

Mineralogical and preliminary fluid inclusion studies of lead-antimony mineralisation in north Cornwall

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Antimony mineralisation in the Port Isaac-St Endellion district of north Cornwall has been investigated in order to clarify its relationship to regional models of polymetallic mineralisation. The mineralisation is stratabound and confined to Middle Devonian to Lower Carboniferous volcano-sedimentary basinal successions. Minor shear structures and veining developed during late Variscan deformation at a high angle to regional cleavage, and these also contain small deposits of lead-antimony sulphosalt minerals. The minerals include species in the plagiomite, jamesonite and boulangerite series associated with replacement carbonate phases and small quantities of gold. Fluid inclusion studies and the discrimination of equilibrium assemblages within sulphide and sulphosalt assemblages indicate a multistage evolution for the deposits. Thermometric data show that the fluids contain variable proportions of H₂O, CO₂ and CH₄. Minimum temperatures for Sb-As- (Au) mineralisation associated with highly deformed quartz microfabrics, (T_H=280-315°C, salinity 3-4wt% NaCl equivs) contrast to those for Pb-Zn-Ag mineralisation associated with relatively unstrained quartz (T_H=180-210°C, salinity 0.5-6wt% NaCl equivs). This may denote more than one mineralising event characterised by differences in structural style. Provisional results indicate that Sb-(Au) mineralisation was controlled by H₂O-CO₂ fluid immiscibility. The evidence allows a new interpretation for antimony mineralisation in south-west England to be proposed.

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Introduction

Antimony deposits in north Cornwall comprise one of largest group of occurrences for that element in the British Isles (Dewey 1920). They are mostly confined to the Trevoise and Jackets Point Slate Formations (Selwood and Thomas 1986) in the Port Isaac district, and are exposed along the coastline and in shallow underground workings (Fig. 1). In addition to antimony these mines also produced subordinate quantities of lead, silver and minor gold (Dines 1956). Mining activity other than for antimony included lead workings at Pentire, Treburgett and Tregardock, where lead-antimony sulphosalt associations are also identified in this work. Mining records for the district date from 1681 (Borlase 1758) with intermittent production continuing until 1947 (Dines 1956).

Scattered workings for antimony are present in east Cornwall, south Devon and north Devon within Middle Devonian to Lower Carboniferous basinal sedimentary and volcanic rocks. Silver-lead-antimony sulphosalts are present in Pb-Zn-Ag deposits within the Lester Slates and Sandstones at Coombe Martin, north Devon. Scrivener and Bennett (1980) determined that these deposits were stratiform in character which demonstrates that the occurrence of antimony mineralisation in the region is not restricted to discordant vein type structures. Such concentrations of sulphides may be important in evaluating source characteristics for late Variscan lead-antimony-gold bearing structures.

Material was collected from coastal exposures, underground workings and abandoned mine dump waste and studied using a combination of petrographic, electron-microprobe and fluid inclusion analytical techniques.

Mineral assemblages and paragenetic relationships

Sulphide bearing vein systems are recognised in the areas around Bounds Cliff, Pentire and Portquin (Fig. 1). These are subdivided into three types in which mineral assemblages commonly display a predominance of one component and which may be classified as arsenopyrite dominated (Type 1), galena (+ base metal sulphide) dominated (Type 2), and lead-antimony sulphosalt dominated (Type 3) (Fig. 2). This subdivision is supported by paragenetic studies and by preliminary thermometric data from fluid inclusions. Quartz, chlorite and carbonate phases are consistently present as gangue minerals: fluorite has not been recorded.

Bounds Cliff vein system

Vein systems at Bounds Cliff (SX 02308140) extend southwards for a distance of 5km and account for the highest concentrations of antimony mineralisation in the study area. Type 1 and Type 3 assemblages are represented, both with minor gold associations. Arsenopyrite-pyrite-marcasite mineralisation is representative of Type 1 assemblages which are zonally arranged about linear fractures. Subsequent reworking during later deformation is demonstrated by brecciated pyrite and arsenopyrite enclosed by chalcopyrite-sphalerite-galena and lead-antimony sulphosalts. Stibnite is less common but is associated with other antimony bearing minerals. Native gold exists in association with various sulphosalts and pyrite, and is estimated from modal analyses to be present in concentrations between 2-20ppm. A narrow compositional range was determined between 90wt%Au-10wt%Ag to 92wt%Au-8wt%Ag (CJS).

Sulphosalt dominated assemblages consist of tetrahedrite (CuAg)₁₀ (FeZn)₂ (SbAs)₄S₁₃, jamesonite (Pb₅FeSb₆S₁₄), bourmonite (CuPbSbS₃), boulangerite (Pb₅Sb₄S₁₁) and plagiomite (Pb₅Sb₈S₁₇) intergrowths developed along intergrain fractures and at vein intersections. In addition, intragrain fractures in quartz and carbonate phases contain fibrous crystals of jamesonite and boulangerite (Fig. 3a) showing that the lead-antimony sulphosalt minerals typify the latest stages of the paragenesis which exhibits complex textural and mineralogical relationships. Tetrahedrite is partially replaced by

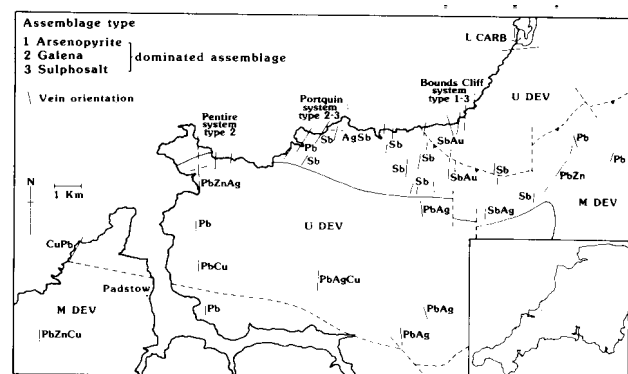


Figure 1. Pb-Ag-Sb mineralisation, north Cornwall.

Dominant mineral present	Association	Inclusion/intergrowth
Galena	Cp Sp Sid Qtz	Ag tetrahedrite bournonite meneghinite polybasite/antimopearcite
Arsenopyrite	Py Qtz	Infrequent fracture fillings of Cp-Aur tetrahedrite
Sulphosalt	Other sulphosalts (Pb-Sb) Sid Qtz	Lamellar intergrowths of bournonite-jamesonite. Reaction fringes of plagioclase Fibrous crystals of jamesonite-boulangerite. Inclusions of meneghinite-bournonite-jamesonite in galena. Inclusion of bournonite in tetrahedrite.
Legend Qtz Quartz Sid Siderite Py Pyrite Cp Chalcopyrite Sp Sphalerite		

Figure 2. Vein assemblages, north Cornwall.

bournonite (+ meneghinite $Pb_{13}CuSb_7S_{24}$) and typically contains lower silver contents (<4wt% Ag) compared with tetrahedrite from the Pentire vein system. Colour variations in reflected light reveal wider variations in silver concentration (Ramdohr 1980), exemplified by tetrahedrite from Treore Mine (SX 02058005) containing up to 21wt% Ag. Little variation in chemistry was found in bournonite, regardless of its textural position in respect to other sulphosalts. Microprobe analyses of jamesonite ($Pb_{4.2}FeSb_{6.2}S_{13.6}$) from Bounds Cliff showed trace amounts of copper and bismuth. Plagioclase is developed as fringes on jamesonite; a recalculated formula gives $Pb_{5.3}(Sb_{0.99}Bi_{0.01})_{8.4}S_{16.3}$ and an Sb/Pb ratio of 1.58, which compares well with the ideal formula ($Pb_5Sb_8S_{17}$) and ratio (1.60), supporting the identification. The paragenetic sequence and microprobe analyses of the lead-antimony sulphosalts (boulangerite + plagioclase), those containing essential extra cations (bournonite + jamesonite) and tetrahedrite (\pm argentine varieties), suggest a partial reaction/replacement sequence characterised by coeval reductions in Cu/Sb and Pb/Sb ratios (Fig. 4).

Pentire vein system

Typical examples of Type 2 assemblages are developed as wallrock impregnations, within minor brecciated zones, and as vein fillings through weakly chloritised metabasite and grey-green metapelites in the underground workings of Pentire Mine (SW 94228112). The paragenesis (Fig. 3b) features contemporaneous deposition of sphalerite and chalcopyrite followed by galena-carbonate mineralisation in dilatant sites within a quartz framework. A few samples of galena show recrystallised textures and have evidently been deformed, indicating late textural restructuring. No significant concentrations of antimony or silver were detected in galena; recalculated analyses indicate approximate stoichiometry (PbS to $Pb_{1.1}S_{0.9}$). Silver-bearing minerals occur as intergrowths in galena, commonly as unorientated rounded or irregular shaped blebs interpreted to be of exsolution origin from solubility data (Amcoff 1976; Hoda and Chang 1975); tetrahedrite in this association was found to contain up to 23wt% Ag. Recalculation gives the formula, $(Cu_{0.6}Ag_{0.4})_{10.2}(Fe_{0.6}Zn_{0.3})_{18}(Sb_{0.95}As_{0.05})_{4.1}S_{12.9}$ for the Pentire specimen. Detailed studies have found that many inclusions comprise two discrete sulphosalt types, in which tetrahedrite is commonly associated with polybasite (or ?antimopearcite). These silver bearing minerals are volumetrically important and contribute largely to the high silver values reported in analyses of galena from north Cornwall (Dines 1956). Argentinian tetrahedrite (freibergite), polybasite, and rarely bournonite may coexist as intergrowths; nucleation evidently showing some preference for sites adjacent to chalcopyrite. Abundant sphalerite may occur in association, or forms monomineralic veins (<30mm width). Pyrite, (arsenopyrite?), tetrahedrite and fibrous needles of jamesonite are also present as primary phases.

East of Pentire Mine (SW 94608020), veins containing a jamesonite-bournonite assemblage are associated with weakly developed sphalerite-galena mineralisation.

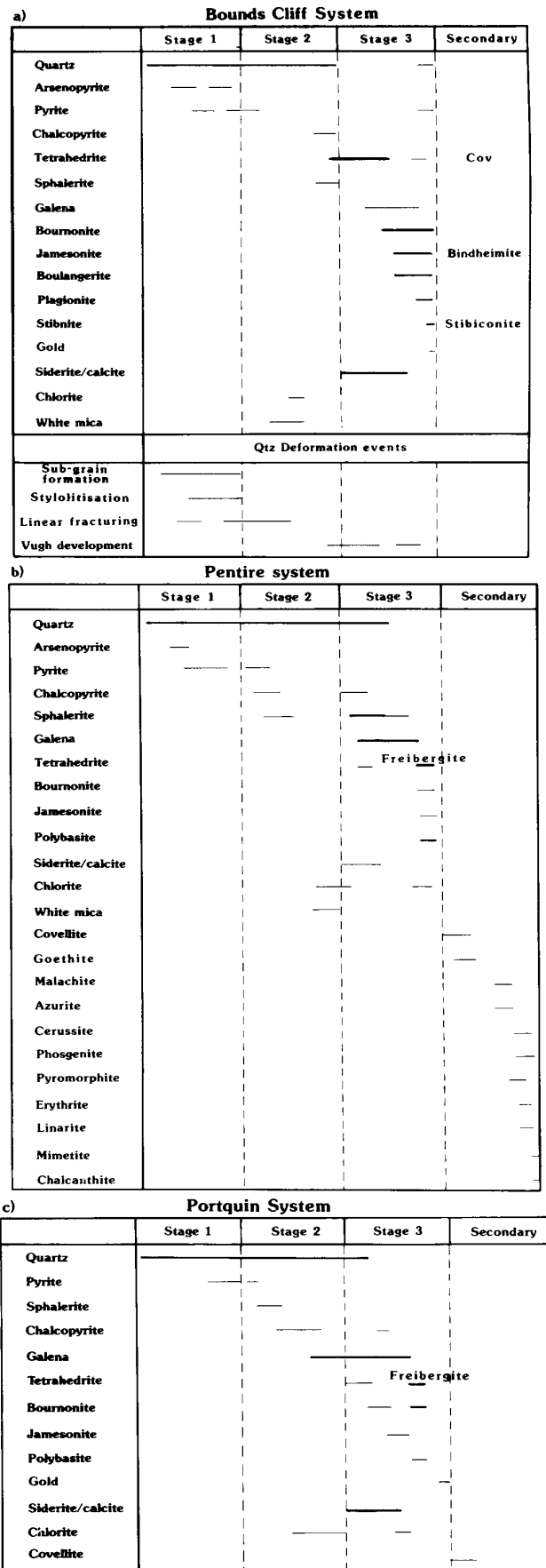


Figure 3. Paragenetic sequence in north Cornwall vein systems.

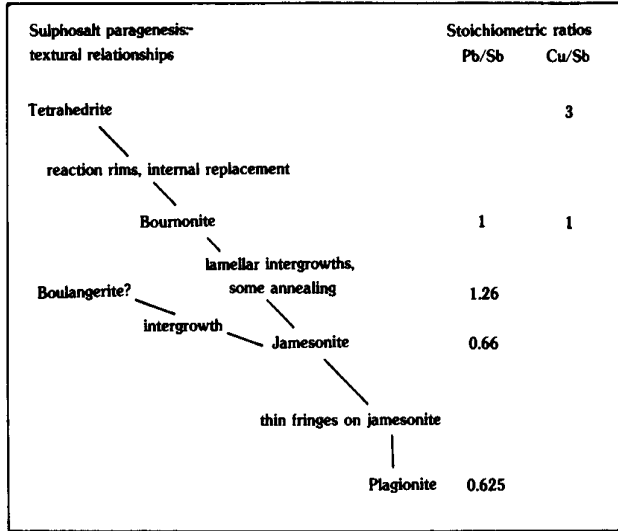


Figure 4. Cu/Sb and Pb/Sb ratio variation in sulphosalt paragenesis.

Portquin vein system

At Portquin (SW 96198058), and Reedy Cliff (SW 97908107), Type 2 and Type 3 assemblages respectively were investigated (Fig. 3c).

In Type 2 veins, lead-antimony sulphosalts occur as intergrowths in galena, however bournonite (+ meneghinite?) intergrowths predominate over argentian tetrahedrite. A consistent association with chalcopyrite was found in galena containing abundant bournonite and tetrahedrite inclusions. Sulphide zonation was developed due to the partial replacement of sphalerite-chalcopyrite by internal zones of galena-sulphosalt association. Pyrite, jamesonite and trace quantities of gold located in tetrahedrite alteration zones were also recorded.

The sulphosalt mineralisation here shares similar features to those in the Bounds Cliff vein system, and is closely related in time to the overprinting of earlier textures by siderite-calcite assemblages. Siderite (FeCO₃) is the predominant carbonate, containing significant Mg and up to 4wt% Mn. It occurs as a pervasive replacement, particularly in the Portquin vein system in which pseudomorphs after quartz are concurrent with extensional modifications to the vein fabric. Textural equilibrium is only recognised between carbonate and lead bearing phases, mostly in dilatant sites occupied by sulphosaltsiderite or galena-siderite associations.

Structural considerations

Preliminary studies on the vein structures suggest that sulphide deposition occurred at various times through a protracted series of deformation events.

Vein geometry indicates that dilation occurred several times in response to brittle shear deformation and so may be used as a strain marker. En-echelon vein arrays have been mapped as parallel-sided, sub-vertical zones transecting cleavage and folds. Individual veins may be linear or sigmoidal and show gently tapering, branched terminations. They are traversed by later, generally smaller veins, inclined at acute angles and representing subsequent shear increments in the deformation.

The strain histories demonstrated by vein fabrics are considered an important factor in understanding the mineralising processes. Various stages of quartz recrystallisation, sub-grain formation, stylolitisation and vugh development have been recognised, suggesting isochemical remobilisation during vein growth. Quartz fabrics associated with Type 1 and Type 3 assemblages are the result of repeated shearing; evidently allowing textural modification to control later sulphide deposition. Mosaic textured domains of recrystallised origin are associated with generations of highly strained quartz. Later quartz generations have retained relatively unstrained fabrics and are restricted to dilatant sites. It may be significant in view of the thermometric data (see below) that textures from the Pentire vein system do not show evidence of such intense deformation.

Whilst these processes have not been quantified, it is recognised that such features indicate remobilisation of components through the vein; that the vein has remained sealed at various stages of its development, and that textural re-adjustment or re-equilibration has occurred in response to strain increments.

Arsenopyrite geothermometry

Arsenopyrite compositions may reflect formation temperatures, provided they belong to an equilibrium sulphur buffering assemblage and satisfy stringent criteria (Kretschmar and Scott 1976). Only well formed grains in contact with coexisting phases are recognised as constituting a buffered assemblage in this study. Microprobe grain traverses indicated acceptable homogeneity. Table 1 shows that the combined minor element contents of arsenopyrite are <0.12wt%, and that analyses sum between 99.32wt%. An arsenopyrite standard calibrated with Asp 200 was used as a standard for Fe, As and S. Repeat analyses were performed and were in good agreement with original runs. Calculated compositions fall within the extrapolated region (fig. 4 Kretschmar and Scott 1976) and conform to FeAs_{0.9}S_{1.1}, (As/S = 0.82). An estimated temperature for arsenopyrite formation was calculated ranging between 310°C and 340°C.

Table 1. Arsenopyrite compositions.

	wt.%	at.%	wt.%	at.%	wt.%	at.%	wt.%	at.%	wt.%	at.%
S	21.87	35.93	21.91	36.1	22.19	36.34	22.16	36.52	22.61	36.91
Fe	35.43	33.41	35.68	33.75	35.86	33.72	35.38	33.47	36.09	33.82
Co	0.07	0.06	0.1	0.09	0.08	0.07	0.07	0.06	0.05	0.04
As	43.51	30.59	42.55	30	42.61	29.86	42.45	29.93	41.82	29.21
Ag	n.d.		0.05	0.03	n.d.		n.d.		n.d.	
Sb	n.d.		0.06	0.03	n.d.		0.04	0.02	0.04	0.02
	100.9	99.99	100.4	100	100.7	99.99	100.1	100	100.6	100

The abstracted use of the arsenopyrite geothermometer without additional independent thermometric data is discredited (Sharp *et al.* 1985); but the fluid inclusion microthermometry detailed below supports these findings and the applicability of the geothermometer may be verified in the present study.

Preliminary fluid inclusion studies

Microthermometric data were obtained from samples of quartz vein material from the Bounds Cliff and Pentire vein systems. Fluid inclusion preservation in highly strained quartz sampled from Bounds Cliff is poor; primary inclusions are small (c.10-15µm) and commonly masked by numerous secondary inclusions. This contrasts with the relatively unstrained quartz fabrics from the Pentire vein system. In addition, samples from Buttspill mine (SX 43756775), in the Tamar Valley, were studied (RCS) to provide a comparison with typical 'crosscourse' mineralisation.

Two types of inclusion were recognised in the Pentire and Bounds Cliff vein systems:- aqueous (A-type) and carbonaceous (C-type). Ctype inclusions contain CO₂ as the major non-aqueous phase together with constant or variable proportions of an aqueous phase, and are abundant in the Bounds Cliff vein system.

Preliminary thermometric analyses are shown in Figs 5 and 6. T_H, data indicate minimum temperatures of fluid trapping and are uncorrected for pressure.

Pentire vein system

The textural relationship of quartz containing A-type inclusions suggests that the contained fluids were trapped immediately prior to or simultaneously with galena-sphalerite mineralisation. Homogenisation temperatures define two groups; at T_H = 280°C and at T_H = 180-210°C. The higher value may correlate with the T_H range observed for C-type inclusions from the Bounds Cliff vein system (see below). Final ice melting temperatures indicate salinities in the range from 0.5-6wt% NaCl equivs and 4wt% NaCl equivs for the lower and

higher temperature groups respectively. All A-type inclusions exhibit eutectic melting temperatures consistent with predominantly H₂O - NaCl fluid compositions.

Bounds Cliff vein system

Fluids associated with arsenopyrite-pyrite and later lead-antimony sulphosalt assemblages typically contain C-type inclusions. These homogenise into the liquid state on heating (T_H = 280-315°C). Variable H₂O-CO₂ ratios in adjacent inclusions with similar homogenisation temperatures suggest entrapment during phase separation. Partial homogenisation of C-type inclusions was always to the liquid state (CO₂ L + V → L), and occurs over the temperature range 22.2-30.2°C, indicating CO₂ densities in the range of 0.6-0.75 gm⁻³ (Shepherd *et al.* 1985). Slightly lowered solid CO₂ melting temperatures and partial homogenisation temperatures of the liquid CO₂ phase suggest that minor proportions of CH₄ may contribute to the fluid (Shepherd *et al.* 1985). The temperatures of final clathrate melting indicate a salinity of approximately 4wt% NaCl equivalents for C-type inclusions. The T_H- salinity plot (Fig. 6), compares the Pentire and Bounds Cliff vein systems with typical 'crosscourse' fluid chemistry (CaCl₂ - NaCl - H₂O). Lower salinities and higher homogenisation temperatures characterise fluids associated with the north Cornwall mineralisation.

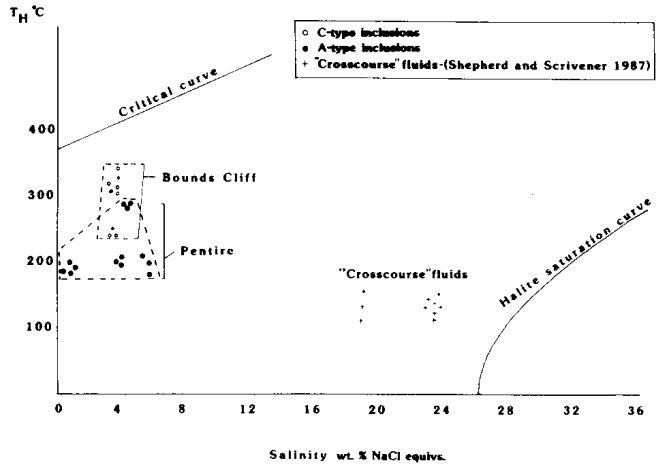
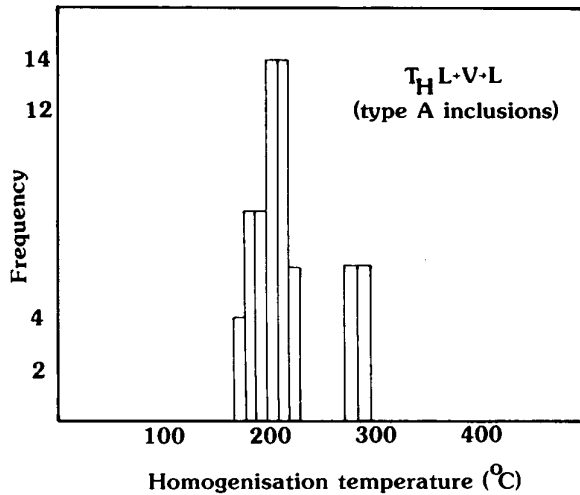


Figure 6. T_H - Salinity plot showing comparative data from north Cornwall and 'crosscourse' fluids.

Pentire system Pb-Zn-Ag



Bounds Cliff system Sb-Au

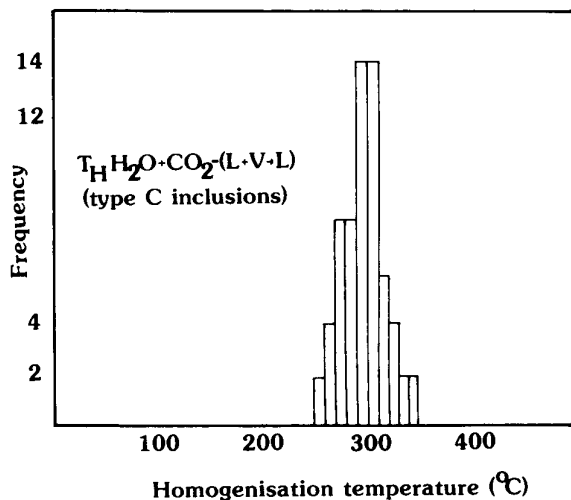


Figure 5. Histograms of fluid inclusion homogenisation temperatures.

Discussion and conclusions

Antimony mineralisation in north Cornwall is principally developed within the Bounds Cliff vein system. Lead-antimony sulphosalt assemblages typify the latter stages of mineralisation and evidently display greater affinity with arsenopyrite-pyrite-dominated than galena-dominated assemblages. It is unlikely that antimony was remobilised from minor element concentrations in earlier sulphides. The association with carbonate phases and minor quartz implies a change in fluid chemistry and disequilibrium with earlier phases, allowing the deposition of sulphides and sulphosalts containing lead to be favoured. T_H for inclusions from Type 2 assemblages in the Pentire vein system indicate minimum trapping temperatures of 210°C, while higher temperatures (T_H=280-315°C) are indicated for mineralisation in the Bounds Cliff vein system. The latter temperature estimate gives an approximate correlation with arsenopyrite geothermometry. This preliminary study indicates differences in mineralising fluid, assemblages and vein microstructures between the Pentire and Bounds Cliff vein systems, which require further investigation to determine their precise temporal relationships.

The abundance of non-aqueous volatiles (CO₂ ± CH₄) and low salinities for fluids associated with mineralisation in the Port Isaac St Endellion district is in direct contrast to 'crosscourse' fluids which are regionally characterised by H₂O - NaCl - CaCl₂ composition (Shepherd and Scrivener 1987) and which are considered to be basinal brines of Permo-Triassic age. CaCl₂ has not been detected in samples from the Pentire or Bounds Cliff vein systems, and suggests a different genetic model for the north Cornwall mineralisation. Present modelling of these deposits employs the derivation of fluid components through metamorphic processes from which the deposition of sulphide-sulphosalt assemblages occurred at various stages in the development of minor shear structures during late Variscan deformation. Earlier studies (Hosking 1964; Edwards 1976) have suggested that the antimony mineralisation in north Cornwall is a low temperature expression of the granite-related ore deposition. It is considered here that this mineralisation was generated earlier, during Variscan thrusting and shear fracturing.

The importance of CO₂ rich fluids to Sb-Au type mineralisation is widely recognised (eg. Bril 1982; Goldfarb 1986). Wallrock-fluid interactions (Seward 1973) can be important in achieving favourable conditions for mineralisation and may explain selective lead-antimony sulphosalt-gold deposition at vein intersections (higher rock/ fluid ratio). The variable/heterogeneous trapping of fluids (ie. variable phase ratios in C-type inclusions) suggest fluid unmixing at T_H = 315°C. Deposition of gold and sulphosalts may then occur by the destabilisation of gold complexes (Naden and Shepherd 1989) and those which control antimony solubility.

Type 3 sulphosalt parageneses are characterised by decreasing Pb and increasing Sb/Cu+Ag. This is commonly identified in studies of sulphosalt mineralisation (Moelo 1983). The sequence is a function of increasing chemical potential of more volatile Sb₂S₃ and its progressive

gressive enrichment in later mineral species (McKinstry and Kennedy 1957). More specifically the sequence could be a typomorphic feature of particular hydrothermal deposits (Mozgova *et al.* 1969). According to Jankovic *et al.* (1977), complex sulphosalts containing additional cations will be deposited first regardless of Pb/Sb ratio. The identification of early bournonite and jamesonite in the Bounds Cliff vein system is in agreement with this model. The importance of reaction/replacement processes has, however remained unclear, yet textural analyses suggest that these ought to be considered in paragenetic studies. A consequence of replacement and breakdown reactions in lead-antimony sulphosalts has been the formation of exsolved phases and further PbS solid solution (Czamanske and Hall 1975; Moelo *et al.* 1988).

Correlation of antimony mineralisation with volcanic sequences in north Cornwall (Edwards 1976) can now be refined using initial results of the BGS remapping programme of the Trevoze Head and Camelford district (1986-1990). It is recognised that lead- and antimony-bearing vein systems are located around centres of Middle to Upper Devonian volcanism at Pentire (SW 93608028), Trelill (SX 04407800) and St Endellion (SW 99757867). Source lithologies for antimony and gold are cited as basaltic lavas and tuffs (Edwards 1976), however interflow metapelites are found to contain up to 100ppm Sb. It is suggested that mobilisation of antimony and gold was by fluids capable of transporting components necessary for carbonate deposition. Important species in such fluids include CO_2 , CO_3^{2-} , H_2CO_3 , HCO_3^- and H^+ (Fournier 1985). Carbonate mineral chemistry reflects the low CaCO_3 but higher (FeMg) CO_3 and Mn (up to 8wt%) of host metapelites, indicating a possible exhalative process of enrichment. The reliance on host lithologies of favourable chemistry may explain the restricted distribution of lead-antimony deposits to basinal sequences with volumetrically significant volcanic contributions in the region. Extensional volcano-sedimentary basins with exhalative contributions to sedimentation are cited as source terrain characteristics for particular types of gold mineralisation (Simpson *et al.* 1989). It is suggested that such a model may apply to fault-controlled alkaline basaltic volcanicity within sequences enriched in metalliferous elements in north Cornwall. The subsequent mobilisation of these metals through fluid-dominated processes was effective in concentrating antimony and gold. These fluids were focussed through NNE-SSW shear structures such as the Bounds Cliff vein system during late Variscan deformation.

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