Cypriot Copper-Bearing Wood

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SYNOPSIS

A portion of a lagging pole, of Pinus brutia, containing numerous, radially disposed, blebs of native copper visible to the naked eye, which was recovered from Roman workings in a Cypriot copper mine, is described and compared with a similar specimen described elsewhere.

Examination of polished sections reveals that each of the larger lenticular blebs consists of a core of native copper commonly containing comparatively small masses of cuprite and distorted tracheids or portions of tracheid walls. The core is more-or-less completely fringed by cuprite, and this is bordered by a zone of distorted tracheids and partly decomposed tracheid walls. Some of the tracheids of this outer zone are stained, or infilled, by a green copper species, whilst others, both within and outside the zone, contain either calcium sulphate or limonite.

Apart from the larger complex blebs there are embryonic ones either consisting of one, or more, tracheids with cuprite-veneered inner walls, or of slender cuprite lenses, with or without kernels of native copper.

The problem of the genesis of these micro deposits is considered, and whilst it is tentatively concluded that reduction of cupric ions by glucose, derived from cellulose, and possibly coniferin, by hydrolysis, played a major rôle, no unique and complete answer to the problem is possible because the necessary chemical data are nowhere available.

Introduction.

In Western Cyprus a number of massive cupriferous pyritic bodies occur. Some of these are presently being mined, but all of them were discovered by Roman or pre-Roman prospectors and were exploited by them. The Roman and pre-Roman miners were able to work these deposits quite extensively because of the deep water-table and because they possessed large slave labour forces. The immensity of some of the operations is indicated by the vast accumulations of ancient slag in the vicinity of Cyprus Mines' Corporation Skouriotissa Mine, and by the scale of the early workings which have, from time to time, been intersected during recent operations at Skouriotissa and elsewhere. The present mining is concerned with the recovery of the ore, which, for one reason or another the Romans and their predecessors did not, or could not, extract.

The early miners were extremely well versed in mining methods and the close timbering which they had to use extensively differed but little from that employed by much more recent miners. One Roman set recovered is shown in figure 1.

Roman sets consisted of vertical posts each terminated by a long and short 'horn' between which the cap rested. A lagging of poles, about 2 in. in diameter, was placed above and alongside the sets in the drifts, and the spaces between it and the rock were infilled with the pole and set trimmings (i.e., twigs and chips).

Some of this wood, when cut, is seen to contain blebs of native copper, ranging from minute specks to ones whose maximum dimension is, c. 0.25 in. According to Mr. Stuthridge (to whom I am indebted for much of the information concerning the early methods of mining, and for the copper-bearing wood which is described in some detail below) " most of the copper wood that has been found occurs as top and side lagging, although the occasional post of a drift set has turned out to be copper-bearing — probably about one in every hundred, if that ".

Mr. Stuthridge also makes the point that most of the Roman timber which is found is very badly charred probably due to the heat produced during oxidation of the sulphides, and this makes copper-bearing wood comparatively rare since it is only found in those copper-rich zones which were not particularly rich in the readily oxidised pyrite.

General Geology and Mineralogy of the Ore Deposits.

A brief description of the geology and mineralogy of the ore deposits is a necessary prelude to an appreciation of the possible genesis of the copper-bearing wood.

The massive cupriferous pyritic bodies which characterise the Cypriot metallogenetic province are spatially — and probably genetically — related to pillow lavas. In some instances they occur more-or-less at the spilite/overlying rock contact, whilst elsewhere they are found largely, or entirely, in the volcanic rock.

At Skouriotissa, for example, the largest of the ancient producers, the orebody lies under a hill which rises c. 1,300 ft. above sea level. This feature is capped by limestone beneath which lie, from top to bottom, chalky marls, tuffs, bentonitic clays, black shales, the orebody and the so-called Upper Pillow Lavas. The orebody, which is 2,200 feet long and 900 feet wide, and occupies a depression in the lavas, is a near-horizontal elliptical (concavo-convex) lens, with the concave surface uppermost.

At Maurovouni, the canoe-shaped orebody lies in altered Upper Pillow Lavas and these are overlain by marls and fanglomerates.

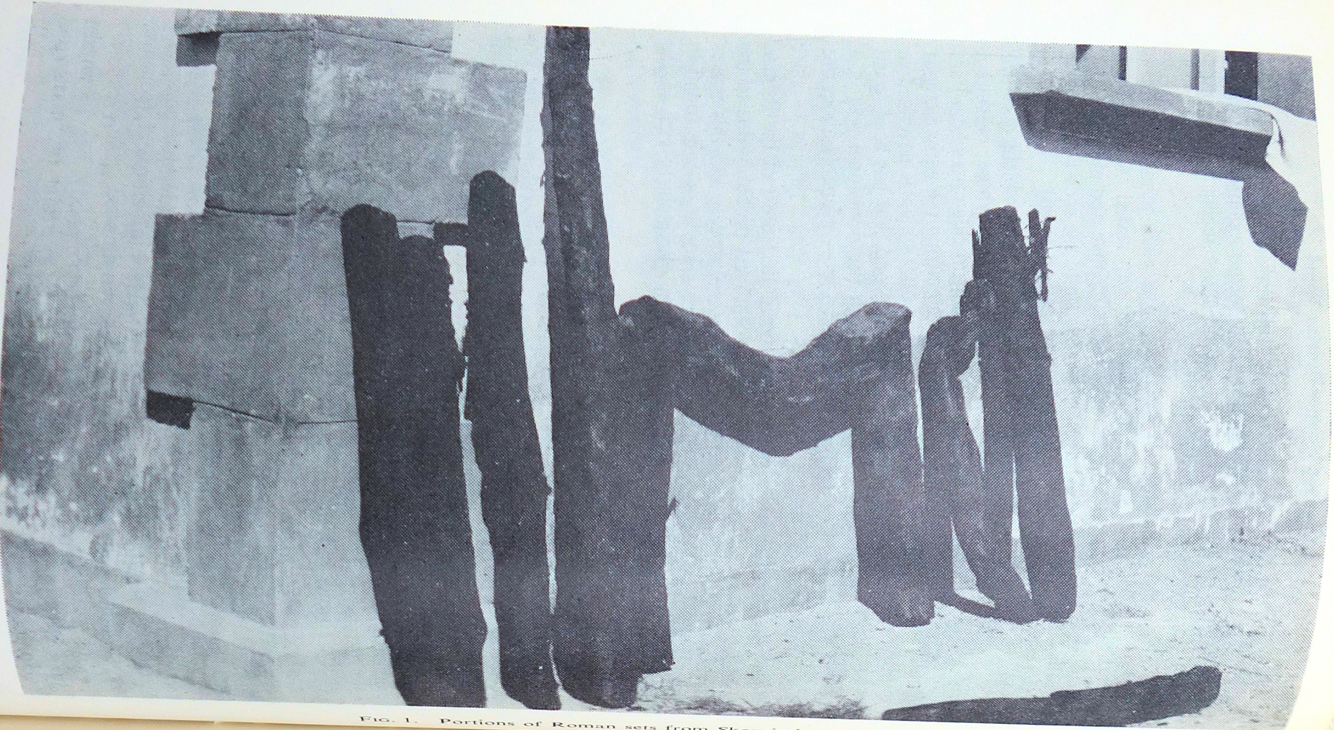


Fig. 1. Portions of Roman sets from Skouriotissa Mine.

The primary ore, which may be conglomeratic or brecciated, consists essentially of pyrite, marcasite and chalcopyrite, the two former species occurring as crystals or as colloform bands. At Apliki, sphalerite is unusually prevalent. The ore generally has been subject to considerable oxidation and secondary sulphide enrichment, and locally much of the chalcopyrite has given way to chalcocite and minor covellite.

Strong gozzans are present, and in consequence of the comparatively dry climate, sulphates form the dominant suite of minerals overlying the sulphides. At Skouriotissa, recent removal of the overburden as a preliminary to opencast mining operations, has presented an unusually good opportunity for further examination of these sulphates, and Bear (1963, p.53) makes the following observations about them :—" Besides the sulphates of copper, iron and zinc, such as chalcanthite, melanterite, brochantite and goslarite, various complex and unstable ferrous and ferric compounds have formed. These include römerite, coquimbite, fibroferrite, copiapite, natrojarosite, alunogen and abundant szomolnokite. The entire western end of the sulphide body was weathered in place and changed to natrojarosite. Between the natrojarosite and main body of sulphide is a zone ranging from nearly pure szomolnokite, to various proportions of sulphide and szomolnokite, to sulphide only slightly oxidised." (See footnote.)

Within certain of the Roman workings Mélon and Donnay (1936, pp.164-165) noted voltaite, coquimbite, alunogen, römerite, chalcanthite, fibroferrite, halotrichite, and tamarugite. (See footnote.)

That the iron-bearing sulphates are present in vastly greater amounts than the copper ones is not surprising when it is realised that the average, essentially sulphide ore, produced from Skouriotissa assays 2.25 per cent copper, 48.0 per cent sulphur and 43.0 per cent iron (Bear, op. cit., p.53).

The Copper-Bearing Wood.

The specimen of copper-bearing wood (fig. 2), which has been examined in some detail by the writer has been identified by Mrs. Turk (Personal Communication) as Pinus brutia. The specimen, which is a portion of a lagging pole, has a marked transverse fracture near one end, due, possibly, to local collapse of the roof of the drift.

Footnote :—The formulae of some of the more unusual species are as follows :— **Römerite,** near $Fe^{\cdot\cdot}\ Fe_2^{\cdot\cdot\cdot}\ (SO_4)_4 .\ 12H_2O$. **Coquimbite,**
$4[Fe_2(SO_4)_3 .\ 9H_2O]$. **Fibroferrite,** $Fe^{\cdot\cdot\cdot}\ SO_4\ OH .\ 5H_2O$. **Copiapite,**
$R^{\cdot\cdot}\ Fe_4^{\cdot\cdot\cdot}\ (SO_4)_6\ (OH)_2 .\ nH_2O$, where $R^{\cdot\cdot}$ includes $Fe^{\cdot\cdot}$, Mg, Al, Cu,
or Na_2. **Natrojarosite,** $[NaFe_3^{\cdot\cdot\cdot}\ (SO_4)_2\ (OH)_6]$. **Szomolnokite,** $FeSO_4 .$
H_2O. **Voltaite,** $20[HK_2Fe_4^{\cdot\cdot}\ (Fe_2Al)_3\ (SO_4)_{10} .\ 13H_2O]$. **Alunogen,**
$Al_2(SO_4)_3 .\ 18H_2O$. **Halotrichite,** $Fe^{\cdot\cdot}Al_2\ (SO_4)_4 .\ 24H_2O$. **Tamarugite,**
$NaAl\ (SO_4)_2 .\ 6H_2O$.

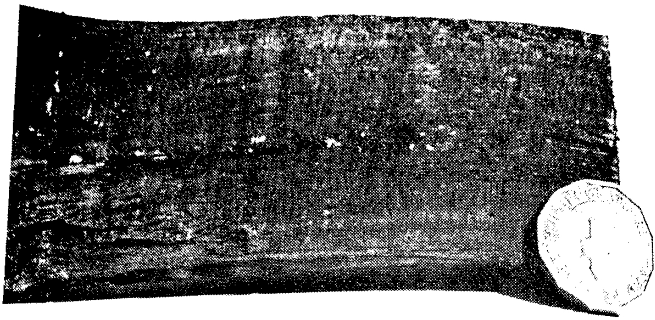


FIG. 2. Longitudinally split portion of lagging pole, of Pinus brutia, showing specks of native copper (white). All the following figures relate to portions of this specimen.

Before removal of material for sections it was c. 7 in. long and c. 1.75 in. in diameter, and about a half of it was covered by loosely attached bark on which there were still some fragments of lichen. The wood, which consists of light golden- and darker-brown annual rings, shows no sign of decay and takes a high polish.

Before the writer acquired the specimen it had, for a part of its length, been sawn longitudinally in order to see if it contained copper, and on the surface so exposed, which was c. 4.5 in. in length, 43 blebs of native copper occurred which were distinctly visible to the naked eye. A line bisecting this saw-exposed plane longitudinally would divide it into two halves which differ in that one contains virtually all the readily visible blebs of copper. In transverse section all the larger copper blebs are so orientated that their long axes are aligned along radii (fig. 3). They occur from almost the centre of the pole to within a quarter of an inch of its outer surface. There is no relationship between the size of a given bleb and its location, and randomly distributed between the larger ones are smaller specks which are just visible to the naked eye. (Under the microscope still smaller copper masses are seen.)

Chromographic contact prints, using 0.880 ammonia as an attacking agent and a 1 per cent alcoholic solution of rubeanic acid as a developing reagent, indicate that copper is scattered throughout the wood but maximum concentration occurs in the half which contains most of the visible copper and which lies beneath that portion which is partly covered with bark and which, therefore, may well have faced the roof of the drift. This distribution of copper differs significantly from that of similar material examined by Biek.

Biek (1963, p.126) records that the specimen examined by him " is a three inches thick uniform section cut (in modern times) from what had reputedly been a pit prop in a Roman part of a copper mine in Cyprus. It was found by R. Emmerson to be part of a young cedar (Cedrus sp.) about four inches in diameter. Some bark still remains. Even more remarkable than its state of preservation, literally perfect, is the abundant presence of copper not only as the usual greenish coloration but also as specks of bright metal. An X-radiograph has confirmed that the metallic copper is concentrated in the outer rings and present uniformly throughout the depth of the specimen . . . there is little variation in copper from bark to pith . . ."

Biek's specimen and the writer's differ in the following important respects :—

i. Biek's is of a size that makes it probable that it was a vertical member of a set : the writer's was a portion of a lagging pole.

ii. The metallic copper in Biek's specimen is concentrated in the outer rings and is more-or-less uniformly distributed in an annular zone : the writer's specimen, as noted above, shows a completely different distribution pattern.

iii. The total copper distribution patterns of the two specimens differ significantly.

iv. Biek's specimen has been identified as cedar whilst the writer's is pine.

The significance of some of these differences will be discussed later.

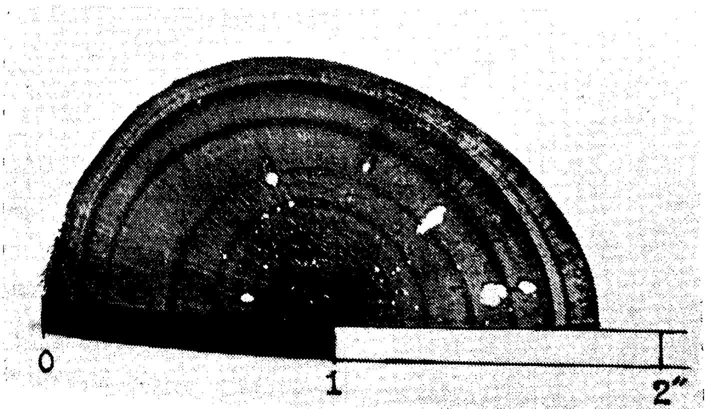


FIG. 3. Transverse section of lagging pole showing blebs of native copper (white) arranged in a crudely radial manner.

Microscopic Examination of the Wood. (Figs. 4-7.)

Examination, under the microscope, of thin and polished longitudinal and transverse sections of the wood reveals the following :—

Each large cupriferous bleb consists of a core of native copper which, in longitudinal sections, is seen to contain elongate inclusions and voids which approximately parallel the long axes of neighbour tracheids, and which are walls of tracheids and distorted tracheids which were not infilled with copper species. Randomly dispersed small aggregates of cuprite also occur in the native metal.

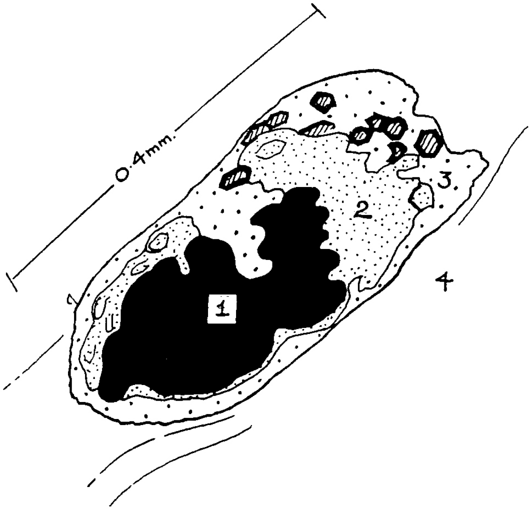


FIG. 4. Sketch of a polished transverse section of a radially orientated copper-bearing pod. The native copper core (1), is surrounded by massive cuprite (2) containing, particularly near the outer margin, portions of tracheid walls which may or may not, be distorted. Zone 3 consists essentially of highly distorted tracheids and a few of the less distorted ones are infilled with a secondary green copper species (hatched in diagram). Zone 4 is normal wood, but near the copper pod the tracheids have been somewhat distorted as if to accommodate the mineral deposit as it was growing.

A cuprite zone, of varying thickness, more-or-less completely invests each mass of copper and the contact between the two species is undulating.

Fringing the cuprite zone is one which contains distorted and fragmentary tracheids in a micro-mottled organic matrix. The tracheids bordering each large bleb have been contorted to accommodate the accumulation of copper species.

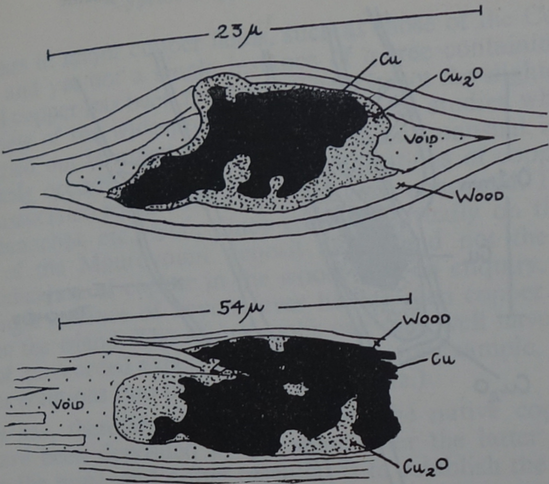


FIG. 5. Sketches of polished tangential longitudinal sections of comparatively simple copper/cuprite pods.

In the various sections examined cupriferous micro-deposits of a wide range of complexity occur. Locally the cavity of a single tracheid is simply smeared with cuprite: elsewhere one, or a few neighbouring tracheids are partly infilled with cuprite, and within some or all of the oxide accumulations one or more kernels of native copper may be present. On other occasions cuprite, with or without native copper, occupies spaces which appear to have been largely developed by the thrusting apart of adjacent tracheids by the developing minerals. Finally, the large complex blebs are to be seen which have been described above.

In the sections examined, green and more rarely blue minerals occasionally stain or fill some of the tracheids immediately fringing the blebs. These species have not been identified but they are probably copper- and possibly copper/iron-bearing sulphates.

Beyond the sites of obvious copper deposition some of the tracheids are infilled with white calcium sulphate, whilst others, both removed from and near the copper deposits, contain an opaque iron species which may be limonite.

Genesis of the Copper Deposits in the Wood.

When considering the genesis of the copper deposits in the wood it is necessary not only to consider the various chemical aspects of the question but also the temporal ones. Although the specimens examined by Biek and the writer came from underground workings this does not necessarily mean that the copper was introduced after the wood had been placed in the mine: it could have accumulated

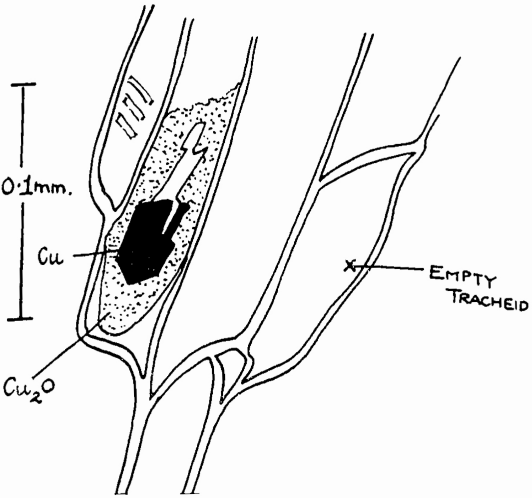


FIG. 6. Sketch of a tangential longitudinal section of a number of tracheids of which one is part infilled with native copper and cuprite.

when the trees were growing : indeed, Biek (op. cit., p.127) briefly mentions such a case (the only one reported in the literature) which was described by Frankforter (1899, pp.44-45). Frankforter (p.45) states that " while removing an oak tree which grew within the limits of the city of Minneapolis, and which had recently died, a gentleman noticed, in cutting up the trunk, a considerable quantity of bright copper-coloured powder disseminated through the pores of the wood. A quantity of this powder was examined, and found to be pure metallic copper. . . . A careful preliminary examination . . . showed certain parts of the tree to be thoroughly impregnated with the granular copper powder. Some of these granules were so large as to take on the form of flakes. The larger ones were partly rolled up so as to fit in the irregular pores of the wood ". Frankforter further notes that " the microscopical examination revealed the fact that only the outer annual rings contained an appreciable quantity of the metal. The last six or seven annual rings were so full of these flakes of copper that they could easily be discerned by the naked eye. The earlier rings showed only a trace under the microscope and by analysis, and those nearest the heart were quite free from the metal ". This copper distribution pattern led the investigator to conclude that the tree assimilated excessive quantities of copper from the soil (for a reason or reasons unknown) during the last few years of its life and that this was probably the cause of its death.

Although Frankforter's observations are very interesting no weight can be attached to them when considering the genesis of the copper in the Cypriot wood. Thousands of trees have been felled in recent years in major copper fields, such as those of the Congo and Zambia, and yet not a single instance of a tree containing visible elemental copper has been recorded. There is not the slightest doubt that in view of the widespread biogeochemical studies which have been carried out in many copper areas such an unusual indication of the likely occurrence of marked concentrations of copper in the soil and associated rocks would not have gone unnoticed. The writer has cut branches, etc., from pines growing virtually on the gossan outcrop of the Maurovouni deposit and found not the slightest visible indication of copper in the wood, nor, on enquiry, could he find a single instance of anyone ever having seen copper in recent timber on the island. The Minneapolis oak may well have been the subject of unrecorded experiments involving, for example, injections of copper solutions into the tree. (See footnote.)

One has, therefore, to consider how the native copper, etc., could have been deposited in the wood after the latter had been placed in the mine, and a first essential is to establish the nature of the underground environment. As a result of vigorous oxidation of the sulphides during the period of early mining operations the underground temperature may well have been locally in excess of 100° F., and the atmosphere would have been humid and charged with sulphur dioxide. (It is worth remembering that comparatively recently underground operations were terminated at Skouriotissa because of the exceedingly high temperatures, the presence of much sulphur dioxide, and the tendency to spontaneous combustion. There is no reason for believing that conditions in the Roman mines were better.

Immediately after the first rains following periods of drought, ground water, charged with sulphates and sulphuric acid, as a result of the flushing out of the oxidised zone, would drip from the roofs of drives on to the lagging and other timber supports, and migrate into the wood, particularly via radial cracks which developed as it dried out in the hot, drier periods. It follows that such events might well cause vertical timber to be saturated throughout by metal-rich liquids, as a result of drops falling on the upper exposed end, whilst only certain sectors of horizontal lagging would be so affected. This accounts adequately for the difference in the distribution of copper in Biek's specimen, whose size indicates that it was a vertical member of a set, and of the metal in the writer's piece of lagging.

Footnote:—On occasion the wood of fallen branches of British oaks is brilliant emerald green. By some this has been erroneously thought to be due to the deposition of copper salts, the trees having absorbed unusually high concentrations of the metal in question from the soil. The colour is, in fact, due to the invasion of a fungus, Chlorosplenium aeruginascens, the "emerald or verdigris rot". In the Tunbridge Wells area this wood was used, together with others, from the seventeenth century until 1934, in the manufacture of wooden articles decorated with mosaic veneers which are generally known as Tunbridge Ware. (See 'Emerald rot used in Tunbridge Ware', The Times, Jan. 21st, 1965.)

The major question as to how the copper and cuprite developed remains to be examined.

The possible modes of cuprite copper genesis present themselves and will be subsequently considered : —

i. The species in question developed from one or more copper-bearing sulphides which may have been deposited from solution in the wood, or may have been washed in — possible as colloids — from the overlying ore. In the latter case the sulphides may be entirely the products of strictly inorganic reactions — in the former case the possibility of their precipitation by biogenic hydrogen sulphide must be considered.

ii. The cuprite and copper were the products of reactions involving sulphate solutions and reducing gases, other than hydrogen sulphide, which were generated by decomposition of the timber by bacteria.

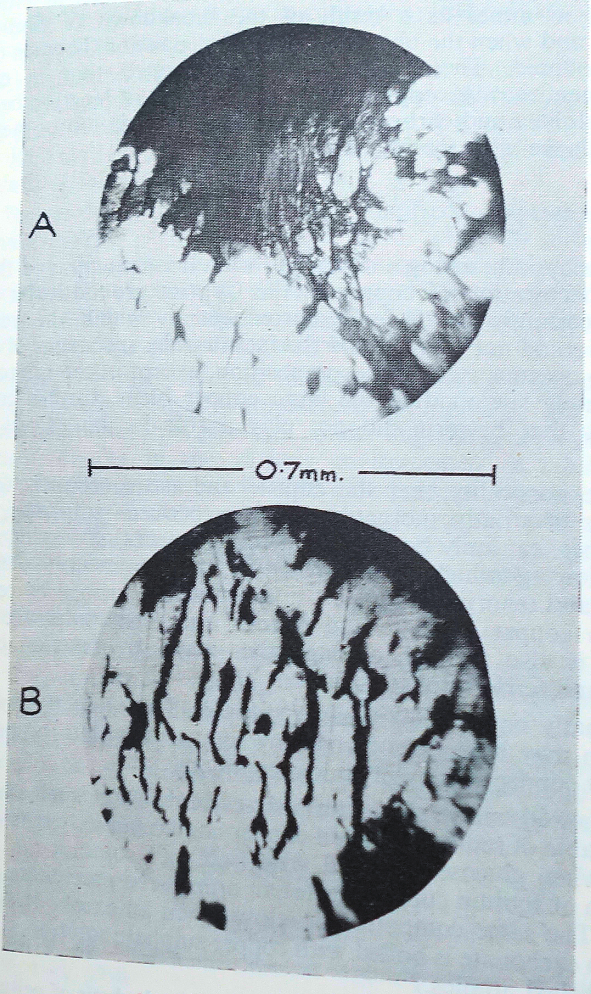
iii. The minerals in question were the outcome of strictly inorganic reactions, essentially between sulphates, which took place within the wood.

iv. The copper and cuprite owe their origin essentially to reactions between copper-bearing sulphate solutions and organic reducing agents which were the products of hydrolysis of one or more of the components of the timber.

In virtually all accounts of the development of cuprite and native copper in the oxidized zones of ore-deposits it is postulated that these species are derived from copper-bearing sulphides, particularly chalcocite, which in its turn commonly formed as a result of replacement of pyrite. Thus Park and Mac Diarmid (1964, p.420) suggest that the following equation may represent the usual mode of genesis :—

$$2Cu_2S + 8Fe^{+3} + 12SO_4^{-2} + 6H_2O + {}^3/_2O_2 \longrightarrow$$
$$2Cu + Cu_2O + 8Fe^{2+} + 12H^+ + 14SO_4^{-2}.$$

However, not a single speck of any sulphide has been identified in any of the sections prepared from the specimen described in some detail above, nor in sections from other specimens from a further number of early workings in Cyprus ; nor has the sodium azide iodine reaction (see Feigl, 1947, p.464) established the presence of sulphide ions in any of the material examined. Although it might be argued that sulphides which had been precipitated in the timber, or had been transported in a finely divided state into it, were the progenitors of the species under review, and that subsequent reactions have erased all traces of their former presence, such a theory is based on such nebulous postulates that it cannot be seriously considered. It also follows therefore, that biogeneic and other theories of sulphide formation need not be pursued further.



FIGS. 7A and B. FIG. 7A. Portion of a complex pod showing native copper (white) partly fringed by tracheids containing cuprite (medium grey). Normal wood is dark grey.

FIG. 7B. Copper core of complex bed containing distorted and crushed tracheids and tracheid walls (black).

(Both figures are of polished tangential longitudinal sections.)

The possibility that the minerals in question might have been the products of reaction between ionic copper, in solution, and reducing gases which were developed by the decomposition of components of the timber by bacteria is, initially, an attractive thought when it is remembered that methane has accumulated in some old workings in mines as a result of the breakdown of timber by bacteria, and when the observations of Douglas and Thomas (1957) are considered. These workers have suggested that in certain environments native copper may be the product of reaction between cupric sulphate and carbon developed by the oxidation of methane, in accordance with the following equations :—

$$CH_4 + O_2 \longrightarrow C + 2H_2O$$
$$CuSO_4 + C \longrightarrow Cu + SO_2 + CO_2.$$

This hypothesis loses its appeal when it is remembered that the high concentration of copper in the Cypriot ground-water would almost certainly create an environment in which the required bacteria could not survive, and the fact that the specimens of timber show no obvious signs of decomposition, except in the micro-zones immediately surrounding the large copper blebs, further confirms the view that bacteria did not play a rôle in the genesis under review.

The possibility that the cuprite and native copper were the products of strictly inorganic reactions between sulphates, within the timber, can only be supported by utterances of despair such as the following which Bateman (1950, p.271) makes :—" Cuprite (Cu_2O) and tenorite (CuO), . . . are possibly formed by reactions between copper sulphate and ferrous and ferric sulphate. Native copper is also formed, among other ways, by reaction between cupric and ferric sulphate solutions ".

Finally one has to consider the possibility that the species in question may have been formed by reduction within the timber of divalent copper ions by organic compounds.

It is well known that certain six-carbon sugars, such as glucose, are capable of reducing certain copper compounds to cuprous oxide. Thus, when glucose is boiled with Fehling's solution (an alkaline solution of sodium cupri-tartrate) an orange-red precipitate of Cu_2O forms. The same compound develops when an excess of powdered calcium carbonate is boiled with copper sulphate solution. (Hosking. Unpublished Studies.)

Glucose may be prepared by the acid hydrolysis of cellulose and of certain glucosides. It seems possible, therefore, that the cuprite in the Cypriot wood may owe its origin to reaction between glucose and copper-bearing solutions, as the acid-rich ground-water would be quite capable, under the locally high temperatures which must have occurred in the mines, of hydrolysing the cellulose walls of the phloem cells of the timber and also the glucoside coniferin which such wood contains. The alkaline conditions which appear to be necessary for the reduction of cupric to cuprous ions by glucose may well have been provided by calcium carbonate which may have been deposited in the wood as a result of the latter being seasoned before use by placing it in local lime-rich streams. The presence of calcium sulphate in the wood supports the contention that calcium carbonate was originally present.

The question as to how the cuprite was converted to native copper still remains open. Possibly this was effected by reducing agents produced by the acid decomposition of lignin in the vicinity of the centres of precipitation : as noted earlier, lignin has certainly been partially decomposed around such centres.

This last theory of genesis, though admittedly wanting in certain important details, is the only one of those which have been considered which lacks weaknesses of such magnitude that necessitates its out-of-hand rejection. It therefore elects itself as the broadly correct one. Quite obviously the testing of the validity of the theory is desirable, but in practise virtually impossible.

Finally, it is not irrelevant to remark that this study will have been worth-while if it has simply served to emphasise that remarkably little is known with certainty concerning the genesis of cuprite and native copper in the various environments in which these minerals occur, and that much more experimental work is required if really meaningful advances are to be made in the understanding of the genesis of ores containing these species and, indeed, in the fundamental chemistry of the oxidised zone generally.

Acknowledgements.

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Presentations

WE are very grateful to Mr. A. J. Stafford, A.C.S.M. (1916-17 and 1920-21) for presenting to the library a large number of bound Proceedings of the Institution of Mining & Metallurgy, and other mining books.

Mr. Stratford was born at Devon Great Consols Mine and was an old boy of Truro School, and after a year at the School of Mines served in H.M. Forces, returning to the School in 1920. He started his career in South Africa at the Crown Mines and later became one of the senior inspectors of the Rhodesian Mines Department. Other A.C.S.M's. in the Department with him were Messrs. Mitchley, James, Wallace and Benham. He is now living in Cornwall.

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We are grateful for a very fine model of Morococala Mine presented anonymously through D. M. Ferguson, A.C.S.M., 1961.

Award of Scholarship

WE are pleased to report that Mr. J. C. Davey, A.C.S.M., 1925, has covenanted an annual payment of £5 for seven years for the provision of a prize in Economic Geology to be called the "J. C. Davey Prize for Economic Geology".