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The paragenesis of cookeite and hydromuscovite associated with gold at Ogofau, Carmarthenshire.

(With Plate XIX.)

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THIS paper describes occurrences of cookeite (not previously recorded as a British species) and hydromuscovite associated with auriferous pyrite and arsenopyrite, blende, galena, carbonates, and free gold at Ogofau, on the left bank of the river Cothi, half a mile ESE. of Pumpsaint, Carmarthenshire.

The hillside around Ogofau is scarred with opencast workings and trenches reputed to be an 'ancient gold mine' dating back to Roman times. Though the mine is described as 'disused' on the Ordnance Survey map (six-inches/mile, Sheet XVII, NW.), these surface workings have been exploited for free gold intermittently within the last fifty years, and recent development work (by the Roman Deep Holdings, Ltd.) from a shaft sunk in 1910 is approaching the producing stage.

The old workings are scattered over a belt which coincides with a mineralized thrust-zone transgressing the boundary between Valentian (Lower Birkhill) beds and the underlying Ordovician (Bala) beds exposed by a minor up-fold on the west flank of the main Towy anticline. In both these horizons the normal facies comprises cleaved blue-black shales and mudstones with occasional thin bands of grit.

The surface workings expose ochreous shales impregnated with pyrite and arsenopyrite, and banded with narrow gossanous zones, quartz veins, and numerous veinlets of white hydromica in intimate association with quartz, auriferous sulphides, and occasionally free gold. The last is frequently obtainable by panning crushed samples taken from quartzose shatter-belts in the gossanous ground. The opencast work appears to have prospected numerous hydrothermal 'blow-zones' and minor arteries of mineralization structurally related to ore-bodies disclosed by recent underground development work.

These ore-bodies are mainly sheet- and saddle-like masses of pyritic quartz intercalated along bedding planes in small drag-folds on the flank of the anticlinal. Typically, an ore-body is traceable from a feather edge sheet low down on the flanks of a minor up-fold into a substantial quartz-body attaining its maximum thickness towards the crest of the fold, where it is usually involved in a steeply inclined shear-zone.

Shales marginal to these ore-bodies are usually impregnated with sulphides over varying thicknesses above and below the contact, and particular bands in the shale are selectively metasomatized to a compact stony aggregate of auriferous sulphides and quartz. Bands of similar 'stone pyrite' occur also in shale-ground remote from any known, or suspected, body of pyritic quartz, and such shales commonly contain conspicuous dice-like crystals of arsenopyrite and pyrite.

Hydromuscovite is habitually associated with sulphides in both shale-ground and ore-bodies. It is specially abundant in veinlets which also carry coarsely crystalline auriferous sulphides, quartz, and usually a little free gold. These micaccous veinlets are of such frequent occurrence in the lower levels of the mine that water drained from a working face is turbid with finely divided white mica.

Cookeite, on the other hand, is more restricted in its distribution. It has not yet been observed in the surface workings and is very rare in the upper levels of the mine. It was first noticed on the third (420-foot) level as a mica-like deposit in small cavities and along shear- and fracture-planes traversing ore-body and shale-ground alike. It was next observed as the material grouting localized masses of shale-breccia. Unlike the hydromuscovite, the cookeite is rarely in intimate admixture with sulphides: its deposition occurred after the emplacement of the sulphide-quartz-hydromica ore-bodies.

The following table gives an approximate assessment of lodecontent based on a considerable tonnage of high-grade pyritic ore taken from a stope on the third (420-foot) level:

| Quartz | ••• | | | | 53.0 % |
|------------|-------|------|-----------|-----|-------------|
| Hydromuse | ovite | | | | 6.0 |
| Cookeite | | | | | 0.1 |
| Carbonates | | | ••• • | ••• | 0.2 |
| Graphite | | | | | trace |
| Blende and | galen | B. | | | $2 \cdot 0$ |
| Auriferous | | 38.7 | | | |
| | | | | | 100.0 |
| | | | | | |

Elements supplementary to those accounted for by the lodeminerals listed above have been determined spectroscopically by Mr. V. L. Aspland, A.R.S.M., whose findings are summarized below:

> Traces ... B,Sn,Sb,Bi. Minute traces ... Ba,Cd,In,Ga,Ge,Cr,V,Ni,Co.

Quartz is mainly the 'milky' variety, and shows marked triboluminescence. It is characterized by abundant minute cavities containing fluid, with mobile bubbles. Water-clear crystals are confined to vughs.

Hydromuscovite is mainly a lustrous silver-white to cream-coloured variety, grading into a pale, prase-green, wax-like, massive variety veneering planes of compression and shear. Both varieties are very soft, easily deformed, and talc-like to the touch. An aggregate of finely divided mica takes on the consistency of clay when wetted.

Cookeite occurs mainly as a patchy film or incrustation on other lode-minerals. Its rarity in the higher levels of the mine, and the frequency of its occurrence in drill-cores drawn from below the present bottom (420-foot) level, suggest that it is both more widespread and more abundant at depths below this level.

Carbonate is mainly calcite, with ferruginous dolomite as an occasional associate. Chalybite and ankerite are known to occur in the matrix of the mineralized mica-rich shales.

Graphite is probably derived from cleaved black-shales, some of which are known to contain finely divided graphite.

Blende and galena together are subordinate to the other sulphides; but they are the main constituents of a vertical cross-course traceable from surface to depth.

Gold is intimately associated with the pyrite and arsenopyrite. Concentrates of these two sulphides yield gold in quantities varying from pennyweights to 3 ounces or more per ton. The Au: Ag ratio in run-of-mine ore is approximately 1:8. Coarse and fine granular gold is occasionally found in lode-quartz. Gold residual after sulphides is represented by granular, arborescent, and 'wire' gold occurring in small ferruginous pockets in lode-quartz and elsewhere.

COOKEITE.

The mineral ultimately identified (by F. A. B.) as cookeite was first observed by the senior author¹ as an incrustation on lodeminerals in an ore-body on the third (420-foot) level of the mine. Preliminary X-ray powder photographs on carefully selected material were taken by Dr. G. Nagelschmidt, who obtained a pattern closely resembling those of dickite and clinochlore. The mineral analysed was fractionated from material grouting disturbed ground on the third level. The raw sample was a creamy-white, compact but friable mass, with a silky sheen on hackly fracture surfaces and a mica-like lustre when dispersed in water. The analysis no. 1, table I, was done on material carefully freed from quartz, and a portion of the sample analysed was subjected to optical and further X-ray investigations. The mineral consists of pale-cream, transparent plates of irregular outline not exceeding 1 mm. across, and rarely more than 1/10 mm, in thickness. A few plates are semicircular in outline and are probably detached from hemispherical aggregates. The specific gravity determined in mixtures of bromoform and benzene is 2.69 + 0.01. Most of the plates give a sharp biaxial positive interference-figure (optic axial plane parallel to the axis [100]). 2E 80°; and the refractive indices determined by the Becke method are a 1.575, β 1.580, γ 1.595 for sodium-light. Both the optical properties and chemical analysis suggest the identity of the mineral with cookeite from Maine, U.S.A.

Cookeite, discovered in 1862 or 1863 on specimens of lepidolite from Hebron, Maine, by J. P. Cooke, was described as a white or

¹ Several specimens, including a portion of the material used for chemical analysis, have been presented to the Trustees of the British Museum.

yellowish-green, pearly, micaceous mineral resembling nacrite which, when heated before the blowpipe, extoliated like vermiculite, at the same time colouring the flame a vivid lithium red. G J. Brush collected sufficient of this mineral for chemical analysis (no. 2, table I),

| | | 1. | 1 <i>a</i> . | 2. | 3. | 4. | 5. | 6. |
|--------------------------------|---------|-------|--------------|---------------|--------------------|-------------------------|--------|-----------|
| SiO, | | 32.46 | 34.68 | 35.20 | 34-00 | 34.81 | 32.00 | 34.52 |
| TiO ₂ | | 0.03 | 0.03 | | | | | |
| Al ₂ O ₃ | | 45.08 | 48.17 | 44 ·91 | 45.06 | 45.90 | 45.87 | 48.82 |
| Fe ₂ O ₃ | • • • • | 0.27 | 0.29 | | 0.45 | 0.72 | _ | · <u></u> |
| FeO | | 0.99 | 0.90 | | | | — | |
| MnO | | — | — | | | $\mathbf{trac}\epsilon$ | — | |
| MgO | | 0.35 | | | — | — | 0.78 | _ |
| CaO | | 2.73 | | | 0.04 | trace | 1.63 | · |
| Na ₂ O | | 0.04 | 0·04 | | 0·19 (| 0.59 | § 0·65 | |
| K ₂ Ō | | | | 2.57 | 0.14 \$ | 0-02 | 10.06 | _ |
| Li ₂ O | | 2.29 | 2.45 | 2.82 | 4.02 | 3.59 | 2.10 | 2.86 |
| $H_2O +$ | | 12.58 | 13.44 | 13-41) | 14.08 | 14.97 | 17.90 | § 13·80 |
| $H_2O -$ | | 0.35 | | 0.38 \ | 14.90 | 14.01 | 17.29 | ! _ |
| CO ₂ | | 2.74 | | | _ | | | _ |
| F | | | | 0.34 | 0.46 | 0.13 | 0.02 | — |
| | | 99.91 | 100.00 | 99.63 | 99.32 | 100.54 | 100.40 | 100.00 |
| Less $\mathbf{O} = \mathbf{F}$ | | | — | 0.14 | 0.19 | 0.05 | 0.01 | — |
| | | | | 99-49 | $\overline{99.13}$ | 100.49 | 100.39 | |
| Sp. gr. | | 2-69 | | 2.70 | 2.675 | 2.67 | | |

| TABLE I. Unemical analyses of cookene | TABLE | Ĭ. | Chemical | analyses | of | cookeite |
|---------------------------------------|-------|----|----------|----------|----|----------|
|---------------------------------------|-------|----|----------|----------|----|----------|

1. Roman Deep mine, Ogofau, Pumpsaint, Carmarthenshire. Minute, palecream curved plates from cavities in quartz. Refractive indices a 1.575, β 1.580, γ 1.595 (sodium-light), positive, 2E 80°. Analyst, Miss Hilda Bennett, M.A., B.Sc

1*a*. Analysis no. 1 recalculated to 100.00 after subtracting lime, magnesia, 0.15% ferrous oxide, and carbon dioxide, which are present as dolomite.

2. Hebron and Paris, Maine. Extremely minute scales and hemispherical aggregates associated with lepidolite and rubellite. G. J. Brush, Amer. Journ. Sci., 1866, ser. 2, vol. 41, p. 246. Brush recorded SiO₂ 34.93 and SiF₄ 0.47%; the latter is equivalent to SiO₂ 0.27 and F 0.34%.

3. Paris, Maine. The material chosen for analysis was associated only with quartz and tourmaline, thus avoiding contamination with lepidolite. S. L. Penfield, Amer. Journ. Sci., 1893, ser. 3, vol. 45, p. 393.

4. Buckfield, Maine. Pale-yellow, pink, and cream cleavages. Refractive indices a 1.576, β 1.579, γ 1.597, positive, 2V maximum 80°. Analyst, Miss Helen E. Vassar in K. K. Landes, Amer. Min., 1925, vol. 10, p. 390. [Min. Abstr., 3-203.]

5. Wait-a-bit Creek, Columbia river, British Columbia. Thin, foliated, greyish to silvery-white and pale apple-green layers in sericite-schist, and in the small cavities of quartz traversing the schist. Analyst, R. A. A. Johnston in G. C. Hoffmann, Ann. Rep. Geol. Surv. Canada, 1895, vol. 6, p. 22.

6. Theoretical figures corresponding to LiAl₄Si₃AlO₁₀(OH)₈.

and concluded it to be a new species intermediate in composition between euphyllite and margarite. He named the new mineral cookeite in 1866 after its discoverer. Further specimens collected by Brush from Paris and Hebron nearly thirty years later yielded abundant material for a re-examination undertaken by S. L. Penfield (anal. 3), who also noted its hexagonal habit and occurrence in hemispherical or globular aggregates up to 3 mm. in diameter. Penfield concluded that cookeite is closely related to the chlorites and micas. An analysis (no. 4) of cookeite from Buckfield, Maine, was carried out by Miss Helen E. Vassar, and a more detailed optical study was made by K. K. Landes upon the same material and upon cookeite from Greenwood, Maine. Cookeite from Columbia river has also been analysed (no. 5); and the mineral is reported from the pegmatites of Haddam Neck, Connecticut,¹ and of the Karibib and Omaruru districts in South-West Africa.²

The identity of cookeite from Ogofau was confirmed by singlecrystal X-ray photographs, typical material from Hebron, Paris, and Buckfield in Maine, being chosen from the British Museum collection for comparison. Rotation photographs of cookeite from Ogofau and Maine are identical, whereas clinochlore, muscovite, dickite, nacrite, and lepidolite give patterns that are readily distinguished (pl. xrx, figs. 1-9).

Laue photographs of cookeite from Ogofau are of two types: (1) a sharp spot and streak pattern revealing hexagonal or pseudohexagonal symmetry (fig. 1); (2) superposed hexagonal spot patterns spread over an angular range of about 10° (fig. 2). Neither type of photograph is characteristic of a single crystal, and since the plates are curved it is impossible to be certain whether the centre of the hexagonal pattern is displaced from the normal to the cleavage face. That is, if the mineral is truly monoclinic, it is impossible to measure the value of β . A rotation photograph of a curved plate yielding a Laue pattern of type 1 shows streaks corresponding to diffractions from (001) and spots from (hk0) (fig. 4), revealing that each curved plate is made up of a large number of small flakes with their *a*-axes in parallel alinement, but with their *c*-axes inclined to each other at small angles. A Laue photograph of type 1, of a flake aggregate

¹ H. L. Bowman, Min. Mag., 1902, vol. 13, p. 118.

² T. W. Gevers and H. F. Frommurze, Trans. Geol. Soc. South Africa, 1930, vol. 32 (for 1929), p. 139. [M.A. **4**-411.] W. P. de Kock, Trans. Geol. Soc. South Africa, 1933, vol. 35 (for 1932), p. 97. [M.A. **5**-421.]

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(a-axes parallel) consists of 'white' radiation streaks and a superposed monochromatic spot pattern (tungsten L radiation) due to flakes near the centre of the aggregate. Hence each curved plate behaves like a section of a spherulite, and shows a characteristic brush in parallel polarized light.¹ The Laue photograph of type 2 corresponds to a flake aggregate which has been deformed. The individual flakes no longer lie on a curved surface with their a-axes parallel, but are now spread fanwise; and the optic axial angle is lowered by amounts dependent on the angular spread resulting from the deformation. The measurement of the optic axial angle of platy aggregates is, of course, far more sensitive to lack of alinement parallel to the a-axis than to a small tilt of the individual flakes to the c-axis. Curved flake aggregates, with their a-axes strictly parallel, are characteristic not only of cookeite but also of chlorites, vermiculites, pyrophyllite, talc, and nacrite. Hendricks has also recorded the same type of flake aggregate for kaolinite. He found 'that the angle of deviation of individual fragments from the cleavage plane is approximately 12° 15°', and notes that in this particular type of crystalline aggregate 'it is interesting that the displacement of the individual fragments was unaccompanied by rotation about the normal to the cleavage surface '.2 On the other hand, dickite, muscovite, and probably all unaltered micas yield true single-crystal X-ray photographs.

One of the plates yielding a good Laue pattern of type 1 was selected for rotation photographs and oriented to give the *a* and *b* spacings respectively. The *c* spacing was deduced from the basal cleavage 'streaks' on the equators of photographs about the *a* and *b* axes. The dimensions obtained are $a 5 \cdot 13$, $b 8 \cdot 93$, $c 28 \cdot 30$ Å. If the mineral is truly monoclinic with $\beta \neq 90^{\circ}$, then the value $28 \cdot 30$ Å. corresponds to the spacing $c \sin \beta$ of the (001) planes and not to the true unit-cell dimension *c*. We may, however, calculate the unit-cell volume $abc \sin \beta$, and, knowing also the specific gravity and percentage of oxygen, proceed in the usual way to calculate the number of oxygen atoms in the unit cell. This number is 72.37; assuming that the unit cell contains 72 oxygen atoms, the specific gravity is calculated to be 2.676, in close agreement with the observed value 2.69. All the available analyses of cookeite have been recalculated

 $^{^{1}}$ No flakes, however, were found with a uniaxial centre surrounded by a biaxial rim, similar to those recorded by Penfield and Landes from Maine.

² S. B. Hendricks, Zeits. Krist., 1929, vol. 71, p. 269. [M.A. 4-160.]

on the basis of 56 oxygen atoms per unit cell, excluding the water content, to give the atomic contents (table 11). There are two quantities which remain appreciably constant, the number of silicon atoms, approximately 12, and the total number of metal atoms including silicon, approximately 36 per unit cell. The lithium content varies, however, from 3.08 to 5.80, and it must be remembered

| TABLE II. | Analys | es of d | cookeite r | ecalculate | d to give | the numb | er of atoms |
|----------------------------|----------|---------|------------|------------|---------------|----------|-------------|
| | | | per | unit cell. | | | |
| | | | 1.* | 2. | 3. | 4. | 5. |
| Si | | | 12.16 | 13.16 | $12 \cdot 20$ | 12.24 | 11.72 |
| Al | • • • | | 19-90 | 17.78 | 19.04 | 19.04 | 19.80 |
| Fe''' | | | 0.08 | | 0.08 | 0.20 | _ |
| $Fe^{\prime\prime}$ | | | 0.26 | _ | _ | _ | |
| Мg | | | | | _ | _ | 0.44 |
| Ca | | | | | <u>-</u> | | 0.64 |
| Na | • • • | | 0.03 | | 0.12 | 0.28 | 0.48 |
| ĸ | | | | 1.13 | 0.08 | _ | 0.04 |
| Li | | | 3.45 | 4.24 | 5.80 | 5.08 | 3.08 |
| F | | | _ | 0.40 | 0.52 | | _ |
| H_2O | | | 15.71 | 16-61 | 17.88 | 17.44 | 21-12 |
| $\Sigma \overline{S}i, Al$ | ,Li, &c. | | 35.88 | 36.31 | 37.32 | 36.84 | 36.20 |

* The chemical analysis of cookeite from Ogofau has been recalculated to a basis of 72(O,OH) atoms per unit cell. Owing to the possible inaccuracy of the water determinations, nos. 2 to 5 have been calculated on the basis of 56 oxygen atoms per unit cell, neglecting the water content.

that the older methods of analysis were liable to give too high values for lithia determinations. Thus we find that Miss Bennett's analysis of cookeite from Ogofau contains a lower lithia figure than Brush's or Penfield's, and also vields a figure nearer to 36 for the total Si + metal atoms per unit cell. Assuming that the crystal-structure of cookeite, like all members of the mica, chlorite, and clay families, is based upon a hexagonal layer of composition (Si,Al)₄O₁₀, the unit-cell contents of cookeite may be written from table II, 4(Li,Al), Si,AlO10(OH), or approximating to 4LiAl₄Si₃AlO₁₀(OH)₈. The three available analyses of cookeite from Maine show that small quantities of sodium, potassium, &c., should also be included with lithium. The relationship of cookeite to similar minerals with layer-structures is shown in the following table III. Deviations from the numbers given in the third column are well known, and a general theory explaining them will be offered at a future date. Pauling¹ and McMurchy² have shown that the structure of chlorite is built up of

¹ L. Pauling, Proc. Nat. Acad. Sci. U.S.A., 1930, vol. 16, p. 578. [M.A. 4-465.] ² R. C. McMurchy, Zeits. Krist., 1934, vol. 88, p. 420. [M.A. 6-45.]

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alternating layers of $[Mg_3Si_3AlO_{10}(OH)_2]^{-1}$ and $[Mg_2Al(OH)_6]^+$. The negatively charged layers are the same as those existing in magnesian micas, but instead of being linked by potassium ions they alternate with brucite-like layers in which one atom of magnesium is replaced

| TABLE III. | Atomic contents and unit-cell dimensions * of cookeite and |
|------------|--|
| | related minerals. |

| | | 1 unit-cell contents. | Total metal atoms per | Cell dimensions : | | | |
|--------------|-----|--|--------------------------|-------------------|------|----------------|--|
| | | - | unit cell. | <i>a</i> . | b. | $c\sin\beta$. | |
| Chlorite | | Mg5AlSi3AlO10(OH)8 | 40 | 5 30 | 9.19 | 28·27 Å. | |
| Cookeite | | LiAl ₄ Si ₃ AlO ₁₀ (OH) ₈ | 36 | 5.13 | 8.93 | 28.30 | |
| Dickite | ••• | $Al_4Si_4O_{10}(OH)_8$ | 32 | 5.15 | 8.95 | 28.68 | |
| Nacrite | | $Al_4Si_4O_{10}(OH)_8$ | 32 | 5.15 | 8.95 | 28.70 | |
| Lepidolite | | K(Li,Al) ₃ (Si,Al) ₄ O ₁₀ (OH) ₂ | 32 | 5.18 | 9.02 | 19-97 | |
| Muscovite | | KAl ₂ Si ₃ AlO ₁₀ (OH) ₂ | 28 | 5.18 | 9.02 | 19.95 | |
| Talc | | $Mg_3Si_4O_{10}(OH)_2$ | 28 | 5.26 | 9.14 | 18.52 | |
| Pyrophyllite | | $AJ_2Si_4O_{10}(OH)_2$ | 24 | 5.14 | 8-90 | 18.37 | |

* Unit-cell dimensions of these minerals have been redetermined on specimens in the British Museum collections, with the exception of chlorite¹ and muscovite.²

TABLE IV. Sequence of atomic planes along a direction perpendicular to the c(001) plane for half the unit-cell height.

| Phlogopite. | Chlorite. | Cookeite. | Muscovite. | Dickite. | Talc. |
|---|--|---|---|--|---|
| Phlogopite. $\uparrow O_{6}$ $\mid Si_{3}Al$ $\mid O_{4}(OH)_{2}$ $\therefore O_{4}(OH)_{2}$ $Si_{3}Al$ $\mid O_{6}$ $\mid K_{2}$ $\downarrow O_{6}$ | Chlorite. $\uparrow O_6$ $\mid Si_3A1$ $\mid O_4(OH)_2$ $\mid Mg_6$ $\sim \lor O_4(OH)_2$ $\ddagger Si_3A1$ $\ddagger O_6$ $\mid (OH)_6$ $\mid Mg_4A1_2$ $\mid (OH)_6$ | Cookeite. $\uparrow O_6$ $\begin{vmatrix} Si_3A1 \\ O_4(OH)_2 \\ Al_4 \\ \checkmark O_4(OH)_2 \\ Si_3A1 \\ \hline O_6 \\ (OH)_6 \\ Li_2Al_4 \\ (OH)_6 \end{vmatrix}$ | Muscovite. $\uparrow O_6$ $\mid Si_3A1$ $\mid O_4(OH)_2$ $\because Al_4$ $\bigtriangledown O_4(OH)_2$ $\Im i_3A1$ $\mid O_6$ $\mid K_2$ $\downarrow O_6$ | Dickite. $\uparrow O_{6}$ $\mid Si_{4}$ $O_{4}(OH)_{2}$ Al_{4} $\forall (OH)_{6}$ $\forall O_{4}(OH)_{2}$ Al_{4} $\mid O_{4}(OH)_{2}$ Al_{4} $(OH)_{8}$ | Talc. $\uparrow O_{6}$ $\mid Si_{4}$ $O_{4}(OH)_{2}$ $\Leftrightarrow Mg_{6}$ $\Re O_{4}(OH)_{2}$ $\Leftrightarrow Si_{4}$ $\mid O_{6}$ $\downarrow O_{6}$ |
| | ↓ V6 | ↓ U6 | | ↓ [∪] 6 | |

by one of aluminium. The probable sequence of layers for cookeite is $[Al_2Si_3AlO_{10}(OH)_2]^{-1}$ a muscovite-like layer, and $[LiAl_2(OH)_6]^+$ a gibbsite-like layer containing lithium. The detailed sequence of the atoms along the *c*-axis of phlogopite, chlorite, cookeite, dickite, and tale (table IV) shows the relationship of these minerals still more clearly. Cookeite is to be regarded as a distinct species, and more closely related to the chlorite group than to the micas, tale, or the

² W. W. Jackson and J. West, Zeits. Krist., 1930, vol. 76, p. 211. [M.A. 4-467.]

¹ R. C. McMurchy, Zeits. Krist., 1934, vol. 88, p. 420. [M.A. 6-45.]

clay minerals. This conclusion is in agreement with the views of Penfield and Hallimond,¹ and depends not only upon physical and chemical data but upon evidence of its formation. Some of the specimens of cookeite from Maine may support the view that it is an alteration product of lepidolite, tourmaline, or spodumene; but others, particularly those from Buckfield, Ogofau, and Columbia river, show that cookeite undoubtedly occurs as a primary mineral.

HYDROMUSCOVITE.

The material analysed by Miss H. Bennett, M.A., B.Sc., comprises: Variety A. A soft, silver-white variety occurring as delicate membranous flakes in narrow veinlets exposed in the surface-workings marked 'Clochdy Gwenno', where it is associated with quartz, coarsely crystalline pyrite and arsenopyrite.

TABLE V. Chemical analyses of hydromuscovite (and hydrobiotite).

| | | | A. | В. | С. | D. |
|--------------------------------|-------|--------------|--------|-------|--------|-------|
| SiO ₂ | | | 46.54 | 48.39 | 44.81 | 41.54 |
| TiO, | | | 0.17 | 0.11 | 0.24 | 1.90 |
| Al ₂ O ₃ | | | 36.37 | 34.64 | 35.26 | 16.55 |
| Fe ₂ O ₃ | ••• | | 0.72 | 1.15 | 0.63 | 12-10 |
| FeO | | | 0.36 | 0.27 | 0.79 | 2.49 |
| MnO | | | nil | nil | 0.01 | 0.18 |
| MgO | | | 0.50 | 0.44 | 0.52 | 9-19 |
| CaO | | | 0.22 | 0.26 | 0.22 | 2.35 |
| Na ₂ O | | | 0.46 | 0.22 | 0.76 | 1.35 |
| K ₂ O | | | 8.06 | 7.82 | 7.64 | 4.48 |
| Li ₂ O | | | trace | trace | n.d. | |
| H2O (a | bove | 105°) | 6.31 | 6.07 | 8.40 | 7.63 |
| H2O (b | oelow | 105°) | 0.52 | 0.44 | 0.75 | n.d. |
| P_2O_5 | | | 0.06 | 0.06 | trace | _ |
| F | ••• | | 0.02 | 0.06 | n.d. | n.d. |
| CO ₂ | ••• | ··· · | nil | nil | nil | — |
| | | | 100.31 | 99.93 | 100.03 | 99.76 |
| Less 0 |) - F | | 0.01 | 0.03 | | |
| | | | 100.30 | 99.90 | | |

Variety B. A pale, prase-green, wax-like variety occurring as a veneer on shear-planes traversing an ore-body on the 420-foot level in the mine.

Variety C. Mica separated from the same source as variety Λ and treated with KCl solution in the manner described on p. 518.

¹ A. F. Hallimond, Min. Mag., 1925, vol. 20, p. 305.

D. Hydrobiotite, deep orange-brown, from biotitized amphibolite, Hayslad quarry, West Malvern.

For mica A, $\beta 1.575$, $\gamma 1.580$ (sodium-light), sp. gr. 2.65 ± 0.02 . Corresponding values for mica B are within the range approximated for mica A. Unit-cell dimensions determined on mica A are a 5.185, b 9.025, $c \sin \beta 20.20$ Å.

These values agree closely with those determined by Jackson and West (1930) for muscovite: $a 5 \cdot 18$, $b 9 \cdot 02$, $c \sin \beta 19 \cdot 95$ Å. The greater *c*-spacing in the hydromuscovite is related to the high hydroxyl content discussed below (p. 518). McMurchy (loc. cit.) has made a similar observation with regard to chlorites, which show a greater variation in respect of both *c*-spacing and hydroxyl content.

| | | | Atomic pro | · | | Metals and | |
|----------------------------------|-------|-------|-------------------------|--------------------|--------------|------------|--|
| | | 0. | Oxygen and fluorine. | Other elements. | | £ | silicon to basis (O,OH,F) ₄₈ . |
| SiO ₂ | | 46.54 | 1.5498 | 0.7749 | Si | | 12.00 |
| Al ₂ O ₃ | | 36.37 | 1.0703 | 0.7135 | Al | | 11.06 |
| Fe ₂ O ₃ · | | 0.72 | 0-0135 | 0.0090 | Fe''' | | 0.14 |
| FeO | | 0.36 | 0.0050 | 6.0020 | Fe'' | | 0.08 |
| Mg() | | 0.50 | 0.0124 | 0.0124 | Mg | | 0.20 |
| CaO | | 0.15 | 0.0026 | 0.0026 | Ca | | 0.04 |
| Na ₂ O | | 0.46 | 0.0074 | 0.0148 | Na | · | 0.22 |
| K ₂ Ō | | 8.06 | 0.0856 | 0.1712 | K | | 2.66 |
| $H_2O +$ | • • • | 6.31 | 0.3502 | 0.7005 | н | | 10.86 |
| | | | 3.0968 | | | | |
| F | ••• | 0.02 | 0.0011 | | \mathbf{F} | ••• | 0.02 |
| | | | 3.0979 | | | | |

As the two micas A and B contain inclusions of extremely minute needles identified as rutile, the TiO₂ content in each case is ignored in computing formulae. So also is the small P_2O_5 content, which has been traced to dust-like inclusions referred to apatite. The usual analytical figures for water do not discriminate between hydroxyl and molecular water. There is warrant, however, for continuing to treat water above 105° C. as affording an approximate measure of (OH) content.

By computation from the analytical data, specific gravity, and unit-cell dimensions, it is demonstrable that in the muscovite cell the total number of (OH) groups, F atoms, and O atoms independent of hydroxyl rounds off to 48; that is to say, $(O,OH,F)_{48}$ is a constant which is characteristic of muscovite. For micas A and B in particular,

the computed totals are 47.4 and 47.5 respectively. The fact that these totals fall below 48 suggests that none of the water above 105° C. is either mechanically held or adsorbed along crystal surfaces. Assuming $(O,OH,F)_{48}$ to be a constant, it follows that if F shows no significant variation, (OH) and O can vary reciprocally. Hence variation with respect to (OH) must involve departure from the standard O_{40} required for the generalized muscovite formula $(OH)_8K_4Al_8(Si_{12}Al_4)O_{40}$. The treatment (table VI, p. 517) of the analytical data for mica A illustrates the method of computing a muscovite formula to the basis of $(O,OH,F)_{48}$.

The formula thus derived for mica A, together with the formulae for micas B, C, and D, are compared with the ideal muscovite formula in table VII.

| Idea muscov | l ite. | | А. | В. | C. | D. |
|---------------------|-----------|-------------|--|--|---|--|
| K ₄ | |) K (Na | $\left. \begin{array}{c} 2 \cdot 66 \\ 0 \cdot 22 \end{array} \right\} 2 \cdot 88$ | $\left. \begin{array}{c} 2 \cdot 58 \\ 0 \cdot 10 \end{array} \right\} 2 \cdot 68$ | $\left. rac{2\cdot 48}{0\cdot 38} ight brace 2\cdot 86$ | $\left. \begin{smallmatrix} 1\cdot54\\0\cdot70 \end{smallmatrix} ight\} 2\cdot24$ |
| | | Ca Mo | 0·04 0·20 | $\left(\begin{array}{c} 0.06\\ 0.16 \end{array} \right)$ | $\begin{pmatrix} 0.06 \\ 0.20 \end{pmatrix}$ | $\begin{pmatrix} 0.68\\ 3.70 \end{pmatrix}$ |
| A1 | | Mn Fo'' | 0.08 7.59 | - 0.08 7.54 | 0.16 6.60 | 0.04 |
| Alg | ••• | Fe''' | 0.14 | 0.08 1.54 | 0.10 | 2.46 |
| | | Ti Al | —) 7·06) | 7·02 |) 6·06 | (0.38) (0.52) |
| $(Si_{12}Al_4)$ | | Al Si | $\frac{4.00}{12.00}$ 16.00 | $rac{3\cdot 52}{12\cdot 48}$ $\left. 16\cdot 00 ight.$ | $4\cdot 56$ 16.00 | $\begin{array}{c} 4.76 \\ 11.24 \end{array}$ 16.00 |
| O ₄₀ | | 0 | 37.12) | 37.52 | 33.70) | 34.22) |
| (OH,F) ₈ | | (OH) (F | $\begin{array}{c} 10.86 \\ 0.02 \end{array}$ 48.00 | $\begin{array}{c} 10.44 \\ 0.04 \end{array}$ 48.00 | $\begin{array}{c} 14.30 \\ - \end{array} \right\} 48.00$ | 13.78 48.00 |

TABLE VII. Atomic contents of hydromuscovite and hydrobiotite.

The two micas A and B diverge from the ideal muscovite constitution in the following respects:

(a) The proportion of hydroxyl groups is high.

(b) The total of the atoms in place of the standard K_4 is low.

(c) The atom-assemblage corresponding to the standard Al_8 is also low.

Features (a) and (c) are accentuated in the case of mica C, which is a variety of mica A modified by experimental treatment to determine its base-exchange capacity as follows: A quantity of the mica was immersed in N/20 solution of potassium chloride for 120 hours at room-temperature (25-28° C.), and after being thoroughly washed with hot distilled water was analysed by Miss H. Bennett, who found that water above 105° C. was nearly 36 % higher than in mica A.

The term 'hydromica' lacks precise definition. In the case of the Ogofau micas, the modification of the $(OH)_8K_4$ group in the direction $(OH)_{8+m}K_{4-n}$ is considerable, and is accompanied by modification of the standard Al_8 in the direction Al_{8-p} . This divergence from standard muscovite is accompanied also by change in physical properties towards those characteristic of the clay species. The authors suggest that the Ogofau micas might serve, provisionally, as the type to which the description 'hydromuscovite' is applicable; and 'hydrobiotite' would also be illustrated by the constitution, given under D in table VII.

The character and extent of the modification in any particular case is probably determined by the growth conditions and subsequent history of the mica. The Ogofau hydromuscovite is clearly the product of a low-temperature hydrothermal (magmatic) phase of mineralization, and the cookeite is referred to a similar phase.

A lode facies such as that described on pp. 507-510 (exclusive of cookeite) is commonly in genetic relationship to igneous rocks of granitic or granodioritic composition. The probability that the Ogofau ore-bodies are so related is further suggested by the occurrence of cookeite and the increased abundance of this mineral in the deeper levels of the mine. But no igneous rocks consistent with this relationship are exposed in the immediate district, though surface indications of mineralization similar to that at Ogofau occur sporadically over a considerable area.

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EXPLANATION OF PLATE XIX.

The Laue photographs were taken with 'white' X-radiation from a tungsten target using an intensifying screen. The crystal-plate distance for figs. 1 and 2 is 3 cm.

The rotation photographs of single flakes or flaky aggregates, figs. 3 to 9, were all taken with unfiltered CuK₀ radiation, $\lambda = 1.539$ Å., in the same cylindrical camera, diameter 6.04 cm. A length of 10 cm. on both the original Laue and rotation photographic films is equivalent to 10 cm. on the reproduced figures.

FIG. 1. Laue photograph of cookeite from Ogofau, Pumpsaint, Carmarthenshire, along the c [001] axis; a [100] axis vertical.

FIG. 2. Laue photograph of cookeite from Ogofau along the c[001] axis; a[100] axis vertical. An imperfect alignment of the individual crystals is revealed.

Fig. 3. Clinochlore about the b [010] axis.

FIG. 4. Rotation photograph of cookeite from Ogofau about the b [010] axis.

FIG. 5. Rotation photograph of cookeite from Hebron, Oxford Co., Maine, set approximately about the b[010] axis.

Frg. 6. Muscovite about the b [010] axis.

FIG. 7. Dickite about the a[100] axis.

FIG. 8. Nacrite about the a [100] axis.

FIG. 9. Lepidolite about the b[010] axis.

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