

# FLUIDS RELATED TO REMOBILIZATION OF MESOZOIC SULFIDE MINERALIZATION IN THE EPTADENDRO-RACHI REGION IN EASTERN RHODOPE, THRACE, GREECE

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**Abstract:** The copper sulfide mineralization in the Eptadendro and Rachi areas is hosted in the Upper Tectonic Unit of eastern Rhodope in Thrace. The orebodies are found along the contacts between granitoid intrusions and meta-ultrabasic-basic rocks, as well as within meta-ultrabasic-basic rocks. Two stages of mineralization have been identified: an initial stratabound stage which is considered to be of submarine volcanosedimentary origin and a later vein-type stage formed during a hydrothermal episode, related to the intrusion of the granitoids (trondhjemites and pegmatites), during Upper Cretaceous-Early Tertiary. It consists of pyrite, chalcopyrite, sphalerite, galena, hessite, bismuthinite, emplectite, tetradymite, aikinite, wittichenite, siegenite, millerite, bornite, pyrrhotite, covellite, magnetite, hematite and goethite, with chlorite, quartz, calcite and sericite being the main syn-ore gangue minerals. The mineralization has been affected at least by a greenschist facies metamorphic episode during Eocene-Oligocene. Although the sulfide mineralization is partly deformed and shows recrystallization textures, the data obtained from fluid inclusions demonstrate well the physical and chemical parameters of ore-forming environment during the latest hydrothermal event, caused by intrusion of the granitoids. Microthermometric studies showed three groups of fluid inclusions, corresponding to the distinct fluids involved in the mineral deposition and the pegmatite formation. The first group of fluid inclusions hosted in syn-ore quartz is characterized by relatively high homogenization temperatures (300° to 380° C, with a peak at 330° C) and low salinities (1.6 to 7.2 wt% NaCl equiv) and corresponds to the fluids of the main ore stage. The second group is distinguished by a drop in  $T_h$  (210° to 260°C) corresponding to the late ore stage associated with calcite formation, and salinities (3.2 to 6.3 wt% NaCl equiv) similar to the first group. The third group of fluid inclusions in the pegmatite is characterized by temperatures ranging from 300° to 390°C, and variable salinities (6.9 to 8.9 wt% NaCl equiv and 34.7 to 58.5 wt% NaCl equiv) suggesting a magmatic origin. The composition of these fluids is dominated by NaCl+KCl. Most probably these fluids were not related to the ore mineralization process.

**Keywords:** Fluid inclusions, hydrothermal fluids, copper sulfide mineralization, trondhjemites, Eastern Rhodope, Greece

## 1. Introduction

The copper sulfide mineralization in the Eptadendro and Rachi areas (Fig. 1) is hosted in the Upper Tectonic Unit (U.T.U.) of eastern Rhodope in Thrace (Chatzikirkou 2003; Chatzikirkou and Michailidis 2004). The rocks of U.T.U. are divided into meta-ultrabasics (serpentinites), metabasites (hornblendites, amphibolites, metagabbros), acidic rocks (granitoids and pegmatites) and marbles (Mposkos et al., 1989; Ricou et al., 1998; Mposkos and Krohe, 2000; Chatzikirkou, 2003). The meta-ultrabasic rocks are metamorphosed harzburgites,

whereas the protoliths of the metabasites are andesitic- to sub-alkaline-basalts of tholeiitic affinity. The granitoids have an igneous origin, a calc-alkaline affinity, and their protoliths are trondhjemites.

According to Ashworth et al. (1988), Nesbit et al. (1988) and Chatzikirkou (2003) the mineralization is related to a fault zone of NE-SW direction. The orebodies are found along the contacts between the granitoid intrusions (mainly trondhjemites) and the meta-ultrabasic-basic rocks, as well as within the

meta-ultrabasic-basic rocks. Two stages of mineralization have been identified: an initial stratabound stage, which is considered of volcano-sedimentary origin and a later one of vein-type formed during a hydrothermal episode, related to the intrusion of the trondhjemites. The pegmatites are free from ore mineralization.

The orebodies exhibit a lenticular to tabular mor-

phology and are concordant with the host rocks. Their length does not exceed 30 meters, while their thickness is less than one meter. The main ore mineral assemblage of the stratabound mineralization consists of pyrite and chalcopyrite, with minor magnetite and pyrrhotite, whereas the main syn-ore gangue minerals are chlorite and quartz. The vein type mineralization consists of pyrite and chalc-

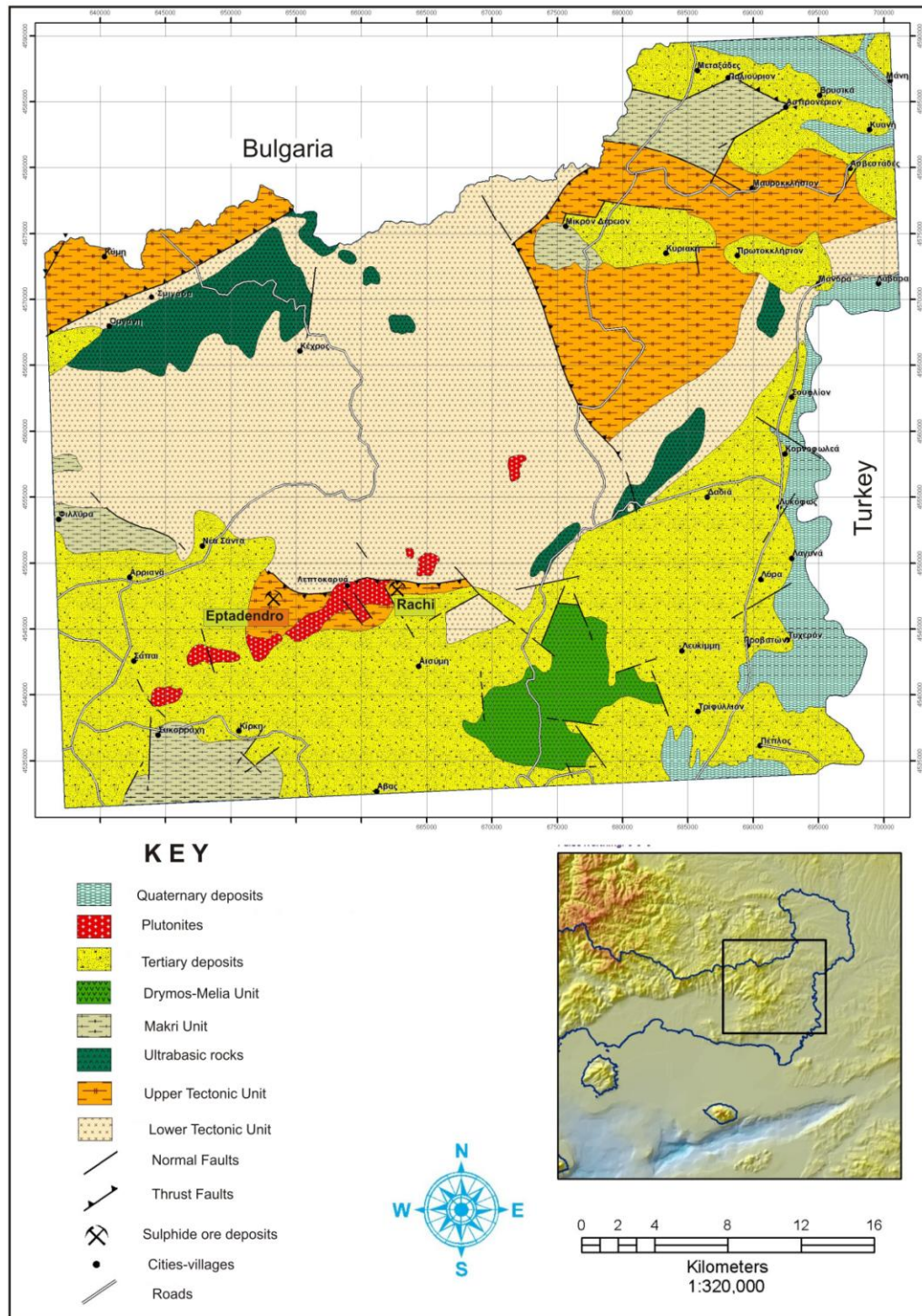


Fig. 1. Simplified geological map of the Eptadendro-Rachi ore district (Chatzikirkou 2003).

pyrite, with minor sphalerite, galena, magnetite and hematite and traces of hessite, bismuthinite, emplectite, tetradymite, aikinite, wittichenite, siegenite, millerite, bornite, pyrrhotite, covellite and goethite (Ashworth et al., 1988; Nesbit et al., 1988; Chatzikirkou, 2003; Chatzikirkou and Michailidis, 2004). Chlorite, quartz, calcite, and sericite are the main gangue minerals.

According to Chatzikirkou (2003) folding of thin mineralized layers, brittle or ductile deformation, annealing or porphyroblast formation of pyrite are typical metamorphic features confirming that the mineralization of both types has been affected by a series of metamorphic and deformation events.

Lead isotope characteristics confirmed that Pb is orogenic and was derived mainly from the crust and to a lesser extent from the mantle (Chatzikirkou, 2003). Besides, lead isotopes give an Upper Cretaceous to Eocene age for the ore mineralization. According to Chatzikirkou (2003) and Chatzikirkou and Michailidis (2004) these data suggest a metallogenetic stage related to the intrusion of the trondhjemites in the area. The  $\delta^{34}\text{S}$  values for pyrite and chalcopyrite presented by Chatzikirkou (2003) vary between 1.2 and 4.6 ‰. These values give a temperature of ore formation of  $293\pm 34^\circ\text{C}$  and indicate that sulfur probably precipitated from a fluid which might have been derived be of magmatic origin (~90%) and partly of seawater (~10%) origin (Chatzikirkou, 2003).

The current study further investigates the fluid inclusions in gangue syn-ore minerals and the acidic intrusions (trondhjemites and pegmatites) in the Eptadendro and Rachi areas, in an attempt to determine more precisely the ore-forming conditions. The mechanism and the physicochemical conditions of sulfide ore deposition are discussed in the context of the fluid inclusion data along with the genetic model proposed by Chatzikirkou (2003) and Chatzikirkou and Michailidis (2004).

## 2. Fluid inclusion study

Microthermometric measurements were conducted on carefully selected fluid inclusions hosted in: hydrothermal syn-ore quartz and calcite from the vein-type ore mineralization (four and two samples respectively), quartz from trondhjemite at the contact with meta-ultrabasic rocks (one sample), rock-forming quartz from the trondhjemite (one sample) and quartz from non-mineralized pegmatite (one sample).

Microthermometric data were obtained using a Leitz SM-LUX-POL microscope, equipped with a LINKAM THM-600/TMS 90 heating-freezing stage, housed at the Department of Mineralogy, Petrology and Economic Geology of the Aristotle University of Thessaloniki, Greece. Calibration of the stage was achieved using organic standards with known melting points (chloroform  $-63.5^\circ\text{C}$ , naphthalene  $80.35^\circ\text{C}$ , Merck 135  $135^\circ\text{C}$ , saccharine  $228^\circ\text{C}$ , Merck 247  $247^\circ\text{C}$ ) and ice ( $\text{H}_2\text{O}$ ). The precision of the heating and freezing measurements were  $\pm 1^\circ\text{C}$  and  $\pm 0.2^\circ\text{C}$ , respectively. Fluid inclusion shapes and sizes, spatial relationships among inclusions, and minerals and number of constituent phases within inclusions were observed in nine doubly-polished thin sections prepared at the Institute of Geological and Mineral Exploration (IGME) of Greece. Routine heating-freezing runs were performed on a total of 421 fluid inclusions, including the 176 fluid inclusion measurements presented by Chatzikirkou (2003). The salinities were calculated based on the equations of state of Potter et al. (1978) and Bodnar (1993), whereas the FLINCOR program (Brown, 1989) was used to calculate salinities and densities from measured ice melting and homogenization temperatures.

## 3. Types of fluid inclusions

Fluid inclusions were evaluated using fluid inclusion types and fluid inclusion assemblages (FIAs) (Goldstein and Reynolds, 1994). The samples contain clear quartz and calcite which are intimately intergrown with the ore minerals. Fluid inclusions have regular or irregular shapes, and are isolated or are arranged in clusters and planes. Inclusions with negative crystal or rounded to elongated isometric shapes typically occur along crystal faces of quartz and calcite. They are assumed to be primary in origin with only a few considered as secondary, according to the criteria of Roedder (1984) and Bodnar (2003). Microthermometric measurements were conducted mainly on primary fluid inclusions and FIAs; inclusions that had been necked down or were secondary in origin were avoided. Fluid inclusions in calcite are rare, so that only a limited number of heating-freezing data were obtained.

At room temperature, only two phase liquid-vapor inclusions (Fig. 2 a-c) were identified: (i) in the ore-related quartz, (ii) at the contact between trondhjemites and meta-ultrabasic rocks and (iii) in the trondhjemite. Inclusions that were analyzed range in diameter between 3 and  $42\ \mu\text{m}$  and homogenize to liquid. The primary fluid inclusions have

relatively consistent liquid to vapor ratios (20-30 volume % vapor). The variability in homogenization temperature data may be due to real variability in the FIAs (Goldstein, 2003).

In the pegmatite's, two types of inclusions were recognized. Type 1 two-phase aqueous liquid-vapor inclusions (20 to 30% vapor) homogenize to the liquid state upon heating. They are the most common type of inclusions and have a maximum diameter of 45  $\mu\text{m}$ . Type 2 inclusions are three phase liquid-vapor inclusions (10 to 15% vapor) that contain a colourless, isotropic cubic daughter mineral that is likely to be halite (Fig. 2 d). Rarely a second daughter mineral exists, being probably sylvite. The type 2 inclusions are up to 50  $\mu\text{m}$  in diameter and homogenize to liquid by either disappearance of the vapor phase (type 2a) or dissolution of the daughter mineral (type 2b). The timing relationship (trapping time) between these two types of inclusions is unclear.

#### 4. Microthermometry results

Microthermometric results and compositional data

from the fluid inclusions are given in Table 1, and depicted in Figures 3 and 4. Homogenization temperatures of the fluid inclusions in syn-ore quartz from the Eptadendro and Rachi ore mineralization range from 275° to 422° C, with a peak at 330° C (Fig. 3a). First observable ice melting of fluids range from -21.6° to -20° C, suggesting that NaCl is the dominant salt component of the fluids (Crawford 1981; Shepherd et al. 1985). The final ice melting temperatures in the same inclusions range from -4.5° to -0.9° C, corresponding to salinities between 1.6 and 7.2 wt% NaCl equiv. Many inclusions in quartz can be grouped into FIAs, which have a constant liquid to vapor ratio (~25 %) and variable shapes, clustering along linear trends that do not cross cleavage or growth boundaries. FIAs showed restricted temperature ranges such as, 325°- 336° C, 331°-353° C and 344°-360° C, showing that the assemblages are true FIAs and, therefore, the inclusions probably represent the original trapping conditions and have not re-equilibrated.

Fluid inclusions in syn-ore calcite from the Epta-

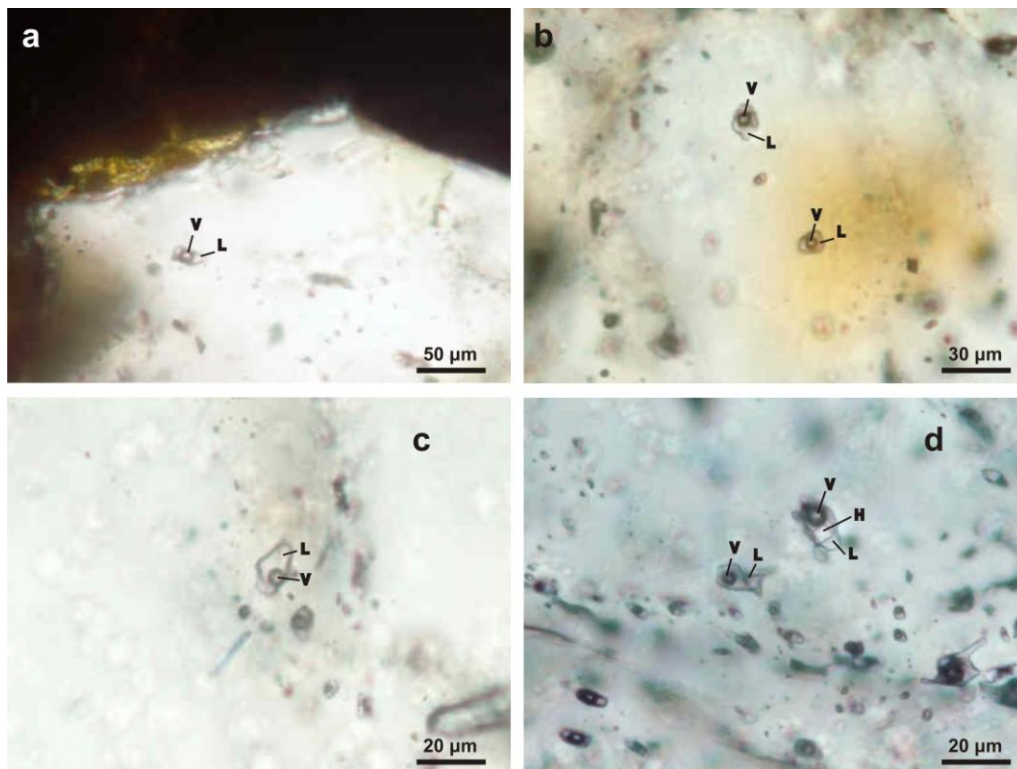


Fig. 2. Fluid inclusions in quartz from the sulfide mineralization at the Eptadendro-Rachi region. (a) Fluid inclusion of type 1 (two-phase aqueous inclusion) in quartz from the ore mineralization (sample E17). (b) Fluid inclusions of type 1 (two-phase aqueous inclusions) in quartz from the ore mineralization (sample E10). (c) Fluid inclusion of type 1 (two-phase aqueous inclusion) in quartz from the trondhjemite (sample F213). (d) Co-existing inclusions from the two types: two-phase aqueous inclusion (type-1) and three-phase halite-bearing aqueous inclusion (type 2). *L* Liquid, *V* vapor, *H* halite.

dendro and Rachi ore mineralizations exhibit  $T_h$  values between 154° and 257° C, with a peak at 240° C (Fig. 3a). These inclusions show final ice melting temperatures between -3.9° and -1.9° C, corresponding to salinities of 3.2 to 6.3 wt% NaCl equiv. An eutectic temperature of ~-21° C indicates that the dissolved salt is NaCl. Multiple populations of two-phase FIAs in calcite indicate restricted  $T_h$ : 224°-238° C, 230°-248° C, and 242°-256° C.

First observable ice melting of inclusions devoid of daughter crystals, in quartz of the pegmatite, range from -24.5° to -23.2° C, suggesting appreciable quantities of KCl in addition to NaCl (Shepherd et al. 1985). Final ice melting temperatures of type 1 inclusions range from -5.8° to -4.3° C (Table 1), which correspond to salinities of 6.9 to 8.9 wt% NaCl equiv. The homogenization temperatures ( $T_h$ ) of type 1 fluid inclusions in quartz of the pegmatite, range from 267° to 388° C (Table 1) and dis-

Table 1. Homogenization temperatures, melting temperatures and salinities of the fluid inclusions from the Eptadendro and Rachi copper sulfide mineralizations.

Sample	Petrography	Area	Host mineral	Fluid inclusion types (homogenize to phase)	Homogenization temp. (°C) Number of measurements (n) (mean) <sup>1</sup>	Last melting temp. (°C) Number of measurements (n) (mean) <sup>2</sup>	Salinity (wt% NaCl eq) (mean)
E10	Ore mineralization	Eptadendro	qtz	1 L+V→L	295 to 422 (91) (342)	-1.8 to -4.1 (48) (-3.1)	4.9 to 6.6 (5.5)
E17	Ore mineralization	Eptadendro	qtz	1 L+V→L	325 to 415 (64) (348)	-2.1 to -4.5 (22) (-3.6)	3.6 to 7.2 (5.9)
F225	Ore mineralization	Rachi	qtz	1 L+V→L	303 to 381 (26) (350)	-0.9 to -3.2 (26) (-2.2)	1.6 to 5.3 (3.8)
F275	Ore mineralization	Eptadendro	qtz	1 L+V→L	275 to 361 (26) (330)	-0.9 to -2.9 (26) (-1.9)	1.6 to 4.8 (3.3)
F230	Ore mineralization	Rachi	ca	1 L+V→L	154 to 257 (17) (229)	-1.9 to -3.9 (12) (-2.6)	3.2 to 6.3 (4.6)
F274	Ore mineralization	Eptadendro	ca	1 L+V→L	155 to 253 (15) (226)	-1.9 to -3.8 (11) (-2.9)	3.2 to 6.1 (4.8)
A7	Contact between trondhjemites and ultrabasics	Eptadendro	qtz	1 L+V→L	302 to 361 (72) (341)	-3.4 to -4.5 (36) (-4.0)	5.6 to 7.2 (6.5)
F213	Trondhjemite	Eptadendro	qtz	1 L+V→L	245 to 429 (53) (341)	-2.0 to -4.2 (22) (-3.0)	3.4 to 6.7 (5.0)
F132	Pegmatite	Eptadendro	qtz	1 L+V→L	267 to 388 (34) (341)	-4.3 to -5.7 (23) (-4.9)	6.9 to 8.9 (7.8)
				2 L+V+S→L	216 to 345 (22) (318)	248 to >600 (23) (445)	34.7 to 58.5 (53.4)

<sup>1</sup> The homogenization temperature corresponds to the temperature of the disappearance of the vapor bubble.

<sup>2</sup> The last melting temperature refers to the temperature of solid dissolution (> 248 °C) or last ice melting in a given inclusion; qtz quartz; ca calcite; l liquid phase, v vapor phase; eq equivalent; n number of microthermometric analyses; avg average; temp temperature

Values of  $T_h$  in the contact between trondhjemite and meta-ultrabasic rocks, as well as in the trondhjemites were obtained from fluid inclusions in quartz (245° to 429° C), with a peak at ~330° C (Fig. 3b). These temperatures overlap with the  $T_h$  values (290° to 390° C) from the syn-ore quartz. Eutectic ice-melting temperatures of inclusions range from -21.0° to -20.1° C, suggesting that the dissolved salt is only NaCl. Final ice melting temperatures range from -4.5° to -2.0° C (Table 1), which correspond to salinities of 3.4 to 7.2 wt% NaCl equiv. Several clusters of fluid inclusions demonstrate FIAs that homogenized between 323° and 342° C, as well as 337° and 356° C, which overlap with the values of  $T_h$  in ore-mineralization stage (Figs 3a,b).

play a bimodal distribution, with peaks at ~320° and ~370° C (Fig. 3c). Homogenization temperatures from 274° to >600° C were observed in the type 2 inclusions either by disappearance of the vapor bubble or dissolution of halite. Sylvite dissolution was observed between 174° and 226° C. Dissolution temperatures of halite in type 2 fluid inclusions (248° to 490° C) indicate a brine salinity of 34.6 to 58.4 wt% NaCl equiv. These data suggest that the hydrothermal fluids were saturated with respect to NaCl and KCl (Cline and Bodnar 1994).

## 5. Discussion

Regarding the genesis of the mineralization in the

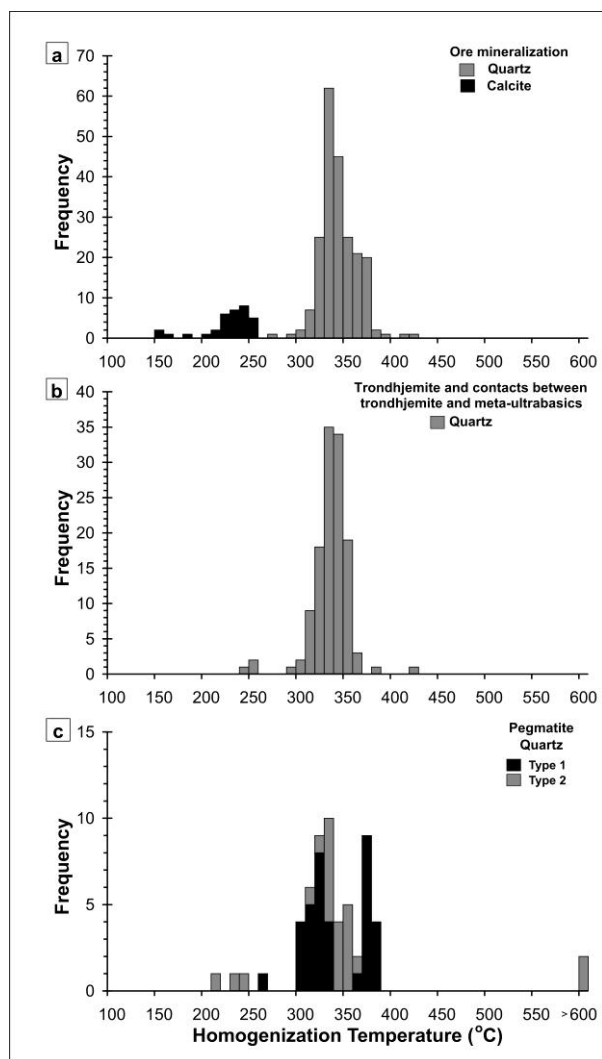


Fig. 3. Homogenization temperatures of fluid inclusions from the Eptadendro-Rachi region in the ore mineralization (a), in the trondhjemite and the contact between trondhjemite and meta-ultrabasic rocks (b) and in the pegmatite (c). *Type 1* two-phase liquid-rich aqueous inclusions. *Type 2* three-phase halite-bearing liquid-rich inclusions.

Eptadendro and Rachi areas, Chatzikirkou (2003) has suggested that the stratabound type mineralization hosted in the meta-ultrabasic-basic rocks is probably of Jurassic age and it was formed in a submarine environment. Seawater and to a lesser extent magmatic water circulating through faults and fissures of the ocean floor rocks has created a “hydrothermal convection system”, which depleted metallic components from the host rocks and deposited them into the ocean floor.

During Upper Cretaceous, the intrusion of granitoids, mainly trondhjemites, created the remobilization of a part of the stratabound mineralization and the formation of the vein-type mineralization

(Chatzikirkou, 2003). This is in agreement with the suggestion of Baziotis et al. (2008) that in the UHP Metamorphic Kimi Complex of East Rhodope, which is 25 km north east of the studied area, the amphibolitized eclogites are crosscut by tonalitic-trondhjemitic dykes of Early Tertiary times, at 65-63 Ma.

In addition, the magmatic fluids of these trondhjemites have added new amounts of mineralization with different elements. Consequently, all the types of mineralization have been affected at least by the greenschist facies metamorphic episode during the Eocene-Oligocene (Chatzikirkou, 2003; Chatzikirkou and Michailidis, 2004).

Fluid inclusions represent the only direct indication of ancient fluids in many crustal rocks. However, the rock may still have a completely different P-T evolution as fluid inclusion contents are seldom inert, and may have been subjected to post-trapping changes. Although the ores in the sulfide mineralization at the Eptadendro and Rachi areas are partly deformed and show some degree of recrystallization, the data obtained from the fluid inclusions demonstrate well the physical and chemical environment during the latest hydrothermal event caused by the intrusion of the trondhjemites during the Upper Cretaceous-Early Tertiary.

Based on the samples studied here, a plot of  $T_h$  values of fluid inclusions and FIAs versus salinity (Fig. 4a,b) suggests three groups, corresponding to the distinct fluids incorporated during the mineral deposition and the pegmatite formation. The first group of fluid inclusions hosted in syn-ore quartz is characterized by high values of  $T_h$  (300° to 380° C, with a peak at 330° C) and low salinities (1.6 and 7.2 wt% NaCl equiv) and represents the main ore stage which is associated with the intrusion of the trondhjemites.

The second group is distinguished by a drop in  $T_h$  (210° to 260 °C), associated with calcite formation, with similar salinities (3.2 to 6.3 wt% NaCl equiv) to the first group of inclusions. The composition of the fluids in these two stages remains the same, dominated by NaCl. The low salinities (1.6 to 7.2 wt% NaCl equiv) of the two ore stages can possibly be attributed to a mixing of ascending hydrothermal fluids and downward migrating dilute meteoric water. Such a process is very likely in a hydrothermal environment of shallow depth, with fault-controlled vein structure and causes ore deposition.

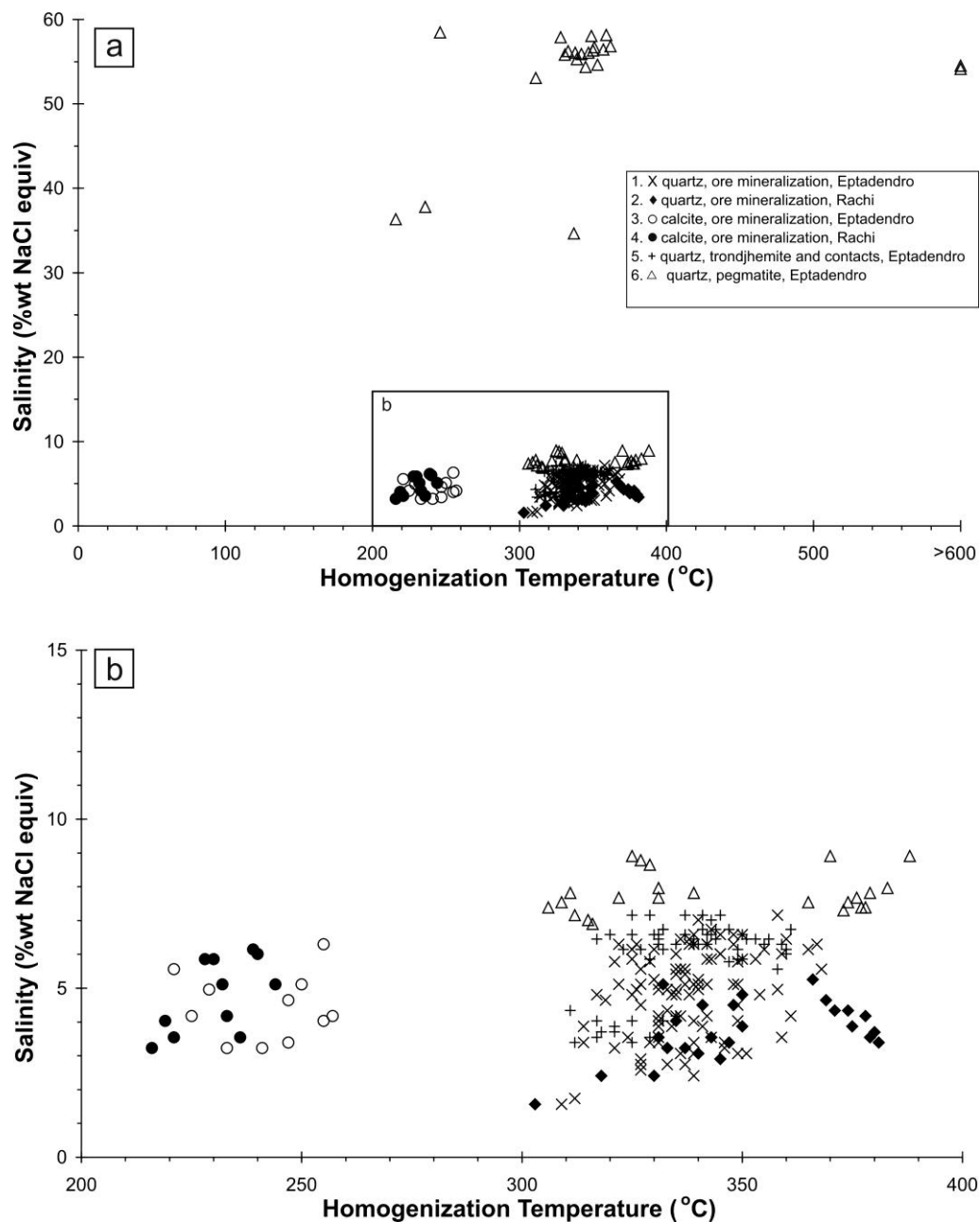


Fig. 4. Homogenization temperatures versus salinity plot for the fluid inclusions from the Eptadendro-Rachi ore mineralizations. Salinity was calculated using the equation of Potter et al. (1978) and Bodnar (1993).

The third group of fluid inclusions is related to the pegmatitic stage, which is free of mineralization. It is characterized by similar temperatures (300° to 390 °C) with the first ore stage, but variable salinities (6.9 to 8.9 wt% NaCl equiv and 34.7 to 58.5 wt% NaCl equiv). The composition of these fluids is dominated by NaCl and KCl documented by the first ice melting temperatures ( $T_e = -24.5^\circ$  to  $-23.2^\circ\text{C}$ ), as well as by the presence of a second daughter mineral in the hypersaline fluids, possibly sylvite, in addition to halite. It is therefore most probable that these fluids were not related to the

ore mineralization, although high salinity fluids can produce ore mineralization. They have a magmatic origin and are responsible for pegmatite formation, which is devoid of ore mineralization.

## 6. Conclusions

In the Eptadendro-Rachi ore district, a stratabound type mineralization hosted in meta-ultrabasic-basic rocks, probably of Jurassic age, was initially formed in a submarine environment. During Upper Cretaceous-Early Tertiary the intrusion of trondjemites partly remobilized this mineraliza-

tion and contributed hydrothermal fluids to the formation of a vein-type mineralization. This mineralization was at least affected by a greenschist facies metamorphic episode during the Eocene-Oligocene.

Fluid inclusion data on gangue syn-ore quartz indicate that the ore mineralization was derived from hydrothermal fluids with low salinity (1.6 and 7.2 wt% NaCl equiv). Values of  $T_h$  from individual FIA vary from 300° to 380° C, with a peak at 330° C. These fluids represent the main ore stage of the vein type, which is related to the intrusion of the trondhjemites. A lower temperature ore stage, associated with the syn-ore calcite formation, is distinguished by a drop in  $T_h$  (210° to 260° C) and having similar salinities (3.2 to 6.3 wt% NaCl equiv) as the first group of inclusions. The composition of the fluids in these two stages is dominated by NaCl. A probable mixing of ascending hydrothermal fluids and downward migrating dilute meteoric waters generated the low salinity fluids.

Fluid inclusions in pegmatite quartz are characterized by temperatures from 300° to 390° C and variable salinities, 6.9 to 8.9 wt% NaCl equiv and 34.7 to 58.5 wt% NaCl equiv. The composition of these fluids is dominated by NaCl and KCl. They have a magmatic origin but probably they are not related to the ore mineralization.

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