

## Microthermometric and O- and H-isotope characteristics of the mineralizing fluid in the Akgüney copper–lead–zinc deposit, NE Turkey

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The Akgüney copper–lead–zinc deposit (Ordu Province) is actively mined in the eastern Black Sea Region of Turkey. This deposit consists of ore-bearing veins emplaced along fault zones and hosted by slightly altered Upper Cretaceous andesites. Seven ore veins, D-1 to D-7, have been identified. These veins strike N 70–80°W with a dip of 50–60°NE, except for D7 which strikes E–W with a dip of 25°S. The ore-bearing quartz vein outcrops are slightly limonitized and hematitized. Two different ore associations are distinguished within the D-1 and D-2 veins. The first contains only pyrite, whereas the second consists of coarse-grained sulphides such as chalcopyrite, galena, sphalerite, tetrahedrite, and secondary limonite veinlets in addition to pyrite. Quartz and calcite are the gangue minerals. The mineral paragenesis has been identified as: pyrite→galena→chalcopyrite (I)→sphalerite→chalcopyrite (II)→(tetrahedrite?)→quartz→calcite. Fluid inclusion data indicate that the ore-forming fluids contained significant concentrations of divalent cations in addition to NaCl. The salinity of the fluids lies in the range 14–24% (ave.=19.0) NaCl equivalent; liquid homogenization temperatures range from 276 to 349°C (ave.=307°C) and decrease to 211°C through the later stages of mineralization. Oxygen and hydrogen isotope data are consistent with the mineralizing fluid being of meteoric origin, with modification of the oxygen isotope composition by exchange with the volcanic host rocks. However, low δD values suggest a magmatic water component in some samples. The combination of fluid inclusion and stable isotope data suggest that the Akgüney Cu–Pb–Zn vein-type deposits were formed by the leaching of metals and sulphur from the surrounding volcano-sedimentary rocks by deep-circulating meteoric water.

**Keywords:** Akgüney; copper–lead–zinc deposit; Eastern Black Sea region; fluid inclusion; stable isotope

### Introduction

The Akgüney copper–lead–zinc deposit is located near to Akgüney Village in the Kabadüz District of Ordu Province within the Eastern Pontide region of Turkey (Figure 1).

The Eastern Pontide region consists of volcano-sedimentary units of Jurassic to Quaternary age, which developed on a pre-Mesozoic basement and were intruded by

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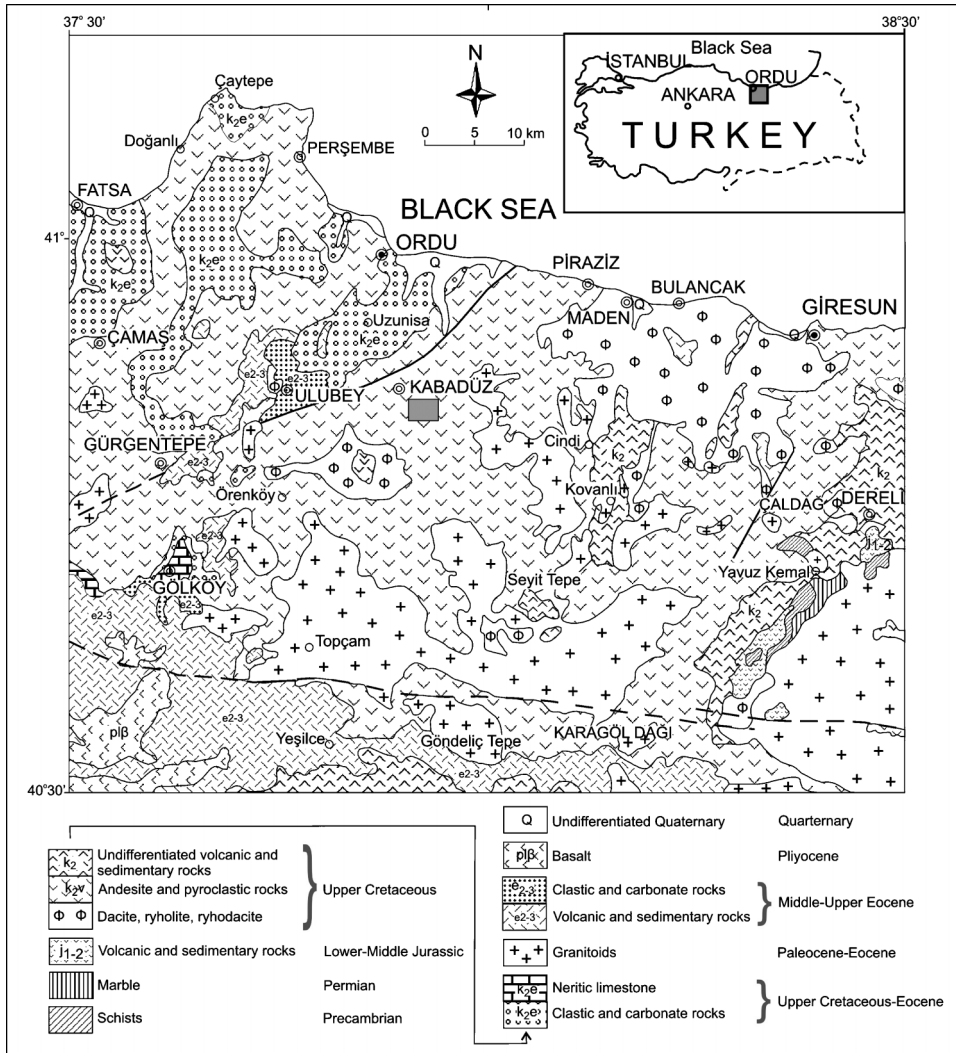


Figure 1. Location and regional geology map of the Akgüney Cu-Pb-Zn deposits (simplified from the 1:500,000 scale geological map of Turkey prepared by MTA, 2002).

Palaeocene (?) granitoids (Figure 1). These volcano-sedimentary units are classified as follows: Lower Basic Series (Liassic to Lower Cretaceous), Dacitic Series (Upper Cretaceous), flysch-like sediments and Upper Basic Series (Eocene), Young Basic Series and late dikes (Oligocene-Pliocene). A widely held view is that the region developed along a convergent plate boundary, possibly as a volcanic arc system (e.g. Tokel 1973, 1992; Eğin and Hirst 1979; Pejatovic 1979; Akıncı 1980, 1985; Manetti *et al.* 1983; Bektaş *et al.* 1984; Gedik *et al.* 1992, 1996; Çamur *et al.* 1994; Yılmaz and Boztuğ 1996; Okay and Şahintürk 1997; Yılmaz *et al.* 1997; Boztuğ 2001; Boztuğ *et al.* 2001, 2002, 2003). This resulted from northward subduction of the northern branch of Neo-Tethyan oceanic crust beneath the Eurasian plate along the İzmir-Ankara-Erzincan suture zone. Granitoids within the area show various mineralogical and geochemical characteristics (calc-alkaline, metaluminous I-type; biotite leucogranites with peraluminous S-type and calc-alkaline to tholeiitic M-type) that

indicate emplacement within various geotectonic environments (arc-related, syn-collisional and post-collisional extension-related) over a period of time from Early/Late Cretaceous to Eocene (Boztuğ *et al.* 2004, 2006; Arslan and Aslan 2006).

The region contains over 400 massive, vein and skarn-type hydrothermal sulphide deposits and mineral prospects. In addition, Cu–Mo mineralizations occur in the porphyry granitic rocks, and Mn occurrences are also known in the volcanic rocks. The dacitic series host Kuroko-type massive sulfide deposits and most of the vein type Cu–Pb–Zn deposits occur in this region.

The Akgüney copper–lead–zinc deposit is a typical example of the volcanic-hosted vein type deposits that occur widely within the Eastern Pontide region of Turkey.

The earliest reported study of the Akgüney Cu–Pb–Zn deposits was carried out by Köse (1987), who interpreted these deposits as ore veins formed along fault zones developed in a NW–SE direction within andesitic and basaltic volcanic rocks. Köse found that the ore veins contain pyrite, chalcopyrite, sphalerite, galena, tetrahedrite, quartz and barite occurring in three different phases of mineralization. This was produced by hydrothermal fluids possibly related to a deep seated granitoid intrusion.

Steinmann and Emerson (2001) concluded that two different types of veins are present in the area; the northern one is rich in zinc, whereas the southern one is rich in copper.

More recently, Ünal (2006) and Ünal and Gökce (2007) carried out detailed geological investigations, including ore petrography and fluid inclusion studies, and their conclusions are summarized below. This paper aims to determine the characteristics and origin of the mineralizing fluids. We report new oxygen isotope data from veins, and hydrogen isotope data of the fluid inclusion water presumed to represent the mineralizing fluid. The chemical composition of the fluids has been constrained from the microthermometric characteristics of these fluid inclusions.

## Geological background

Andesitic rocks crop out around the Akgüney Cu–Pb–Zn deposits (Figure 2). These volcanic rocks consist of agglomerates and interbeds of massive lavas. Agglomerates contain tuffaceous matrix and show well developed bedding. Massive lavas are slightly brecciated. In addition, they have dark grey and black colours in unaltered deeper levels, whereas they are highly altered under surficial conditions and have off-white, yellowish and greenish pale colours representing argilization, limonitization and chloritization at the surface. The volcanic rocks of the area were mapped as Cretaceous on the 1/500,000 scale regional geology map of Turkey. Akıncı (1985) investigated these volcanic rocks in the Upper Cretaceous Dacitic Series, which was mentioned in the introduction section, and Terlemez and Yılmaz (1980) named them as the Upper Cretaceous Mesudiye Formation.

Microscopic investigation of selected samples shows that they contain plagioclase, quartz, biotite, muscovite and hornblende and plot within the andesite field on the diagram of Streckeisen (1976, 1979). Hypo-hyaline porphyritic texture is widespread and pilotaxitic and amigdaloidal textures are seen in some of the samples. Sericitization, argilization and silicification of plagioclases, chloritization of biotites and hornblendes are widespread. Some of the samples show brecciation, with calcite and quartz infillings developed between the brecciated rock fragments.

Strikes and dips of cracks and fractures are randomly dispersed on rose diagrams with minor concentrations of trend between 70 and 115° (N70E and N65W),

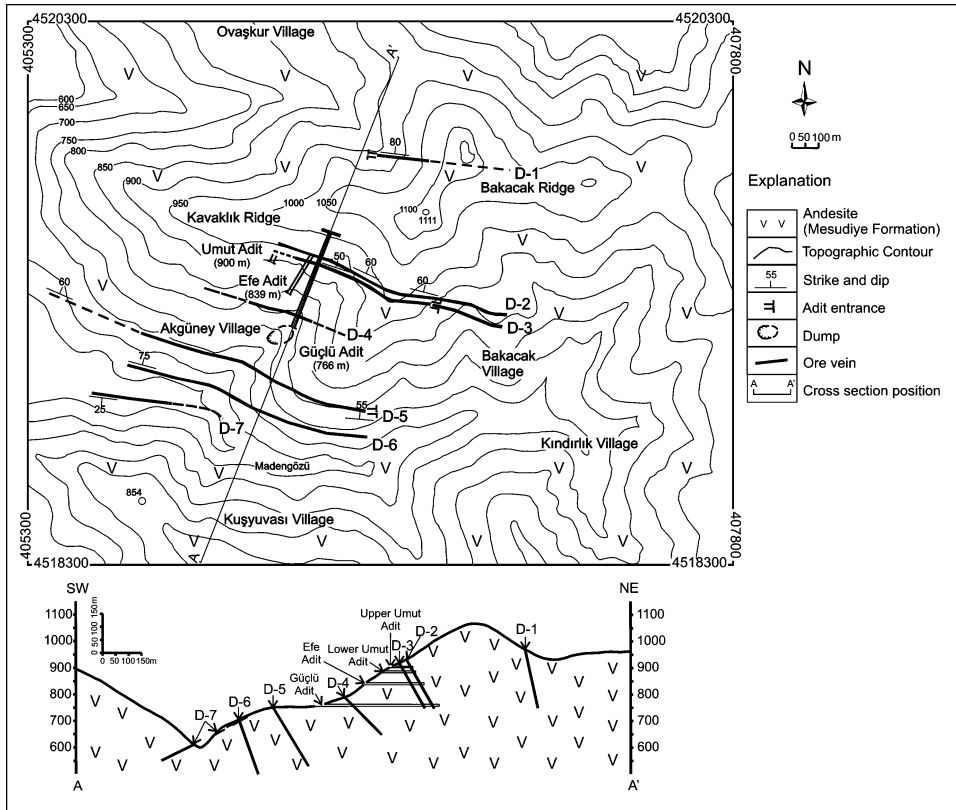


Figure 2. Geological map and cross-section of the Akgüney area.

indicating that these tectonic features developed under a tensional regime in a N–S direction. In addition, some lineaments and ring-like structures were identified on the Landsat images (174/32 ETM) of the investigated area, and these features were suspected to represent a dome-like structure related to a buried granitoid intrusion (Ünal 2006), but the lack of the radial lineaments does not support this idea. Field control of these features showed that the outer ring corresponds with the contact between the massive volcanics of the inner zone and tuffaceous volcanoclastics of the outer zones.

### Ore geology

Cu–Pb–Zn mineralizations within the Akgüney area are developed as ore veins occurring along the fault zones developed in the Upper Cretaceous andesitic volcanic rocks. The veins crop out as slightly limonitized, yellowish-brownish quartz veins. The known ore veins are numbered as D-1 to D-7. These ore veins, strike at  $N70\text{--}80^\circ\text{W}$  with a dip of  $50\text{--}60^\circ\text{NE}$ , except for D-7, which strikes E–W with a dip of  $25^\circ\text{S}$  (Figure 2).

The D-2 and D-3 veins are the only ones which were being mined during the field investigations undertaken for this study. Mining operations start with cross-cutting adits until reaching the veins, then continue as wings in the direction of the ore veins at different levels (Upper Umut Adit, at 902 m elevation; Lower Umut Adit, at 886 m elevation; Efe Adit, at 839 m elevation; Güçlü Adit, at 766 m

elevation). Both of the veins are cut and replaced by the younger faults in a NW–SE direction.

Two different types of mineralization were identified in D-2 and D-3 veins. The first type of mineralization is characterised by fine-grained pyrite, whereas the second one is characterized by a coarse-grained texture and a variety of ore minerals including chalcopyrite, sphalerite, galena, tetrahedrite and limonite as well as pyrite. The second type mineralization cuts the first one and they are classified as early and late stage (the first and second phase) mineralizations. Only the second type of occurrence was mined and the second type was left as pillars. In addition, the upper parts of the veins are rich in galena and sphalerite, whereas the lower parts are rich in chalcopyrite and pyrite. Quartz and calcite are the gangue minerals and are seen to be formed later than the ore minerals. The mineral paragenesis is determined as: pyrite→galena→chalcopyrite (I)→sphalerite→chalcopyrite (II) ± tetrahedrite→quartz→calcite.

### **Fluid inclusion studies**

#### ***Sampling and methodology***

Fluid-inclusion studies were carried out on quartz crystals closely associated with the sulphide minerals. Samples were collected from the D2 and D3 ore veins at the 766 m level; samples OC-4 and OC-5 were taken from the D2 vein, the others from the D-3 vein.

Fluid inclusions were observed using a petrographic microscope, and the following parameters were determined by microthermometry: first melting temperature ( $T_{FM}$ ), last ice melting temperature ( $T_{ICE}$ ), and temperature of homogenization into liquid ( $T_H$ ).

These microthermometric measurements were performed using a LINKAM THMS-600 and TMS-92 freezing–heating stage and a long focal length LW × 40 objective. Liquid nitrogen was used for freezing. The heating rate was controlled manually with the help of a TMS-92 system in the range of 0.1–5.0°C/min. The stage was calibrated using heptane ( $T_m = -90.61^\circ\text{C}$ ) and potassium nitrate ( $T_m = 335.0^\circ\text{C}$ ) at the beginning of daily studies. Repeated measurements showed that the reproducibility of the temperature determinations was better than  $\pm 0.5^\circ\text{C}$  for all three types of determinations.

#### ***Morphology and inclusion types***

Fluid inclusions are developed as either irregularly dispersed primary inclusions within the crystals or linearly dispersed secondary inclusions along the small cracks. Primary inclusions are more abundant than the secondary ones. Both types of inclusions are two phase (liquid + vapour) inclusions with mostly ellipsoidal and pear shaped morphologies.

The size of the inclusions is very small (5–15  $\mu\text{m}$  for primary inclusions, generally smaller than 2.5  $\mu\text{m}$  and rarely larger than 5  $\mu\text{m}$  for secondary inclusions). The volume of the gas phase is generally in the range of 5–10 % and rarely reaches 20%.

#### ***Microthermometric measurements***

First melting temperature ( $T_{FM}$ ) values are dispersed in the range from  $-53.5$  to  $-50.5^\circ\text{C}$  (average:  $-52.0^\circ\text{C}$ ,  $n=90$ ) for primary and secondary inclusions. These temperature values are significantly lower than for the NaCl–H<sub>2</sub>O system and very

close to the eutectic temperature of ‘ $\text{H}_2\text{O}-\text{NaCl}-\text{CaCl}_2$ ’, ‘ $\text{H}_2\text{O}-\text{MgCl}_2-\text{CaCl}_2$ ’, and ‘ $\text{H}_2\text{O}-\text{KCl}-\text{CaCl}_2$ ’ ( $-55.0$ ,  $-52.2$  and  $-50.5^\circ\text{C}$ , respectively; Shepherd *et al.* 1985), and suggest that the mineralizing hydrothermal fluid contains the salts of bivalent ions such as  $\text{CaCl}_2$ ,  $\text{MgCl}_2$ , besides the  $\text{NaCl}$  and possibly  $\text{KCl}$ .

The last ice melting temperature ( $T_{\text{mICE}}$ ) values range from  $-23.0$  to  $-10.0^\circ\text{C}$  (average:  $-15.0^\circ\text{C}$ ,  $n=67$ ) in primary inclusions, whereas they range from  $-23.0$  to  $-11.0^\circ\text{C}$  (average:  $-16.0^\circ\text{C}$ ,  $n=25$ ) in secondary inclusions (Figure 3). The average salinity values, calculated using the equations of Bodnar (1993), for primary and secondary inclusions, are 18.6‰ (24.3–13.9‰) and 19.4‰ (24.3–15.00‰)  $\text{NaCl}$  equivalent, respectively. Although the salinity values cover a wide range, the similarity of the values in primary and secondary inclusions indicates that the salinity of the mineralizing fluids was similar during the various stages of mineralization.

The pressure uncorrected homogenization temperature ( $T_{\text{H}}$ ) values range from 276 to  $349^\circ\text{C}$  (average:  $307^\circ\text{C}$ ,  $n=98$ ) in primary inclusions, whereas they form two different populations in secondary inclusions, in the ranges of  $265.0$ – $309.0^\circ\text{C}$  (average:  $290.0^\circ\text{C}$ ,  $n=38$ ) and  $211.0$ – $233.0^\circ\text{C}$  (average:  $221.0^\circ\text{C}$ ,  $n=18$ ) (Figure 4). These  $T_{\text{H}}$  values indicate that the ore veins were formed in hypothermal and

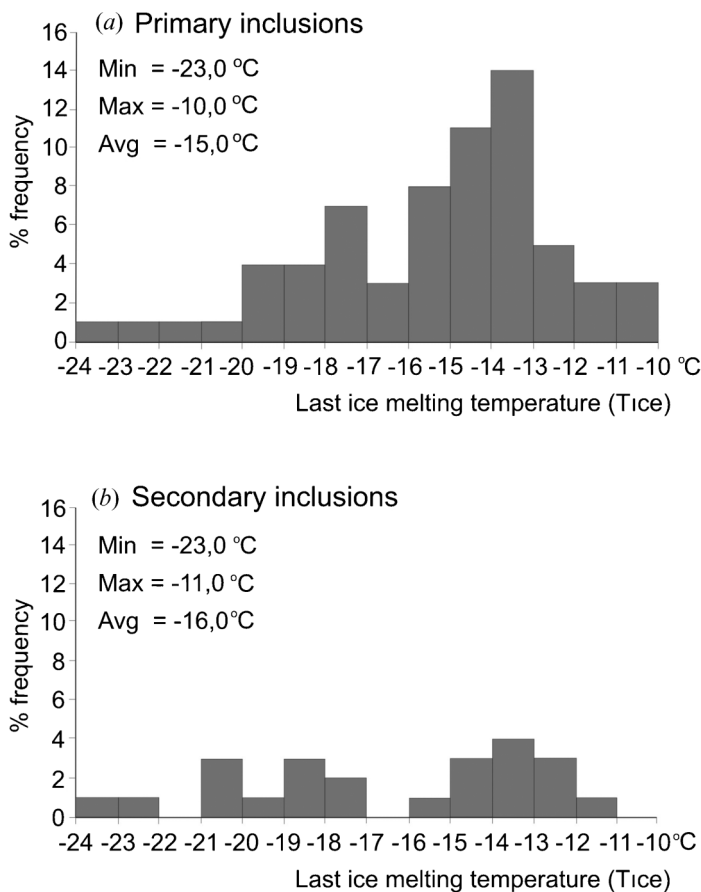


Figure 3. Statistical distribution diagrams of the highest temperature of ice melting ( $T_{\text{ICE}}$ ) values in primary (A) and secondary (B) fluid inclusions.

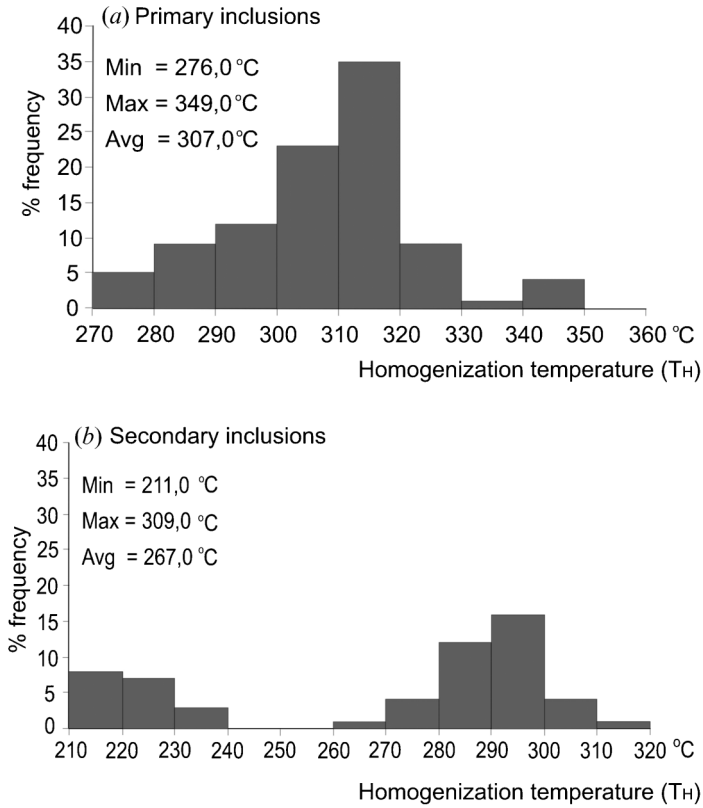


Figure 4. Statistical distribution diagrams of the homogenization temperature ( $T_H$ ) values in primary (A) and secondary (B) fluid inclusions.

mesothermal conditions, and the temperature of the hydrothermal fluids decreased through the later episodes of mineralization.

Plotting the primary and secondary inclusions on the salinity versus homogenization temperature diagram shows a large area of dispersion (Figure 5). Although,

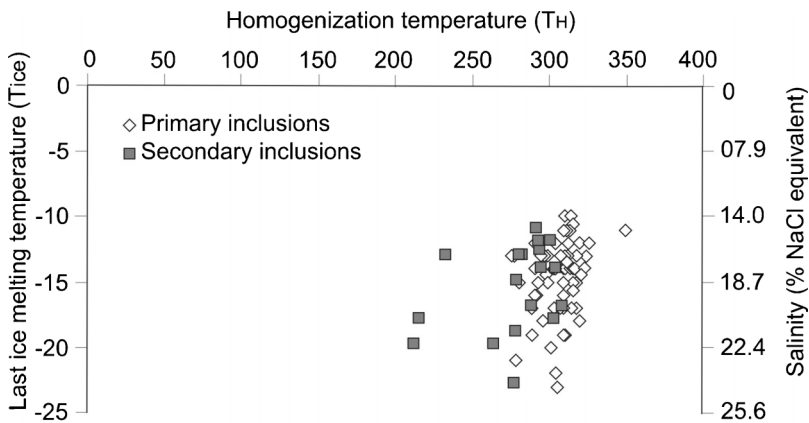


Figure 5. Distribution of fluid inclusions on the salinity–homogenization temperature diagram (only inclusions for which both  $T_{ICE}$  and  $T_H$  values were recorded).

there is no significant separation between the points of primary and secondary inclusions, it may be said that the secondary inclusions are scattered within the low temperature parts of the diagram.

## Oxygen- and hydrogen-isotope studies

### *Analytical methods*

Representative samples were chosen on the basis of fluid-inclusion results, and quartz separates were prepared by hand-picking under a stereo-microscope.

All stable isotope measurements were made at the University of Cape Town (UCT). Oxygen isotope ratios of quartz were determined using conventional extraction methods after reaction with  $\text{ClF}_3$ . The  $\text{O}_2$  was converted to  $\text{CO}_2$  using a hot platinized carbon rod, and isotope ratios were measured using a Finnigan MAT Delta XP mass spectrometer. The data are reported in the familiar  $\delta$  notation where  $\delta = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000$  and  $R = {}^{18}\text{O}/{}^{16}\text{O}$ . The quartz standard NBS28 was run twice with each batch of eight samples and the average difference between 18 pairs of NBS-28 standards run during the course of this work was 0.13‰. This represents the typical precision of the analyses. Data were normalized to a value of NBS28 of 9.64‰, but the difference between normalized and unnormalized data is <0.5‰. Further details of the methods employed for extraction of oxygen from silicates at UCT are given by Vennemann and Smith (1990) and Harris *et al.* (2000). Hydrogen-isotope analyses were carried out on inclusion fluids extracted from the quartz separates by thermal decrepitation at 1000°C. Extracted water vapour was converted to hydrogen gas, and the  $\delta\text{D}$  of each sample was determined. Approximately 2 g clean chips of vein quartz were loaded into a 9 mm (ID) oven dried (800°C) quartz glass tube and attached to the vacuum line. The loaded sample was degassed at about 200°C using a hot air gun until no further rise in the pressure was noticed. The samples were then gently flamed causing the fluid inclusions to decrepitate. The  $\text{H}_2\text{O}$  and  $\text{CO}_2$  liberated from the sample were collected in a liquid nitrogen trap. The  $\text{CO}_2$  was separated from the  $\text{H}_2\text{O}$  and  $\text{CO}_2$  mix using an alcohol trap. The H-isotope composition of the  $\text{H}_2\text{O}$  was determined using the closed tube Zn reduction method of Vennemann and O'Neil (1993). An internal water standard (CTMP,  $\delta\text{D} = -9\text{‰}$ ) was used to calibrate the data to the SMOW scale and a second water standard DML ( $\delta\text{D} = -300\text{‰}$ ) was used to correct for scale compression (e.g. Coplen 1993).

### *Results*

The  $\delta^{18}\text{O}$  values for quartz, the calculated  $\delta^{18}\text{O}$  values for inclusion water in equilibrium with quartz, and  $\delta\text{D}$  values for inclusion water are all given in Table 1. The  $\delta^{18}\text{O}$  values of water in equilibrium with quartz were calculated using the quartz–water fractionation equation of Sharp and Kirschner (1994) and the average homogenization temperature values of primary inclusions for each sample.

The calculated  $\delta^{18}\text{O}$  values of the fluid vary in a narrow range from +1.1 to +4.0‰. The  $\delta\text{D}$  values for the inclusion water range from -94 to -38‰ (average:  $-57 \pm 37\text{‰}$ ).

### *Discussion*

The estimated compositions of the mineralizing fluids plot between the global meteoric water line and the magmatic water box on the  $\delta\text{D}$  versus  $\delta^{18}\text{O}$  diagram

Table 1. Hydrogen and oxygen isotope compositions of the fluid inclusions.

Sample no.	$\delta^{18}\text{O}$ values of quartz	Average homogenization temperature values measured for primary inclusions ( $^{\circ}\text{C}$ )	$10^3 \ln \alpha^*$	Calculated $\delta^{18}\text{O}$ values of the inclusion fluid in equilibrium with quartz	$\delta\text{D}$ values of the inclusion fluid
OC-04	10.0	309	6.6	3.4	-94
OC-05	10.6	296	7.0	3.6	-62
OC-07	10.1	310	6.5	3.6	-45
OC-11	8.8	289	7.3	1.5	-52
OC-12	7.8	304	6.7	1.1	-40
OC-13	8.5	313	6.4	2.1	-49
OC-14	9.0	316	6.3	2.7	-38
OC-17	9.2	319	6.2	3.0	-63
OC-21	8.6	311	6.5	2.1	-51
OC-24	11.2	292	7.2	4.0	-71

Note:  $*10^3 \ln \alpha$  values were calculated from the results of quartz using the  $T_H$  values measured in fluid inclusions and fractionation factors of Sharp and Kirschner (1994).

(Figure 6). The O-isotope composition of the fluid was calculated using the homogenization temperature. The trapping temperature of the fluids is not known precisely because the mineralization depth is unknown. The estimated  $\delta^{18}\text{O}$  values of the mineralizing fluids are, therefore, systematically lower than the real values.

The palaeolatitude of the Pontide region during the time of mineralization (Tertiary) was around  $30^{\circ}$  based on the palaeomagnetic data of Orbay and Bayburdi (1979) and Baydemir (1990). This result suggests that the region was further south than at present, and the  $^{18}\text{O}$  and D values of ambient meteoric water is, therefore,

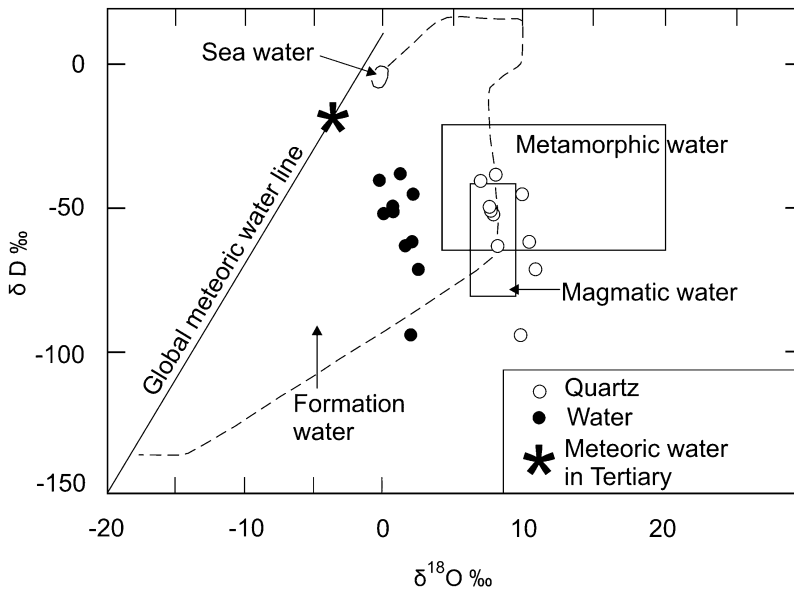


Figure 6.  $\delta\text{D}$  versus  $\delta^{18}\text{O}$  plot (Sheppard, 1986) for the mineralizing fluid that formed the Akgüney deposits. The  $\delta^{18}\text{O}$  fluid data were calculated from quartz values using fluid homogenization temperatures.

estimated to be  $-4$  and  $-22\%$ , respectively, based on the global map of Clark and Fritz (1997, p. 66–67). This point is plotted in Figure 6.

It is suggested on the basis of Figure 6 that either there was involvement of both meteoric and magmatic water within the mineralizing hydrothermal fluid, or that significant modification of the O-isotope composition of meteoric water occurred. The first possibility may be explained by the mixing of the meteoric and magmatic waters during the mineralization processes. Although, there is no evidence of contemporaneous magmatic activity close to the mineralization area, hydrothermal fluids derived from an unexposed plutonic intrusion may be involved in the mineralization processes. The second view is more in accordance with the data from similar deposits within the Pontide region. The data for the vein type Kurşunlu and Inler Yaylası deposits, which are located in close proximity to the Akgüney deposits and hosted by the same Upper Cretaceous and Eocene volcano-sedimentary units, and those for VHMS type Çakmakkaya and Damarköy deposits, which occur in the same volcano-sedimentary unit in eastern Black Sea region, are similar those of Akgüney deposits (Gökce *et al.* 1993; Gökce and Spiro 2002; Gökce and Bozkaya 2003). These authors concluded that meteoric water was dominant in the mineralizing fluid and that its oxygen isotopic composition was profoundly changed as a result of interaction with volcano-sedimentary rocks.

The dispersion of data points shows a vertical trend because of the large width of the range of  $\delta D$  values. Campbell and Larson (1998) indicate that this is a typical feature of most deposits of this type. The calculated  $\delta^{18}O$  values of fluid based on quartz values probably represent more closely the depositing fluid, whereas the  $\delta D$  measured from extracted fluids is a mixture of both primary and secondary fluid inclusions. The contribution of the secondary inclusions to the bulk extracted fluid will change the  $\delta D$  values of the fluid if the  $\delta D$  value of the secondary inclusions is significantly different. It is possible that the vertical lineation of the points is an artifact of the higher uncertainty in  $\delta D$ , specifically those resulting from the incorporation of both primary and secondary fluid inclusions within the extracted bulk.

### Summary and conclusions

The Akgüney copper–lead–zinc deposit consists of ore-bearing veins emplaced along fault zones and is hosted by slightly altered, Upper Cretaceous andesites. The ore-bearing quartz veins show two different ore associations. The first one consists of pyrite, whereas the second one consists of coarse-grained sulfide minerals such as chalcopyrite, galena, sphalerite, tetrahedrite, and secondary limonite veinlets in addition to pyrite.

Fluid inclusion data indicate that the ore forming fluids contain significant quantities of divalent cations ( $CaCl_2$  and/or  $MgCl_2$ ), along with NaCl and possibly KCl. The salinity of the fluids was in the range of 14–24% (average=19%) NaCl equivalent. The homogenization to liquid temperatures are in the range 276–349°C (average: 307°C) and decrease down to 211°C for the later stages of mineralization.

Oxygen- and hydrogen-isotope data suggest that the fluids from which the quartz veins formed contain a significant proportion of meteoric water. The generally low D values suggest magmatic water involvement in some samples. The metals and sulphur in the deposit were most likely derived from the surrounding volcano-sedimentary rocks and transported by the mixture of deeply circulated meteoric water and possible magmatic water.

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