

FE-CR SPINEL AND ILMENITE MASSIVE MINERALIZATION IN METAMORPHIC ULTRAMAFICS FROM THE ASKOS AREA, NORTHERN GREECE

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ABSTRACT

The development of Fe-Cr-spinels and ilmenite mineralization in the ultramafic rocks of the Askos area is studied. Textural and mineralogical features, showed that these rocks have undergone regional metamorphism and recrystallization to upper greenschist-lower amphibolite facies. Meta-ultramafics are characterized by the mineral assemblages: antigorite+Fe-Cr-spinels+ilmenite+chlorite+talc+tremolite and traces of carbonates and quartz. Extensive metasomatic reaction zones of chlorite-, talc-, tremolite- and vermiculite-rich rocks have been developed between the contacts of the ultramafics and the country rocks. Fe-Cr-spinels exhibit usually a zonal texture consisting of: aluminian chromite core, ferritechromite intermediate zones and Cr-magnetite rims. They are considered as the final product of a multistage metasomatic process on primary chromite enclosed in the ultramafic protolith. This process started during the serpentinization and advanced to the present state by the regional metamorphism overprint.

ΣΥΝΟΨΗ

Μελετάται η ανάπτυξη μεταλλοφορίας Fe-Cr-σπινέλλων και ιλμενίτη μέσα στα υπερβασικά πετρώματα της περιοχής Ασκού. Τα ιστολογικά και ορυκτολογικά χαρακτηριστικά έδειξαν ότι αυτά τα πετρώματα έχουν υποστεί γενική μεταμόρφωση και ανακρυστάλλωση στην ανώ πράσινοσχιστολιθική-κάτω αμφιβολιτική φάση. Τα μετα-υπερβασικά χαρακτηρίζονται από τις παραγενέσεις: αντιγορίτης+Fe-Cr-σπινέλλοι+ιλμενίτης+χλωρίτης+τρεμολίτης και ίχνη ανθρακικών και χαλαζία. Εκτεταμένες ζώνες πλούσιων σε χλωρίτη, τάλκη, τρεμολίτη και θερμικουλίτη πετρωμάτων έχουν αναπτυχθεί από μετασωματικές αντιδράσεις στις επαφές των υπερβασίτων με τα γειτονικά πετρώματα. Οι Fe-Cr-σπινέλλοι συνήθως παρουσιάζουν ζωνώδη δομή που αποτελείται από: αργιλιούχο χρωμιτικό πυρήνα, ενδιάμεσες ζώνες οξείδωχρωμίτη και περιθωρια Cr-μαγνητίτη. Θεωρούνται ως το τελικό προϊόν μιας πολλαπλού σταδίου μετασωματικής διεργασίας πάνω στον πρωτογενή χρωμίτη που εγκλειόταν στον υπερβασικό πρωτόλιθο. Αυτή η διεργασία άρχισε κατά τη serpentinίωση και προχώρησε στη σημερινή μορφή με την προσθετική δράση της γενικής μεταμόρφωσης.

K. M. ΜΙΧΑΗΛΙΔΗΣ. Συμπύληση μεταλλοφορία Fe-Cr-σπινέλλων και ιλμενίτη στα μεταμορφωμένα υπερβασικά πετρώματα της περιοχής Ασκού, Βόρειο Θεσσαλονίκης, Β. Ελλάδα.

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1. INTRODUCTION

The variation in composition of chromian spinels during progressive regional and contact metamorphism of ultramafic rocks has been studied by many investigators (SPRINGER 1974; EVANS and FROST 1975; FROST 1975; PINSANT and HIRST 1977; MATTHES and KNAUER 1981 and others). Besides, the alteration of chromite to ferritechromite and the development of optically zoned crystals attracted many studies up to now and different views have been proposed to explain the conditions of this transformation (e.g. GOLDING and BAYLISS 1968; BEESON and JACKSON 1969; ENGIN and AVCOTT 1971; MITRA 1972; PETERS and KRAMERS 1974; SPRINGER 1974; BLISS and MACLEAN 1975; HAMLIN 1975; PINSANT and HIRST 1977; PARASKEVOPOULOS and ECONOMOU 1981; TAKLA 1982; MICHAELIDIS 1982, 1990; ZAKRZEWSKI 1989 and others). The term "ferritechromit" was first used by SPANGENBERG (1943) to describe, not a mineral of specific composition, but an alteration product of chromite having intermediate colour, reflectivity and chemistry between chromite and magnetite.

In most of these studies chromian spinel constitutes an accessory mineral of the ultramafic protolith. It is associated with small amounts of secondary magnetite which formed during the serpentinization of the silicate minerals (olivine, pyroxenes). Apart from these cases massive magnetite mineralizations, associated or not with chromite, are rarely found in Alpine-type ultramafics and have been described by few authors (COMPAGNONI et al. 1980; GHOSE 1980; PARASKEVOPOULOS and ECONOMOU 1980; ZHELYASKOVA-PANAYIOTOVA et al. 1980).

This paper deals with the textural features, chemistry and origin of Fe-Cr spinels and ilmenite in the massive mineralization cropping out within the regionally metamorphosed ultramafic rocks of the Askos area, Northern Greece.

The interpretation of the P,T metamorphic conditions of the meta-ultramafics is also attempted on the basis of textural and mineralogical features.

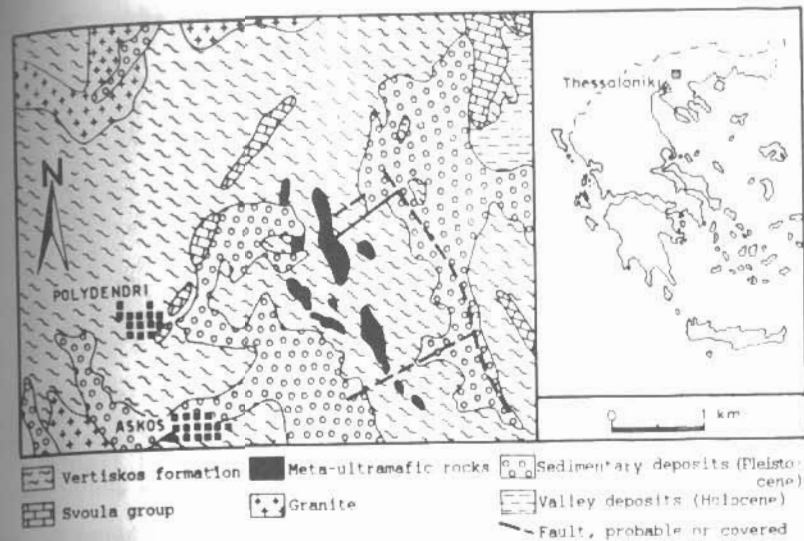


Fig. 1. Simplified geological map of the Askos area based on the Sochos sheet geological map 1:50,000.

Σχ. 1. Απλοποιημένος γεωλογικός χάρτης της περιοχής Ασκος με βάση το γεωλογικό φύλλο Σόχος 1:50.000.

2. GEOLOGICAL SETTING

The Askos area (Fig. 1) belongs to the upper Vertiskos series of the Serbo-Macedonian massif (KOCKEL et al. 1971, 1977), which consists mainly of two mica gneisses and amphibolites alternating with gneisses to the west and an extensive sequence of amphibolites to the east. Intrusive magmatic rocks of Mesozoic age accompanied with pegmatite- and quartz-sills and dykes are widespread in the studied area. Serpentinized ultramafic rocks form concordant lensoidal or elongated bodies within the gneisses. Their size ranges from 100m to more than 1.000m along the longer axis and 50 to 200m along the shorter axis. The contacts between the rock units are always sharp and sheared. Thermal aureoles are absent.

Extensive metasomatic zones of chlorite-, talc-, tremolite- and vermiculite-rich rocks have been developed at the contacts between serpentinite bodies and country rocks (DABITZIAS and PERDIKATIS 1990), or along sheared zones within the serpentinites. Thin bands, layers, stringers or stocks and nodular massive bodies of Fe-Cr spinel and ilmenite mineralizations were found hosted in the meta-ultramafics.

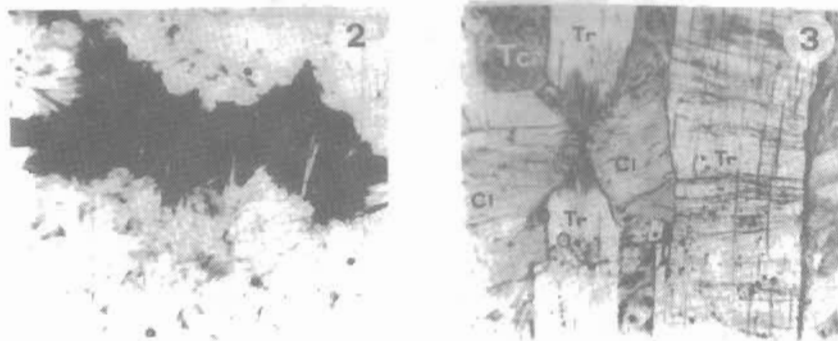


Fig. 2 and 3. 2: Radiating crystals of chlorite (white) as a matrix of the oxide mineralization (black). 3: Tremolite crystals (Tr) replaced by chlorite (Cl) and talc (Tc). Transmitted light, // Nicols, X40.

Σχ. 2. και 3. 2: Ακτινωτής διατάξης κρύσταλλοι χλωρίτη (λευκό), ως κύρια μάζα της μεταλλοφορίας οξειδίων (μαύρο). 3: Κρύσταλλοι τρεμολίτη (Tr) εκτοπίζονται από χλωρίτη (Cl) και τάλκη (Tc). Διερχόμενο φως, // Nicols, X40.

3. PETROGRAPHIC AND MINERALOGIC DESCRIPTION

The ultramafic protoliths suffered hydrothermal metamorphism and Alpine regional metamorphism followed by solid-state deformation which produced massive to schistose antigoritic serpentinites. Excluding the relict chromite cores in the zoned Fe-Cr spinels primary textures and mineralogical relics of the protolith were not preserved. Besides, the mineralogy of the rocks has been locally changed as a result of metasomatism. The following mineral assemblages are found within the studied meta-ultramafics: antigorite+Fe-Cr spinels+ilmenite+chlorite+talc+tremolite and traces of carbonates and quartz.

Massive antigorite serpentinite consists of feathery to coarse fibrolamellar fringes of antigorite. An obvious layering is produced by the concentration of talc flakes at sites of the rock near the metasomatic zones. Chlorite forms massive aggregates of coarse platy to elongate crystals. In many cases radiating clusters of the mineral are found as a matrix of the mineralization (Fig. 2). Talc is observed as a prograde metamorphic mineral forming coarse

large flakes, as well as a retrograde secondary finegrained mineral (TROMMSDORF and EVANS 1972; SPRINGER 1974). Tremolite forms prismatic megacrysts exceeding 10cm along or small acicular crystals. Tremolite crystals are intensely bent due to deformation. In most cases tremolite formation seems to precede the formation of talc and chlorite (Fig. 3).

Fe-Cr spinel and ilmenite (Fe-Cr-Ti oxides) mineralizations form usually massive bodies varying in size from some cm³ to 0.01m³ and show fracturing, folding and deformation. Besides, crystals of these minerals are sometimes aligned as small stringers suggesting a kind of layering. Rhythmic layering of the Fe-Cr-Ti oxides with the silicate minerals is also usually seen. Fe-Cr spinels in the massive type mineralization usually exhibit optical zonation (Fig. 4). Four spinel phases were optically distinguished from reflectivity and colour differences:

(a) altered chromite core.

(b)-(c) ferritechromite zone - magnetite rim around the chromite (in some cases (b) and (c) zones are indistinguishable).

(d) individual magnetite crystals.

Magnetite is the most widespread oxide mineral and appears in polygonal granoblastic aggregates with nearly 120° triple grain boundaries (Fig. 5), a characteristic feature of recrystallization during metamorphism. Chromite cores observed in magnetite crystals constitute a remnant of the ultramafic protolith which survived metamorphism. These cores have characteristically rounded shape (Fig. 4) which indicates that the alteration of primary chromite advances from all directions.

Euhedral megacrysts (blasts) of magnetite (octahedrons) up to 5cm across are found in a chlorite matrix.

Magnetite is characterized by the absence of lamellar ilmenite oxidation exsolutions. However, composite grains of magnetite+ilmenite are observed as a result of intimate intergrowth. Incipient martitization is discernible around grain boundaries and along cleavage planes. The martite lamellae are

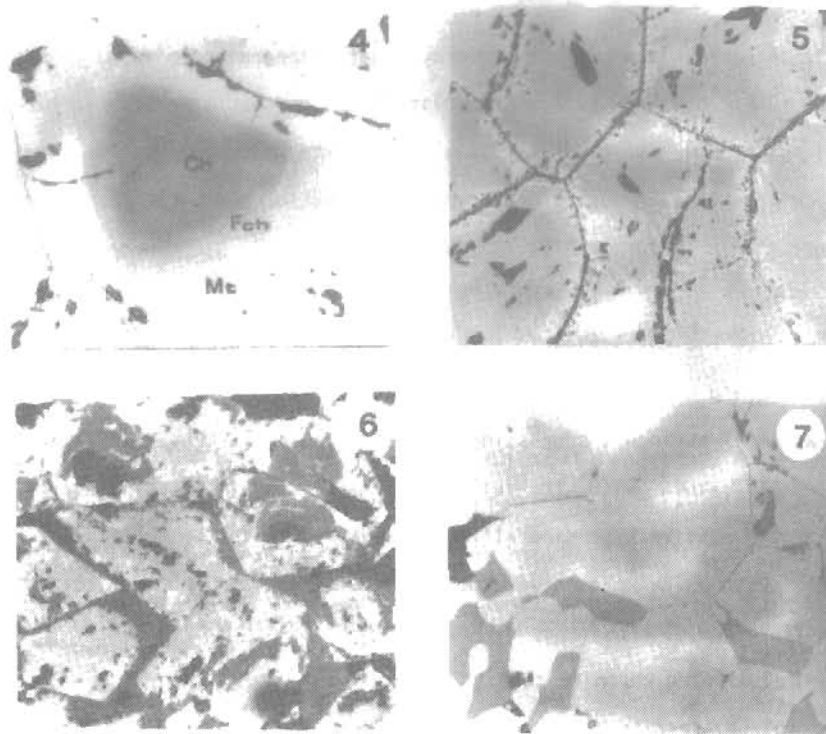


Fig. 4 to 7. 4: Zoned Fe-Cr spinel consisting of a chromite core (Ch) a ferritechromite (Fch) intermediate zone and a magnetite (Mt) rim. 5: Polygonal granoblastic aggregates of magnetite (grey white) showing martite lamellae (white) // to (111) planes. 6: Magnetite crystals (grey white) with incipient martitization (white), partly converted to goethite (gray). 7: Ilmenite (grey) as an interstitial phase between magnetite crystals (grey white). Incident light, oil immersion, X150.

Σχ. 4 ως 7. 4: Ζωνώδης Fe-Cr-σπινελλίος που αποτελείται από χρωμιτικό πυρήνα (Ch), ενδιάμεση ζώνη σιδηροχρωμίτη (Fch) και εξωτερικό περιθώριο μαγνητίτη (Mt). 5: Πολυγωνικά συσσωματώματα κρυστάλλων μαγνητίτη (τεφρόλευκο) με ελάσματα αιματίτη (λευκό) // στα (111) επίπεδα. 6: Κρυστάλλοι μαγνητίτη (τεφρόλευκο) με αρχική μαρτιτίωση (λευκό) που έχουν μερικώς μετατραπεί σε γκαϊτίτη (τεφρό). 7: Ιλμενίτης (τεφρό) ως διακρυσταλλική φάση μεταξύ των κρυστάλλων μαγνητίτη (τεφρόλευκο). Ανακλώμενο φως, ελαιοκατάδυση, X150.

predominantly oriented parallel to the (111) planes (Fig. 5) of the spinel. Magnetite from weathered outcrops is usually partly converted to goethite (Fig. 6). Ilmenite forms anhedral grains which are usually found as an interstitial phase between the Fe-Cr spinel crystals (Fig. 7). Discrete grains of the mineral are also found in a silicate matrix.

4. Methods of investigation.

Rock and ore samples were collected from different sites of the meta-ultramafics and some 50 of them were studied under the petrographic and the ore microscope. X-ray powder diffraction patterns were obtained for 10 samples to determine the serpentine "polymorph(s)" and to aid in the identification of fine-grained mineral phases.

Energy dispersive analyses of the mineral assemblages in the meta-ultramafics were performed in a CAMECA CAMEBAX electron microprobe at the Geological department of the Manchester University, England. Operating conditions were 15KV accelerating potential and 3nA beam current. Natural minerals or pure metals were used as standards. Element concentrations have been calculated with a ZAF-4/FLS software.

The Fe-Cr spinels in study exhibit usually an optical zoning (see previous section). Besides, cryptic zoning was always detected on the spinel crystals. For this reason step scanning analyses were carried out to show the chemical variation of both optically and cryptically zoned Fe-Cr spinels.

The spinel analyses were recalculated to structural formulae assuming perfect spinel stoichiometry ($RO:R_2O_3=1$). Ilmenite formula was assumed to be stoichiometric and calculated from the microprobe analyses on the basis of 4 cations with as much Fe^{2+} converted to Fe^{3+} as is necessary to bring the oxygen total to 6. In the resulting structural formulae Fe^{2+} , Ni and Ca are combined and represent the ilmenite component. Magnesium and Mn represent the geikielite and pyrophanite components respectively. The combined Fe^{3+} , Cr and Al represent the hematite component. Microprobe analyses of amphiboles were recalculated for the estimation of Fe^{3+} and Fe^{2+} cations using the normalization 13 cations except Ca, Na and K (13cCNK).

5. MINERAL CHEMISTRY

Electron microprobe analyses performed on the main minerals of the meta-ultramafics from the Askos area are tabulated in Tables 1,2 and 3 in the form of compositional range limits.

Fe-Cr spinels

A gradual decrease in Cr_2O_3 , Al_2O_3 , MgO and increase in FeO_{total} , NiO was always traced from the chromite core to ferritechromite-magnetite rim in the zoned Fe-Cr spinels (Table 1). The MnO content in the altered chromite cores is unusually very high (2.47-3.51%), it is higher in the ferritechromite zone (3.25-4.25%) but decreases in the magnetite rim (0.53-1.64%). The TiO_2 content

Table 1. Electron microprobe analyses (range limits) of Fe-Cr-spinels from the Askos area.

Πίνακας 1. Αναλύσεις με ηλεκτρονικό μικροαναλυτή (όρια κύμανσης) Fe-Cr-σπινελλίων από την περιοχή Ασκού.

	Chromite core (15)*	Ferritechromite zone (10)*	Magnetite rim (12)*	Magnetite crystals (35)*
SiO ₂	0.28-0.57	0.28-0.40	0.14-0.47	0.08-0.58
TiO ₂	0.13-0.42	0.09-0.53	0.06-0.53	0.03-1.00
Al ₂ O ₃	4.74-7.31	0.01-1.53	0.01-0.22	0.03-0.20
Cr ₂ O ₃	52.33-56.11	18.07-49.45	7.80-14.58	1.06-13.24
Fe ₂ O ₃ **	4.58-9.76	16.56-50.08	53.17-60.77	54.91-67.48
FeO	25.43-27.66	26.36-28.48	28.27-29.31	28.34-30.46
MnO	2.19-3.51	3.25-4.25	0.53-1.64	0.08-1.24
MgO	1.10-3.07	0.77-1.30	0.43-1.01	0.15-0.92
NiO	0.10-0.46	0.30-0.94	0.30-0.95	0.24-0.96
CaO	0.02-0.05	0.01-0.11	0.05-0.14	0.01-0.17
Total	100.00	100.00	100.00	100.00

* Number of analyses-Αριθμός αναλύσεων.

**Estimated from total iron by assuming perfect spinel stoichiometry-Υπολογίστηκε από τον ολικό σίδηρο θεωρώντας το σπινέλλιο με τέλεια στοιχειομετρία.

shows almost the same range limits in the three areas of the zoned spinels with an abundance up to 0.53%. The Cr₂O₃ content of the discrete magnetite crystals being very high (1.06-13.24%) shows always a decrease from the core to the margins, while the NiO content (0.24-0.96%) shows usually an opposite trend. The MnO content is relatively lower than in magnetite of optically zoned crystals (0.08-1.24%), while TiO₂ content was found between 0.03 and 1% (only one analysis 1.84%).

Compositional variations of chrome spinels are best illustrated by projections within the spinel prism, according to IRVINE'S (1965) method (Fig. 8 and 9). In the Cr X 100/(Cr+Al) vs Mg X 100/(Mg+Fe²⁺) face of the spinel prism (Fig. 8a) the studied Fe-Cr spinels plot outside of both the compositional fields and towards the low Mg/Mg+Fe²⁺ side. In the Fe³⁺X100/R³⁺ vs MgX100/(Mg+Fe²⁺) diagram (Fig. 8b) the chromite cores show Fe³⁺/R³⁺ ratios lower than 0.10. Ferritechromites show higher than 0.10 with a trend towards the magnetite composi-

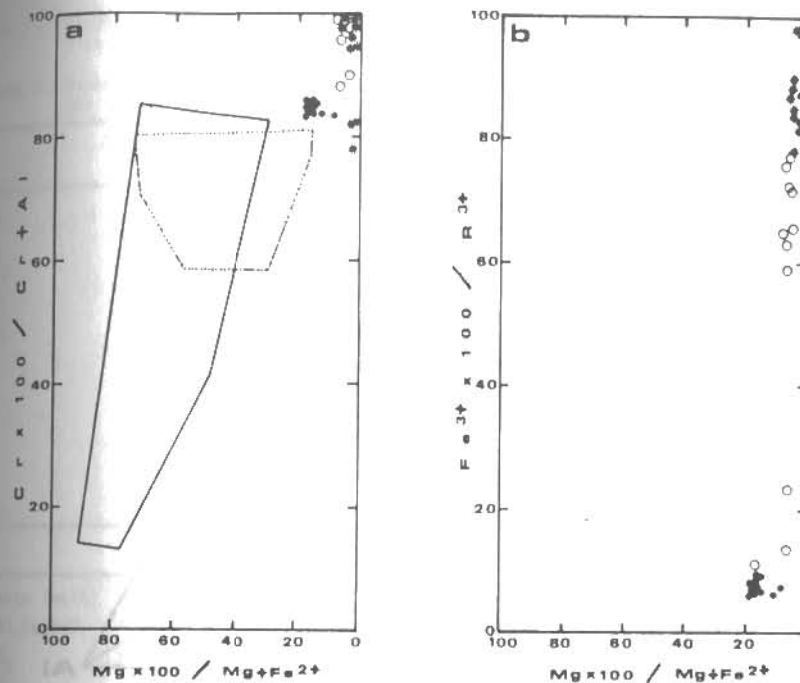


Fig. 8. Variation of: (a) CrX100/(Cr+Al) vs MgX100/(Mg+Fe²⁺) and (b) Fe³⁺X100/R³⁺ vs MgX100/(Mg+Fe²⁺) ratios of the Askos area Fe-Cr spinels. Dots are for chromite cores, open circles for ferritechromites and stars for magnetites. Fields for chromian spinels from alpine (solid line) and stratiform (dotted line) complexes after IRVINE and FINDLAY (1972) are also given.

Σχ. 8. Μεταβολή των λόγων: (α) CrX100/(Cr+Al) προς MgX100/(Mg+Fe²⁺) και (β) Fe³⁺X100/R³⁺ προς MgX100/(Mg+Fe²⁺) στους Fe-Cr-σπινελλίους της Περιοχής Ασκού. Οι τελείες είναι για τους χρωμιτικούς πυρήνες, οι κύκλοι για τους σιδηροχρωμίτες και οι αστέρισκοί για τους μαγνητίτες. Δίνονται επίσης τα πεδία σύστασης των χρωμιούχων σπινελλίων από αλπικού (συνεχής γραμμή) και στρωματόμορφου (γραμμή με τελείες) τύπου συμπλέγματα κατά τους IRVINE and FINDLAY (1972).

tion. In the trivalent cation projection (Fig. 9) the chromite cores plot within the field of aluminian chromite, ferritechromites within the ferrian chromite and the chromian magnetite while magnetites within the chromian magnetite field.

Ilmenite.

The chemical composition of the studied ilmenite confirms that the mineral constitutes a solid solution of the end members FeTiO₃ (ilmenite), MgTiO₃ (geikielite) and MnTiO₃ (pyrophanite). Ilmenite compositions are variable within individual samples of different mineral associations (Table 2):

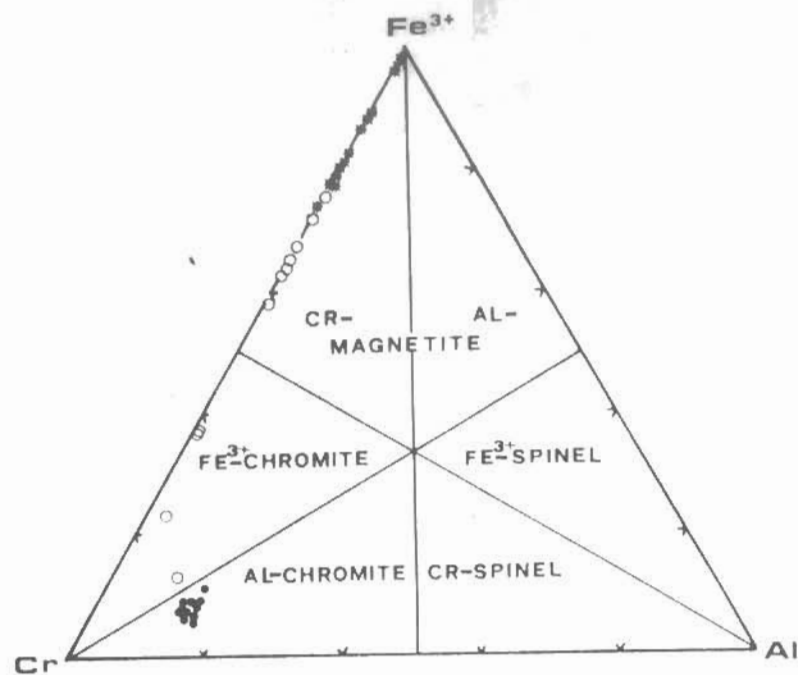


Fig. 9. Distribution of the Askos area Fe-Cr spinels in the Cr-Al-Fe³⁺ triangular classification of STEVENS (1944). Explanation of symbols as for fig. 8.

Σχ. 9. Κατανομή των Fe-Cr-σπινελλίων της περιοχής Ασκού στο τριγωνικό διάγραμμα Cr-Al-Fe³⁺ ταξινόμησης του STEVENS (1944). Συμβολισμός όπως στο σχήμα 8.

Figures 10a-b illustrate the compositional range of ilmenite expressed as: a) ternary percentages of (Fe, Ni, Ca) TiO₃, MgTiO₃ and MnTiO₃ molecules and b) ternary percentages of (Fe, Ni, Ca) TiO₃, MgTiO₃ and (Fe³⁺, Cr, Al)₂O₃ molecules.

Different groups of ilmenites can be distinguished on the basis of the MgTiO₃ and MnTiO₃ component contents (Fig. 10a). Ilmenites associated with chlorite or chlorite+tremolite show the highest MgTiO₃ component content, while those associated with antigorite the lowest one. On the contrary ilmenites have the highest MnTiO₃ component content in association with antigorite or chlorite+tremolite and the lowest in chlorite.

Table 2. Electron microprobe analyses and molecular percentages (range limits) of ilmenites from the Askos area in different silicate matrix.

Πίνακας 2. Αναλύσεις με ηλεκτρονικό μικροσυνάλι και μοριακές αναλογίες (όρια κλίμακας) ιλμενιτών από την περιοχή Ασκού, σε διαφορετική κυρία μάτσα πυριτικών.

	Antigorite matrix (8)*	Chlorite matrix (7)*	Antigorite+Chlorite matrix (6)*	Chlorite+Tremolite (13)*
SiO ₂	0.20-0.47	0.00-0.28	0.15-0.46	0.22-0.40
TiO ₂	51.30-53.40	52.57-53.78	52.40-53.58	51.85-53.23
Al ₂ O ₃	0.00-0.08	0.00-0.22	0.06-0.10	0.01-0.13
Cr ₂ O ₃	0.12-0.47	0.15-0.37	0.25	0.11-0.20
Fe ₂ O ₃	0.00-2.52			0.95-4.26
FeO	34.25-35.08	36.88-38.33**	36.95-38.93**	30.37-33.50
MnO	9.47-10.55	4.28-4.86	5.37-7.75	8.31-9.95
MgO	0.59-1.60	3.18-4.36	1.11-2.07	3.46-4.05
NiO	0.00-0.32	0.11-0.34	0.18-0.69	0.00-0.39
CaO	0.00-0.11	-	-	0.00-0.09
Total	99.35-100.99	99.09-99.99	99.54-100.55	99.91-100.93
Molecular percentages				
Ilmenite (mol%)	92.77-99.72	92.81-98.31	94.85-99.68	98.90-96.89
(Fe,Ni,Ca)TiO ₃	73.72-74.85	74.07-79.06	77.03-81.96	64.43-69.41
MnTiO ₃	20.28-23.08	9.06-10.37	11.48-16.64	17.71-20.98
MgTiO ₃	2.28-6.01	11.88-16.08	4.21-9.40	12.82-15.30
Hematite (Fe,Al,Cr) ₂ O ₃	0.28-7.23	1.69-7.19	0.37-5.15	3.11-11.10

*Number of analyses - Αριθμός αναλύσεων

**Total iron as FeO - Ολικός σίδηρος ως FeO.

Coexisting magnetite and ilmenite

Data on the partitioning of elements between coexisting mineral phases provide insight into the attainment of chemical equilibrium by the mineral assemblages (RUMBLE 1979). The partitioning of elements between ilmenite and magnetite is here emphasized. Therefore, electron microprobe analyses performed on coexisting magnetite and ilmenite grains in the same rock sample and adjacent to each other serve to compare the substitution behavior of the elements into these mineral phases. From Fig. 11a to d results that:

(a) there is a preferential concentration of MgO into ilmenite (note the 1:1 ratio line). This behavior is consistent with the experimental data of PINCKNEY and LINDSEY (1975) and the observations of GASPAR and WYLLIE (1983).

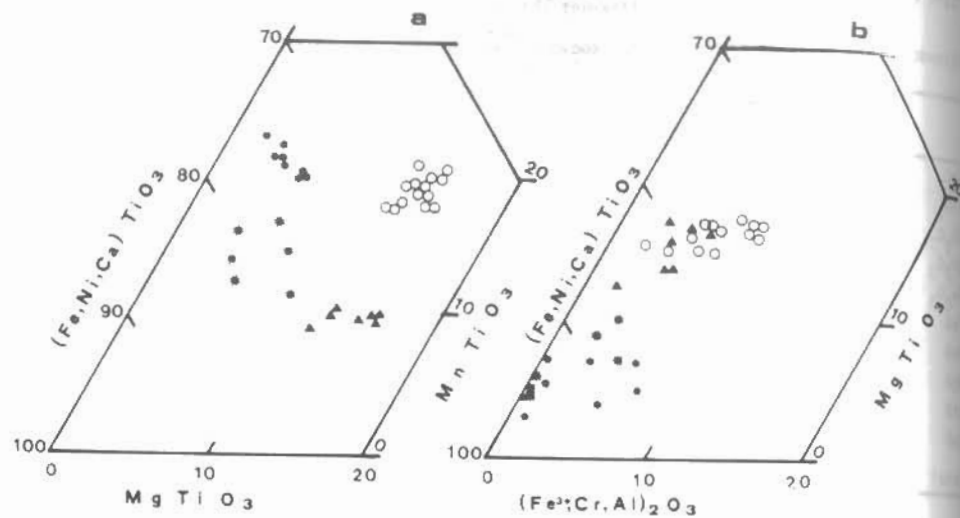


Fig. 10. Ilmenite composition of the Askos area expressed as ternary percentages of the molecules: (a) $(\text{Fe,Ni,Ca})\text{TiO}_3 - \text{MnTiO}_3 - \text{MgTiO}_3$ and (b) $\text{MgTiO}_3 - (\text{Fe,Ni,Ca})\text{TiO}_3 - (\text{Fe}^{3+},\text{Cr,Al})_2\text{O}_3$. Dots are for ilmenite in a matrix of antigorite, circles in chlorite+tremolite, stars in chlorite+antigorite and triangles in chlorite.

Σχ. 10. Σύσταση του ιλμενίτη της περιοχής Ασκού εκφρασμένη σε τριαδικά διαγράμματα μοριακών αναλογιών: (α) $(\text{Fe,Ni,Ca})\text{TiO}_3 - \text{MnTiO}_3 - \text{MgTiO}_3$ και (β) $\text{MgTiO}_3 - (\text{Fe,Ni,Ca})\text{TiO}_3 - (\text{Fe}^{3+},\text{Cr,Al})_2\text{O}_3$. Οι τελείες είναι για ιλμενίτη σε κύρια μάζα αντιγορίτη, οι κύκλοι σε χλωρίτη+τρεμολίτη, οι αστερίσκοι σε χλωρίτη+αντιγορίτη και τα τρίγωνα σε χλωρίτη.

(b) there is a preference of Mn for the ilmenite structure as it is also stated by NEUMANN (1974), HAGGERTY (1976) and GASPARD and WYLLIE (1983). Ilmenite is strongly enriched in Mn relative to magnetite, even, in cases where magnetite surrounds the chromite-ferrite-chromite cores of high MnO-content. This preference shows that Mn as well as Mg fit better into the ilmenite, than into the magnetite structure.

(c) on the contrary Ni and Cr show a preference for the magnetite lattice. This regular partitioning of the elements confirms an at least local chemical equilibrium in the meta-ultramafics during the metamorphic recrystallization.

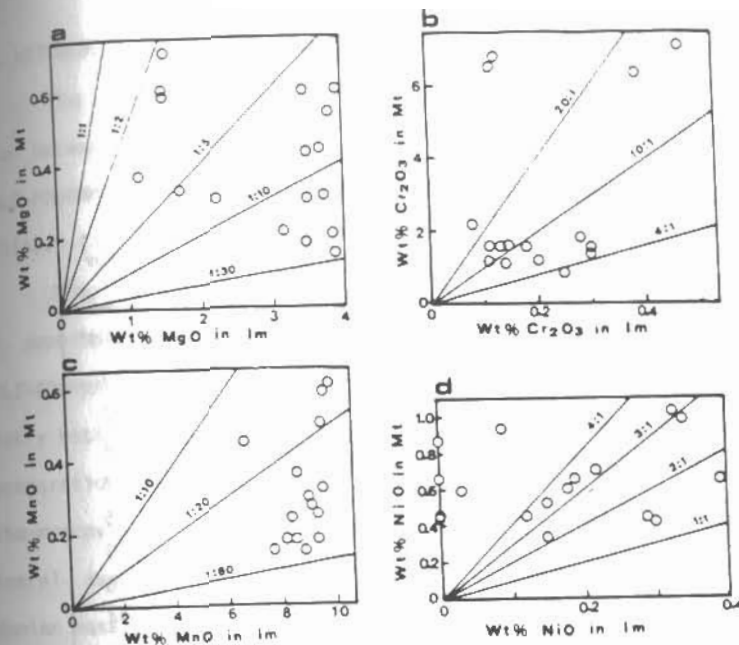


Fig. 11. Distribution of: (a) MgO, (b) Cr_2O_3 , (c) MnO and (d) NiO wt% between coexisting ilmenite (Im) and magnetite (Mt) in the Askos area meta-ultramafics (ratio lines are given for comparison).

Σχ. 11. Κατανομή της % περιεκτικότητας στα οξείδια: α. MgO, (β) Cr_2O_3 , (γ) MnO and (δ) NiO μεταξύ ιλμενίτη (Im) και μαγνητίτη (Mt) που συνυπάρχουν στην περιοχή Ασκού (δίνονται σχέσεις λόγων για σύγκριση).

Silicate minerals

Antigorite (Table 3) is slightly SiO_2 -rich and MgO-poor compared to the serpentine formula, owing to its alternating wave structure (WHITTAKER and WICKS 1970; MELLINI et al. 1987; PEACOCK 1987). It is also characterized by relatively high Al_2O_3 contents up to 2.46%. The Cr_2O_3 and NiO abundances in the mineral vary between 0.14-0.41% and 0.15-0.31% respectively.

Chlorites forming the matrix of the oxide mineralization have the composition of clinochlore-pennine according to the classification of HEY (1954). The formula positions occupied by Si (tetrahedral sites) vary between 6.11 and 6.39, while the ratio $\text{Fe}_{(\text{total})}/\text{Fe}+\text{Mg}$ of these chlorites varies between 0.065 and

Table 3. Microprobe analyses of silicate minerals from Askos area (range limits).

Πίνακας 3. Αναλύσεις με ηλεκτρονικό μικροαναλυτή πυριτικών ορυκτών από την περιοχή Ασκού (όρια κύμανσης).

	Antigorite (8)*	Chlorite (13)*	Talc (5)*	Tremolite (10)*
SiO ₂	42.03-43.54	31.15-33.10	61.20-62.78	54.24-56.77
Al ₂ O ₃	0.10- 2.46	11.90-16.05	0.10- 0.18	0.87- 2.20
TiO ₂	0.01- 0.10	0.11- 0.13	0.04- 0.08	0.01- 0.14
Cr ₂ O ₃	0.14- 0.41	0.22- 0.51	0.12- 0.15	0.03- 0.27
FeO _t	2.82- 4.56	4.27- 5.45	1.56- 2.44	5.40- 7.35
MnO	0.07- 0.25	0.12- 0.29	0.05- 0.09	0.26- 0.63
MgO	36.68-38.23	32.79-36.11	28.95-30.06	19.35-20.88
NiO	0.15- 0.31	0.16- 0.49	0.09- 0.22	0.03- 0.34
CaO	0.01- 0.15	0.10- 0.19	0.02- 0.04	10.96-12.01
Na ₂ O	<0.20	0.12- 0.40	0.08- 0.12	0.37- 1.35
K ₂ O	-	0.01- 0.11	-	0.01- 0.09
Total	84.01-87.28	86.75-88.07	94.25-94.98	95.91-97.95

*Number of analyses - Αριθμός αναλύσεων.

The Cr₂O₃ and NiO contents in the studied chlorites vary between 0.15%-0.52% (on the average 0.33%) and 0.16-0.52% (on the average 0.31%) respectively.

Chromian chlorites are considered as a usual mineral associated with ferritechromite. However, in our case it is clear that Cr liberated from the chromite to ferritechromite alteration enters the magnetite lattice and not that of chlorite.

Analyzed amphiboles are characterized as tremolites according to LEAK'S (1978) nomenclature, with Si in tetrahedral sites varying between 7.60 and 7.87 and the ratio Mg/(Mg+Fe²⁺) varying between 0.91 and 0.98.

Talc analyses show only little systematic chemical variation. Al₂O₃ is low and ranges from 0.10 to 0.18 and FeO from 1.56 to 2.44%wt.

6. METAMORPHIC EVOLUTION OF THE META-ULTRAMAFICS FROM THE ASKOS AREA.

The metamorphic evolution of the Serbo-Macedonian massif has been studied between others by HÖCKEL et al. (1977) KASSOLI-FOURNARAKI (1981), KASSOLI-FOURNARAKI et al. (1985) PAPADOPOULOS and KILIAS (1985), DIXON and DIMITRIADIS (1987), SAKELLARIOU (1988).

Prograde metamorphism was found to extend between upper greenschist to upper amphibolite facies for the main lithologies of Serbo-Macedonian massif. KASSOLI-FOURNARAKI et al (1985) on the basis of plagioclases' and amphiboles' chemistry estimated a P=2-4Kb and T=600-630°C for the amphibolites, while KOUROU (in preparation) using the garnet-biotite geothermometer and the plagioclase-biotite-muscovite-garnet geobarometer found T=450-550°C and P=5Kb as conditions of mineral assemblages equilibration for the mica schists of the Serbo-Macedonian massif.

The use of coexisting magnetite and ilmenite pairs in the meta-ultramafics from the Askos area, for the deduction of the metamorphic conditions according to the proposed models of BUDDINGTON and LINDSLEY (1964), SPENCER and LINDSLEY (1981) unfortunately is not reliable. Both minerals deviate in composition substantially from the system FeO-Fe₂O₃-TiO₂. Magnetite is characterized by very high Cr₂O₃ content and ilmenite by very high MnO content. This results in a great uncertainty in the calculation of T and f_{O₂} conditions, even if we use the model suggested by POWELL and POWELL (1977). Besides, as stated by RUMBLE (1976) the magnetite-ilmenite geothermometer and oxygen geobarometer is severely limited in its usefulness in low- and intermediate-grade metamorphic rocks. In this case the composition of natural magnetite remains fixed at Fe₃O₄ and unable to accommodate appreciable FeTiO₄ in solid solution. Magnetite in study has Fe₂TiO₄ always lower than 2.5% in solid solution.

Therefore, we made use of the silicate mineral assemblages for the interpretation of the metamorphic conditions in the Askos meta-ultramafics. These rocks are antigorite serpentinites containing the assemblages: antigorite + Fe-Cr spinels+ilmenite+chlorite+tremolite+talc+carbonates+quartz, whereas brucite,

diopside and olivine are absent. The PT metamorphic conditions can be discussed in terms of reaction curves and thermodynamic data of the multisystem $MgO-SiO_2-H_2O (\pm CaO \pm Al_2O_3 \pm FeO \pm CO_2)$.

The PT-stability conditions of the studied mineral assemblages lie between those defined by the reactions: $Ant+Di=Fo+Tr+H_2O(1)$ and $Ant=Fo+Ta+H_2O(2)$. The first reaction takes place at 486°C for $P_{total}=P(H_2O)=2Kb$ and about 522°C for 4Kb (OTERDOOM 1978 p. 134, fig. 5). The second reaction corresponds to the upper stability limit of antigorite (513°C for 2Kb and about 550°C for 4Kb; EVANS et al. 1976, p. 89 fig. 4). Actual temperatures were probably slightly lower because of dilution of metamorphic fluids by CO_2 .

Tremolite + olivine is stable at temperatures between 450° and 825°C at $P_t=P(H_2O)=5Kb$ (JENKINS 1983). Tremolite could have formed solely within the antigorite stability field.

Aluminous serpentine minerals could occur in place of chlorite as there is an extensive solid solution between chlorite and serpentine at low T (FAWCETT and YODER 1966). Experimental work in the system $MgO-Al_2O_3-SiO_2-H_2O$ (VELDE 1973) has shown that below 450°C the 7Å lattice structure of serpentine is stable over much of the compositional range between serpentine sensu stricto and amesite. Serpentine and Al-rich serpentine form a solid solution series. Above 450°C this breaks down and an end member 7Å serpentine coexists with a 14Å chlorite, as happens in the studied meta-ultramafics.

Therefore, the silicate mineral assemblage in the Askos meta-ultramafics denote metamorphic conditions of $T=450-550^\circ C$, and $P=4Kb$ corresponding to the transition from upper greenschist to lower amphibolite facies.

7. DISCUSSION AND CONCLUSIONS

Serpentinization of an ultramafic rock yielding a mesh-textured lizardite-bastite serpentinite commonly produces a magnetite rim around accessory chromite, as well as minute discrete magnetite crystals and aggregations of

crystals (BLISS and MACLEAN 1975; PINSENT and HIRST 1977; MAHLBURG KAY 1983 and others). Magnetite results from iron liberated from primary silicate minerals (olivine, pyroxenes) during the hydration process (ASHLEY 1975; ECKSTRAND 1975; MOODY 1976; MACFARLANE and MOSSMAN 1981). As stated by SPRINGER (1974), BLISS and MACLEAN (1975) in antigorite serpentinites produced by regional metamorphism in a grade between upper greenschist to amphibolite, magnetite rims react with chromite cores to produce Al- and Mg-poor ferritechromite zones. The stable spinel phase in antigorite serpentinites is Al-poor magnetite, Cr-magnetite or ferritechromite depending on the local Cr/Fe³⁺ ratio in the rock (EVANS and FROST 1975).

The origin of the Fe-Cr spinel and ilmenite massive mineralization in the Askos meta-ultramafics is related to the complex evolution of the ultramafic protolith. In the studied area the following series of events can be proposed:

(a) tectonic emplacement of the ultramafic protoliths (serpentinized before or during their transport from middle oceanic ridges towards the continent).

(b) alpine regional metamorphism resulting in the antigorite serpentinization and the formation of the Fe-Cr spinel and ilmenite mineralization.

During serpentinization manganese rich fluids affect chromite enclosed in the ultramafics as an accessory mineral, or as thin layers and massive chromite bodies. This resulted in the alteration of chromite, its transformation to a manganese one in a first stage and consequently to ferritechromite - chromian magnetite (MICHAILIDIS 1990). This transformation was a volume for volume metasomatic process as described by BEESON and JACKSON (1969), actually a "solid state diffusion" in which Fe³⁺ substitutes for Al (and Cr) and Fe²⁺ and Mn (with minor Ni) for Mg. Therefore, ferritechromite is considered as an intermediate chemical step in the transformation of chromite to Cr-magnetite and may be finally to pure magnetite. This means that once primary chromite has been al-

tered to ferritechromite, it is still susceptible to further transformation and Cr is progressively replaced by Fe^{3+} , pushing the composition towards that of magnetite sensu stricto (Fig. 8b and 9).

As stated by HARKER (1974) the diffusion of ions and consequently the metasomatic process is promoted along with the increase of pressure and temperature conditions. Therefore, regional metamorphism in the studied area has played a key role as to the acceleration of the chromite alteration.

The transformation of chromite to ferritechromite has released Al and Mg into surrounding serpentine which was transformed to Al-serpentine and finally to chlorite. Gradual transformation of ferritechromite to Cr-magnetite has liberated more Cr and Mn. Chromium was redistributed mainly in chlorite which contains up to 0.52% Cr_2O_3 , while Mn was fixed mainly in the ilmenite structure.

The following observations confirm the above processes:

- (a) the relict chromite cores in the zoned Fe-Cr spinels
- (b) the high Cr - content of magnetite crystals and the decrease of the element outwards
- (c) the relatively high Al - content of antigorite and the common association of chlorite with the Cr-magnetite massive mineralization.

The chemical inhomogeneity of the zoned Fe-Cr spinels, being found only in the massive type mineralization, is explained by the incomplete equilibration of primary chromite and secondary magnetite during the metamorphic recrystallization (FROST 1975; PINSENT and HIRST 1977). Densely packed chromite in the massive bodies has smaller amounts of solid and fluid phases with which to react (LIPIN 1984).

The TiO_2 contents of the relict chromite cores do not exceed 0.52% and they are the same in ferritechromite and magnetite zones. Therefore, the formation of the discrete ilmenite grains cannot be attributed to the transformation of chromite.

Ilmenite formation can be explained by the following processes:

(a) primary magmatic segregations (HAGGERTY 1976). Magmatic ilmenite, as a rule, is connected with basic rocks or the pyroxene rich members of peridotites. The normal sequence of oxide crystallization is chromite, followed by ilmenite, followed by titanomagnetite. In alpine-type peridotites, however, ilmenite is rarely found as magmatic segregations. Besides, no textural evidence was found for "external granule exsolution" of ilmenite from a ulvospinel-magnetite solid solution (BUDDINGTON and LINDSLEY 1964).

(b) exsolution lamellae in chromite (RAMDOHR 1969, p. 928, fig. 550a), which during metamorphism suffered separate recrystallization. However, relict chromite cores do not contain any exsolution bodies of ilmenite.

(c) a result of the reaction $rutile+spinel+serpentine$ (or $olivine+water$) = $chlorite+ilmenite + Fe-Cr spinel$ as stated also by COOK (1973) and FROST (1975).

This last case of formation is considered as the more probable, because rutile was found as discrete mineral grains and exsolution lamellae in chromite from the Gomati area chromitites, of Serbo-Macedonian massif (SCARPELIS and ECONOMOU 1978; CHRISTODOULOU 1980).

The tremolite-, chlorite-, talc-rich zones being connected with the thrust-contact zones suggest that the changes in mineralogy are a consequence of metasomatism and not the result of original bulk compositional differences. Calcium, Al and Si were added to the ultramafics via an infiltrating fluid phase (PEACOCK 1987). The adjacent country rocks and mainly the gneisses and the acid intrusive suite were a significant source of the elements which take part in the infiltration metasomatism.

These zones were formed before or at least late in the thrusting event as it is shown by the intensively banded tremolite megacrysts.

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