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A FLUID INCLUSION STUDY OF QUARTZ, CALCITE & BARYTE FROM DIFFERENT LOCALITIES OF CARBONATE-HOSTED GOLD AND BASE METAL MINERALIZATION IN THE WESTERN RHODOPE AND SERBOMACEDONIAN MASSIFS, MACEDONIA, GREECE

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ABSTRACT

Several deposits of carbonate hosted base metal and gold mineralization in the Western Rhodope and Serbomacedonian massif were sampled for fluid inclusion microthermometry. Five different types of fluid inclusion were identified. It is concluded that the deposits are spatially or genetically related to mid-Tertiary calc-alkaline magmatism and tectonism. The following styles of mineralization are identified: (i) Skarn mineralization, (ii) porphyry style, (iii) mesothermal-metamorphic shear zone, (iv) Epithermal systems and (v) MVT style mineralization. Processes responsible for ore deposition include: boiling/phase separation, and reaction of the mineralizing fluid with the host marbles.

INTRODUCTION

Fluid inclusion analyses have been completed on a suite of auriferous, base metal and barren veins from the Western Rhodope (Palia Kavala, Thassos and Pangaeon) and the Serbomacedonian massif (Madem Lakkos, Piavitsa, Stratoniki, Zepko and Varvara), Macedonia, Greece. Only summary data are presented here, more detailed descriptions are given in the following unpublished Institute of Geology and Mineral Exploration Open File Reports: Nimfopoulos *et al.* (1993), Constantinidou (1993), Eliopoulos (1993) and Dimitroula (1993). The fluid inclusion studies were undertaken in conjunction with a more general metallogenic study of the region (Hellingwerf *et al.*, 1994) which provides a framework within which the fluid inclusion data can be interpreted. The location of the study area is illustrated in Figure 1.

GENERAL GEOLOGICAL SETTING

The Rhodope Massif comprises a sequence of intermixed para- and orthogneisses, schists and marbles. Rb-Sr geochronology of an intrusive granitoid complex within the massif gives and age of 342±27Ma, which indicates that the massif is at least Lower Carboniferous in age (Moorbath and Zagorcev, 1983). The rocks have been divided into two main units:

1) A lower unit of basement gneisses intercalated with mica schists and rare amphibolites (local amphibolite-eclogite facies metamorphism, [Mboskos *et al.*, 1990]).

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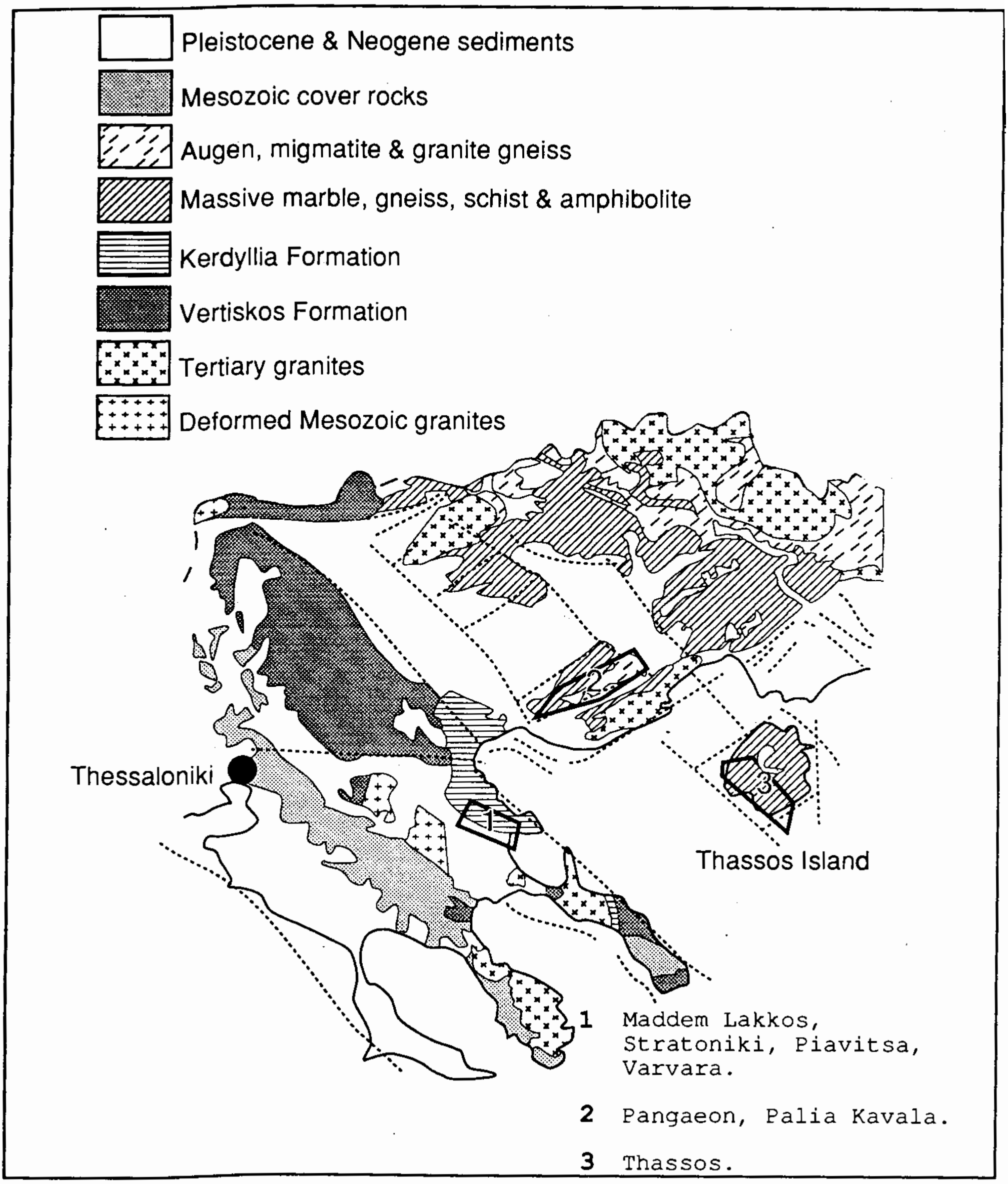

Fig. 1: Geological map of the Chalkidiki region showing the location of the variou deposits studied (Modified from Constantinides, 1993; Bornovas and Rondoyianni 1983).

2) An upper carbonate unit comprising marbles, calcareous schists and minor cherts (middle-upper amphibolite metamorphism, [Mboskos *et al.* (1990)]).

The Serbomacedonian massif comprises similar rock types to those in the Rhodope. Again the age is uncertain, but zircon U/Pb dating of meta-rhyolites suggests a Cambrian age (Frei, 1989). The massif contains two major stratigraphical units (Kockel *et al.*, 1977):

1) The Kerdyllia formation which comprises biotite gneisses interbedded

with hornblende gneiss, amphibolite, and local marbles. This is thought to represent an original volcano-sedimentary sequence (Dimitriadis, 1974; Fournaraki, 1981).

2) The Vertiskos formation overlies the Kerdyllia formation and consists mainly of two mica gneisses quartz feldspar gneiss and ortho-amphibolites. Metamorphosed basic and ultra-basic rocks commonly occur at the contact between the two formations.

The contact between the two systems is in part gradational but the contact relationship in usually obscured by the major Stratoni-Varvara fault system. The rocks of the Serbomacedonian massif were metamorphosed to almandine-amphibolite facies with partial anatexis (Dimitriadis, 1974). K-Ar radiometric age determinations indicate that this metamorphism took place between the Upper Jurassic (Kalogeropoulos *et al.*, 1990) and the Oligocene (Harre *et al.*, 1968). This period of amphibolite facies metamorphism was followed by at least one retrogressive phase.

The Serbomacedonian and Rhodope massifs are cut by major low angle thrusts and extension faults, which are superimposed on a network of large scale horst-graben structures (cf. Frei, 1992, for a detailed discussion on plate tectonic reconstructions in the region).

During the Tertiary there was widespread volcanic activity in the Balkan and northern Aegean region (Fytikas *et al.*, 1980). This volcanism was accompanied by widespread calc-alkaline plutonic magmatism with the development of large scale acidic intrusives in the form of monzodiorites, granodiorites and granites. Locally some of these granites are accompanied by skarn mineralization (e.g. Madem Lakkos). The plutonic intrusives are generally Eocene to Miocene in age (Nimfopoulos 1988; Kockel *et al.* 1977). Associated with these plutons are sub-volcanic stocks and dykes, this suite is mainly located in the Vertiskos formation. Most of the acidic porphyry suite are accompanied to a greater or lesser degree by metalliferous mineralization (e.g. Skouries syenite porphyry, Eliopoulos and Economou-Eliopoulos, 1991). Apart from the Tertiary magmatism and associated mineralization, there is a north-west trending belt of Jurassic (136-155Ma) granites which were probably emplaced during a period of late Jurassic, upper greenschist facies metamorphism. The granites themselves are commonly highly deformed and are generally unmineralized.

STYLES OF MINERALIZATION

In the Serbomacedonian and Rhodope massifs base and precious metal mineralization, mostly small scale is widespread. However, the polymetallic Olympias and Madem Lakkos mines show that there is economic potential in the region. Mineralization appears to be related to Tertiary igneous and tectonic activity, and several different styles of mineralization have been identified. The more important of these are listed below:

1) Vein and replacement polymetallic sulphide deposits within marbles (e.g. Olympias: Kalogeropoulos *et al.*, 1989).
2) Porphyry copper deposits (e.g. Skouries: Eliopoulos and Economou-Eliopoulos, 1991; Tarkian *et al.* 1991).
3) Skarns with magnetite and various metal sulphides hosted in marbles and carbonates, intruded by Oligocene granitoids(e.g. Madem Lakkos: Nebel *et al.* 1991; Gilg 1993).
4) Gold-bearing veins, with quartz, carbonates, and sulphides (e.g. Palia Kavala, Pangaeon: Spathi *et al.*1982; Arvanitides *et al.* 1989; Nimfopoulos, 1990; Nimfopoulos *et al.* 1991; Baker *et al.* 1992).

5) Epithermal precious and base metal mineralization (e.g. Stratoniki and Zepko: Diakakis and Stefanides, 1992).
6) Karst controlled Zn-Pb±Fe deposits (e.g. Thassos: Vavelidis and Amstutz, 1983; Vavelidis, 1984, Omenetto, 1985).

ANALYTICAL TECHNIQUE

Samples, representative of each type of mineralization, were prepared for fluid inclusion microthermometry as doubly polished 100-150µm thick

Tab. 1: Summary of the microthermometric data for the different prospects.

Deposit name	**Style of mineralization**	**Host mineral**	**Th (°C) range**	**Salinity (wt% NaCl Equiv.)**	**Notes**
Palia Kavala	Shear-zone controlled & replacement	Quartz	280-320 (Mean 305; n = 34)	0-13 (Mean 6; n = 34)	Phase separation and P-T fluctuations. 3-phase CO_2-rich inclusions predominate
		Calcite	~100 (n = 9)	~0 (n = 9)	
Pangaeon	Shear-zone controlled & replacement	Quartz	275-330 (Mean 302; n = 29)	~4 (n = 35)	Minor amounts of CH_4 and N_2 are indicated by the depression of the CO_2 triple point.
Thassos	Stratigraphic and tectonic control: MVT?	Baryte	L-only fluid inclus.	0-22 (n = 10)	Fluids are Na-Ca-Mg brines with Ca to Na ratios of ~1:9
Madem Lakkos	Skarn-type massive or disseminated	Quartz	124-291 (Mean 291; n = 32)	1-28 (Mean 12; n = 42)	Data for lower temperature L+V inclusions (TYPE I FI)
			209-600 (Mean 340; n = 43)	32-74 (Mean 48; n = 43)	Data for higher temperature multiphase high salinity inclusions (TYPE III and IV FI)
Piavitsa	Replacement hydrothermal	Quartz	168-338 (Mean 258; n = 94)	1-9 Mean 3; n = 34)	Type I inclusions predominate,
Stratoniki	Skarn and replacement	Quartz	158-272 (Mean 211; n = 32)	3-5 (Mean 4; n = 32)	No CO_2 detected. Data for Type I inclusions
Zepko	Fault controlled replacement	Quartz	204-242 (Mean 221; n = 20)	0-4 (Mean 1; n = 12)	No CO_2 detected. Data for Type I inclusions
Varvara	Fault controlled replacement	Quartz	317-379 (Mean 342; n = 25)	0-6 (Mean 1; n = 16)	3-phase CO_2-rich (Type II inclusions predominate

wafers. Microthermometric analysis was undertaken using a LINKAM THM600 heating/freezing stage. The stage was attached to a LEITZ ORTHOLUX microscope carrying a NIKON PLAN 40 long working distance objective. The operating procedure is described in Shepherd (1981) who commissioned the fluid inclusion laboratory at IGME (Athens) where the analyses were undertaken. Calibration of the heating-freezing stage was done using synthetic fluid inclusion standards. Accuracy is estimated at ~0.5°C (between -100 and -20°C), ~0.2°C (between -20 and 30°C), ~1°C (between 30 and 200°C) and ~5°C (between 200 and 500°C). Thus, analytical errors are insignificant with regard to any geological interpretation. Data reduction were undertaken using FLINCOR (Brown and Lamb, 1987); a software package relating fluid inclusion microthermometry to fluid composition and P-T conditions of fluid trapping.

FLUID INCLUSION MICROTHERMOMETRY

Optical examination of quartz and carbonate from representative fluid inclusion samples from all localities revealed the presence of the following five inclusion types:

Type I: 2-phase inclusions: L + V aqueous
  a. 2-phase with L>V (PK-PG-TH-ML-PV-ST-ZP).
  b. 2-phase with V>L (TH-ML).
Type II: 3-phase CO_2-rich inclusions: L1 + L2 + V (PK-PG-VR)
Type III: 3-phase aqueous inclusions: L + V + S ± opaque.
  a. 3-phase with S1=halite (ML-PV)
  b. 3-phase with S2=sylvite (ML). Sylvite was determined from morphology (spheroid as opposed to cuboid) and its higher coefficient of solubility than sylvite (NaCl).
Type IV: Multiphase inclusion (ML)
  a. Multiphase with L + V + S1 + S2 ± opaque and solid <50%
  b. Multisolid with L + Solids (+V) and Solid >50%
Type V: Monophase inclusions.
  a. Only Vapour (V) (PK-TH-ML-VR
  b. Only Liquid (L) (PK-TH-ZP-VR)

L=liquid, V=vapour, S=solid, S1=halite and S2=sylvite

PK: Palia Kavala, PG: Pangaeon, TH: Thassos, ML: Madem-Lakkos, PV: Piavitsa, ST: Stratoniki, ZP: Zepko, VR: Varvara.

Microthermometric data for each of the mineralised localities are summarised in Table 1 (for further information see Nimfopoulos *et al.*; 1993, Constantinidou, 1993; Eliopoulos, 1993 and Dimitroula, 1993).

DISCUSSION OF FLUID INCLUSION DATA

Rhodope Massif

Palia Kavala and Pangaeon

The ore-forming fluids for the gold-bearing veins of Palia Kavala and Pangaeon are characterised by low salinity CO_2-rich fluids. Microthermometric data (Nimfopoulos *et al.* 1993; Eliopoulos, 1993) indicate that phase separation characterises some of the mineralization and that mineralization took place between 275 and 330°C at about 2-3kb. These conditions of mineralization are typical of many gold deposits. For example, the metamorphic gold deposits of Canada (e.g., Kerrich, 1977), as well as for mesothermal systems in Nevada and Colorado, USA, Mexico, Peru and New Zealand (Rye and Sawkins, 1974; Tsui and Holland, 1979; Nash and Cunningham, 1973; Paterson, 1987).

Large quantities of CO_2-and H_2O-rich fluid were released during greenschist to lower amphibolite facies of metamorphism in the Rhodope massif during back-arc thrusting (e.g., Boccaletti *et al.*, 1974; Fytikas *et al.* 1984; Dercourt *et al.* 1985; Frei (1992)). These fluids infiltrate to higher crustal levels prior to vein formation (cf. Imbrahim and Kyser,1991). The ubiquitous presence of CO_2 in fluid inclusions from all of the studied areas in Palia Kavala and Pangaeon is probably the product of decarbonatisation and devolatilisation of pelitic sedimentary rocks under greenschist to lower amphibolite metamorphic conditions (Kerrich and Fryer, 1979; Goldfarb *et al.*, 1989; Elder and Cashman, 1992). The association of gold with rocks of greenschist metamorphic facies (greenstones) has been regarded as a critical factor in the genesis of metamorphic quartz Au-veins. Precambrian to Lower Palaeozoic gneisses and schists (metapelites) are speculated to be the best candidate source bed for gold (e.g. Keays, 1984; Curti, 1987; Lattanzi *et al.*, 1989). These source rocks do not necessarily have to be enriched in gold, as the ore-forming fluids in metamorphic gold districts appear to be able to produce extreme enrichment of gold with respect to other, geochemically more abundant, metals. These fluids are able to extract, transport, and precipitate economic amounts of gold from source rocks containing no more than Clarke values of gold (Saager *et al.*, 1982; Fyfe and Kerrich, 1984).

Thassos

The fluid inclusion data refer only to baryte from Mavrolakkos. The majority of inclusions were one phase L-only indicating low temperatures of formation as no vapour bubble has nucleated (<100°C). Occasionally these had necked to produce a 2-phase (L+V) system allowing salinity measurements to be made. No daughter minerals were observed. Salinities range between between 0-22 wt% equiv. NaCl. Hydrohalite melting temperatures reflect Ca/ Na ratios of approximately 1:9. Only a small number of measurements were made on samples from Thassos. Thus, the fluid inclusion data are only indicative. However, Ca to Na ratios coupled with the range in salinity tentatively indicate a mixing of two fluids: firstly a moderate to high salinity calcic brine, analogous to basinal brines, mixing with a dilute fluid, which perhaps represents groundwater.

Serbomacedonian Massif

Madem Lakkos

The Madem Lakkos mineralization is characterised by an extremely hot saline fluids with homogenization temperatures ranging between 300 and > 600°C, very high salinities (> 70% equiv. NaCl dissolved salt) and highly variable inclusions with different types of daughter minerals. The variation in T_h and salinity indicates the presence of three inclusion populations: (i) a high-T (400-600°C) and high-salinity (60-70 wt%) group, (ii) a lower-T (260-360°C) and low-salinity (0-4 wt%) population and (iii) a group with low T (200-250°C) and moderate to high salinity (35-45 wt%). The least evolved of the fluids appear to be those where the fluid inclusions contain two daughter minerals (sylvite + halite). This fluid is thought to represent a highly saline Na-K brine, of probable, magmatic origin. These high salinity fluids are, also, commonly, characterised by the presence of a small opaque phase, probably sulphide, indicating that these fluids also carried metals.

A later fluid was of lower T and low salinity, and may represent boiling within the system. Further evolution of the system generated cooler, but higher salinity fluids. These are Ca-rich and have probably resulted from fluid-rock interaction with host marbles. This released calcium into the fluid during the formation of skarn type minerals.

The physical and chemical composition of the fluids associated with the Madem-Lakkos skarn mineralization are similar to the fluids described for porphyry copper deposits (Spooner, 1981) and for skarn deposits (Roedder, 1984). The similarities between porphyry copper and skarn deposits are documented by the occurrence of early, high temperature skarn mineralization, which is commonly overprinted by later porphyry-Cu mineralization. At lower temperatures, and in distal parts of the system, porphyry style mineralization gives way to vein type Pb-Zn-Ag deposits (Roedder,1984).

Fluid inclusion studies of skarn deposits indicate relatively high temperatures (400-650°C),intermediate salinity 10-45 wt % equiv. NaCl) and relatively low CO_2-content ($XCO_2 < 0.1$). Boiling only occurring in the upper parts of the system (Einaudi 1982). Wilson *et al*. (1980) reported very high salinities (up to 74.6% with 56.8 wt % equiv. NaCl+17.8 wt % equiv. KCl) from the Granisle deposit in British Columbia, this range of salinity is similar to the high-temperature, high-salinity fluid observed at Maddem Lakkos. The extreme variation in salinity, in these types of deposit, probably results from various mixtures of highly saline magmatic fluids and later circulating groundwaters (Roedder, 1984).

The fluid inclusion study on Madem Lakkos samples also showed that there is a good correlation between mineralogy and type of inclusions: samples from the early Cu-Fe mineralization show a predominance of high salinity high temperature fluid inclusions while samples containing later polymetallic mineralization, including gold, have a predominance of lower temperature lower salinity fluid inclusions. Suggesting that Cu-Fe mineralization is associated with hot saline, magmatic, fluids and polymetallic±Au mineralization is associated with cooler more dilute calcic fluids, which have probably interacted with meteoric waters.

The above data contrast with Gilg (1993) who studied the mineralization at Maddem Lakkos and recorded salinities in the range 30-45 wt%, with homogenization temperatures between 120-320°C. Gilg (1993) also records that the occurrence of sylvite (KCl), as a daughter mineral, is rare. This is not the case in one of the samples analysed, where sylvite is relatively common. The data of Gilg (1993) are in agreement with the lower end of our high-salinity data, and our lower-temperature data (cf. Table 1). Hence, there is the possibility that we have observed an early high-salinity high-temperature magmatic phase of mineralization not recorded by Gilg (1993). However, the data of Gilg (1993) include information from other deposits in the Kassandra Mining district which make a direct comparison between the two data sets difficult.

Piavitsa, Stratoniki and Zepko

The mineralization at Piavitsa, Stratoniki and Zepko formed from fluids similar in origin (magmatic) to those of Madem Lakkos. However, the fluids have more distal characteristics, i.e. lower salinities, lower temperatures and generally no daughter minerals (rare daughter minerals were observed at Piavitsa). These temperatures and salinities (Table 1) are also similar to Palia Kavala and Pangaeon in the western Rhodope, but CO_2, detectable by microthermometry, is absent. The skarn and replacement type of mineralization at Stratoniki also shows similar temperatures and salinities (Table 1) to Piavitsa with no daughter minerals and no visible CO_2. The ore forming fluids from Zepko were similar to those of Piavitsa Stratoniki, but were of lower temperature and salinity (Table 1).

Varvara

Fluid inclusions from the Varvara deposit are characterised by low salinities, average homogenization temperatures and the presence of CO_2 (Table 1). As stated for Palia Kavala and Pangaeon, this type of fluid is typical for many gold deposits in quartz veins hosted in low-medium grade metasediments (turbidite-hosted gold). Phase separation into CO_2-rich and H_2O- rich fluid was not observed. Microthermometry did not indicate the presence of other volatiles (e.g., CH_4, N_2). This is unusual as most fluids associated with Au mineralization, in these terrains, generally contain minor to significant amounts of other volatiles such as CH_4 and N_2 (Bottrell *et al.*, 1988; Craw, 1990; Naden and Shepherd, 1990 Shepherd *et al.*, 1992).

CONCLUSIONS

On the basis of microthermometric data (homogenisation temperatures, salinity, bulk density, volatile [CO_2, CH_4] content) the following conclusions are suggested:

1) The deposits can be classified according to distance from possible heat sources as: a) proximal skarn and replacement type (Madem Lakkos) b) porphyry style (Piavitsa) c) mesothermal-metamorphic shear zone controlled (Palia Kavala, Pangaeon, and Varvara) d) distal fault controlled epithermal systems (Stratoniki and Zepko) and finally e) Thassos where there heat sources which can be related to mineralization.

2) The main causes of ore deposition are thought to be: a) Phase separation (Palia Kavala) b) reaction with the host marbles (Madem Lakkos, Palia Kavala, Pangaeon, Stratoniki, Piavitsa, Varvara, Zepko) c) the introduction of cooler, less saline meteoric water into the fault zones, which results in a complex zone of fluid mixing and temperature reduction (Palia Kavala, Pangaeon, Thassos, Madem Lakkos, Stratoniki, Piavitsa, Varvara and Zepko)

3) The presence of CO_2 in the ore solutions has probably played a key role, as it makes Au-complexes more stable, relative to those of other metals (Schmidt-Mumm, 1985). Because, CO_2 phase separation has played an important role in depositing gold from the mineralizing fluids its identification could be used as one exploration tool for locating favourable areas of mesothermal gold mineralization (e.g. Palia Kavala, Pangaeon).

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