determinations, petrographical descriptions, and geological notes are by Dunham, except where the last-mentioned were supplied by the donors of specimens. Some of the specimens were of a high order of purity; they were powdered and washed, adventitious lighter matter being removed by floatation in heavy liquids. Others were extremely complex and only yielded pure minerals after laborious fractional separation. The purity of the powders was checked by rigorous optical examination of immersions before analysis.

All the samples contained a little chloride, estimated at 0.04 % in the Stonecroft example (no. 15, below). In most cases ample material was available, but there was insufficient of sample no. 4 for carbon dioxide determination, and the value given in table I is a calculated one. Samples nos. 15 and 17 (table IV) contain respectively 9.48 % and 2.15 % material insoluble in hydrochloric acid. Dr. F. A. Bannister and Dr. G. F. Claringbull have kindly examined the insoluble residues by means of X-rays, and they report as follows:

Residue from sample no. 15 consists mainly of white fragments (quartz and kaolinite) with a few fragments of a buff colour which contain quartz and in addition a phase not so far identified. Residue from sample no. 17 consists mainly of transparent fragments of quartz; goethite and polycrystalline quartz are the constituents of the brown, opaque fragments in the residue.

We are indebted to Mr. W. Anderson and Mr. A. Fowler of the Geological Survey, and to Professor S. E. Hollingworth, who kindly presented the specimens from Easington, Newbiggin, and Carrock Fell respectively. Mr. Anderson also collected the rock from Evenwood. To Mr. H. Berry, of the Fuel Research Coal Survey, Chester, we are grateful for the ankerite from Scremerston. To the Non-ferrous Minerals Development Control, and the Home Ore Department (Iron and Steel Control), of the Ministry of Supply, we tender our thanks for facilities in connexion with their Nenthead and Carricks operations respectively.

VEIN ANKERITES FROM THE COALFIELD.

The widespread occurrence of ankerite in partings and 'cleat' of coal seams has long been recognized, though the only investigation of the composition of the mineral in the Northumberland-Durham coalfield so far made has been that of the washery sludges mentioned above. The

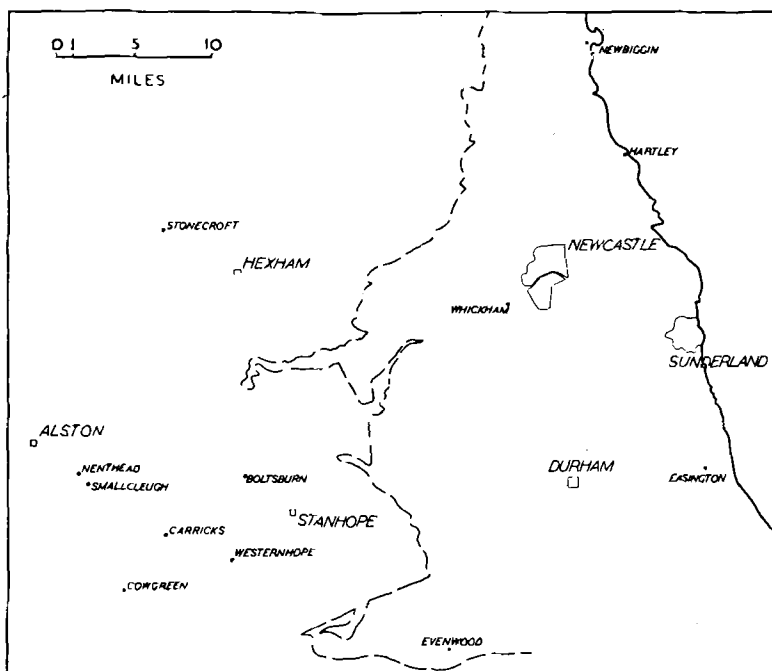


FIG. 1. Sketch-map of the north of England, to show ankerite and chalybite localities to which reference is made. The broken line represents the western margin of the Durham-Northumberland coalfield.

mineral was clearly introduced after the compaction and jointing of the coal, and if further confirmation is needed that the carbonate liquors were from an outside source, it is provided by the occurrence of veins of ankerite in other Coal Measure rocks. Six new examples of vein ankerites are described below.

Localities and geological relations (fig. 1).—1. Scremerston Colliery, near Tweedmouth, Northumberland (1-inch map, 1).¹ The sample consisted of plates of the 'white parting' from the cleat of the Scremerston Main coal, of Bernician (Lower Carboniferous) age, which averages 4 feet thick (Fowler 1926, p. 10).

¹ One-inch sheet numbers refer to the New Series maps of the Geological Survey.

2. Newbiggin-by-the-Sea, Northumberland (1-inch map, 10); veinlets from a sandstone of the Upper Group of the Coal Measures, exposed on the foreshore near the south end of the promenade, $\frac{3}{4}$ mile south-west of the church.

3. Easington Colliery, Co. Durham (1-inch map, 27); up-boring from the shaft; ankerite from vertical veinlet 0.5 mm. wide in 'micaceous sandstone', 10 feet above the Low Main seam (Middle Coal Measures).

4. Easington Colliery, Co. Durham; down-boring from the shaft; thin vein of ankerite in 'clay ironstone', 15 feet below the Hutton seam (Middle Coal Measures).

5. Easington Colliery; undersea boring, 3 miles east of the coast, 1500 feet below sea-level; thin ankerite vein in 'hard, grey, fine-grained sandstone', 127 feet above the Low Main seam.

6. Newbiggin Colliery, Northumberland (1-inch map, 10); ankerite filling a vertical joint found in a boring into 'tough, fine-grained sandstone' above the Brockwell seam (base of Middle Coal Measures).

Composition.—In table I the chemical composition, ordinary refractive index, and specific gravity of these ankerites are compared with those from the coast at Hartley, from Hartley Burn, and from Ashington, lettered C, A, and B respectively in Hawkes and Smythe (1935, p. 68). Although not showing the same uniformity as the earlier examples, the range in composition of the new ankerites from the Coal Measures is small, covering, for example, only 4% MgO. On the other hand, the Scremerston ankerite differs widely from those in the younger rocks.

TABLE I. Analyses of ankerites from the north-east coalfield.

	1.	2.	3.	4.	C.	B.	A.	5.	6.
Al ₂ O ₃	nil }		nil }	0.56	0.83	0.80	1.90 }		{ nil
Fe ₂ O ₃	nil }	1.20	nil }			0.08	0.61 }	1.16	{ nil
FeO	7.84	10.30	12.20	11.54	13.54	13.04	12.53	14.72	15.68
MnO	1.70	0.88	n.d.	0.50	0.75	0.75	0.61	n.d.	0.80
MgO	14.91	13.95	13.74	12.72	11.45	11.02	9.98	10.65	10.94
CaO	29.70	23.75	28.90	30.60	29.15	27.94	26.62	29.20	29.00
CO ₂	45.10	44.50	45.16	44.10	43.50	42.40	39.65	42.66	43.40
Insol.	0.60	0.07	nil	nil	0.80	3.75	7.80	1.80	nil
	99.85	99.65	100.00	100.02	100.02	99.78	99.70	100.19	99.82
t_D^{20}	2.93	2.95	n.d.	2.96	3.01	2.99	3.04	3.04	3.01
d_D^{20}	1.692-1.706	1.701-1.711	1.707-1.713	1.709-1.717	1.708-1.713	1.703-1.717	1.703-1.717	1.711-1.714	1.716-1.717
FeCO ₃	12.7	16.9	19.7	18.5	22.1	22.1	22.6	24.2	25.0
MnCO ₃	2.8	1.4	—	0.8	1.2	1.3	1.1	—	1.3
MgCO ₃	31.3	29.6	28.7	26.4	24.2	24.2	23.3	22.7	22.6
CaCO ₃	53.2	52.1	51.6	54.3	52.5	52.4	53.0	53.1	51.1
	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
FeCO ₃ .CaCO ₃ ...	23.6	31.4	36.7	34.4	41.1	41.2	42.0	45.1	46.6
MnCO ₃ .CaCO ₃ ...	5.2	2.7	—	1.5	2.3	2.4	2.1	—	2.3
MgCO ₃ .CaCO ₃ ...	68.6	64.7	62.7	58.6	52.8	52.9	50.9	49.6	49.4
CaCO ₃	2.6	1.2	0.6	5.5	3.8	3.5	5.0	5.3	1.7
	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

COAL MEASURE ROCKS CONTAINING FERRIFEROUS CARBONATE.

Localities and Geological Relations.—7. Easington Colliery; rock from the down-boring containing vein of ankerite (sample no. 4).

8. Newbiggin Colliery; rock containing vein of ankerite (sample no. 6).
 9. Boring 400 yards north-east of Evenwood Colliery, Co. Durham (1-inch map, 32); 'very hard sandstone', 60 feet below the Hutton seam.
 10. Easington Colliery; rock from the undersea boring containing vein of ankerite (sample no. 5).
 11. Easington Colliery; rock from the up-boring containing vein of ankerite (sample no. 3).
 W. Boring at Green's farm, $1\frac{1}{2}$ miles east-south-east of Whickham, Co. Durham (1-inch map, 20); 'limy band in sandstone', 7 inches thick, 11 feet 3 inches below the Hutton Seam. This rock contained the open joint with millerite crystals, described by Anderson and Smythe (1942).

TABLE II. Analyses of ferriferous rocks from the Coal Measures.

	7.	8.	9.	10.	11.	W.
Al ₂ O ₃	0.36	0.95	nil	2.28	3.10	1.97
Fe ₂ O ₃						
FeO	7.77	10.20	1.66	8.65	32.92	25.41
MnO	n.d.	n.d.	0.06	n.d.	n.d.	0.10
MgO	8.26	6.40	0.40	0.76	3.98	2.83
CaO	18.05	18.80	1.50	0.65	7.55	4.70
CO ₂	27.33	27.40	2.66	5.65	31.08	22.38
H ₂ O	0.35	0.05	trace	1.30	0.50	0.60
Insol.	37.45	35.60	93.65	80.42	21.15	42.10
	<u>99.57</u>	<u>99.40</u>	<u>99.93</u>	<u>99.71</u>	<u>100.28</u>	<u>100.18</u>
Total carbonate	60.4	61.8	6.27	14.1	76.6	55.4

Petrographic descriptions.—7. (E. 20742)¹ Although described as a fine-grained sandstone in the boring log, the proportion of carbonate in this rock is so high that it should be regarded as a gritty ankerite mudstone. It consists of angular quartz grains averaging 0.1 mm. set in a cryptocrystalline carbonate matrix which is not resolved even at the highest magnifications. The carbonate of the matrix appears to have replaced some of the quartz. Adjacent to the ankerite vein, which contains crystals up to 4 mm. diameter, incipient formation of rhombs in the matrix of the rock is displayed.

8. (E. 20739) The Newbiggin rock is closely similar to that described above. Sharp quartz grains averaging 0.1 mm. are set in a fine-grained carbonate matrix, the ordinary refractive index of the carbonate being approximately 1.715. A little clastic mica is present. There is again a vague suggestion of coarse rhombs in the matrix.

9. (E. 20847) In the Evenwood rock the proportion of carbonate is very much less than in the two preceding rocks. Subangular grains of quartz and shreds of white mica of average length 0.15 mm. make an

¹ Numbers in brackets refer to slides in the Slice Rock collection of the Geological Survey and Museum.

interlocked mosaic in which small patches of fine-grained carbonate occur. A little feldspar is present.

10. (E. 20741) Carbonate is a minor constituent in the rock from the undersea boring at Easington, which is a shaly sandstone consisting of rounded quartz grains and shreds of mica averaging 0.06 mm. in a dark streaky matrix. The presence of carbonate here, apart from a few 'pods' seen at high magnification, is not evident under the microscope. Possibly it forms part of the dark matrix.

11. (E. 20740) A normal chalybite mudstone, largely composed of carbonate with average grain-size near 0.005 mm. Opaque minerals including pyrite averaging 0.025 mm. are scattered through the rock and there are rare fragments of quartz and mica. A satisfactory determination of the refractive index of the carbonate has not been obtained, but ω seems to be near 1.820.

W. (E. 20738) From Whickham comes a gritty chalybite mudstone, with angular quartz grains averaging 0.2 mm. in a chalybite matrix varying from cryptocrystalline to 0.1 mm. in grain size. Shreds of clastic mica and a little oligoclase are present. A few rounded structures in the matrix may represent the remains of ooliths, but these are rare. The ordinary-ray refractive index of the carbonate is 1.830.

Composition of the rock carbonates.—In table III the compositions of the rock carbonates, recalculated to 100 %, are compared with those from the veins for which they are the hosts. In the rocks from the down-

TABLE III. Comparison of vein and rock carbonates.

	Easington shaft down-boring.		Newbiggin.		Evenwood (no vein).
	4. Vein.	7. Rock.	6. Vein.	8. Rock.	9. Rock.
FeCO ₃ ...	18.5	18.0	25.0	24.1	42.7
MnCO ₃ ...	0.8	—	1.3	—	1.4
MgCO ₃ ...	26.4	28.6	22.6	21.6	13.2
CaCO ₃ ...	54.3	53.4	51.1	54.3	42.7
Sp. gr. ...	2.96	2.84	3.01	2.86	n.d.
ω ...	1.709	n.d.	1.715	1.715	n.d.
	Easington undersea boring.		Easington shaft up-boring.		Whickham (millerite vein).
	5. Vein.	10. Rock.	3. Vein.	11. Rock.	W. Rock.
FeCO ₃ ...	24.2	80.5	19.7	71.5	74.2
MgCO ₃ ...	22.7	11.3	28.7	10.9	10.7
CaCO ₃ ...	53.1	8.2	51.6	17.6	15.1
Sp. gr. ...	3.04	n.d.	n.d.	n.d.	3.10
ω ...	1.712	—	1.710	1.820	1.830

boring at Easington shaft and from Newbiggin, the compositions of the rock and vein carbonates are so close as to justify the assertion that they are of common origin and it seems probable that the rocks have been permeated by carbonate liquors from an extraneous source. Whether a previously existing matrix has been replaced by ankerite, or whether this mineral has been emplaced by replacement of quartz grains is not certain, even though there is clear evidence of attack upon the quartz by the carbonate, for similar relations are normally found in unaltered sedimentary chalybite mudstones. On the other hand, the two other Easington rock carbonates contrast markedly with the vein-ankerites and there is no reason to consider that any genetic relation exists between them. The carbonates in the Easington up-boring and Wickham rocks can readily be matched among the normal chalybite mudstones of the Coal Measures.

ANKERITES FROM THE OREFIELD.

The northern part of the northern Pennine orefield, lying south-west of the Northumberland-Durham coalfield (fig. 1), between the main Tyne valley and the natural gap of Stainmore, is an area of Lower Carboniferous rocks in which thin limestones are interbedded with shales and sandstones. Intruded into the series is the Whin Sill, a system of sheets of quartz-dolerite at various stratigraphical levels. Ankerite is one, if not the most abundant, of the minerals accompanying the metalliferous veins of the field, from which galena, blende, fluorite, baryte, witherite, and iron ores are produced, particularly where they traverse limestone or dolerite. In limestone, ankerite is, in many cases a dominant constituent of the flat deposits, of metasomatic origin, usually extending far beyond the limits of the portions workable for lead, zinc, or iron ores. In the Whin Sill, it appears both in the veins and as a constituent of 'white whin', produced by metasomatism of the dolerite adjacent to them. Ankerite is the chief constituent of the poor-grade 'coarse grey' ore, with which there is reason to think the richer limonitic iron ores of Weardale were formerly diluted for technical purposes (Dunham, 1941). The quantities of ankerite are much greater in the orefield than in the coalfield, and, as a result of metalliferous mining operations, the occurrences are more readily accessible. Nine full analyses of ankerites from the orefield are given below; in addition, one sample has been examined from the Carrock Fell mine in the Lake District, where ankerite is associated with wolframite and scheelite.

Localities and Geological Relations.—12. Cowgreen barytes mine, 2½ miles west of Langdon Beck, Co. Durham (1-inch map, 25). Here baryte is being worked from

the Winterhush Vein between walls of Whin Sill and overlying Melmerby Scar Limestone; the orebody varies up to 16 feet wide in the limestone, and contains a mixture of baryte, sugary quartz, and ankerite, with a little galena and blende. In the Whin Sill ankerite is not an important constituent of the vein, but it is abundantly present in the altered dolerite. The sample was taken from the coarse tailings of the dressing plant.

13. Carricks ironstone mine, Wearhead, Co. Durham (1-inch map, 25). The deposits worked consist of replacement flats in the Great Limestone, related to weak NE.-SW. and E.-W. veins. Beneath the oxidation zone, the primary minerals are chalybite, ankerite, and quartz, with minor quantities of galena and fluorite. Within the oxidation zone, both ankerite and chalybite are replaced by limonite (this term being used here to mean hydrated ferric oxide, with subsidiary manganese oxides). The ankerite of sample no. 13 is particularly associated with a belt of NNW.-SSE. cross veins lying west of the main orebodies; it would normally be classified in the field as fine-grained, iron-stained dolomite. The typical ankerites associated with the orebodies are represented by samples nos. 19-21, below.

14. Carrock Fell wolfram mine, 7 miles NE. of Keswick, Cumberland (1-inch map, 23). Here a small complex of wolframite-quartz veins occurs, the wall-rocks being greisen and gabbro. Associated minerals include molybdenite, scheelite, arsenopyrite, pyrite, and chalcopyrite; carbonates, recorded as calcite and dolomite, occur according to Hitchen (1934, p. 191) as the latest phase of the mineralization, associated with sulphides. The ankerite sample examined was from Harding Vein, where a new level has recently (1942-3) been driven; it was separated from a specimen in which calcite (with ω 1.664) and quartz were present.

15. Stonecroft mine, 2 miles WNW. of Fourstones, near Hexham, Northumberland (1-inch map, 13). An abandoned lead mine, Stonecroft worked galena-baryte veins between Whin Sill walls. Pale brown coarse ankerite is abundantly present on the extensive dumps, forming according to an estimate based on the magnesia-content of representative samples taken during the Geological Survey investigation, at least 20 % of their total mineral contents. Other minerals present in the veins included quartz, blende, witherite, and pyrite; Sir Arthur Russell informs us that he has found harmotome here. The ankerite sample was collected from the coarse dump near Stonecroft winding shaft.

16. Smalleugh mine, half a mile south of Nenthead, Cumberland (1-inch map, 25). One of the most extensive metasomatic galena deposits of the northern Pennines, the Smalleugh flats are replacements of the lowest 10-15 feet of the Great Limestone in a horst bounded to the south-west and north-east by Smalleugh and Handsome Mea Great cross veins respectively. An excellent description of the deposit, with a block diagram, has been given by Wallace (1861, pp. 135-6, plate XIV), who has estimated that not less than five million cubic feet of limestone have undergone decomposition here. Although the workings have long been abandoned, it was found possible to examine parts of them in 1931. Ankerite forms coarse white streaks, roughly parallel to the bedding of the limestone; it is also abundantly present, with quartz, in the fine-grained hard matrix of the deposit. Other minerals in the flats included blende, pyrite, fluorite, chalybite, and calcite, in roughly that order of abundance. The sulphides were deposited either contemporaneously with, or later than, the ankerite. The sample examined was derived from one of the white streaks in the deposit. Recently the mine dumps have been treated by flotation for recovery of their zinc-content. Separations of ankerite were made from a representative sample of the feed to the mill;

it is estimated that ankerite forms approximately 35 % of the total mineral contents.

17. Westernhope 'New' mine, $2\frac{3}{4}$ miles SSE. of Westgate in Weardale, Co. Durham (1-inch map, 25). Metasomatized Great Limestone, related to the Ash Hill and Westernhope Great galena-fluorite veins is exposed in an open cut on the east bank of Ash Cleugh, the eastern headwater of Westernhope Burn. Like most of the occurrences in Weardale, the ankerite forms a medium-grained rock composed of interlocking idiomorphic rhombs, with individual crystals up to 2 mm. long. The sample was separated from this rock, which also contains some chalybite.

18. Carricks ironstone mine; underground boring to no. 4 Cross Vein, south of Dawson's Vein, no. 4, 9 feet. The sample consisted of white rhombs from a cavity in highly ankeritized limestone.

19. Boltsburn mine, Rookhope, Co. Durham (1-inch map, 25). The metasomatic flats in the Great Limestone associated with Boltsburn Vein, worked for galena from 1898 to 1931, contained fluorite, chalybite, ankerite, quartz, and calcite, with minor quantities of blende, pyrite, and chalcopyrite. Between the workable portions of the flats and the vein there was generally a belt of 'rider' (described by Carruthers as 'close-grained, ferruginous and dolomitic limestone', 1923, p. 11, footnote) varying from a few feet up to 20 feet wide. D. F. Hewett (1928, p. 836) has described the flat at Boltsburn as 'a tabular mass of dolomitized limestone breccia' and has stated that 'the dolomite is uniformly light gray in color and coarsely crystalline'. Microscopic investigation of specimens collected before the mine was flooded has now shown that the supposed dolomite is in fact ankerite with a high proportion of FeCO_3 . The sample represents the 'rider' between the vein and the workable flat after removal of chalybite, which was also present.

An estimate of the quantitative importance of ankerite at Boltsburn may be gained from the fact that nearly 40 % of the crude ore, of which a little over half a million tons were mined between 1901 and 1931, consisted of ankerite.¹ Great quantities of the mineral remain in the unworked portions of the deposit.

Carbonaceous material is present in the Boltsburn sample, presumably remaining from the limestone which has been replaced. Determinations on the mineral concentrate gave C 0.6 %; H 0.8 % and on the dried residue of material insoluble in hydrochloric acid, C 4.2 %, H 5.9 %. Some of the hydrogen may be from hydrous minerals.

20. Carricks ironstone mine; white rhombs from a cavity in the metasomatized Great Limestone associated with Lowe's Vein. The sample was pure and free from chalybite.

21. Carricks ironstone mine; Lowe's Vein West boring no. 1, 190 feet. The core specimen was chosen as representing the typical carbonate iron ore of this mine; it consisted of a mixture of coarse, pale grey, interlocking rhombs of ankerite, with yellow chalybite in streaks and irregular masses. The sample is a concentrate of ankerite separated from this mixture; a concentrate of chalybite was also prepared and analysed (no. 22, below). A small quantity of chalybite remains in the ankerite concentrate, but it is considered not to exceed 3 %.

Composition.—The analyses in table IV indicate a wide range in composition among the northern Pennine ankerites, with corresponding ranges in refractive index and specific gravity. The Cowgreen mineral,

¹ This estimate is based on recent samplings of the tailings heaps.

TABLE IV. Analyses of ankerites from the northern Pennine orefield and the Lake district.

	12.	13.	14.	15.	16.
Al ₂ O ₃	0.63	3.01	0.69	0.69	0.60
Fe ₂ O ₃	3.30	5.20	7.50	13.90	13.72
FeO	1.18	1.39	3.44	0.42	1.90
MnO	18.71	14.60	14.26	8.52	10.33
CaO	29.85	31.20	29.05	27.20	29.20
CO ₂	45.72	44.23	44.71	39.45	43.33
Insol.	0.25	0.30	trace	9.48	0.55
	99.64	99.93	99.65	99.66	99.63
$d_{4^{\circ}}^{t^{\circ}}$	2.86	2.91	2.96	2.95	3.00
ω	1.685-	1.690-	1.694-	1.709-	1.702-
	1.689	1.695	1.700	1.711	1.725
ϵ	1.507	n.d.	n.d.	1.523	n.d.
FeCO ₃	5.3	8.7	12.1	25.0	22.4
MnCO ₃	1.9	2.3	5.6	0.8	3.1
MgCO ₃	39.3	31.5	30.1	19.9	21.8
CaCO ₃	53.5	57.5	52.2	54.3	52.7
	100.0	100.0	100.0	100.0	100.0
FeCO ₃ .CaCO ₃ ...	9.9	16.2	22.8	46.6	41.2
MnCO ₃ .CaCO ₃ ...	3.6	4.3	10.5	1.4	5.8
MgCO ₃ .CaCO ₃ ...	85.9	68.9	65.8	43.6	47.1
CaCO ₃	0.6	10.6	0.9	8.4	5.9
	100.0	100.0	100.0	100.0	100.0
	17.	18.	19.	20.	21.
Al ₂ O ₃	0.73	0.66	nil	2.13	1.96
Fe ₂ O ₃	14.92	20.20	20.88	23.30	0.57
FeO	2.53	2.38	1.25	n.d.	22.61
MnO	8.01	6.24	5.34	4.88	2.24
MgO	29.25	27.80	27.85	27.62	3.01
CaO	42.22	42.07	40.82	41.00	28.30
CO ₂	2.15	0.35	3.40	0.67	40.91
Insol.	99.81	99.70	99.54	99.60	0.83
	99.81	99.70	99.54	99.60	100.43
$d_{4^{\circ}}^{t^{\circ}}$	3.03	3.08	3.07	3.05	3.11
ω	1.716-	1.720-	1.721-	1.722-	1.727-
	1.722	1.729	1.727	1.733	1.734
ϵ	1.529	n.d.	n.d.	n.d.	1.536
FeCO ₃	24.8	32.9	34.9	38.7	37.6
MnCO ₃	4.2	3.9	2.1	—	3.7
MgCO ₃	17.2	13.2	11.5	10.5	6.5
CaCO ₃	53.8	50.0	51.5	50.8	52.2
	100.0	100.0	100.0	100.0	100.0
FeCO ₃ .CaCO ₃ ...	46.2	61.3	65.0	72.2	70.1
MnCO ₃ .CaCO ₃ ...	7.8	7.3	3.9	—	6.9
MgCO ₃ .CaCO ₃ ...	37.6	28.8	25.2	23.0	14.2
CaCO ₃	8.4	2.6	5.9	4.8	8.8
	100.0	100.0	100.0	100.0	100.0

with only 5.3 % FeCO_3 , is not far removed from the dolomite end of the series, while the white crystals from Carricks mine, with 38.7 % FeCO_3 , contain 72.2 % ferrodolomite. Refractive index measurements showed some variation of ordinary-ray index within individual samples. In some instances this was as little as 0.002, and only one sample, that from Smalleleugh, showed a greater range than 0.011. In some instances this variation is due to zoning, such as the Carricks mine crystals (no. 20) display. At Cowgreen two distinct ankerites were found; that of the analysed sample (no. 12), with ω 1.685–1.689, and another with ω 1.712. The latter was carefully removed in the purification process, but it did not prove possible to obtain a sufficiently clean concentrate for analysis. Ankerites intermediate in composition between the two were not present. At Smalleleugh, on the other hand, evidence was obtained of a range with many intermediate stages present, from ω 1.682 up to ω 1.728. The analysed sample, separated in Sonstadt's solution (mercuric iodide in potassium iodide) was freed from the lighter dolomitic ankerite, and the full range represented by this sample is ω 1.702 to 1.725, with very rare grains reaching 1.728. The light crop from the separation (not analysed) contained ankerites ranging down to ω 1.682.

In an examination of typical mill-feed to the flotation plant at Nenthead, derived from the dumps at Smalleleugh and other adjacent mines, the same range in refractive indices was noted. In separations it was found that the heaviest fraction, sinking in Sonstadt's solution, contained ankerite with ω 1.710 to 1.723, together with blende, galena, pyrite, and fluorite. Middlings from this separation consisted of ankerite with ω 1.695 to 1.718, the bulk lying between 1.700 and 1.710. Floats carried ankerite ranging down to ω 1.682, with calcite, quartz, and micaceous minerals. Repeated separations failed to produce samples of ankerite sufficiently pure to warrant analysis.

CHALYBITES FROM THE OREFIELD.

Three samples of chalybite from Carricks mine have been isolated from spathic ores in which it is the most important constituent. Sample 22 is from Lowe's Vein West boring no. 1 at 190 feet from the same core-specimen as the ankerite concentrate, no. 21. Sample 23 comes from the same boring, at 242 feet; sample 24 from Lowe's Vein West boring no. 11 at 155 feet. In each case the chalybite is present in the metasomatized limestone in the form of irregular yellow streaks, associated with more or less ankerite. Zoning was not found when the powders were examined microscopically, but in the case of sample 22 a small quantity of ankerite,

present as minute inclusions of low refractive index, was noted. In samples 23 and 24, however, the purity of the powders is such as to preclude the possibility that appreciable amounts of free ankerite are present.

TABLE V. Analyses of chalybites from Carricks mine, Weardale, Co. Durham.

	22.	23.	24.
Al ₂ O ₃ ...	0.55	2.83	0.40
Fe ₂ O ₃ ...	0.94		
FeO ...	47.20	52.00	51.87
MnO ...	3.45	n.d.	3.17
MgO ...	2.86	2.78	2.50
CaO ...	4.80	3.62	3.60
CO ₂ ..	37.74	38.21	38.28
Insol. ...	2.65	trace	trace
	<u>100.19</u>	<u>99.44</u>	<u>99.82</u>
$d_{40}^{t_0}$...	3.58	3.73	3.77
ω ...	1.838	1.839	1.841
ϵ ...	n.d.	n.d.	1.611
FeCO ₃ ...	79.1	87.2	83.3
MnCO ₃ ...	5.8	—	5.1
MgCO ₃ ...	6.2	6.1	5.2
CaCO ₃ ...	8.9	6.7	6.4
	<u>100.0</u>	<u>100.0</u>	<u>100.0</u>

MINERALOGICAL DISCUSSION.

Chemical composition.—The whole suite of sixteen new and three earlier analyses (Hawkes and Smythe, 1935) of ankerite from north-eastern England provides a progressive series in which the contents of MgCO₃ range, in small steps, from 39.3 to 6.5 %, while those of FeCO₃ vary from 5.3 to 38.7 %. The analyses confirm the tendency, often noted, for the approximate constancy in content of CaCO₃ which led Doelter (1912, p. 371) to the conclusion that the mineral consists of isomorphous mixtures of dolomite and ferrodolomite. It is clear from older analyses, as well as from the new data here presented, that this is too simple a view of the series. The present analyses show contents of manganese carbonate varying between 0.8 and 5.6 %, while calcium carbonate is present in every sample in excess of that required to satisfy dolomite, ferrodolomite, and mangandolomite, varying in amount unsystematically up to 10.6 %. As shown by the data summarized by R. G. Wayland (1942, p. 616), the ionic radius of iron is nearer to that of manganese than are the radii of magnesium or calcium; it is thus most probable that the manganese is present as a substitution of iron, but

it may be noted that Wayland's work also suggests extensive miscibility between MnCO_3 and CaCO_3 . A convenient picture of the ankerite series may be constructed in terms of *ferromangandolomite* $(\text{Fe,Mn})\text{CO}_3 \cdot \text{CaCO}_3$, *dolomite*, and *calcite* (fig. 2).

No natural occurrence of pure ferrodolomite or magnesia-free ferromangandolomite has ever been recorded, and the samples from Carricks

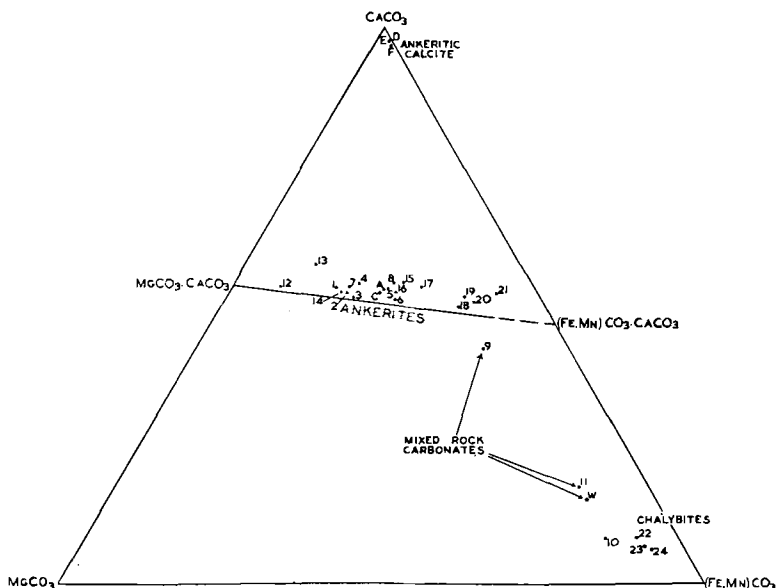


FIG. 2. Diagram to illustrate the compositions of the ankerites, chalybites, and ankeritic calcites in terms of CaCO_3 , MgCO_3 , $(\text{Fe,Mn})\text{CO}_3$.

and Boltsburn mines here described represent, with one exception,¹ the nearest approach to this end of the supposed series so far recorded. The ferromangandolomite-contents, disregarding sample 21, are 68.6 (18) and 68.9 % (19), while in sample 20, in which owing to paucity of material manganese was not determined, there was 72.2 % ferrodolomite. In view of the association with chalybite of contemporaneous or nearly contemporaneous origin in these occurrences it is possible that the absence of carbonates nearer in composition to ferrodolomite may have a real significance, and that the isomorphous series ends between 65 and

¹ For which E. S. Larsen and H. Berman (Bull. U.S. Geol. Surv., 1934, no. 848, p. 88) give the following data: CaCO_3 48.3, MgCO_3 11.3, FeCO_3 37.9, MnCO_3 2.5 %; ω 1.749, ϵ 1.547.

75 % (Fe,Mn)CO₃.CaCO₃. The Smalleleugh sample provides evidence of continuity from dolomite to this stage, and it is considered that the other samples, in which the range of composition is small, represent the results of local equilibria. The Evenwood rock carbonate (no. 9) perhaps supports the contention of limited miscibility. Its composition suggests a mixture of ankerite and chalybite, though unfortunately the material is so fine-grained that unequivocal microscopical confirmation of this has not been obtained. The occurrence without associated ankerite veins makes it unlikely that the carbonate cement has been subjected to later replacement by ankerite. The composition may be expressed as follows: FeCO₃ 13.0; FeCO₃.CaCO₃ 55.4; MnCO₃.CaCO₃ 2.7; MgCO₃.CaCO₃ 28.9 %. The ankerite portion, assuming that the chalybite is pure FeCO₃, would contain 66.7 % ferromangandolomite.

It is tentatively suggested that ankerite may be defined as an isomorphous series containing the carbonates of magnesium, calcium, iron, and manganese not containing more than approximately 70 % ferrodolomite, in which a limited proportion of manganese carbonate may be present,¹ and in which calcium carbonate in excess of that required to form dolomite, ferrodolomite, and mangandolomite may occur in amounts up to 20 %.² The series is not linked either with the ankeritic calcites or the chalybites through homogeneous intermediate compounds. The structure of the ankerites, the manner in which substitution takes place, and the way in which the 'excess' CaCO₃ is accommodated remain to be investigated by X-ray methods.

Of the three analyses of chalybites from Carricks mine, two contain MgCO₃ and CaCO₃ in the proportions required to form dolomite, with a slight excess of MgCO₃ in one case. A third analysis (no. 22) of the sample known to carry a little adventitious ankerite can be expressed as containing dolomite plus 3.9 % ankerite with 70 % ferrodolomite (table VI). It is remarkable, therefore, that these three analyses can be interpreted in terms of chalybite and non-ferriferous dolomite. The small quantities of manganese in these samples are assumed to occur as substitutions of iron in the chalybite.

The rock carbonates from the Coal Measures are not capable of explanation in terms of chalybite, rhodochrosite, and dolomite. One of them (no. 10) contains excess of MgCO₃, pointing to a member of the

¹ An ankerite described by Wayland (1942, p. 628) from Butte, Montana, contains 6.9 % MnCO₃.

² See H. W. Foote and W. M. Bradley, Amer. Journ. Sci., 1914, ser. 4, vol. 37, p. 343.

TABLE VI. Recalculated compositions of chalybites.

	22.	23.	24.	10.	11.	W.
(Fe,Mn)CO ₃ ...	83.3	87.2	88.4	80.5	71.9	74.1
{ FeCO ₃ .CaCO ₃ * ...	3.1	—	—	—	—	—
{ MgCO ₃ .CaCO ₃ * ...	0.8	—	—	—	—	—
MgCO ₃ .CaCO ₃ ...	12.8	12.4	11.6	15.1	23.1	23.4
MgCO ₃ ...	—	0.4	—	4.4	—	—
CaCO ₃ ...	—	—	—	—	5.0	2.5
	100.0	100.0	100.0	100.0	100.0	100.0

* In the proportions 3 dolomite : 7 ferrodolomite.

chalybite-magnesite series, stages in which have been named by T. Crook (1919, p. 75); there is, however, no evidence as to the stage which is represented. The other two (no. 11 and W) contain a sufficient excess of CaCO₃ and could be calculated as mixtures of ankerite and chalybite; but in the absence of satisfactory optical evidence as to the homogeneity of the carbonates it is not proposed to discuss this further.

Refractive index.—The determinations recorded in tables I, III, and IV were made by the immersion method, using yellow monochromatic illumination. Values for the extraordinary-ray index quoted were obtained on grains oriented on an improvised microscope goniometer. It was not considered justifiable to attempt determinations of ϵ except in the case of samples where the range of ω was known to be small.

The refractive index measurements indicate that none of the analysed ankerites is completely homogeneous in the sense that all the material present has constant indices; each sample represents a smaller or greater part of the isomorphous group. In fig. 3 the values for ω are plotted against dolomite-content. In many samples showing a range in refractive indices the bulk of the grains in the immersions examined were found to correspond to a much more restricted range; for example, in no. 2 ω ranges from 1.701 to 1.711, but the great majority of the grains showed ω 1.706. Where predominant values could reasonably be determined these are shown on the diagram. Comparing the ranges of ω determined for the present material with the line joining the accepted value for pure dolomite (1.680) and that deduced from the literature by Hawkes and Smythe (1935, p. 72) for pure ferrodolomite (1.765) all the values are low and would appear to be lower than substitution by manganese and the presence of 'excess' CaCO₃ can fully explain.

It will be evident that, from a practical point of view, determination of refractive index gives only a very approximate guide to composition in this complex system. W. E. Ford (1917) has shown that refractive indices, as well as specific gravities of the minerals of the calcite group

calculated from the percentages of the simple carbonates corresponds reasonably accurately with the determined values. Confirmation of this was obtained in the case of the ankerites and chalybites described here.

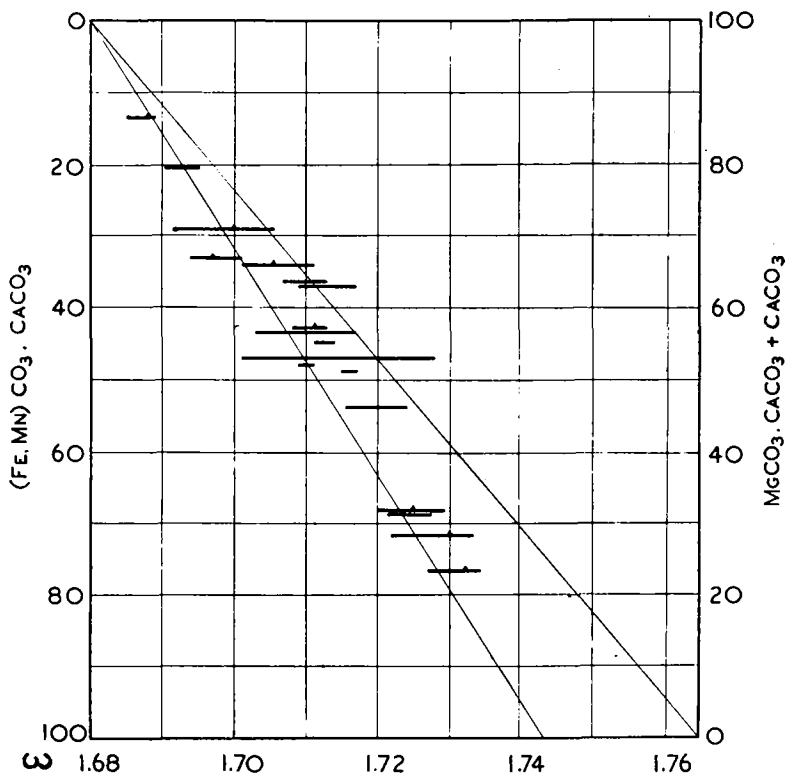


FIG. 3. Ordinary-ray refractive indices of ankerites, plotted against dolomite plus 'excess' calcite and ferromangandolomite. The cross-mark indicates the probable mean index where this can be determined.

Specific gravity.—This property gives a slightly more accurate guide to composition than refractive index, but even so it only serves to define certain limits as possibilities. In fig. 4 the relation between specific gravity and composition for the ankerites here described is shown; values to which small corrections for impurities (on the assumption that these had a specific gravity of 2.65) had been applied were used in the construction of this diagram. The substitution of manganese for iron has only a very small effect upon the specific gravity.

Textural relations.—In all parts of the orefield, ankerite appears to have been the earliest, or one of the earliest, products of the mineralization process. In the replacement deposits limestone was converted into a rock composed of interlocked sharp rhombs of ankerite before the deposition of sulphides. In examples like Boltsburn the selvedge of ankeritized limestone alongside the vein was unfavourable to further replacement, and galena-fluorite deposits were formed only where the solutions were able to break through this selvedge to reach fresh limestone beyond. Deposition of ankerite continued, however, and was partly

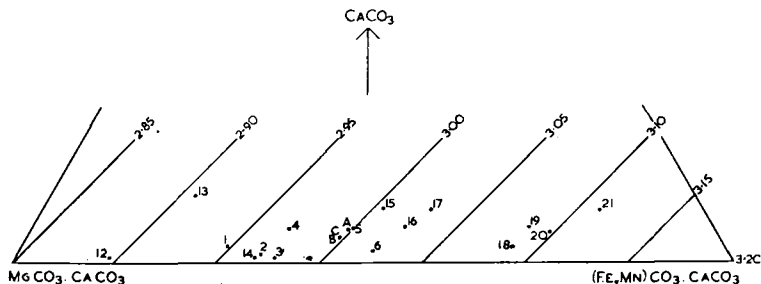


FIG. 4. Specific gravities of ankerites, after correction for impurities, in relation to composition.

contemporaneous with the sulphides. The Smalleugh material provides evidence that deposition of ankerite commenced at the dolomite end of the series, and became more ferriferous and less magnesian at later stages; the cloudy cores of the crystals here range between ω 1.682 and 1.700; the outer zones, clearer of inclusions, are also of higher refractive index. By the use of the staining method devised by Hallimond (1925, p. 39), in which a chip of rock is treated with hydrogen peroxide before sectioning, it was found that ankerite could be preferentially stained, the more ferriferous zones taking a darker stain. Applied to material from Middlehope mine, Weardale, this showed dolomitic inner zones and ferriferous outer zones in ankerite from replacements of the Great Limestone (pl. III, fig. 1). On the other hand, sections of the core from Carricks mine, from which samples 21 and 22 were extracted, failed to show zoning in the ankerite, confirming the small range of composition indicated by refractive index measurements. Of the two ankerites at Cowgreen mine that with the lower refractive index (no. 12) crystallized first.

There is equally definite evidence that in the metasomatized limestone chalybite crystallized later than ankerite, wrapping itself about

the ankerite rhombs and in places cutting through them in tiny veinlets. The process of formation of the carbonates may thus be envisaged as starting with dolomite and proceeding towards the ankerite with 65–75 % ferrodolomite. This was reached if adequate supplies of iron were available, but after this more ferriferous ankerite was not formed; instead, chalybite with dolomite in solid solution was deposited.

It is interesting to note that whereas in the case of the low-temperature Pennine mineralization ankerite is an early mineral, in the Carrock Fell paragenesis, where minerals considered to be of high-temperature origin are present, the ankerite is a late-stage product.

Oxidation of ankerite and chalybite.—Within the ‘oxidation zone’, above the groundwater table, waters of surface origin carrying dissolved oxygen and carbon dioxide act upon the carbonates. In the orefield the depth of this zone varies from zero adjacent to the valley-bottoms to a maximum of about 300 feet beneath the hills. The product of oxidation of both ankerite and chalybite is a mixture of hydrous ferric and manganese oxides; crystalline goethite is rare. Chalybite is more responsive to oxidation than ankerite, as might be expected, and at most of the Weardale ironstone mines specimens of dense limonite (after chalybite) containing sharp rhombs of little-altered ankerite can be picked up. At a more advanced stage, however, ankerite is itself attacked, and preferential oxidation shows up the ferriferous zones in much the same way as the staining process mentioned above. The alteration of ankerite to limonite involves the release of calcium and magnesium bicarbonates; the substantially lower magnesia- and lime-contents of the oxidized ‘brown haematite’ ores as compared with the primary spathic ores (Dunham, 1941, pp. 7, 8) strongly suggest that these constituents are carried away. No doubt they are, however, redeposited elsewhere;

TABLE VII. *Comparison of primary and oxidized iron ores, Carricks mine.*¹

		‘Fine steel grey’ (Chalybite).	‘Coarse grey’ (Ankerite).	‘Brown haematite’ (Limonite).
FeCO ₃	...	73.4	31.2	0.0
MnCO ₃	...	7.0	4.6	0.0
MgCO ₃	...	3.8	10.0	0.3
CaCO ₃	...	8.5	28.0	1.3
(Fe, Mn) ₂ O ₃ .xH ₂ O	...	0.0	0.0	81.4
FeS ₂	...	1.3	0.3	0.0
SiO ₂	...	5.6	22.1	16.1
		<u>99.6</u>	<u>96.2</u>	<u>99.1</u>

¹ Recalculated from analyses by L. C. W. Birkett, 1940, quoted in Dunham, 1941, table III, nos. 2–4.

possibly the dolomitic ankerite found as an iron-stained, fine-grained 'dolomitization' zone along the cross veins at Carricks mine (sample 13) is the product of such a process.

Origin of the carbonates.—Ankerite is widespread throughout the northern part of the northern Pennine orefield, north of Stainmore.¹ Chalybite, in quantity, is of more restricted range, the principal deposits being in Upper Weardale. It is a striking fact that the most ferrous ankerite is always associated with chalybite, and that its refractive index ω does not exceed 1.734. In relation to the zonal arrangement of minerals in the orefield (Dunham, 1934) the iron-rich areas are found in the inner, fluorite-galena zone east of the Burtreford disturbance. In the outer zones more magnesian ankerites occur.

In the southern part of the northern Pennines, south of Stainmore, covering the west Yorkshire dales, on the other hand, ankerite is very rare, though otherwise similar suites of metalliferous minerals occur. The most important difference between the northern and southern parts of the orefield is the presence in one and the absence in the other of the Whin Sill. L. R. Wager (1929, p. 105) has made the interesting suggestion that the lead vein solutions in the northern area derived their iron through the metasomatism of the Whin Sill. The alteration process adjacent to the veins consisted of the conversion of ferromagnesian minerals into carbonates and feldspars into micaceous minerals. The chemical data already available (Finlayson, 1910; Wager, 1929; Smythe, 1930) show that iron is abstracted in the process, together with, in certain cases but not in others, magnesia. Possibly, therefore, the metasomatism of the Whin Sill is sufficient to explain the abundant ankerite and chalybite of the northern area. It should be made clear, however, that the hydrothermal solutions responsible for the Pennine mineralization were not themselves the product of the Whin Sill; this dolerite has its own suite of hydrothermal products, and ankerite is not one of them.

It is tempting to regard the ankerites of the coalfield as products of the same widespread mineralization as those of the orefield. Baryte and witherite deposits are, indeed, worked in the Coal Measures, and there can be little doubt that these represent the outer zone of the Pennine mineralization; but they are not accompanied by ankerite. A small galena-calcite vein, described by H. Louis (1903, p. 128) from the shale and sandstone roof of the Maudlin Seam at Wearmouth Colliery,

¹ Refractive index measurements on crystals from numerous localities are given in the forthcoming Geological Survey Memoir on the orefield.

Sunderland, had carbonate mineralization associated with it in the coal itself; the composition of the carbonate, in spite of the author's unsupported conclusion to the contrary, strongly suggests ankerite. Recalculated from the analysis quoted, it contains ferrodolomite 50, dolomite 38, calcite 12%; or if the zinc present is assumed to be in the sulphide state, then the ferrodolomite figure is slightly increased in proportion. Possibly some of the ankerite in the Coal Measures came from the same hydrothermal source as that in the orefield; but as yet there is no evidence as to the origin of the widespread partings of this mineral in the coals.

SUMMARY OF CONCLUSIONS.

1. Sixteen new analyses of ankerite from the Northumberland-Durham coalfield and the northern part of the northern Pennine orefield, taken in conjunction with three earlier analyses (Hawkes and Smythe, 1935), provide evidence of a range of solid solutions from dolomite up to 65-75% ferromangandolomite; calcium carbonate in excess of that required for dolomite and ferromangandolomite is always present. Ordinary ray refractive indices ω range from 1.682 to 1.734.

2. Since the most ferrikerous ankerite of the series occurs in intimate association with chalybite it is suggested that the ankerite series ends at 65-75% ferrodolomite.

3. Chalybites from the orefield contain dolomite, but not ferrodolomite, in solid solution.

4. Crystallization of ankerite from hydrothermal solutions proceeded from dolomite towards ferrodolomite; after the limit of solid solution had been reached, chalybite with dolomite in solid solution appeared instead of ankerite.

5. The distribution of ankerite in the northern Pennines is consistent with Wager's view (1929) that the iron was derived through the metasomatism by lead vein solutions of the Whin Sill; it is suggested that magnesia was supplied to the solutions from the same source.

6. In two rocks from the Coal Measures contiguous with ankerite veinlets ankeritization is demonstrated; in three other Coal Measure rocks carbonate cements, presumably of sedimentary origin, consist of chalybite possibly mixed with ankerite.

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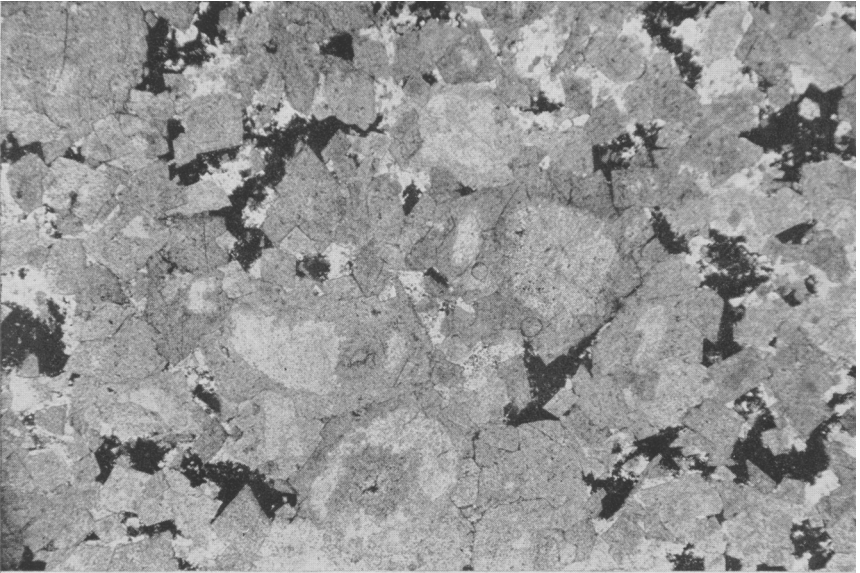
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EXPLANATION OF PLATE III.

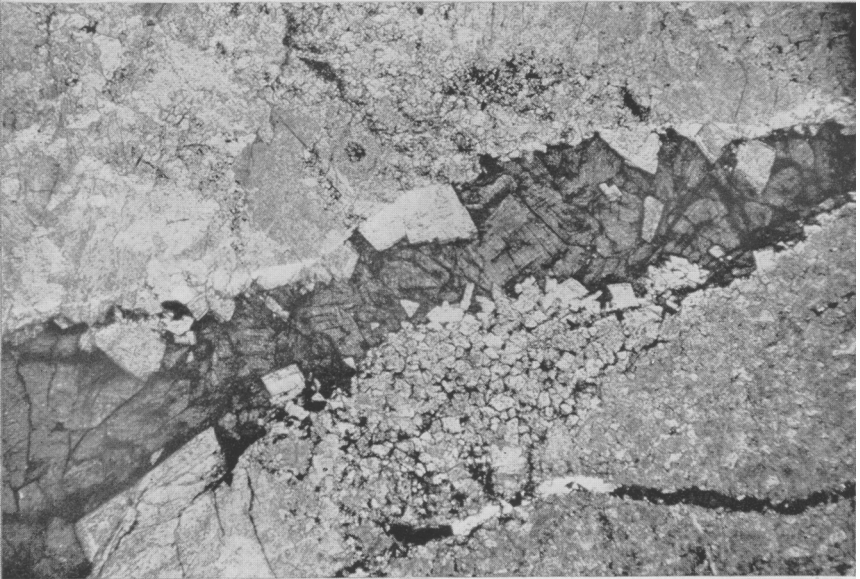
Photomicrographs of thin sections of Great Limestone after staining with KOH and H_2O_2 . Black and dark grey areas are chalybite. The tint of the ankerite varies according to its iron-content.

FIG. 1. Middlehope mine, $1\frac{1}{4}$ miles north of Westgate in Weardale, Co. Durham. (Section E. 18903A; Geological Survey Photograph M. 2295.) To show dolomitic cores in ferriferous ankerite. In some instances the cores preserve the form of fossil fragments. Late chalybite is present. Ordinary light, $\times 15$.

FIG. 2. Carricks ironstone mine, no. 2 borehole, Lowe's Vein West, at 66 feet, Weardale, Co. Durham. (Section E. 19999A, Geological Survey Photograph M. 2296.) To show a gash lined with zoned ankerite rhombs and filled with chalybite. Ordinary light, $\times 15$.



1



2

J. A. SMYTHE AND K. C. DUNHAM: ANKERITE AND CHALYBITE.