

GEOCHEMICAL AND THERMOBAROMETRIC CHARACTERISTICS OF FLUIDS ASSOCIATED WITH QUARTZ-SCHEELITE VEINS OF METAGGITSI-SALONIKIO, E. CHALKIDIKI PENINSULA

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ABSTRACT

Primary aqueous CO₂-bearing fluid inclusions with variable CO₂/H₂O volumetric ratios coexist in quartz and scheelite from vein systems hosted by two-mica gneisses and schists of the Vertiskos Formation in the Pirgadikia, Metagitsi-Salonikio area, E. Chalkidiki. Inclusions with low CO₂ volumetric percentage of 5-25 vol% homogenize to the H₂O-phase at temperatures between 230 and 325°C, whereas those with high CO₂ volumetric percentage of 55-85 vol% homogenize to the CO₂ phase at temperatures ranging from 220 to 331°C. Salinities are less than 10 equiv wt.% NaCl. These veins formed from boiling CO₂-laden fluids at temperatures of 250-400°C and pressures between 1 and 2 kbars during retrograde greenschist facies metamorphism.

ΣΥΝΟΨΗ

Πρωτογενή ρευστά εγκλείσματα περιέχοντα CO₂ με μεταβλητή αναλογία του CO₂ προς το H₂O κατ'όγκο, συνυπάρχουν σε χαλαζία και σεελίτη από συστήματα φλεβών που φιλοξενούνται μέσα σε διμαρμαρυγιακούς, γνευσίους και σχιστολίθους της σειράς του Βερτίσκου, στην περιοχή Πυργαδίκια-Μεταγγίτσι-Σαλονικιό στην Αν. Χαλκιδική. Εγκλείσματα με χαμηλή κατ'όγκο αναλογία σε CO₂ (5-25%) ομογενοποιούνται στην υδατινή φάση σε θερμοκρασίες μεταξύ 240-325°C, ενώ εκείνα με υψηλή κατ'όγκο αναλογία σε CO₂ (55-85%) ομογενοποιούνται στην φάση του CO₂ σε θερμοκρασίες 250-328°C. Αλατότητες είναι μικρότερες των 10% κ.β. ισοδύναμο NaCl. Οι φλέβες αυτές σχηματίστηκαν από αναβράζοντα διαλύματα πλούσια σε CO₂ κάτω από θερμοκρασίες 250-400°C και πιέσεις 1-2 kb κατά την διάρκεια ανάδρομης μεταμόρφωσης πρασινοσχιστολιθικού βαθμού.

ΕΙΣΑΓΩΓΗ - INTRODUCTION

Scheelite mineralization of the Pirgadikia-Metagitsi-Salonikio area in E. Chalkidiki Peninsula (Fig. 1) occurs within quartz veins and veinlets developed in two-mica gneisses and schists of the Vertiskos Formation. The Vertiskos, and Kerdilia Formation to the east, comprise the Paleozoic or older (Kockel et al, 1977) Sercomacedonian Massif (Fig. 1). Intense exploration for tungsten in the area was carried out during the years 1963-1972 by various mining companies, and IGME since 1984 (Veranis and Bitzios, 1984).

Σ.Π. ΚΙΛΙΑΣ. Γεωχημικά και θερμοβαρομετρικά χαρακτηριστικά ρευστών που σχετίζονται με φλέβες χαλαζια-σεελίτη της περιοχής Μεταγγίτσι-Σαλονικιό, Α. Χαλκιδική

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Ψηφιακή Βιβλιοθήκη "Θεόφραστος" - Τμήμα Γεωλογίας, Α.Π.Θ.

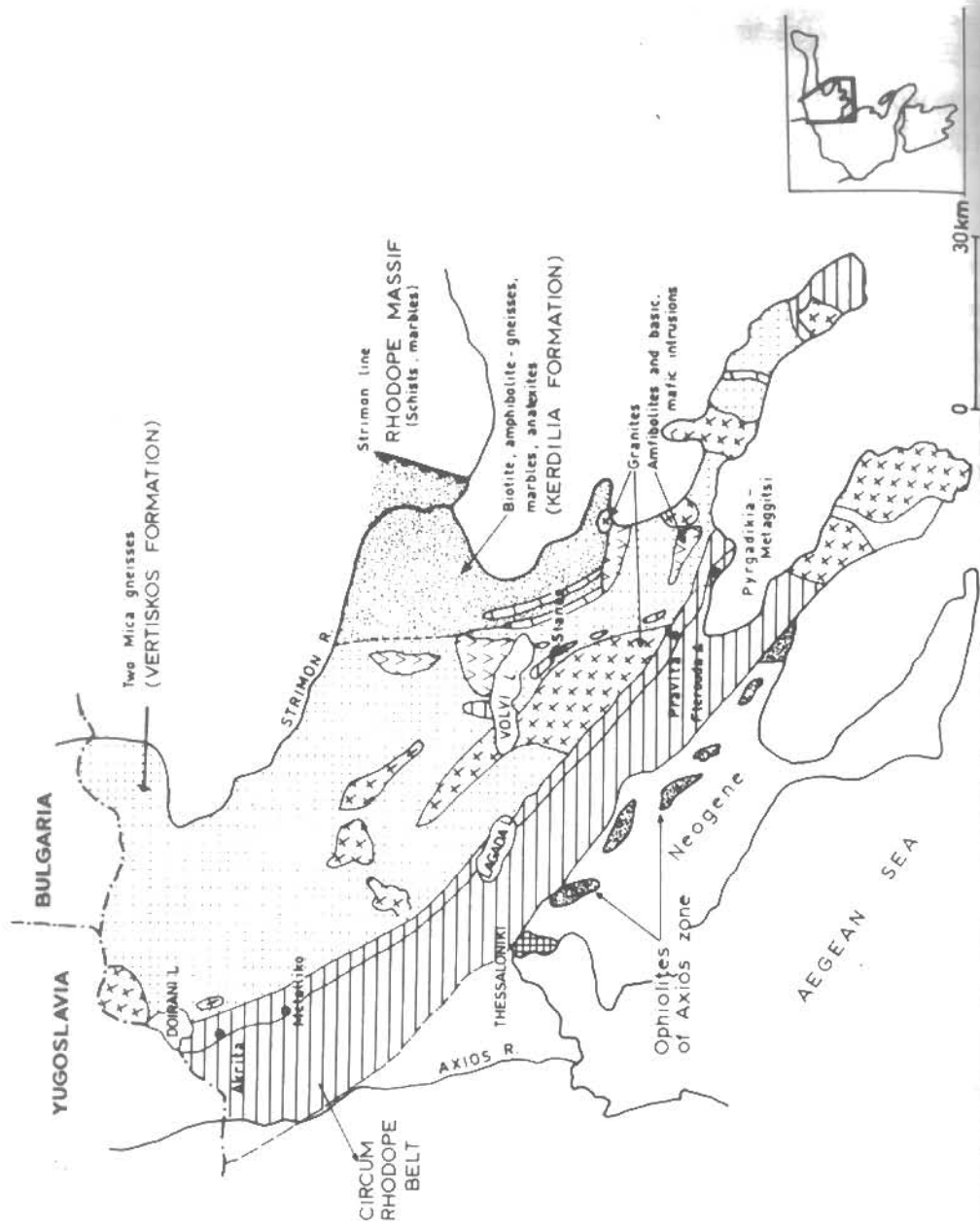


Fig. 1. Geotectonic map of the Serrovomacedonian Massif (Veranis, 1989, modified after Kockel et al., 1977). Γεωτεκτονικός χάρτης της Σερρομακεδονικής ζώνης (Kockel et al., 1977, συμπληρωμένος από Veranis, 1989).

This paper reports fluid inclusion data in an attempt to decipher the geochemical and thermobarometric characteristics of the fluids responsible for the genesis of the quartz-scheelite veins in the area.

ΓΕΝΙΚΗ ΓΕΩΛΟΓΙΑ - GENERAL GEOLOGY

Vertiskos formation is a heterogeneous assemblage consisting of lithological units of the Upper-Paleozoic to Triassic Perirhodope Zone to the west (Fig. 1), two-mica gneisses/schists, amphibolites, portions of metaophiolitic units, granitic bodies (i.e. Arnea), and few occurrences of marble.

These rocks are considered to be of volcanosedimentary, sedimentary, and igneous parental origin (Fournaraki, 1981; Papadopoulos, 1982; Dixon and Dimitriadis, 1985; Kougoulis, 1986; Kougoulis et al., 1987). Regarding the deformation-metamorphic history of this heterogeneous rock assemblage, and despite recent developments (Patras et al., 1986; Dixon and Dimitriadis, 1985; Sakellariou, 1988) a complete and thorough description of the successive events is not possible at this stage. However, K/Ar dating of amphiboles from amphibolites and gneisses (E. Simos, 1989, writ. commun.) gives an upper limit of 95-110 m.y. (Upper Cretaceous) for the regional amphibolite facies metamorphism (T=550°C, P=4-5 kb) event (which was followed by retrograde greenschist facies metamorphism) which must have commenced in Jurassic since it has also affected rocks of Upper Paleozoic-Lower Mesozoic age (S. Kaloogeropoulos, pers. commun.).

Vertiskos Formation in the area of Metaggitsi-Salonikio where scheelite mineralization occurs is underlain by E-W trending two-mica gneisses of possible para-origin, metabasic rocks of igneous origin, tourmaline-and/or garnet-bearing mica schists, cherts with banded iron formation, and a limited number of dykes of granitic composition (Veranis and Bitzios, 1984) (Fig. 2).

Veranis and Bitzios (1984) have observed two major stages of deformation (D₁ and D₂), and two minor secondary (D₃ and D₄), of probable Alpine age in the area: D₁ is characterized by numerous closed and recumbent isoclinal folds with b₁ axes plunging 0 to 15° from NW to SE. D₂ is accompanied by S-type fracture, strain-slip or crenulation cleavage and microfolds. This second stage is also characterized by folding with b₂ axes parallel to b₁ plunging to higher angles of 0-60°, suggesting that the two deformation stages belong to the same tectonic process.

Two types of faults have been observed in the area: (a) normal, and (b) reverse, trending E-W and dipping less than 45°. Shear zones in the area

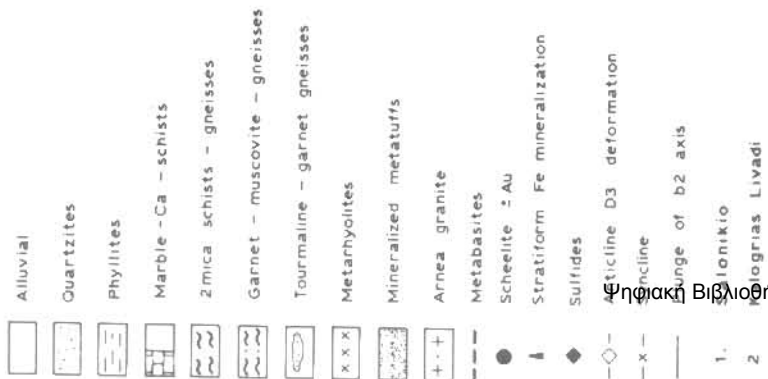
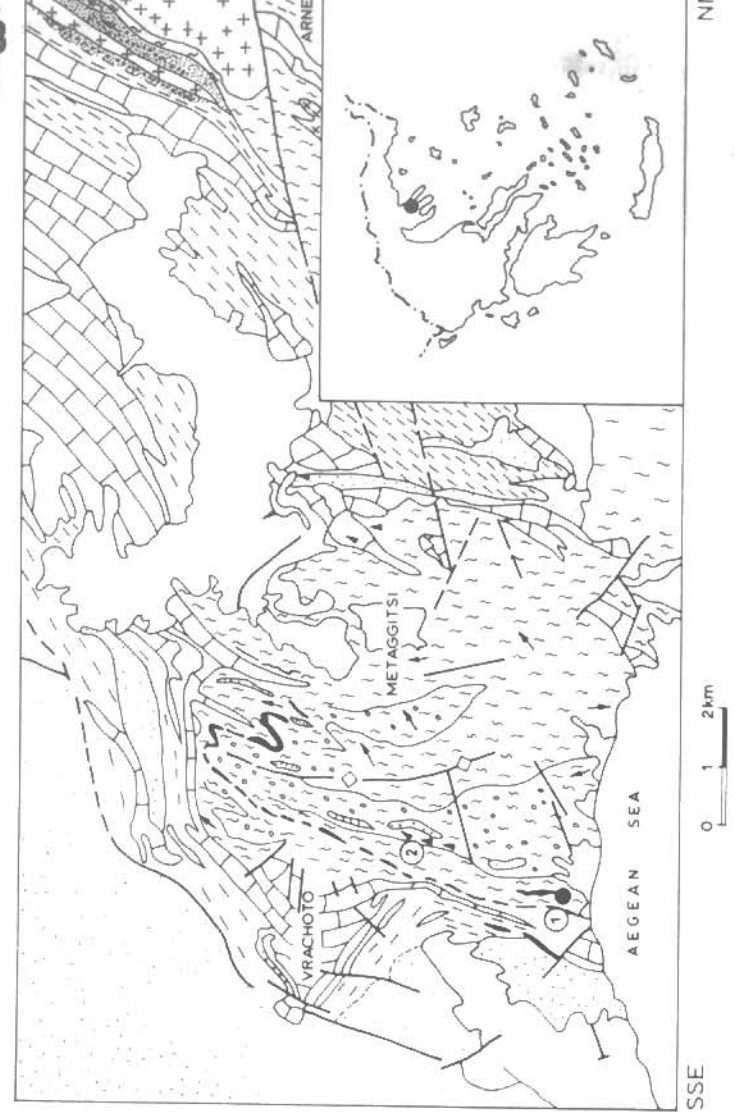


Fig. 3. Geological map of S.E. Chalkidiki Peninsula (Veranis and Bitzios, 1984, modified after Kockel et al., 1977) Γεωλογικός χάρτης της Σερβομακεδονικής Ζώνης (Kockel et al., 1977, τροποποιημένος από Βεράνη και Μπιτζίο, 1984)

are probably due to the second (Bitzios, 1984).

Scheelite mineralization in the Salonikio area is contained within systems of quartz veins and veinlets trending from NE to E and dipping 50 to 70° from NW to N. They follow the S-type fracture cleavage which are related to the D₂ deformation event. The veins are hosted by two-mica gneisses and schists, biotite/epidote schists, and locally metabasites. The veins have sharp contacts and measure up to 1 km length. The width of the veins varies from a few cm to 1 m.

Scheelite is the main tungsten-bearing mineral, accompanied by traces of powellite, and stolzite. The two wolframates are associated with traces of arsenopyrite, molybdenite, and goethite, and gangue calcite and white mica.

ΜΕΛΕΤΗ ΡΕΥΣΤΩΝ ΕΓΚΛΕΙΣΜΑΤΩΝ - FLUID INCLUSION STUDY

Fluid inclusions were studied in double-polished chips of 200-300 μ thickness from vein quartz samples. Microthermometric measurements were conducted on carefully selected fluid inclusions using a temperature programmable Linkam TH600 heating-freezing system (Shepherd, 1981). The stage was calibrated in the range -100 to 600°C using various melting point standards according to the method of Macdonald and Spooner (1981). The uncertainty in the measurements of the temperatures of the phase changes described is ±0.5°C.

On the basis of the microscopy and heating-freezing studies four types of fluid inclusions in quartz and scheelite were distinguished :

- Type A, mixed CO₂-H₂O inclusions (CO₂, 5-25 vol.%; modal value 10 vol.%) with low salinity (\bar{x} : 3.8 equiv. wt% NaCl) and average bulk density of 0.93 g/cm³ (Fig. 3A).
- Type B, mixed CO₂-H₂O inclusions (CO₂, 55-85 vol.%, modal value 80 vol.%) with low salinity (\bar{x} : 2.1 equiv. wt% NaCl), and average bulk density 0.88 g/cm³ (Fig. 3A,B).
- Type C, pure CO₂ inclusions with average bulk density 0.80 g/cm³ (Fig. 3C).
- Type D, pure aqueous inclusions (gas 5-20 vol.%) (Fig. 3D).

Inclusion types A, B, and C are generally small (10-50 μ in the longest dimension), and they are either randomly distributed in the host crystal or occupy healed micro-fractures. Inclusion types A, B, and C with no obvious planar distribution which were considered primary (Roedder, 1984) and co-genetic with the host mineral and each other, were exclusively studied. Type D inclusions

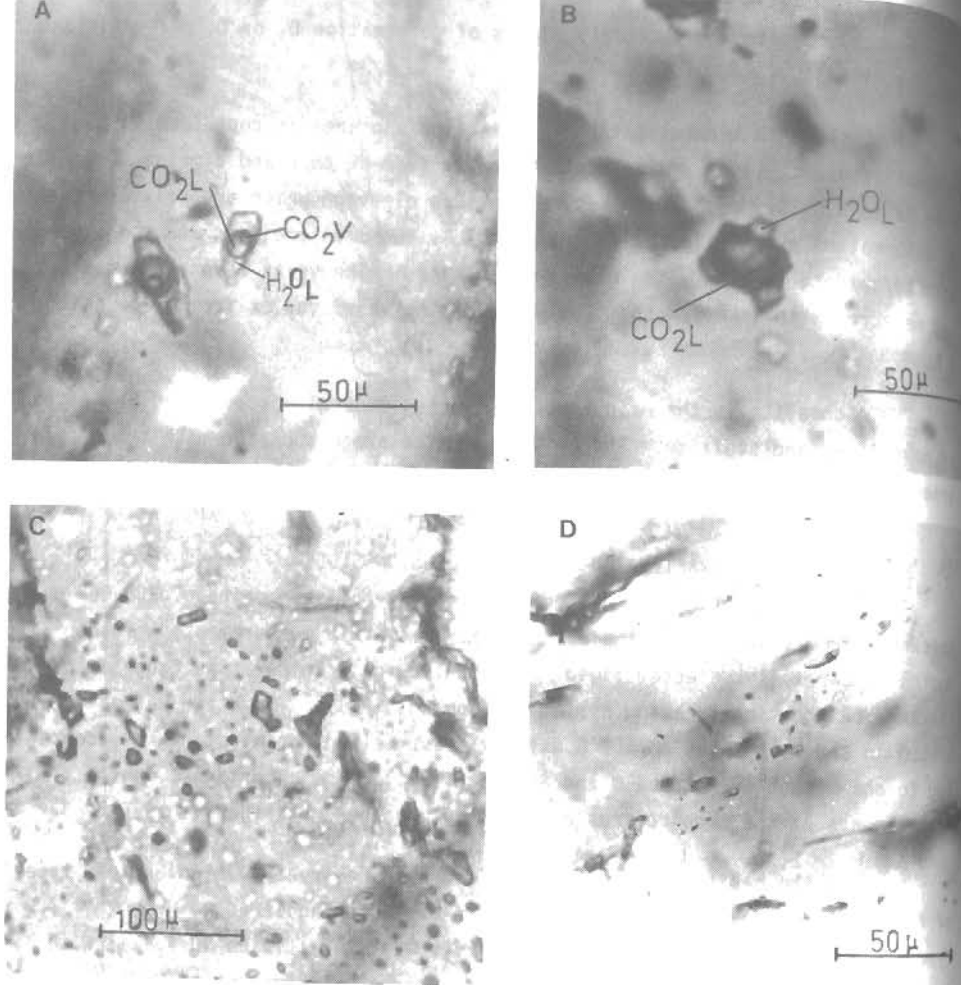


Fig. 3. Fluid inclusion types.

- Coexisting primary A and B type inclusions. CO₂ (liquid+vapor)-phase representing about 25% (A), and about 70% (B) of the inclusion volume (CO₂L:CO₂ liquid; CO₂V:CO₂ vapor; H₂O:L:H₂O liquid).
- Representative primary B type inclusion. CO₂-phase occupying more than 70% of the inclusion volume.
- Three-dimensional cluster of monophasic C type inclusions. CO₂ (liquid)-phase representing 100% of the inclusion volume.
- Healed fracture containing secondary D type aqueous inclusions.

Εικ. 3. Τύποι ρευστών εγκλεισμάτων

- Συνοπάρχοντα πρωτογενή εγκλείσματα τύπου Α και Β. Η φάση του CO₂ (υγρό και αέριο) καταλαμβάνει περίπου 25% (Α), και περίπου 70% (Β) του όγκου του εγκλείσματος (CO₂L:CO₂ υγρό; CO₂V:CO₂ αέριο; H₂O:L:H₂O υγρό).
- Αντιπροσωπευτικό πρωτογενές εγκλείσμα τύπου Β. Η φάση του CO₂ καταλαμβάνει περισσότερο από 70% του όγκου του εγκλείσματος.
- Τρισδιάστατη κατανομή μονοφασικών εγκλεισμάτων τύπου C. Η φάση του CO₂ (υγρή) καταλαμβάνει το 100% του όγκου του εγκλείσματος.
- Επουλωμένη ρωγμή περιέχουσα δευτερογενή εγκλείσματα τύπου D.

were not found outside extensive bands that occupy healed microfractures, are considered secondary and non-related to the tungsten-mineralizing event, and will be not dealt further.

ΣΤΟΙΧΕΙΑ ΚΡΥΟΜΕΤΡΙΑΣ - FREEZING DATA

The CO₂ phase in type A, B, and C inclusions melted at temperatures between -57.5 and -56.3°C, indicating that the non-condensable gas in these inclusions is CO₂ with traces of CH₄ (Swanenberg, 1979; Heyen et al., 1982). The CO₂ clathrate melted at temperatures between 6.8 and 10°C (average value, 8.0°C) (Fig. 5A), and 5.5-10°C (average value 8.5°C) (Fig. 5B) for inclusion types A and B respectively.

Salinities of the inclusion fluids, expressed in terms of equivalent wt percent NaCl were estimated in the system H₂O-CO₂-NaCl from the melting temperature of clathrate (Fig. 5) (Collins, 1979).

CO₂ molar composition is calculated in the system H₂O-CO₂-NaCl from the density of the CO₂ fluid phase (Anquas et al., 1976), salinity and density of the aqueous liquid phase (Potter and Brown, 1977) combined with estimates of volume proportions of phases observed in fluid inclusions using equations given by Nicholls and Crawford (1985).

ΣΤΟΙΧΕΙΑ ΘΕΡΜΟΜΕΤΡΙΑΣ - HEATING DATA

The CO₂ phases in type A inclusions homogenized at temperatures between 18.4 and 28°C (average value, 23.6°C), the gaseous phase disappearing (Fig. 6A). The CO₂ and aqueous phases homogenized into a liquid (aqueous) phase at temperatures in the range 230-325°C (average value, 270°C) (Fig. 7A). The CO₂ phases in type B inclusions homogenized into a liquid CO₂ phase at temperatures between 10 and 24°C (average value, 17.3°C) (Fig. 6B). The total homogenization of these inclusions occurred with the disappearance of the liquid (aqueous) phase at temperatures in the range 220-331°C (average value, 292°C) (Fig. 7B). A large number of type A and B inclusions decrepitated before total homogenization denoting high internal pressures. The CO₂ phases in type C inclusions homogenized into the liquid CO₂ phase at temperatures between 6.5 and 23.9°C (average value, 15.6°C) (Fig. 6C). The secondary type D inclusions homogenized into a liquid phase at temperatures in the range 110-135°C (average value, 115°C). Freezing and heating data are summarized in Table 1.

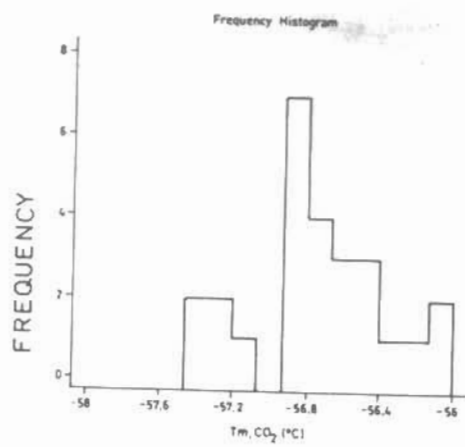


Fig. 4. Histogram of CO₂ melting temperatures, T_m, CO₂, of inclusion types A and B.
Εικ. 4. Ιστογράμμο θερμοκρασιών τήξης του CO₂, T_m, CO₂, εγκλεισμάτων τύπου Α και Β.

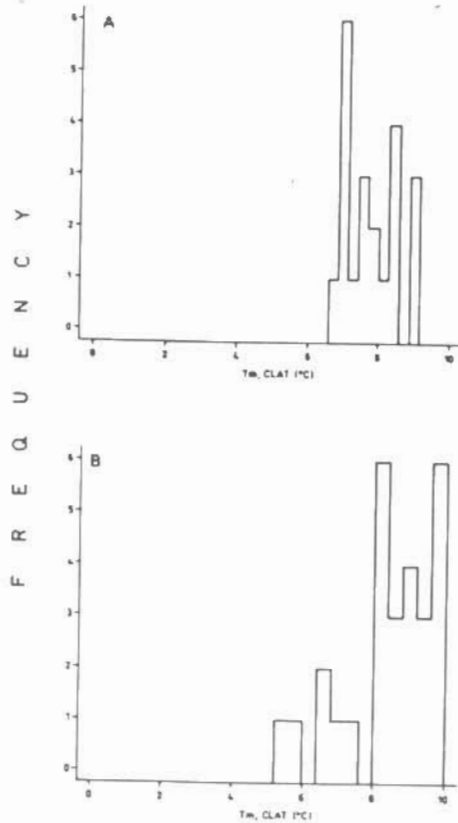


Fig. 5. Histograms of CO₂-hydrate (clathrate) melting temperatures T_m, CLAT of inclusion types A (5A) and B (5B).
Εικ. 5. Ιστογράμμοτα θερμοκρασιών τήξης του clathrate, T_m, CLAT, εγκλεισμάτων τύπου Α (5Α) και Β (5Β).

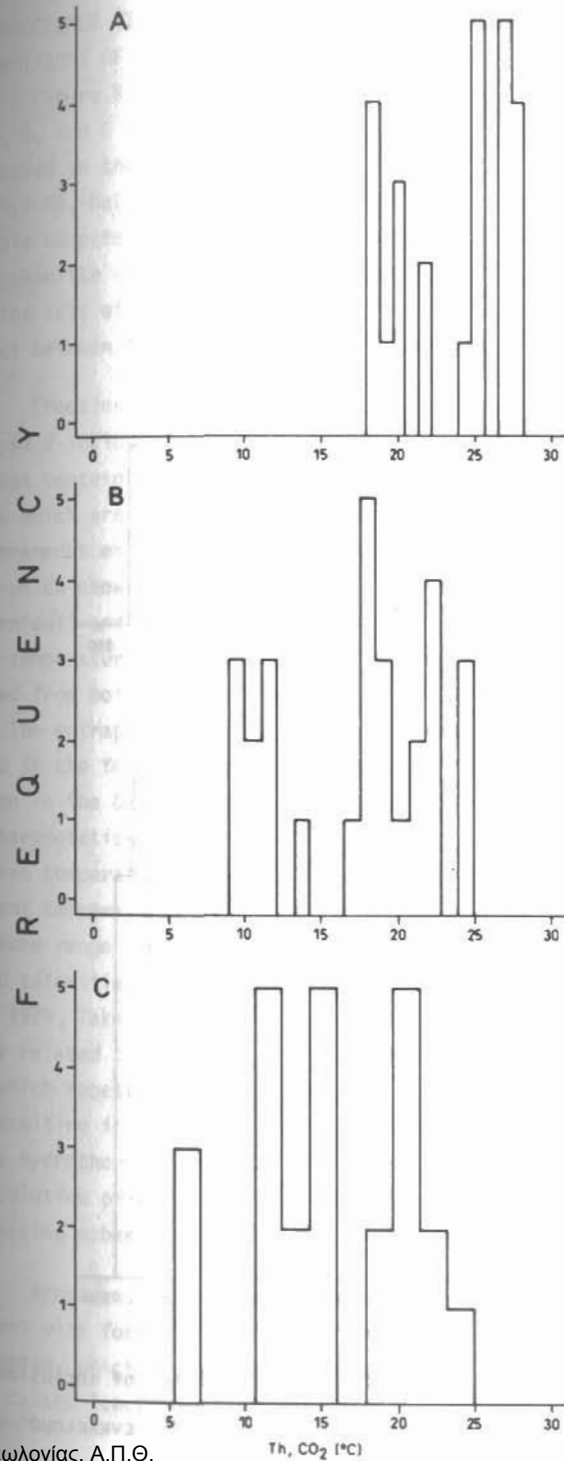


Fig. 6. Histograms of CO₂-phase (liquid and vapor) homogenization temperatures, T_h, CO₂, of inclusion types A (6A), B (6B) and C (6C).
Εικ. 6. Ιστογράμμοτα θερμοκρασιών ομογενοποίησης της φάσης του CO₂ (υγρό και αέριο), T_h, CO₂, εγκλεισμάτων τύπου Α (6Α), Β (6Β) και C (6C).

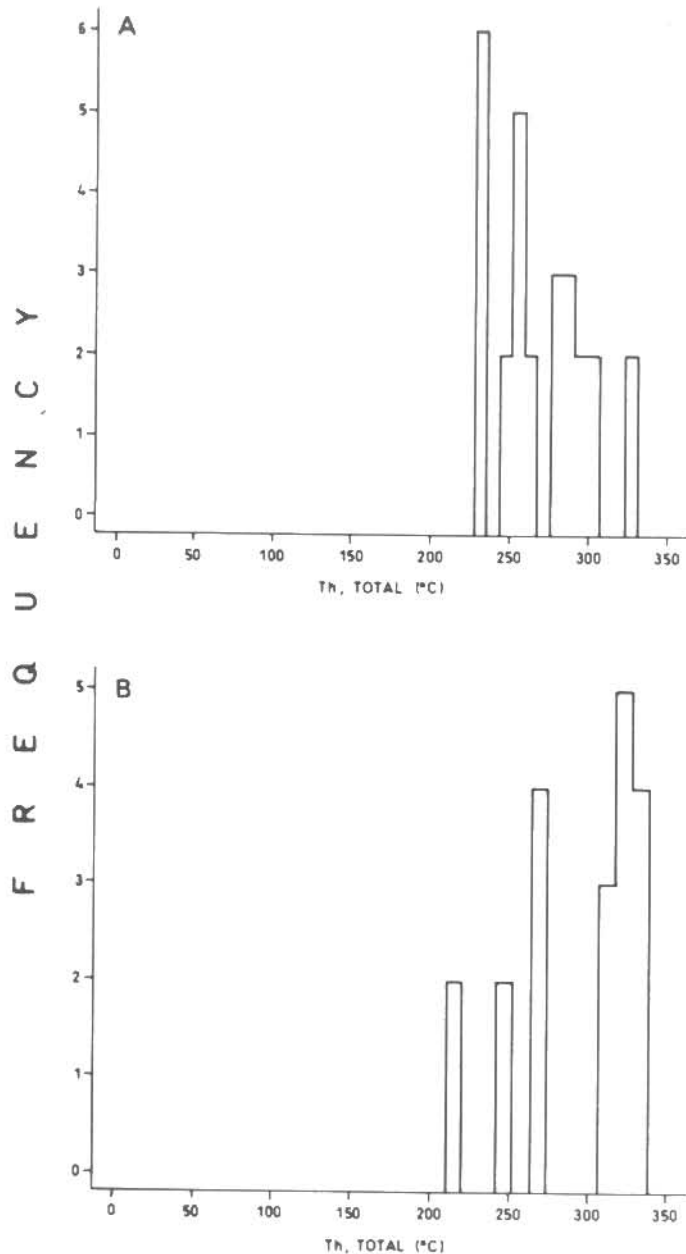


Fig. 7. Histograms of total homogenization temperatures T_h , TOTAL, of inclusion types A and B (7B).
 Εικ. 7. Ιστογράμματα θερμοκρασιών ολικής ομογενοποίησης T_h , TOTAL, ενκλεισμάτων τύπου A και B (7B).

Figure 8 shows groups of calculated isochores of selected inclusion types A, B, and C with molar volumes (types A and B) and densities (type C) as shown, based on the microthermometry data and calculated properties in the system H_2O-CO_2-NaCl and H_2O-CO_2 , with the use of Fortran programs in an IBM compatible computer (Nicholls and Crawford, 1985). Common fluid entrapment, during scheelite deposition, is indicated by the intersection of the three corresponding sets of calculated isochores at temperatures of 250-400°C and pressures between 1 and 2 kbars as indicated by the shaded area of Figure 8.

Freezing studies have shown that type A inclusions contain 3-12 mole% CO_2 . Type B inclusions are richer in CO_2 and contain 20-45 mole% CO_2 . Type C inclusions contain almost pure CO_2 . The apparently primary inclusion types A, B, and C which are cogenetic are characterized by highly variable composition. Contemporaneous entrapment of inclusions, with large variation in the CO_2/H_2O ratios, which show both homogenization in the CO_2 fluid phase (type B, high CO_2 molar content) and the aqueous phase (type 1, low CO_2 molar content), at almost similar temperatures (Table 1), suggests that the fluids in these inclusions were entrapped from boiling hydrothermal solutions at the above mentioned P-T conditions. The entrapment of different proportions of two immiscible fractions resulted in the formation of a large number of mixed inclusion which exhibit variation in the CO_2/H_2O ratio and salinity as in inclusion types A, B, and C. This interpretation is supported by the large range of overlapping final homogenization temperature observed for type A and B inclusions which coincide with entrapment temperatures indicated by intersecting isochores, and the wide temperature range over which CO_2 and saline H_2O are immiscible for pressure around 1 kb and salinities up to 6 equiv. wt% NaCl (Todheide and Franck, 1963; Gehrig et al., 1979, Takenouchi and Kennedy, 1965; Bodnar et al., 1985). Boiling is probably related to the tectonic development of the scheelite-bearing veins during which repeated hydraulic fracturing caused significant pressure fluctuations resulting in CO_2-H_2O unmixing. In addition CO_2 separation from the low salinity hydrothermal fluid may have caused rapid cooling and had a marked effect on the solution pH with a shift from weakly acid to weakly alkaline at around 300°C causing scheelite deposition (Higgins, 1985).

Pressure, temperature and composition of the mineralizing fluids are consistent with formation during the last stages of retrograde greenschist facies metamorphism, which probably either remobilized preexisting tungsten mineralization or caused leaching of tungsten from the surrounding rocks, and deposition in

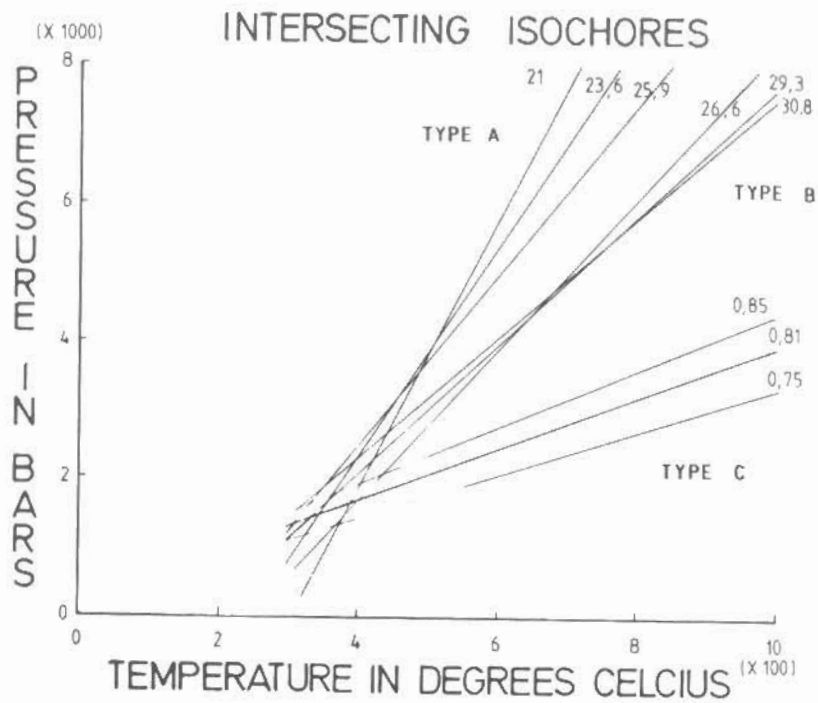


Fig. 8. Calculated isochores of selected type A, B, and C inclusions. Numbers correspond to molar volumes (cm^3/e) (Types A, B), and densities (g/cm^3) (Type C).

Εικ. 8. Υπολογισμένες ισοχωρές ευθείες από επιλεγμένα εγκλείσματα τύπου Α, Β, και C. Οι αριθμοί αντιστοιχούν σε μοριακούς όγκους (cm^3/e) (τύπος Α και Β), και πυκνότητες (g/cm^3) (τύπος C).

TABLE 1. Microthermometry data and properties of inclusion types A, B, C; and D.
ΠΙΝΑΚΑΣ 1. Στοιχεία μικροθερμομετρίας και ιδιότητες εγκλεισμάτων τύπου Α, Β, C, και D.

INCLUSION TYPE	MELTING TEMPERATURES T_m, CO_2 (°C)		APPARENT SALINITY (EQUIV WT% NaCl)	HOMOGENIZATION TEMPERATURES T_h, CO_2 (°C)		CO_2 MOLE% T_h, TOTAL	DENSITY BULK (g/cm^3)
	R	X		R	X		
A	R: -56.3 to -57.5 X: -56.7 ± 0.36 M: -56.8	R: 6.8-10 X: 8.0 ± 0.98 M: 7	R: 0.0-6.12 X: 3.8 ± 1.10 M: 3.1	R: 18.4-28.0 X: 23.6 ± 3.60 M: 28	R: 230-325 X: 270 ± 28 M: 260	R: 3-17 X: 9 ± 4 M: 13	R: 0.81-0.99 X: 0.93 ± 0.03 M: 0.92
B	R: 5.5-10 X: 8.5 ± 1.2 M: 10	R: 0.0-8.3 X: 2.2 ± 1.0 M: 2	R: 10.0-24.0 X: 17.3 ± 4.8 M: 21.9	R: 220-331 X: 292 ± 37 M: 318	R: 20-58 X: 33 ± 10 M: 25	R: 0.83-0.92 X: 0.88 ± 0.02 M: 0.91	R: 0.83-0.92 X: 0.88 ± 0.02 M: 0.91
C	-	-	R: 6.5-23.9 X: 15.6 ± 5.1 M: 14.6	-	100	R: 0.75-0.88 X: 0.80 ± 0.04 M: 0.75	R: 0.75-0.88 X: 0.80 ± 0.04 M: 0.75
D	-	R: 10-15 X: 11 ± 2 M: 12	-	R: 110-135 X: 115 ± 5 M: 115	-	R: 1.04-1.08 X: 1.04 ± 0.04 M: 1.04	R: 1.04-1.08 X: 1.04 ± 0.04 M: 1.04

¹Data are from Figures 4, 5, 6, and 7; R=range, X=average, ±=standard deviation

²Melting temperatures of CO_2 refer to both inclusion types A and B.

T_m, CO_2 : Melting temperature of solid CO_2

T_m, CLAT : Melting temperature of CO_2 -hydrate (clathrate)

T_h, CO_2 : CO_2 -phase (liquid and vapor) homogenization temperature

T_h, TOTAL : Total homogenization temperature of the CO_2 - and aqueous- phases

the veins. Hydrothermal solutions rich in CO₂ are associated with tungsten deposits in various geologic environments many of which contain evidence of a heterogeneous CO₂-H₂O hydrothermal fluid (e.g. Shepherd et al., 1976; Kelly and Rye, 1979; Cheilletz, 1984), and CO₂ is known to play a very important role in transportation of tungsten in solution in carbonic complexes, and its deposition (Ivanova and Khodakovskii, 1968; Higgins and Kerrich, 1982; Higgins, 1985).

Recognition of such fluids accompanied with deposition mechanisms, such as boiling, based on fluid inclusion studies, constitute very important evidence which may be used in exploration for tungsten deposits.

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