

Occurrence and formation of cuprostibite in a Zn-Pb-Ag mineralized siliceous dolomite at Långsjön, Central Sweden

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With 8 figures and 2 tables in the text

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Abstract: The first occurrence of the rare mineral cuprostibite, Cu_2Sb , in a sulphide mineralized metamorphic dolomite is reported. The major sulphide minerals in the mineralized zones are sphalerite and galena. Cuprostibite occurs in small amounts together with minor quantities of a variety of Sb-rich minerals in different mineral assemblages and textural modes. The formation of cuprostibite took place through subsolidus decomposition of preexisting complex Cu-Pb-Sb sulphosalts and through crystallization from Cu-Ag-Sb-Pb rich fluids under conditions of low sulphur fugacity. Composite grains of cuprostibite + dyscrasite + gudmundite + galena, which show intense graphic intergrowth textures, crystallized from the last portion of these fluids during rapid cooling. This mineral assemblage, which also comprises arsenopyrite, was formed at temperatures below 280°C and at $\log a_{\text{S}_2} \leq -14.4$, as determined from the gudmundite stability and by means of arsenopyrite geothermometry. During the ore forming stages the prevailing temperatures were lower than during the peak of the regional metamorphism of the area ($\sim 600^\circ\text{C}$), which implies that the observed Sb-rich ore mineral assemblages postdate this metamorphic event. However, this observation does not rule out a premetamorphic concentration of the ore metals in the siliceous dolomite.

Key words: Cuprostibite, genesis, dyscrasite, native antimony, gudmundite, texture, microprobe analysis, sphalerite, galena; Sweden (Långsjön).

Introduction

Cuprostibite (Cu_2Sb) has so far been reported from the type locality, Taseq (SØRENSEN et al. 1969) and two adjacent localities within the Ilimaussaq intrusion, Greenland (KARUP-MØLLER et al. 1978). At these localities the mineral is found in sulphide bearing ussingite and analcime-aegerine veins (KARUP-MØLLER et al. 1978). Conditions for the formation of cuprostibite in these igneous rocks has been determined to $T = 360-430^\circ\text{C}$ and $\log a_{\text{S}_2} = -17.2$ to -11.8 (KARUP-MØLLER 1978). Cuprostibite has also

been observed in one magnetite-rich specimen from Långban, Central Sweden (BURKE 1980). No information on the cuprostibite bearing host rock at this locality was given and the conditions for cuprostibite formation were not investigated.

The present paper describes the mineralogy of a cuprostibite-bearing sulphide mineralization in a new type of host rock: a metamorphic siliceous dolomite. The aim of this study is to define the conditions of cuprostibite formation at this new locality and to relate these conditions, in time, to the regional metamorphism of the area.

Geology and regional metamorphism

The Långsjön Zn–Pb–Ag mineralization is situated in Central Sweden, approximately 50 km SW of Stockholm. The mineralization occurs in metamorphic siliceous dolomites which are surrounded by pelitic gneisses (Fig. 1). Younger granitic rocks, pegmatites and doleritic dykes are minor rocks in the vicinity of the mineralized area. All rock types are of Precambrian age. The ore mineral-bearing dolomite is mainly composed of

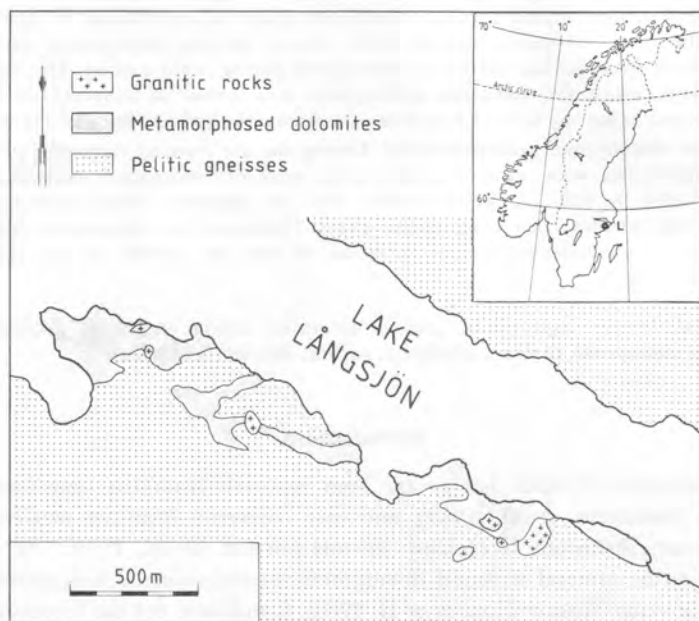


Fig. 1. Location of the Långsjön Zn–Pb–Ag mineralization and the geology of the area (simplified after PETERSSON 1980).

calcite, dolomite, diopside, serpentine and olivine. Serpentine is a product of extensive retrograde alteration of olivine.

The regional geology of the area has been described by STÅLHÖS (1975). On the basis of observed silicate mineral assemblages in the pelitic gneisses in combination with the andalusite-sillimanite univariant (RICHARDSON et al. 1969) and the muscovite + quartz = K-feldspar + andalusite/sillimanite univariant (KERRICK 1972), STÅLHÖS (1975) determined the conditions of regional metamorphism of the area to $P_{\text{tot}} = 3-4$ kbar and $T = 675-725$ °C. A recent thermodynamic analysis of the aluminium silicate triple point (DAY & KUMIN 1980) has shown that the triple point of HOLDAWAY (1971) describes the largest body of experimental and calculated data for this system in a better way than the triple point of RICHARDSON et al. (1969). Considering this observation and the fact that dolomite, calcite, diopside and olivine are coexisting phases in the dolomitic rocks at Långsjön, an estimation of the P and T parameters of the regional metamorphism may be obtained from the intersection of the andalusite-sillimanite univariant and the univariant for the reaction 1 diopside + 3 dolomite = 2 forsterite + 4 calcite + 2 CO₂ (KÄSE & METZ 1980). The intersection of these two univariants, in P-T space, lies at $P = 2$ kbar and $T = 600$ °C. These P and T values still fulfil the conditions for the reaction muscovite + quartz = K-feldspar + andalusite/sillimanite to go to the right. Furthermore, this temperature estimate is in better agreement with results obtained, when applying the garnet-biotite geothermometer (THOMPSON 1976; HOLDAWAY & LEE 1977; FERRY & SPEAR 1978) on reported microprobe analyses of garnet-biotite pairs in the present metapelites (STÅLHÖS 1975). This geothermometer yields a temperature of 600 ± 50 °C (depending on calibration) for the composition of these mineral pairs.

Mineralization

The general structural and chemical character of the Zn-Pb-Ag mineralization at Långsjön has been reported by PETTERSSON (1980). The mineralized zones, which are dominated by sphalerite and galena, occur in a siliceous dolomite and have an affinity for the silicate rich layers of this rock. Large-scale structural concordance between the mineralization and host rock is prevailing. In detail these zones are composed of narrow sulphide-rich veins, weak disseminations and microbreccias. The average Zn : Pb : Ag wt-% ratio of the mineralization is approximately 300 : 100 : 1, and the Ag/Cu wt-% ratio is 5-10. The content of Bi is below 5 ppm and the Ag/Sb wt-% ratio is higher than unity. The silver content correlates with the lead content in the mineralized zones (Fig. 2).

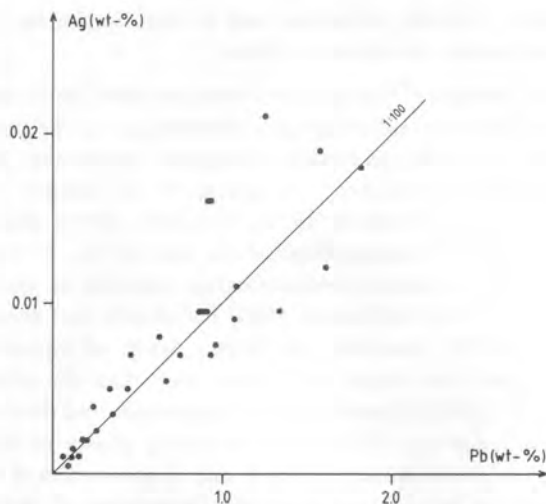


Fig. 2. Correlation plot of the Pb- and Ag-content in mineralized drill-core sections from Långsjön (analyses from PETERSSON 1980).

Mineralogy, textures and mineral chemistry

Apart from the major ore minerals, sphalerite and galena, a number of Sb-rich minerals have been observed in the present mineralization. These phases comprise native antimony (Sb), dyscrasite (Ag_3Sb), cuprostibite (Cu_2Sb), breithauptite (NiSb), gudmundite (FeSbS), tetrahedrite ($(\text{Cu}, \text{Ag})_{10}\text{Fe}_2\text{Sb}_4\text{S}_{13}$) and an unknown sulphosalt A (approx. $\text{Cu}_6\text{Fe}_6\text{Sb}_4\text{S}_{13}$). Other minor ore minerals occurring are native silver (Ag), arsenopyrite (FeAsS) and chalcopyrite (CuFeS_2). Tetrahedrite and the unknown sulfosalt A are exclusively found in the central part of large galena grains or in large galena-sphalerite composite grains. These mineral inclusions frequently show partial (Fig. 3 A) to complete (Fig. 3 B) decomposition to chalcopyrite + native antimony \pm dyscrasite. Chalcopyrite is only found in this textural situation. The galena grains, in which these inclusions are found, are also rich in uniformly distributed, microscopic inclusions ($\leq 5 \mu\text{m}$) of an Ag-Sb rich phase. Sharply defined areas, rich in minute ($\leq 10 \mu\text{m}$) rounded inclusions of Sb-rich minerals, are also found particularly in the rim zone of these galena grains. The Sb-minerals occurring in these areas are (1) native antimony (Fig. 3 C) or (2) cuprostibite + dyscrasite \pm antimony (Figs. 3 D and 4). Galena is always a constituent of these inclusion-rich areas. Cuprostibite and the other antimony rich minerals occur in three additional textural modes: (3) cuprostibite + dyscrasite + galena \pm

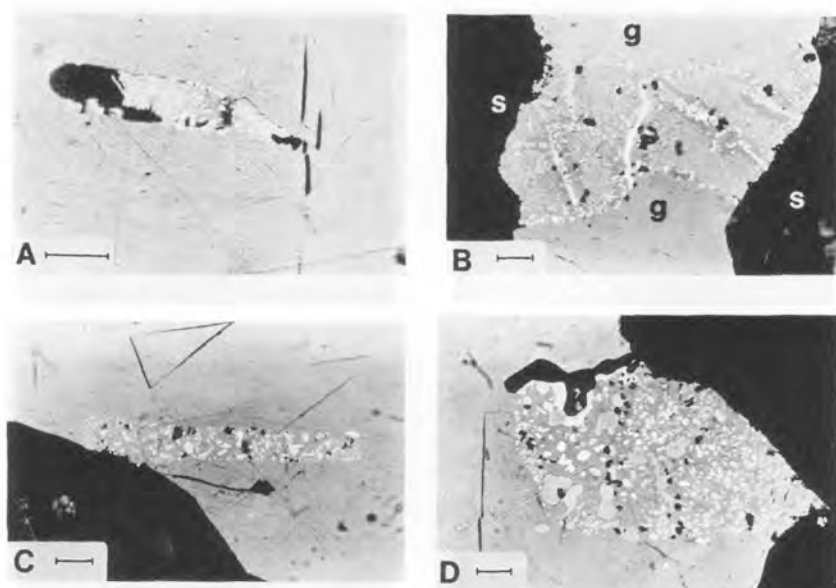


Fig. 3. Microphotographs. Reflected light, oil immersion, one nicol. Scale bar equals $10\ \mu\text{m}$. A: Partially decomposed sulphosalt A (dark grey) in galena. Decomposition products are chalcopyrite (grey), antimony (white) and very minor dyscrasite (white). B: Decomposition texture composed of chalcopyrite (grey) and antimony (white) in a composite galena (G) - sphalerite (S) grain. C: Lath-shaped area in galena showing a concentration of antimony inclusions (white). D: Inclusion rich area in the rim of a large galena grain. Mineral inclusions are cuprostibite (grey), antimony (white) and minor dyscrasite (very light grey).

antimony as interstitial infillings in dense galena-sphalerite matrix (Fig. 5 A), (4) infillings of cuprostibite + dyscrasite in fractured sphalerite grains, (5) composite grains ($\leq 40\ \mu\text{m}$) of gudmundite + galena + dyscrasite + cuprostibite which show graphic intergrowth textures (Fig. 5 B). These composite grains are found along microfractures in carbonate-silicate matrix or as thin rims crystallized around large sphalerite grains (Fig. 5 C). The most frequent mode of occurrence of cuprostibite is as small interstitial grains ($\leq 40\ \mu\text{m}$) in galena-sphalerite matrix (texture 3). Simple gudmundite-galena intergrowth textures have been observed in a small number of small ($\leq 100\ \mu\text{m}$) galena grains. One grain of breithauptite has also been identified. Breithauptite forms the central part of a composite cuprostibite-breithauptite grain. The grain displays an irregular intergrowth between the two phases (Fig. 6). Arsenopyrite is relatively rare and has been observed in association with the gudmundite-bearing assemblages. Native

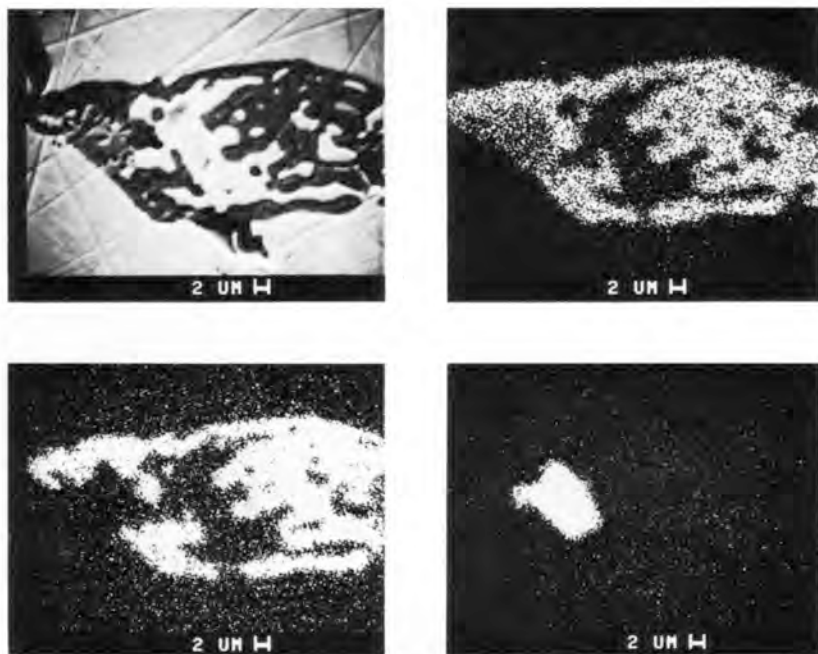


Fig. 4. SEM micrograph (top left) of a break-down texture composed of cuprostibite (dark grey), dyscrasite (medium grey) and galena (light grey), in a large galena grain and X-ray emission images for Sb-radiation (top right), Cu-radiation (bottom left) and Ag-radiation (bottom right) of this area.

silver is still rarer and occurs as corroded monomineralic grains in the gangue mineral matrix or as very small ($\leq 10 \mu\text{m}$) inclusions in dyscrasite.

Electron microprobe analyses of the ore minerals were made with an ARL-SEM-Q instrument. The operating conditions were an acceleration voltage of 20 kV and a probe current of $0.02 \mu\text{A}$. Pure metals were used as standards for Fe, Ni, Cu and Ag, and natural stibnite, galena, sphalerite and arsenopyrite served as standard samples for Sb, S, Pb, Zn and As. Data were reduced using the Magic IV computer program (COLBY 1968). Representative analyses of the Sb-rich minerals and arsenopyrite are summarized in Table 1. The compositional variations observed for the various phases are very small. The composition of cuprostibite was carefully monitored by microprobe analyses of a larger number of grains. The results of these analyses (Table 2) indicate a slight Cu-deficiency in cuprostibite. This deviation from ideal Cu_2Sb -composition could be due to evenly

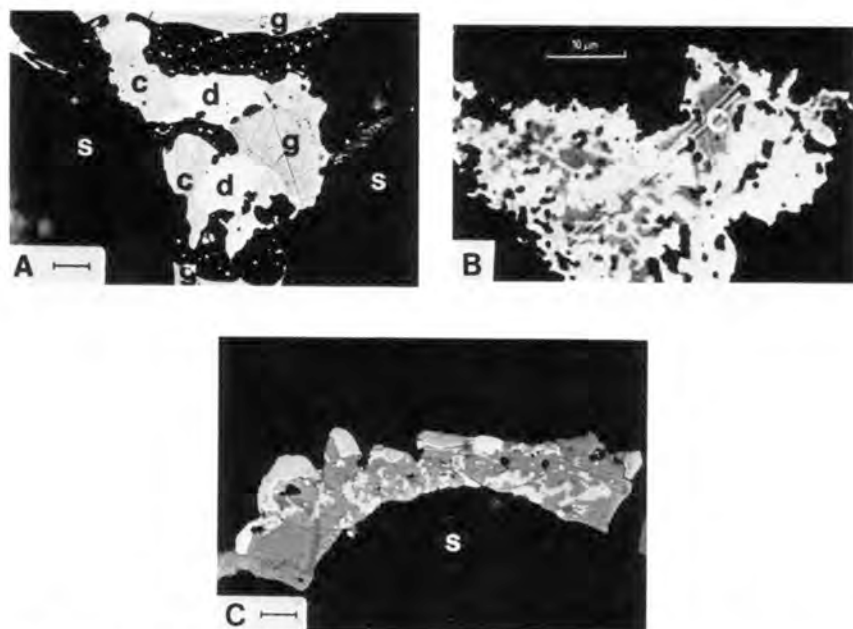


Fig. 5. Microphotographs. Reflected light, oil immersion, one nicol. Scale bar equals $10\ \mu\text{m}$. A: Interstitial grains of cuprostibite (C), dyscrasite (D) and galena between large grains of galena (G) and sphalerite (S). B: Composite grain of dyscrasite (white), gudmundite (light grey), galena (dark grey) and cuprostibite [dark grey (C)] occurring in a microfracture in carbonate-silicate matrix. C: Thin rim of galena (dark grey), cuprostibite (medium grey), gudmundite (light grey, myrmecitic) and dyscrasite (white) on a large sphalerite grain (S).

Table 1. Microprobe analyses of Sb-rich minerals and arsenopyrite from Långsjön.

	1	2	3	4	5	6	7
Pb (wt-%)	0.93	0.22	—	0.03	—	0.58	0.76
Ag	0.06	—	73.10	0.09	—	6.14	0.60
Zn	0.04	0.10	—	0.16	—	1.52	0.09
Cu	0.23	1.61	0.09	0.06	—	34.89	24.12
Co	0.05	0.02	—	—	0.29	0.05	0.05
Ni	0.05	30.91	0.07	0.07	0.06	0.02	0.04
Fe	0.03	0.02	0.02	26.37	34.37	3.63	20.18
Sb	97.56	66.81	24.73	57.82	0.36	28.31	28.13
As	0.37	1.03	0.19	0.30	44.36	0.14	0.08
S	0.04	0.06	0.03	14.95	20.51	23.35	25.01
	99.36	100.78	99.32	99.85	99.95	98.63	99.06

1: Native antimony; 2: Breithauptite; 3: Dyscrasite; 4: Gudmundite; 5: Arsenopyrite; 6: Tetrahedrite; 7: Mineral A (approx. $\text{Cu}_6\text{Fe}_6\text{Sb}_4\text{S}_{13}$).

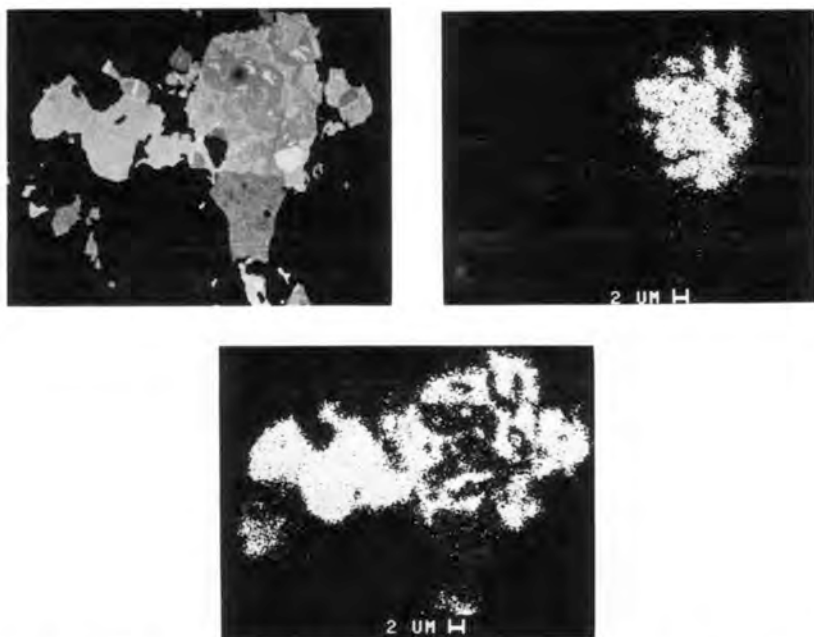


Fig. 6. Microphoto (top left) of a breithauptite-cuprostibite intergrowth and X-ray emission images for Ni-radiation (top right) and Cu-radiation (bottom) of the grain.

Table 2. Composition of cuprostibite from Långsjön based on 31 analyses of 16 different grains.

	Range (wt-%)	Mean (wt-%)	Standard dev.	Atomic proportions
Cu	47.88 – 50.05	49.24	0.62	0.775
Zn	0.00 – 0.88	0.29	0.30	0.004
Ag	0.08 – 0.28	0.16	0.04	0.001
Pb	0.00 – 0.32	0.08	0.07	—
Fe	0.00 – 0.08	0.02	0.02	—
Sb	48.68 – 50.81	49.36	0.47	0.405
As	0.26 – 0.61	0.44	0.08	0.006
S	0.06 – 0.45	0.16	0.08	0.005

Concentrations of Ni and Co were below the detection limits.

Chemical formula: $(\text{Cu}_{1.861}\text{Zn}_{0.011}\text{Ag}_{0.004})(\text{Sb}_{0.974}\text{As}_{0.014}\text{S}_{0.012})$ or $\text{Cu}_{1.876}\text{Sb}$.

distributed and extremely small grains of native antimony in cuprostibite. Another possibility is that cuprostibite is stable within a narrow composition range and that the chemical formula for the mineral should be Cu_{2-x}Sb . In fact, experimental work on the pure Cu-Sb system has shown that the composition of cuprostibite varies from $\text{Cu}_{2.07}\text{Sb}$ to $\text{Cu}_{1.94}\text{Sb}$ (MAVRODIEV & TOLEV 1978). Concentrations of Ag and Sb in galena could not be detected with the electron microprobe.

Discussion

Considering the observed mineral assemblages and their textural characteristics, the ore minerals can be divided into two major groups: (I) sphalerite and galena with inclusions of a variety of Sb-rich minerals, and (II) interstitial and fracture controlled grains of Sb-rich minerals + galena + arsenopyrite.

Regarding the relationship between the mineral assemblages of these two groups, the results obtained in a number of experimental investigations and theoretical approaches on systems containing sulphur, base metals and noble metals are of specific interest especially with respect to conditions of formation (T and a_{S_2}) and crystallization sequence. Although there exists no experimental work describing the phase relations in a system of the chemical complexity characterizing the group II assemblages ($\text{Ag} + \text{Sb} + \text{Cu} + \text{Pb} + \text{Fe} + \text{S} + \text{As}$) a considerable number of studies on ternary and pseudoternary systems of different combinations of these elements have been performed. The results of these studies, when applied to the present mineral assemblages, could be expected to yield information on possible T - a_{S_2} fields for the cuprostibite formation.

The experimental investigation of the system Cu-Sb-S (SKINNER et al. 1972) has shown that cuprostibite is formed under conditions of very low sulphur fugacity and that Cu_2S is the only sulphur-bearing phase with which it can coexist (in the Cu-Sb-S system). This implies that tetrahedrite (treated as $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$) cannot be in equilibrium with Cu_2Sb , which indicates that the tetrahedrite-bearing association I has formed under conditions different from those prevailing during the cuprostibite-formation (association II). The fact that tetrahedrite and the Cu-bearing sulphosalt A are only observed as inclusions in large galena grains, and that they are often partially or completely decomposed even in such protected positions, indicates that these Cu-bearing minerals formed prior to cuprostibite. This observation is further supported by the fact that cuprostibite grains never show features of decomposition or other mineral reactions, irrespective of texture and mineral assemblages.

The existence of uniformly distributed microinclusions of an Ag-Sb rich phase in some galena grains is a feature often observed in Zn-Pb-Ag

mineralizations. These types of inclusions have been interpreted as ex-solutions of Ag-Sb rich phases from galena, with an initial composition on the PbS-AgSbS₂ join, during cooling events (AMCOFF 1976; SANDECKI & AMCOFF 1981). On the basis of results from annealing experiments on such galena grains, it is believed that they have formed at temperatures of 350-400 °C (HALL & CZAMANSKE 1972; SANDECKI & AMCOFF 1981). Assuming that all the silver in the present mineralization was initially dissolved in galena and that this galena was a PbS-AgSbS₂ solid solution phase (galena_{ss}), it is likely to have contained ~ 1 wt-% Ag (see Fig. 2) or ~ 2 mole-% AgSbS₂. According to the experimentally determined phase relations in the system PbS-AgSbS₂ (HODA & CHANG 1975; AMCOFF 1976) a galena_{ss} of this composition must have formed at temperatures above 220 °C. However, the solubility of silver in galena is not just an effect of formation temperature. AMCOFF (1976) has pointed out that if the system is characterized by an Ag/Sb molar ratio which is higher than one and if it contains other elements (e.g. Cu and Fe) that are soluble in galena, then the solubility of Ag in galena will be suppressed. This means, in the present case, that an initial galena_{ss} could have crystallized at temperatures well above 220 °C, as Cu is present in the system and the Ag/Sb ratio is higher than unity.

Apart from the previously discussed mineral inclusions, galena also contains large numbers of minute rounded Sb-mineral grains, which are concentrated in small sharply defined areas. Due to the spatial distribution of these areas they cannot be interpreted as exsolution minerals formed from a chemical homogeneous phase. These textures are more likely to be the result of decomposition, through desulphidation, of unknown Cu-Pb-Ag-Sb and Pb-Sb sulphosalts. The observed cuprostibite + antimony + dyscrasite + galena breakdown textures (Fig. 3 D) could have resulted from decomposition of Ag-bearing bournonite, and the antimony + galena textures (Fig. 3 C) could have been produced through desulphidation of boulangerite. The fact that these breakdown textures are found in the rim-zones of the large galena grains indicates that the formation of Pb-bearing sulphosalts occurred after the initial galena_{ss} formation. Hence, the sulphur fugacity must have been above the boulangerite sulphidation curve at the time of Pb-sulphosalt formation.

The cuprostibite occurring as interstitials and fracture infillings is characterized by small ($\leq 40 \mu\text{m}$) monomineralic grains, which are partially in contact with dyscrasite and grains of galena of approximately equal size. The contact surfaces between the Sb-rich minerals are simple (straight or smoothly rounded), but when in contact with large galena or sphalerite grains, the latter are typically corroded. The textural, mineralogical and chemical simplicity of this assemblage makes it difficult to

deduce its condition of formation. However, the presence of cuprostibite indicates that the prevailing sulphur fugacity was very low. Also, the equal and somewhat larger grain size of the minerals of this assemblage and their internally smooth contact surfaces indicate that they were formed in a relatively extended crystallization period during which the temperature decreased slowly. Furthermore, these geometric observations, combined with the observed corrosion occurring at the larger galena and sphalerite grains in contact with the Sb-rich minerals, suggest that these minerals crystallized from a Cu-Ag-Sb-rich fluid rather than through direct (in situ) breakdown of pre-existing sulphosalts.

The composite, mineralogically and/or texturally complex grains of Sb-rich minerals and galena, which occur along microfractures in carbonate-silicate matrix or as rims on sphalerite, are more informative with respect to their conditions of formation. Gudmundite, which is constantly observed in this assemblage, has a maximum thermal stability of 280 °C (CLARK 1966). This implies that the assemblage has crystallized at temperatures below 280 °C. Furthermore, gudmundite is only stable at low sulphur fugacities (BARTON 1971). Small grains ($\leq 50 \mu\text{m}$) of arsenopyrite have occasionally been observed in the gudmundite-bearing assemblages. These arsenopyrites are chemically homogeneous and they contain only minor amounts of Co and Ni (analysis 5 in Table 1). Application of the arsenopyrite geothermometer (KRETSCHMAR & SCOTT 1976) to these arsenopyrites, in combination with the sulphidation curve for stibnite (CRAIG & BARTON 1973), yields a temperature of formation of 360 °C at a $\log a_{\text{S}_2}$ of -10.4 for this assemblage (Fig. 7). The choice of the stibnite sulphidation curve is obviously an overestimation of the sulphur fugacity, as the obtained temperature of formation exceeds the thermal stability for gudmundite. However, there exist no experimental data on the sulphidation curve for gudmundite or cuprostibite. Lacking these data a more likely estimation of the conditions of formation for the gudmundite-bearing assemblage may be obtained by combining the upper thermal stability of gudmundite (280 °C) with the arsenopyrite geothermometry. These combined data yield a $\log a_{\text{S}_2}$ of -14.4. The persistent occurrence of graphic intergrowths on a very small scale indicates that the minerals of this assemblage have formed during a stage of rapid cooling.

Finally, the occurrence of native silver, native antimony and dyscrasite in the present mineralization cannot be explained by any model which assumes a precipitation of these phases from a common source. The occurrence of monomineralic and corroded grains of silver is probably an effect of desulphidation of pre-existing silversulphides. This decomposition must have taken place previous to the initial cuprostibite forming stages.

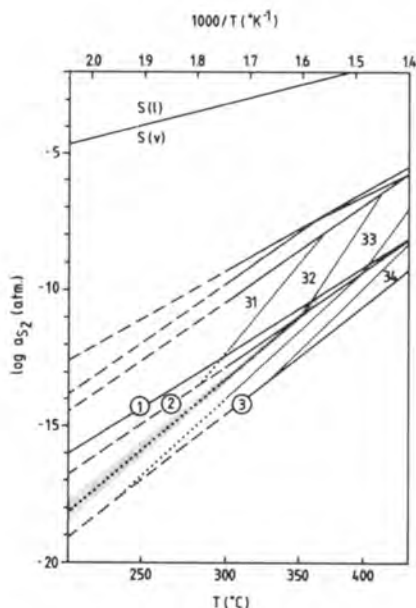


Fig. 7. Activity of S_2 -temperature projection of the stability field of arsenopyrite below 400 °C, contoured in atomic-% of arsenic (nos 31 to 34). Solid lines down to 300 °C are after KRETSCHMAR & SCOTT (1976), and dotted and dashed lines represent extrapolations of their data to lower temperatures. Nos 1, 2 and 3 designate the reactions $2/3 \text{Sb}_2\text{S}_3 = 4/3 \text{Sb} + \text{S}_2$ (CRAIG & BARTON 1973), $2 \text{FeAsS} + 2 \text{As} = 2 \text{FeAs}_2 + \text{S}_2$ (KRETSCHMAR & SCOTT 1976) and $4 \text{FeAsS} = 2 \text{FeS} + 2 \text{FeAs}_2 + \text{S}_2$ (KRETSCHMAR & SCOTT 1976), respectively. Shaded area is a $\log a_{S_2}$ - T projection for the compositions of the present arsenopyrites.

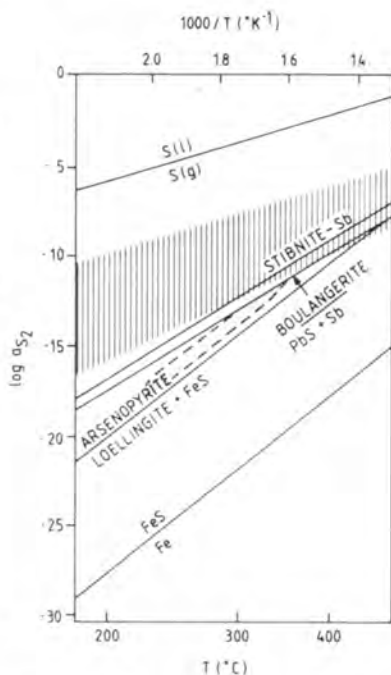


Fig. 8. The conditions for cuprostibite formation at the Långsjön locality (shaded area) in relation to some sulphide reaction curves (CRAIG & BARTON 1973; KRETSCHMAR & SCOTT 1976) and to the main-line (hatched area) of ore mineral forming environments (BARTON 1973).

Conclusions

When related to experimentally derived parameters, the observed ore mineral assemblages and their characteristic textures can be interpreted by a four-step model:

- (1) Crystallization of sphalerite and PbS-AgSbS_2 solid solution phases (galena_{ss}) at relatively high temperature and sulphur fugacity. The galena_{ss} also contained minor amounts of Cu.
- (2) Slow cooling rate produces exsolution bodies of Ag-Sb rich phases and Cu-bearing sulphosalts in existing galena_{ss}. Continuing crystallization produces Cu-Ag-Sb poor galena and a variety of Sb-Pb rich sulphosalts. The sulphur fugacity during this stage was above the Ag_2S sulphidation curve.
- (3) Slowly decreasing temperature and decreasing sulphur fugacity causes breakdown of existing sulphosalts and silversulphides. The decomposition of these phases, when occurring in carbonate-silicate matrix or in other less protected positions, gives rise to local metal rich fluids. The prevailing sulphur fugacity was below the boulangerite sulphidation curve (CRAIG & BARTON 1973).
- (4) Decreasing temperature, with a rapid cooling at the end of the stage, combined with very low sulphur fugacities leads to the crystallization of dyscrasite + cuprostibite + galena + antimony and later gudmundite-bearing assemblages from the locally derived fluids. At the end of the stage the temperature was below 280 °C and the $\log a_{\text{S}_2}$ was below -14.4.

The sulphur fugacity during the cuprostibite forming stages was obviously considerably lower than what is normal during sulphide ore forming events (Fig. 8).

The major part of the ore mineral forming event at the present locality took place after the peak of the regional metamorphism. However, this conclusion does not rule out a premetamorphic or primary concentration of Zn, Pb and Ag in the dolomitic host rock.

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