CENTENAIRE DE LA SOCIÉTÉ GÉOLOGIQUE DE BELGIQUE
GISEMENTS STRATIFORMES ET PROVINCES CUPRIFÈRES, LIÈGE, 1974, 203-213

ON THE DIAGENETIC FORMATION OF ORES IN SEDIMENTARY BEDS, WITH SPECIAL REFERENCE TO KAMOTO, SHABA, ZAÏRE

by Paul BARTHOLOMÉ*

ABSTRACT

Based on observations pertaining to the stratiform copper-cobalt deposit at Kamoto, Zaire, an analysis is presented of the various ways in which metal may be brought to and fixed into sedimentary beds during sedimentation and diagenesis.

Petrological criteria are shown to exist by which the time of metal fixation may be established relative to pyrite growth and other diagenetic events. It is concluded that the concepts of syngenetic and epigenetic deposition are quite inadequate to describe the phenomena responsible for metal fixation.

INTRODUCTION

In a paper published in 1972, Garlick summarizes the history of ideas concerning the genesis of stratiform copper deposits in the central African Copperbelt. He notes that, for many years, "hydrothermal solutions emanating from adjacent intrusive granites" were held responsible for these deposits, although Schneiderhöhn had put forward a quite different interpretation. He also explains how he himself became convinced that:

1. the granites and pegmatites are nowhere intrusive into what is now called the Katangan but were truncated by the erosional surface at the base of the Katangan;
2. the mineralization was emplaced before folding.

Then he describes various features of the rocks, some of which suggest that sulfides behaved in the same manner as zircon and tourmaline during sedimentation or resedimentation. He finally concludes: "The copper, cobalt, and iron may be derived from either:

1. Erosion of a vast hinterland with perhaps only normal concentrations of the metals, with perhaps local rich concentrations.

(*) Laboratoire de Géologie appliquée, Université de Liège, Belgium.

2. Supply by upwelling oceanic currents.
3. Volcanic exhalations, which in view of the lack of recognizable volcanic debris in the Lower Roan, were probably distant from the Copperbelt."

Garlick's views are actually quite comparable with Schneiderhöhn's. However, as shown from the following extract, translated from a paper published in 1937 (p. 286-287), the latter author differed in:

a) allowing more explicitly for a possible concentration of the metals during early or even late diagenesis and

b) asserting categorically that the copper originated from the same source as the detritals constituting the sedimentary sequence, i.e. the Basement, although after perhaps considerable delays and lengthy detours during diagenesis:

"Primary origin of the mineralized horizon: It is in my opinion syngenetic-sedimentary. The metal content comes from the groundwaters of the sedimentary sequence during gradual accumulation. In these groundwaters, were contained the metals originating from the weathering of the innumerable chalcopyrite-pyrite veinlets that one still finds even today in many places in the Basement. Furthermore in the groundwaters, metallic solutions flowed that came from the alteration of the fragments of ore incorporated as such among the detritus.

"The precipitation of the metals took place at any time during the sedimentary cycle: either simultaneously with the other precipitated minerals of host rock, or soon afterwards, or perhaps also much later; or, which is perhaps the most probable, scattered indissolved sulfide remnants inside a bed behaved as nuclei, and the small amount of metal present in the groundwater gathered on them during a long period of time so that the metal initially highly dispersed in time and space became concentrated into a small thickness (of strata)"?

My own experience lies essentially with the Kamoto ores, and more specifically with their mineralogy, petrology and geochemistry. And my purpose in this paper will be to discuss the information obtained from Kamoto in the light of the theories put forward by Schneiderhöhn, Garlick and others.

It is recognized of course that all the stratiform copper deposits in the Copperbelt are not identical. In particular, considerable differences are known to exist between those of Northern Zambia and Southern Shaba, the two sub-provinces which constitute the Copperbelt. The following are especially noteworthy:

1. The landscape where the mineralized beds and formations were deposited was more rugged in Zambia than in Shaba. In the latter sub-province, none of these palaeohills so often mentioned by Zambian geologists have as yet been discovered; the stratigraphy of the ore-bearing succession is accordingly much more uniform,

2. The detritals brought to the basin of deposition were distinctly coarser and more arkosic in Zambia,

3. The geochemical sedimentary environment was different, although presumably hypersaline in both sub-provinces: anhydrite, which is widespread and abundant in Zambia, is not found in the ore-bearing succession in Shaba. On the other hand, the latter is highly magnesian: some beds contain much magnesite; others contain an abundance of authigenic chlorite devoid of iron, which may have resulted from the transformation of palygorskite; dolomite is a major component of almost all beds; calcite is never found.

In addition to differences between the two sub-provinces, there are obviously variations among individual deposits. However, some kind of relationship also exists between them as it has been recognized by all authors. The relationship is evidenced by:

1. the metal content: all these deposits are mined for copper; some of them are also mined for cobalt, but for no other chalcophile element,
2. the extremely well marked stratiform character,
3. the location of the orebodies at a small distance above an unconformity at a level where the succession ceases to be essentially detrital.

Kamoto is one of the stratiform deposits of the Copperbelt. It cannot be taken as a representative sample of this group of deposits. But one of its most interesting features is that the Kamoto ores have suffered little deformation and essentially no metamorphism. This is especially striking when one compares it with the Zambian deposits. It appears therefore likely that the Kamoto ores are especially reliable sources of information concerning problems of genetic interest such as the composition of mineralizing solutions, and the environment where sulfides precipitated.

SOME MAJOR FEATURES OF THE KAMOTO ORES

The beds which constitute the two orebodies mined at Kamoto are made of the following components (Oosterbosch, 1962; Bartholomé, 1962; Bartholomé *et al.*, 1972; Dimanche, this volume; Katekesha, 1974):

Major minerals: quartz (in part detrital, in part authigenic; small amounts of chalcedony are also present); muscovite (detrital but modified during diagenesis); chlorite (in part detrital but mostly authigenic); dolomite (perhaps in part detrital); magnesite (entirely authigenic).

Minor minerals: chalcocite; digenite or anilite; bornite; chalcopyrite; carrollite.

Trace minerals: pyrite, covellite, rutile, tourmaline, zircon, graphite. The absence of sphalerite and galena even as trace minerals is noteworthy.

The beds are quite often laminated, especially where mineralized, the lamination resulting either from changes in grain size or in mineralogy. Most individual laminae are made of dolomite, shale or authigenic quartz. Some of them contain 25 % sulfides by weight.

Detailed petrological study has demonstrated a complex suite of diagenetic events in the Kamoto ores, as evidenced by pseudomorphs and nodules as well as by the great abundance and variety of authigenic minerals. Sulfide minerals were obviously involved in the diagenetic processes.

On the other hand, a general agreement seems to have been reached about an essentially pre-folding and pre-faulting age of mineralization as suggested by Schneiderhöhn and by Garlick for the entire Copperbelt. The observations presented by Oosterbosch (1962), Demesmaeker, François, Oosterbosch (1963) and François (1973) are most significant in this respect.

We are therefore led to analyze in detail the history of the mineralized sequence during the *diagenetic time interval.*

AN ANALYSIS OF METAL FIXATION IN SEDIMENTARY ROCKS

I define the *diagenetic time interval* as follows: the beginning of it is taken as the moment where the clastic components of the rock are deposited, which of course varies from one horizon to the next; the end is taken as the moment when the primary porosity and permeability of all the beds in the sequence have decreased down to the very low values now observed.

Obviously, throughout the diagenetic time interval, the chemical composition of any bed in the sequence may change because of chemical reactions with the fluids flowing through it or through adjacent beds, or else with chemical compounds diffusing through these fluids. Variable quantities of a chemical element may be added or removed according to the solubility and the rates of flow involved.

It is of course recognized that, during the same diagenetic time interval, recrystallization of a mineral species may also take place, the size and shape of the grains as well as their precise location in the rock being changed in the process. This phenomenon is important in particular because it may obliterate earlier textures (see for example Brown and Bartholomé, 1972). However, we are not directly concerned with it here since, by the definition that has just been given, recrystallization in itself does not change the metal content of the rock.

Any chemical element may be brought to the rock along with terrigenous grains and other allochemical or orthochemical material settling at the interface between the free water and the sediment. I shall call *allothigenic* the mineral grains incorporated in the sediment by being brought mechanically to the interface, since they were not formed in the sedimentary rock where we find them (*allothi* means *elsewhere* while *allos* means *other*; *authi* means *here*).

A chemical element may also be incorporated in the rock by entering authigenic minerals growing at the water-sediment interface. I shall call *strictly sedimentary* both the allothigenic and authigenic mineral grains brought to the rock at this interface, i.e. in direct contact with the overlying water.

On the other hand, a chemical element may become fixed in the rock below the interface because of some chemical reaction taking place there. As a result of such reactions, new authigenic mineral grains may form; allothigenic ones may be altered as well as earlier formed authigenic grains. But since interstitial waters are never concentrated with respect to the chemical elements considered here, high elemental contents become possible only if material is imported from some external reservoir.

During early diagenesis, under a small thickness of sediment, importation is possible from the overlying water body, either by diffusion (mostly vertically) or by flow (mostly horizontally as in sabkhas); at that time, the interstitial water is a slightly modified sea, or lake, or lagoon water.

During later diagenesis, after a sufficient thickness of sedimentary layers of varying permeability have been deposited above the bed, the exchange of matter is no longer possible directly with the overlying water. However the interstitial water flowing through the bed or through adjacent beds may supply chemical elements and remove others. This water may again originate from the overlying sea or lake or lagoon as in the seepage refluxion model (Adams and Rhodes, 1960), or it may be local meteoric water rising through the sedimentary pile as it happens nowadays in the Red Sea and Lake Kivu (Degens, Ross, 1969; Degens *et al.*, 1973), or no doubt may have several other origins.

Presumably waters of somewhat different origins, compositions, and temperatures may flow successively through the same beds, resulting in successive generations of authigenic minerals and successive alterations of earlier formed mineral grains.

If we now consider a sulfide mineral, which obviously involves at least two chemical elements, the following possibilities arise:

a) metal and sulfur are both truly sedimentary. This may happen if the sulfide grains are allothigenic as nowadays in the Black Sea sediments (Spencer and Brewer, 1971), or if they form authigenically from truly sedimentary material, e.g. pyrite from gypsum and goethite coatings on sand grains.

b) metal is truly sedimentary while sulfur is imported and fixed in the bed during diagenesis. This appears to be the usual way in which microscopic pyrite is formed in sediments (Berner, 1970; etc.): iron is brought as a component of the terrigenous fraction; sulfur is borrowed from the interstitial water where bacterial reduction of sulfate proceeds and is replenished by diffusion from the overlying water.

c) sulfur is truly sedimentary while metal is imported and fixed in the bed during diagenesis. This might happen if allothigenic gypsum reacted after deposition with a metal-bearing brine.

d) sulfur and metal are both imported and fixed in the bed during diagenesis. Since the length of the diagenetic time interval may be ten or one hundred million years, this does not mean, obviously, that sulfur and metal are brought at the same time.

CRITERIA CONCERNING THE RELATIVE AGE OF AUTHIGENIC MINERALS

According to the distinctions suggested above, authigenic minerals may be truly sedimentary, early or late diagenetic, according to whether they formed from the overlying water, from a modified overlying water, or from a water that is distinctly different in composition and/or origin. However we often lack operational criteria that are sufficiently sensitive to establish such distinctions in practice.

In fact the best criteria that are available to us concern the time relationships between metal fixation on one hand and pyrite formation on the other. In 1962 I used these criteria to suggest that in the Kamoto ores most of the copper and cobalt were imported and fixed after pyrite formation. Since then, many new observations have demonstrated the usefulness and versatility of these criteria in the study of Copperbelt ores where traces of pyrite are ubiquitous (Bartholomé, Katekesha and Lopez Ruis, 1971; Bartholomé *et al.*, 1972; Cailteux, this volume; Dimanche, this volume). Furthermore, other authors have applied them to quite different deposits (e.g. Arnold, Maucher, Saupé, 1972).

These criteria use the fact that in many sulfide ores hosted in sedimentary rocks, iron has gone in part or completely into the sulfide phases. In general at least part of this iron is truly sedimentary and belongs initially to terrigenous minerals, from which it is extracted in order to react with sulfur.

Of course we cannot determine how late after deposition pyrite formation begins and how long it lasts. But the interesting fact is that, in some ores, mineral grains containing another metal were present in the sediment at the beginning of pyrite growth (Arnold, Maucher, Saupé, 1972; Cailteux, this volume) while in others, for instance Kamoto, pyrite growth essentially preceded metal fixation.

Other authigenic minerals such as quartz, magnesite, apatite may be used in the same manner.

SUMMARY OF OBSERVATIONS MADE AT KAMOTO

Pyrite is present in trace amounts in the Kamoto orebodies and becomes abundant in some beds of the hangingwall. Some of it is framboidal but most is subidiomorphic. In the hangingwall, small concretions of irregular shapes and sizes (most often a few millimeters across) are found.

Microprobe examination has shown that pyrite grains are zoned with respect to cobalt, this element being enriched in a thin outer rim and occasionally also along a growth zone inside the grain. This was first discovered by examining a few samples from the hangingwall (Bartholomé, Katekesha, Lopez Ruiz, 1971). But now it appears to be true of all pyrite grains within and around the orebodies, whatever their shape and precise location. The concretions observed in the hangingwall display the same zoning as microscopic pyrite (fig. 1). This zoning does not ressemble at all the one observed in garnet and resulting from metamorphic recrystallization in a closed system (Hollister, 1966). It results most likely from an increase in the cobalt content of interstitial waters at the end of pyrite growth. Furthermore, this increase occurred before most of the other diagenetic phenomena

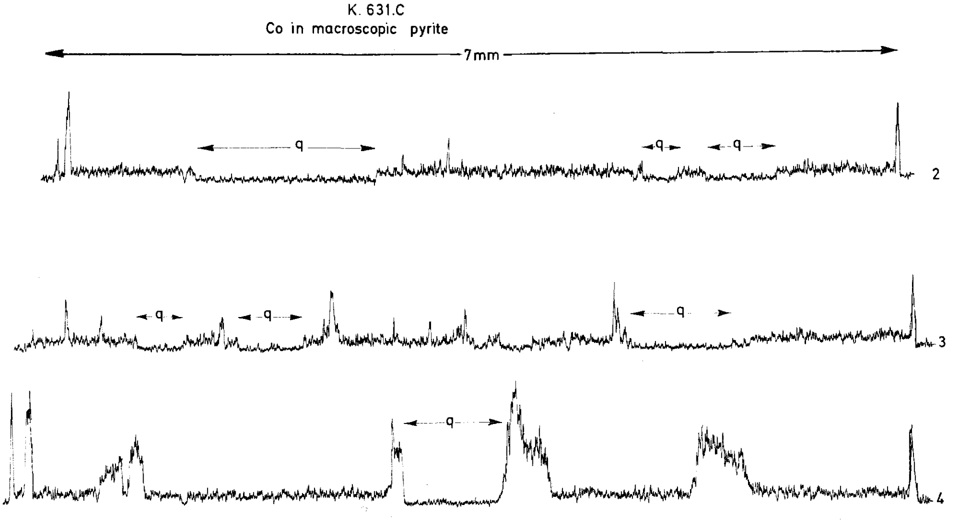

FIG. 1. — *Three microprobe traces through a pyrite nodule found in the hangingwall of the upper orebody at Kamoto. Quartz inclusions or embayments within the nodule are shown by the letter q. The cobalt-rich rim is clearly visible.*

took place, since the same cobalt-rich rim is observed around pyrite grains enclosed within authigenic quartz or phosphate or chalcopyrite.

On the other hand, within the orebodies where digenite or anilite, chalcocite, bornite and carrollite are the major sulfides, pyrite is found exclusively as inclusions in authigenic minerals, mostly quartz, and magnesite (Bartholomé, 1962; Bartholomé *et al.*, 1972). This observation suggests that during diagenesis the chemical composition of the interstitial water changed in such a way as to make pyrite unstable.

We are therefore led to imagine a two-stage model for the diagenetic evolution of the Kamoto deposit.

During the first stage, pyrite grew at the expense of truly sedimentary iron on one hand, and on the other hand of sulfur that was imported from overlying water or perhaps borrowed from earlier formed anhydrite. Some chalcopyrite may well have been contemporaneous with this pyrite and have originated in the same way, copper being imported from the overlying water. The fact is that the pyrite-chalcopyrite association is present in several carbonaceous beds of the hangingwall at Kamoto but the copper content in these beds does not rise much above 1 %.

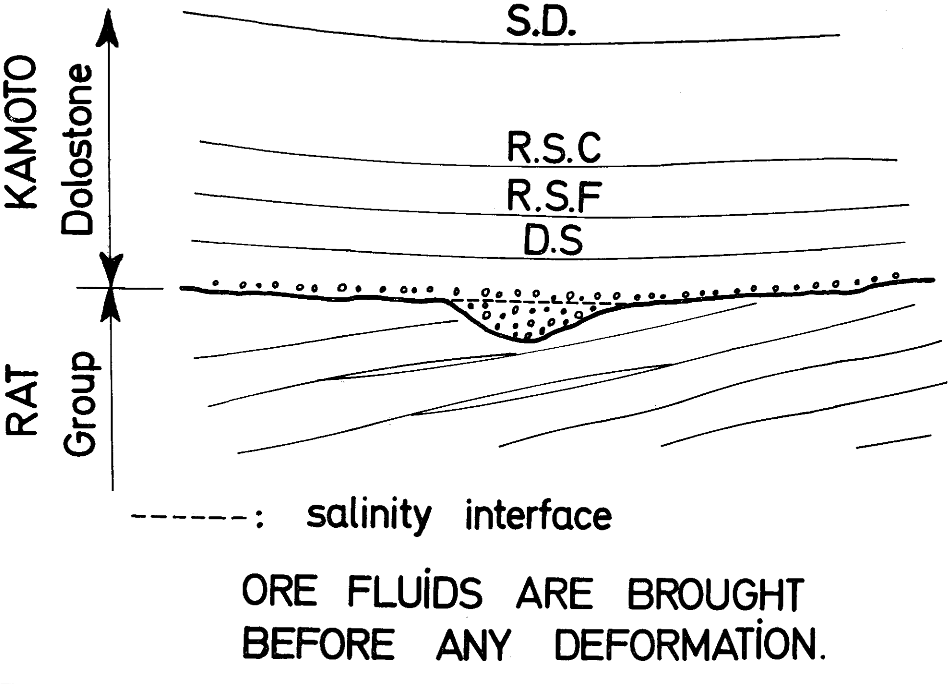

FIG. 2. — *A hypothetical cross-section through the RAT Group and the Kamoto Dolostone at the start of the second stage of diagenesis. The Kamoto Dolostone, which is the lowermost formation of the Mines Group, consists of: a) a red-colored, unmineralized conglomerate at the base, b) a laminites-bearing member which, where mineralized, constitutes the lower orebody. This member is subdivided into several units named R.S.F., D.S., etc., c) a non-laminated member, named R.S.C., which is nowhere mineralized. The lower part of the S.D. Formation above may also be mineralized and constitutes the upper orebody. It is noteworthy that this upper orebody is found only where the lower one is well developed (Oosterbosch, 1962; François, this volume).*

First stage diagenetic processes modified the beds of the Mines Group over a wide area and an important thickness.

During the second stage, which developed only locally in the lower part of the Mines Group, copper and cobalt were imported with a brine that invaded the lower part of this Group. Pyrite reacted with the copper-rich brine and transformed into bornite. In addition, large amounts of copper sulfides, bornite and carrollite precipitated so that the grade reached several percent of copper and half a percent of cobalt. The necessary sulfur was also imported with the brine as sulfate or else was borrowed from earlier formed anhydrite. The reaction ceased when organic matter became depleted.

What was the source of this brine? Bartholomé *et al.* (1972) have suggested that it originated in a lagoon existing some time after the deposition of the Mines Group or at the end of it; the lagoon water would have flowed through the sediments according to the seepage refluxion model. No doubt other origins are possible.

Whatever their origin, the brines had a high density, as evidenced by the presence of halite and sylvite as daughter minerals in fluid inclusions (Pirmolin, 1970). Perhaps they followed pebble filled palaeochannels at the base of the Mines Group (fig. 2) and from there spilled over into the overlying beds, rich in carbonaceous matter, where the metal was trapped. More probably, they filled incipient synclinal depressions after a slight warping had occurred (fig. 3). There are several ways in which a stratified structure of the interstitial waters may explain the particular distribution of orebodies observed in the Mines Group by François (this volume).

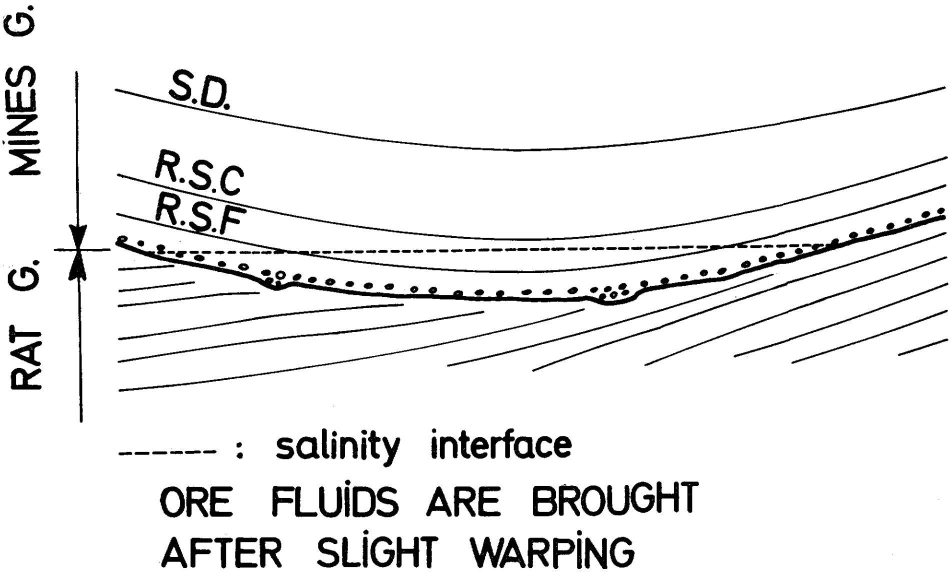

FIG. 3. — *Another hypothetical cross-section across the same units and at the same time as in figure 2.*

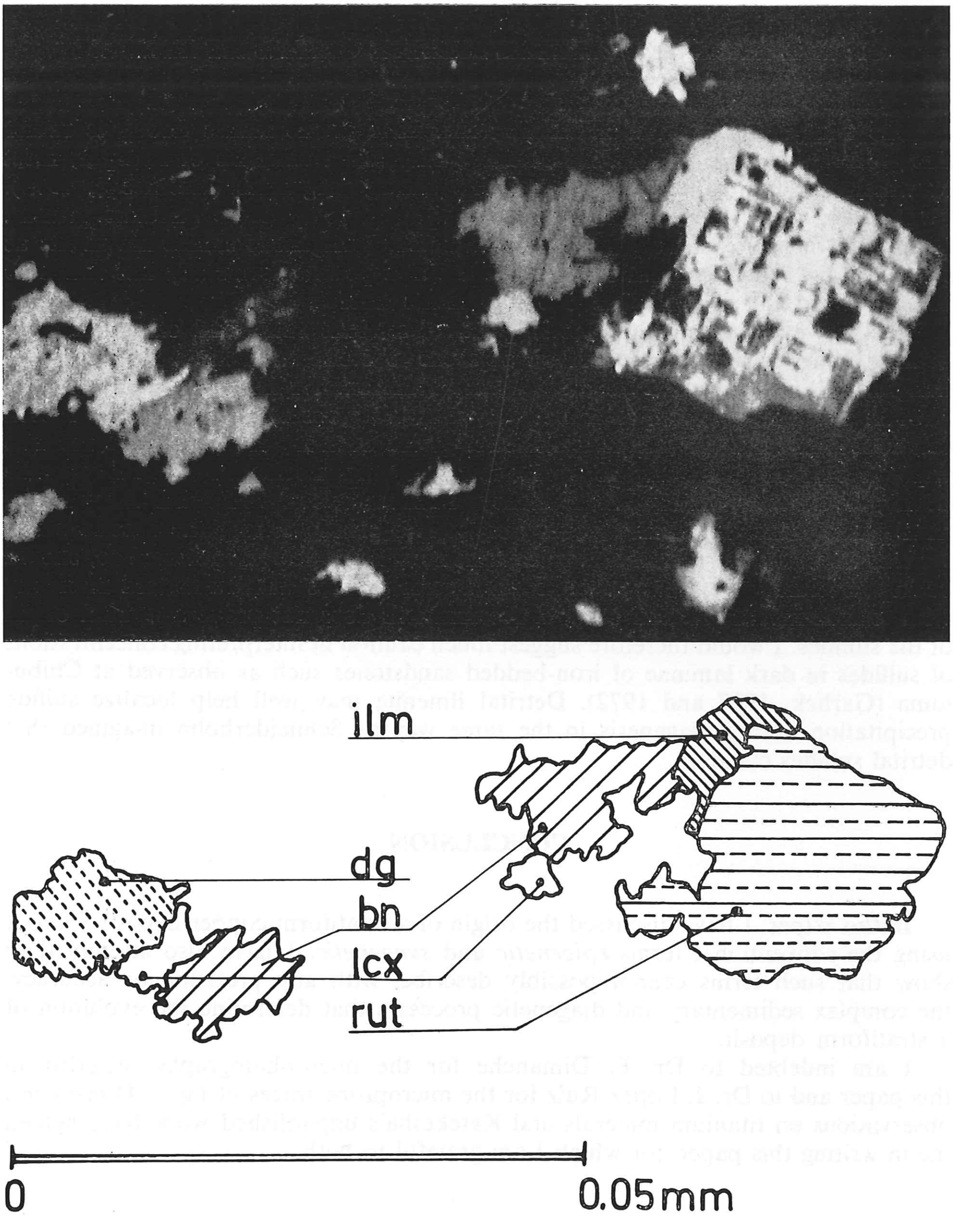

FIG. 4. — *Composite grains where bornite (bn) and digenite (dg) are intimately mixed with leucoxene (lex), altered ilmenite (ilm), and rutile (rut). The latter mineral forms a grid suggesting pseudomorphism after ilmenite exsolutions in magnetite. Microprobe work has shown that the iron content of titanium minerals never reaches much above 1 %. This element has gone almost entirely into the sulfide phases. Polished section, imm., photographed by F. Dimanche. Other examples of such composite grains are given by Cailteux and Dimanche (1974).*

ON THE LACK OF DETRITAL SULFIDE GRAINS AT KAMOTO

The terrigenous fraction in all the formations of the Mines Group is rather fine-grained and very mature. It contained only small amounts of feldspar, most of which has been altered into muscovite during diagenesis (Katekesha, 1974). Tourmaline, zircon and titanium minerals were the only dense minerals present. It appears therefore unlikely that there were any terrigenous sulfide fragments as suggested by Schneiderhöhn and in fact detailed petrological study has failed to bring forward any evidence that such fragments existed.

It is however noteworthy that in polished sections rutile and other titanium-bearing minerals are often observed adjacent to sulfides. In the beds above the hangingwall, rutile + pyrite assemblages are common. Within the orebodies, rutile + bornite and rutile + copper sulfide are exclusively observed (fig. 4). The reason for these particular textures is as follows: grains of more or less altered ilmenite were present in the terrigenous fraction; they transformed into rutile + pyrite during the first stage of diagenesis as they did in many common sedimentary rocks; then pyrite transformed into bornite or copper sulfide during the second stage.

But whatever the reason, the fact itself, i.e. the texture just referred to, should be kept in mind as evidence that association of sulfides with rutile and indirectly with tourmaline, zircon and other dense minerals does not prove a detrital origin of the sulfides. I would therefore suggest much caution in interpreting concentrations of sulfides in dark laminae of iron-bedded sandstones such as observed at Chibuluma (Garlick, 1967 and 1972). Detrital ilmenite may well help localize sulfide precipitation during diagenesis in the same way as Schneiderhöhn imagined that detrital sulfides could.

CONCLUSION

In this paper, I have discussed the origin of a stratiform copper deposit without using the conventional terms *epigenetic* and *syngenetic*. I have also attempted to show that such terms cannot possibly describe, with any precision or accuracy, the complex sedimentary and diagenetic processes that determine the evolution of a stratiform deposit.

I am indebted to Dr. F. Dimanche for the microphotographs included in this paper and to Dr. J. Lopez Ruiz for the microprobe traces of fig. 1. Dimanche's observations on titanium minerals and Katekesha's unpublished work have helped me in writing this paper, for which I am grateful to both.

References

Adams, J. E. and Rhodes, M. L. (1960). — Dolomitization by seepage refluxion. *Bull. Amer. Ass. Petr. Geol.*, Vol. 44, pp. 1912-1920.

Arnold, M., Maucher, A. and Saupe, F. (1972). — Diagenetic pyrite and associated sulphides at the Almadén mercury mine, Spain. In: *Ores in Sediments*, ed. by G. C. Amstutz and A. J. Bernard, Berlin, Springer-Verlag, p. 7-19.

BARTHOLOMÉ, P. (1962). — Les minerais cupro-cobaltifères de Kamoto (Katanga-Ouest). I. Pétrographie. *Studia Universitatis "Lovanium", Faculté des Sciences (Kinshasa)*, n° 14, p. 40.

BARTHOLOMÉ, P., KATEKESHA, F. and LOPEZ-RUIZ, J. (1971). — Cobalt zoning in microscopic pyrit from Kamoto, Republic of the Congo (Kinshasa). *Mineral. Deposita (Berl.)*, Vol. 6.

BARTHOLOMÉ, P., EVRARD, P., KATEKESHA, F., LOPEZ-RUIZ, J. and NGONGO, M. (1972). — Diagenetic ore-forming processes at Kamoto, Katanga, Republic of the Congo. In: *Ores in Sediments*, ed. by G. C. Amstutz and A. J. Bernard, Berlin, Springer-Verlag, p. 21-41.

BERNER, R. A. (1970). — Sedimentary pyrite formation. *Am. Jour. Sci.*, 268, p. 1-23.

BROWN, A. C. and BARTHOLOMÉ, P. (1972). — Inhomogeneities in cobaltiferous pyrite from the Chibuluma Cu-Co deposit, Zambia. *Mineral. Deposita (Berl.)*. Vol. 7, p. 100-105.

CAILTEUX, J. et DIMANCHE, F. (1974). — Examen des oxydes de fer et titane dans l'environnement du gisement de cuivre de Musoshi (Shaba, Rép. du Zaïre). *Bull. Soc. fr. Minéral. Cristallogr.*, 96, p. 378-382.

DEGENS, E. T. and KULBICKI, G. (1973). — Hydrothermal origin of metals in some East African Rift Lakes. *Mineral. Deposita*, 8, p. 388-404.

DEGENS, E. T. and ROSS, D. A., ed. (1969). — *Hot Brines and Recent Heavy Metal Deposits in the Red Sea.* Berlin, Springer-Verlag, p. 600.

DEMESMAEKER, G., FRANÇOIS, A. et OOSTERBOSCH, R. (1963). — La tectonique des gisements cuprifères stratiformes du Katanga. In : *Gisements stratiformes de cuivre en Afrique*, 2^{e} partie, ed. by J. Lombard and P. Nicolini. Association des Services géologiques africains, Paris, p. 47-115.

FRANÇOIS, A. P. (1973). — *L'extrémité occidentale de l'arc cuprifère shabien. Etude géologique.* Lubumbashi, Département géologique de la Gecamines, p. 120.

GARLICK, W. G. (1967). — Special features and sedimentary facies of stratiform sulphide deposits in arenites. *Proc. of the 15th Inter-University Geol. Cong.*, Univ. Leicester, Eng., p. 107-169.

GARLICK, W. G. (1972). — Sedimentary environment of Zambian copper deposition. *Geologie en Mijnbouw*, vol. 51 (3), p. 277-298.

HOLLISTER, L. S. (1966). — Garnet zoning: an interpretation based on the Rayleigh fractionation model. *Science*, 154, p. 1647-1651.

KATEKESHA, WA MUSANGANA (1974). — Doctorate thesis (in preparation).

OOSTERBOSCH, R. (1962). — Les minéralisations dans le système de Roan au Katanga. In: *Gisements stratiformes de cuivre en Afrique*, 1re partie, ed. by J. Lombard and P. Nicolini. Association des Services géologiques africains, Paris, p. 71-136.

PIRMOLIN, J. (1970). — Inclusions fluides dans la dolomite du gisement stratiforme de Kamoto (Katanga occidental). *Ann. Soc. Géol. Belg. (Liège)*, 93, p. 193-202.

SCHNEIDERHÖHN, H. (1937). — Die Kupferlagerstätten von Northrhodesia und Katanga. *Geol. Rundschau*, Bd. 28, p. 282-291.

SPENCER, D. W. and BREWER, P. G. (1971). — Vertical advection diffusion and redox potentials as controls on the distribution of manganese and other trace metals dissolved in waters of the Black Sea. *J. Geophys. Res.*, Vol. 76, No. 24, pp. 5877-5892.