

Research Paper

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DOI number:

<http://dx.doi.org/10.12681/bgsg.20777>

Keywords:

Tinos, ophiolite complex, chromite, mineralization, ultramafic rocks

Citation:

Kokkaliari M., Seymour St. K., Tombros F. S., Koutsopoulou E. (2019), Study of the Chromite Mineralization Associated to Ophiolites from Tinos Island, Attico-cycladic Massif. Bulletin Geological Society of Greece, 55, 185-201.

Publication History:

Received: 07/07/2019

Accepted: 12/11/2019

Accepted article online: 14/11/2019

The Editor wishes to thank two anonymous reviewers for their work with the scientific reviewing of the manuscript and Ms Emmanouela Konstantakopoulou for editorial assistance

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STUDY OF THE CHROMITE MINERALIZATION ASSOCIATED TO OPHIOLITES FROM TINOS ISLAND, ATTICO-CYCLADIC MASSIF

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Abstract

This paper aims to study the chromitites, as well as their host rocks (meta-peridotites, meta-dunites and serpentinites) of the ophiolite complex of Mount Tsiknias, in Tinos Island. Recognition of their mineralogy and their textural features was carried out through detailed petrographic study. The mineral chemistry analysis contributed to the evaluation of the analyzed chromites, the chemical composition of which provides important information about the petrogenetic evolution of the chromitite ores. The chromites were in equilibrium with boninite melts derived from Supra-Subduction Zone, e.g., a depleted mantle wedge. In the binary classification diagram for spinels, the Tinos samples extend in the fields of Mg-chromite and chromite sensu strictu. In the TiO₂ vs Al₂O₃ diagram, the chromites plot in the field of Supra-Subduction Zone (SSZ) peridotites and partly overlap the field of chromites in Back-Arc Basalts (BABB), however the same samples plot in the field of chromite of boninites. In the Al₂O₃ vs Cr₂O₃ diagram both groups of Tinos chromites plot in the field/extremity of "mantle chromites".

Keywords: Tinos, ophiolite complex, chromite, mineralization, ultramafic rocks

Περίληψη

Η παρούσα εργασία έχει στόχο την μελέτη των χρωμιτιτών, καθώς επίσης και των πετρωμάτων-ξενιστών τους (μετα-περιδοτίτες, μετα-δουνίτες και σερπεντινίτες), στο οφιολιθικό σύμπλεγμα του Όρους Τσικνιά, στην νήσο Τήνο. Ο προσδιορισμός της ορυκτολογικής σύστασης καθώς και ο ιστολογικός χαρακτηρισμός των δειγμάτων επιτεύχθηκε μέσω λεπτομερούς πετρογραφικής μελέτης. Η ορυκτοχημική ανάλυση συνέβαλε στην αξιολόγηση των αναλυθέντων χρωμιτιτών, η χημική σύσταση των οποίων παρέχει σημαντικές πληροφορίες σχετικά με την πετρογενετική εξέλιξη των χρωμιτικών κοιτασμάτων. Οι χρωμίτες βρίσκονται σε ισορροπία με μπονινιτικά μάγματα, που είναι προϊόντα περιβαλλόντων καταβύθισης ζωνών ‘Supra-Subduction’, π.χ. απεμπλουτισμένης μανδυακής σφήνας. Στο δυαδικό διάγραμμα ταξινόμησης των σπινελίων, τα δείγματα προβάλλονται στο πεδίο των μαγνησιούχων χρωμιτιτών, αλλά και χρωμιτιτών με την στενή έννοια του όρου. Στο πετροτεκτονικό διάγραμμα $TiO_2 - Al_2O_3$, οι χρωμίτες εμπίπτουν στο πεδίο των ‘Supra-Subduction’ (SSZ) περιδοτιτών και επικαλύπτουν μερικώς το πεδίο χρωμιτιτών σε βασάλτες όπισθεν του τόξου (BABB), ωστόσο τα ίδια δείγματα από τους χρωμίτες της Τήνου εμπίπτουν στο πεδίο των χρωμιτιτών σε ισορροπία με μπονινιτικά μάγματα. Στο διάγραμμα $Al_2O_3 - Cr_2O_3$ και οι δύο ομάδες των χρωμιτιτών της Τήνου προβάλλονται στην άκρη του πεδίου ‘μανδυακοί χρωμίτες’.

Λέξεις κλειδιά: Τήνος, οφιολιθικό σύμπλεγμα, χρωμίτης, μεταλλοφορία, υπερβασικά πετρώματα

1. INTRODUCTION

Ophiolites are remnants of oceanic lithosphere which are tectonically emplaced along continental margins in accretionary prisms during orogenic processes (Rogkala et al., 2019). Also, they represent important targets for exploration of chromite and related PGE ore deposits. An illustrative example of such chromite metallogenetic setting is represented by the ophiolite complex of Mount Tsiknias in Tinos Island. The Tsiknias ophiolite complex has been emplaced before ca. 18 Ma, i.e. the age of Tinos granodiorite pluton which intrudes both the ophiolite and the tectonically underlying Cyclades Blueschist Unit (Avigad & Garfunkel, 1989; Bröcker & Franz, 2000; Ring et al., 2001). Its metamorphic sole comprises of greenschists and amphibolites. The “mantle” sequence of the Tsiknias ophiolite is represented as highly tectonized

serpentinites, wherever they can be discerned, with protoliths comprising dunites, harzburgites and lesser pyroxenes and gabbros. The meta-dunites and -peridotites represent the host rocks of the chromite mineralization. In order to contribute to the understanding of the genesis of Greek chromitites, the present work aims to study the lithotypes, paragenesis and mineral chemistry of the host rocks of Tinos chromitites. In combination with the mineral chemistry of chromite we also attempted to depict their possible genetic environment.

2. GEOLOGICAL SETTING

The Tinos ophiolite complex has been mapped by Melidonis (1980) and on a scale of 1:5000 by Vakondios (1997), whereas the age, the origin and the emplacement of the ophiolite has been recently studied by Lamont et al. (2019). Tinos chromitite orebodies are exposed in Mount Tsiknias (Fig. 1), the host rocks of which are mostly serpentinites, meta-dunites, -peridotites, with minor -pyroxenites and locally-gabbros.

Tinos Island is part of Cyclades Blueschist Unit (CBU), a nappe overlying a Pre-Alpidic anatectic basement covered by meta-volcanic and meta-sedimentary rocks (Bröcker and Franz, 2005; Rabillard et al., 2015). The whole sequence is underlain by the lowermost Basal Unit (BU), which comprises of Late-Triassic to -Cretaceous neritic meta-dolomites and -flysch (Xypolias et al., 2012), and overthrust by an ophiolite slice (Katzir et al., 2000). These rocks have been metamorphosed at eclogite to blueschist facies conditions (Brichau et al., 2010) at 53-40 Ma, and retrograded at greenschist to amphibolite facies at ≥ 25 and ~ 12 Ma (Bolhar et al., 2010).

Lamont et al. (2019) described that the Tsiknias ophiolite even though was formed at ~ 162 Ma, it was overthrust at ~ 75 Ma. At the eastern part of Tinos, the entire tectonostratigraphy as well as the Tsiknias ophiolite is intruded by a ~ 14 to 18 Ma monzogranite to granodiorite pluton (Brichau et al., 2007) and a boron- and fluorine-rich leucogranite (Mastrakas and St. Seymour, 2000). The intrusion of the pluton resulted in contact metamorphism comprising of pyroxene and amphibole hornfelses and pyroxene-garnet skarns close to the pluton, and more distant albite-epidote hornfelses (Stolz et al., 1997).

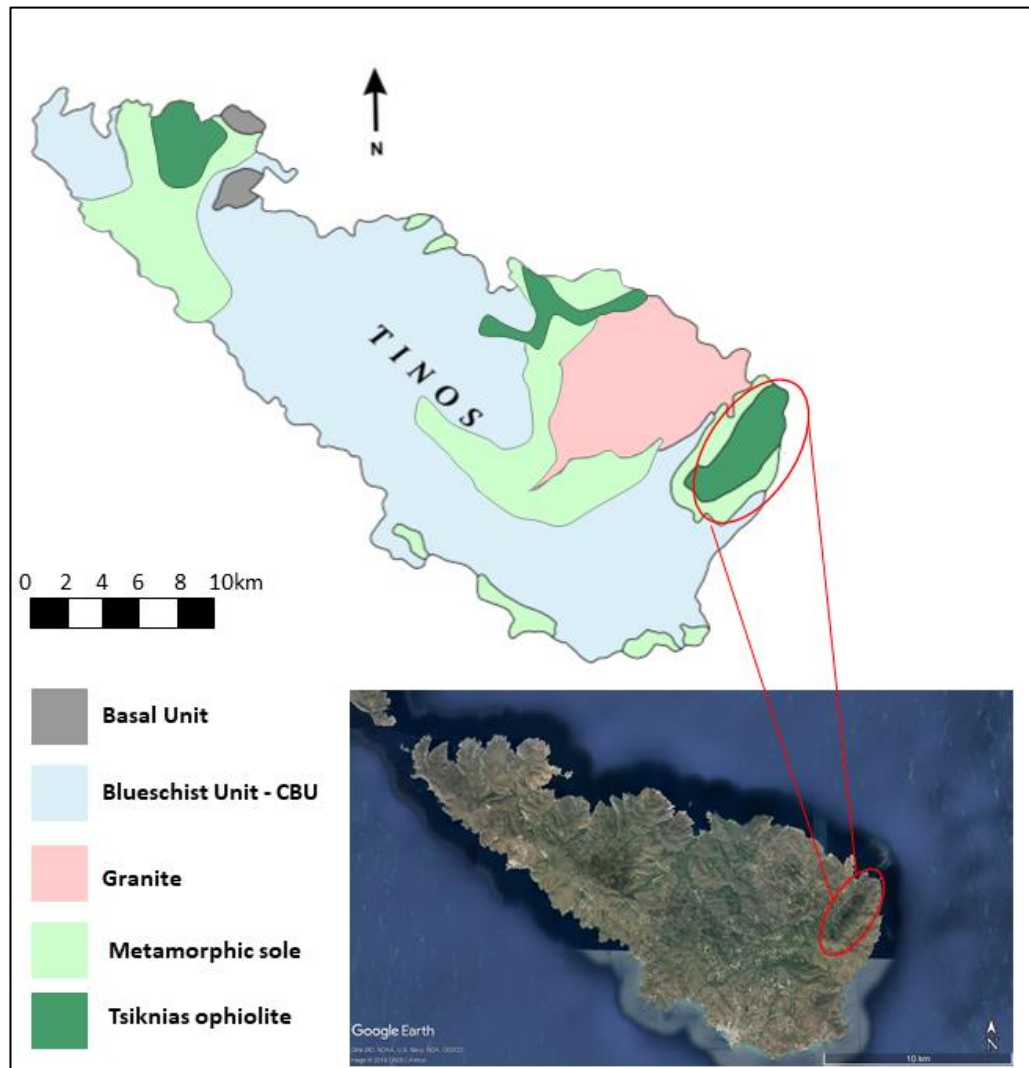


Fig. 1: Simplified geological map of Tinos Island (modified after Melidonis, 1980; Lamont et al., 2019).

3. MATERIALS AND METHODS

Sampling included the ultramafic host rocks, i.e., serpentinites, meta-dunites, -peridotites as well as the chromitites of the ophiolite complex. From these samples a total of 48 polished thin sections have been studied for their mineralogical and textural features using optical microscopy and Scanning Electron Microscopy (SEM). Determination of the chemical composition of the minerals was conducted using a JEOL JSM-6300 Scanning Electron Microscope, equipped with energy and wavelength dispersive spectrometers (EDS and WDS), at the Laboratory of Electron Microscopy and Microanalysis, University of Patras, Greece. The chemical

composition of the minerals was determined using natural and synthetic standards and the operating conditions were 20 kV accelerating voltage, 1.0 nA beam current and 60s counting time. The analyzed areas were selected using back-scattered electron images to avoid contamination. Measured contents were automatically corrected for atomic number, absorption in the sample, fluorescence and dead time (the ZAF correction), using the INCA software. Selected samples were also analyzed in the Department of Earth and Planetary Sciences Mc Gill University, Montreal, Canada using a JEOL Superprobe 8900 instrument equipped with energy and wavelength dispersive spectrometers (EDS and WDS, respectively) and an xClent system for ppm-level resolution. On line ZAF corrections were applied utilizing CAMECA software. Operating conditions for the ore minerals were an accelerating voltage of 20 kV and a beam current of 30 nA. Twenty seconds counting time was used for all elements except Ag (100s) and As (50s). Pyrite, sphalerite, galena, chalcopyrite, spessartine, stibnite and synthetic CoNiAs were used as standards. Detection limits in wt. % were: 0.042 for Ag, 0.054 for Zn, 0.048 for Fe, 0.061 for Sb, 0.022 for S, 0.066 for As, 0.072 for Cu, 0.037 for Au, 0.046 for Co, 0.207 for Pb, 0.049 for Ni, 0.036 for Cd and 0.031 for Mn.

4. RESULTS

4.1. PETROGRAPHIC ANALYSIS OF THE ULTRAMAFIC ROCKS

Meta-peridotites have porphyroclastic and sometimes boxwork texture and they comprise mainly olivine, orthopyroxene, chromite and minor clinopyroxene, having the assemblage of typical harzburgites. Secondary minerals include serpentine, chlorite, magnesite, iddigsite and metallic minerals. A large percentage of the samples is rich in serpentine with hourglass, mesh and bastitic texture, however the percentage of serpentinization varies in the samples, reaching up to 40%. The chromite crystals are mainly euhedral to subhedral, with characteristic brecciated texture, often replaced by chlorite and serpentine (Fig. 2A), whereas relict olivine and pyroxenes are mostly anhedral. Sometimes fingerprints of deformational processes such as crystal twinning, kink-band and mylonitic textures are observed. In every case, olivine and pyroxenes are altered to chlorite, iddigsite and serpentine (Fig. 2B). Serpentine veins, which crosscut the initial development of serpentine, were also identified. Metallic minerals usually occur in micro-fractures, with a specific orientation against pyroxene.

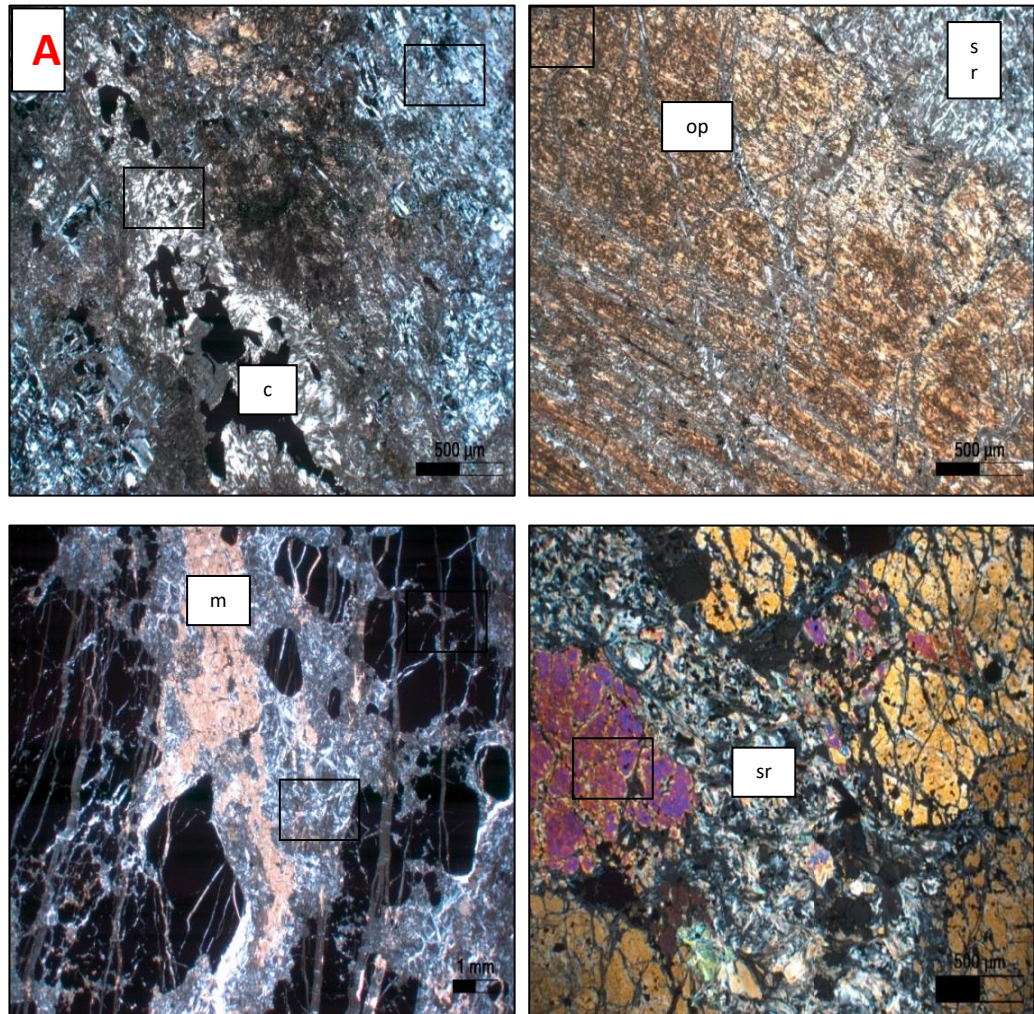


Fig. 2: Cross-polarized (PX) images of the ultramafic rocks (A) crystals of chromite, which is surrounded by chlorite, in a serpentine mass in the meta-peridotite, (B) altered crystals of orthopyroxene in the meta-peridotite. (C) crystals of chromite with brecciated texture in the serpentinite, (D) crystals of brecciated olivine, intensely altered to serpentine in a meta-dunite. Abbreviations according to Whitney & Evans (2010).

Serpentinities consist of serpentine, chromite, chlorite and magnesite. The chromite crystals are euhedral to subhedral and often intense brecciated. The cracks are filled with serpentine and magnesite (Fig. 2C). Meta-dunites occur with holocrystalline, porphyroblastic and in some cases boxwork texture, mainly comprising olivine, chromite and minor pyroxene. Olivine is the main mineralogical component, often characterized by elongation of its crystals. Secondary minerals include serpentine, chlorite and magnesite. Meta-dunites are characterized by the presence of micro-

brecciated zones and twinings due to extensive deformation (Fig. 2D). Chromite is euhedral to subhedral and mainly distributed at the margins of olivine but rarely appear as inclusion within it. Magnetite is part of the primary assemblage of the meta-dunites; however, it may also be present due to serpentinization. Serpentine presents boxwork and often intersertal texture and the degree of serpentinization varies depending on the sample, but usually does not exceed 30%.

4.2 PETROGRAPHIC ANALYSIS OF THE CHROMITITES

A detailed petrographic study was carried out in order to examine the texture of the chromite samples and the related metallic phases. The chromite crystals (Table 1) appear subhedral to anhedral and contain inclusions of silicate minerals, such as serpentine and chlorite due to alteration of olivine crystals, resulting in a poikiloblastic texture.

Networks of brecciated chromite and relict crystals probably of olivine and pyroxenes, within the groundmass of serpentine were also observed (Fig. 3A). In these cases, the groundmass of serpentine displays hourglass texture, whereas granular texture dominates the chromites. The latter are often characterized by bright luminous areas, especially at their rims, which are attributed by the increase of iron oxide content, hence the development of a transitional composition to ferrochromite (Table 1) that sometimes is followed by the presence of magnetite (Fig. 3B).

Metallic constituents were also detected in chromite samples which are divided in two groups: the first set contains base metal (Fe, Ni, and Cu) oxides, alloys and sulfides, whereas the second group comprises incompatible element phases (Pb, Sb, Mo, Sn and Zn) i.e. oxides, sulfides and antimonides. Irregular-shaped Ni-Fe alloys, as well as oxides, occur in the groundmass of serpentine, interstitial to the chromitite crystals. In the areas where Fe content increases, the brightness of the crystals decrease, and such compositional changes occur mainly at the crystal margins. Oxides containing Pb as well as antimony phases (Pb-Sb, Pb-Fe-Sb, Pb-Sb-As) were also observed (Fig. 3C). In general, the above phases occupy the open spaces between the chromite crystals. The identified base metal sulfides i.e., pyrite, pyrrhotite, chalcopyrite and heazlewoodite, are either dispersed in the serpentine groundmass or interstitial to the chromite crystals. The maximum sulfide crystal sizes do not exceed 10 μm , however,

in most of the cases the sulfide crystals are less than 5 μm . In some cases, when the crystals have a suitable size for observation, it appears that they have developed a microfracture structure. Heazlewoodite occurred in the form of small (euhedral) crystals (up to a few μm in size) embedded in a matrix composed of serpentine (Fig. 3D) and galena, wurtzite and sphalerite were identified included in the serpentine mass between the chromite crystals or adhered at their margins. Galena was also detected in the cracks between the chromite crystals.

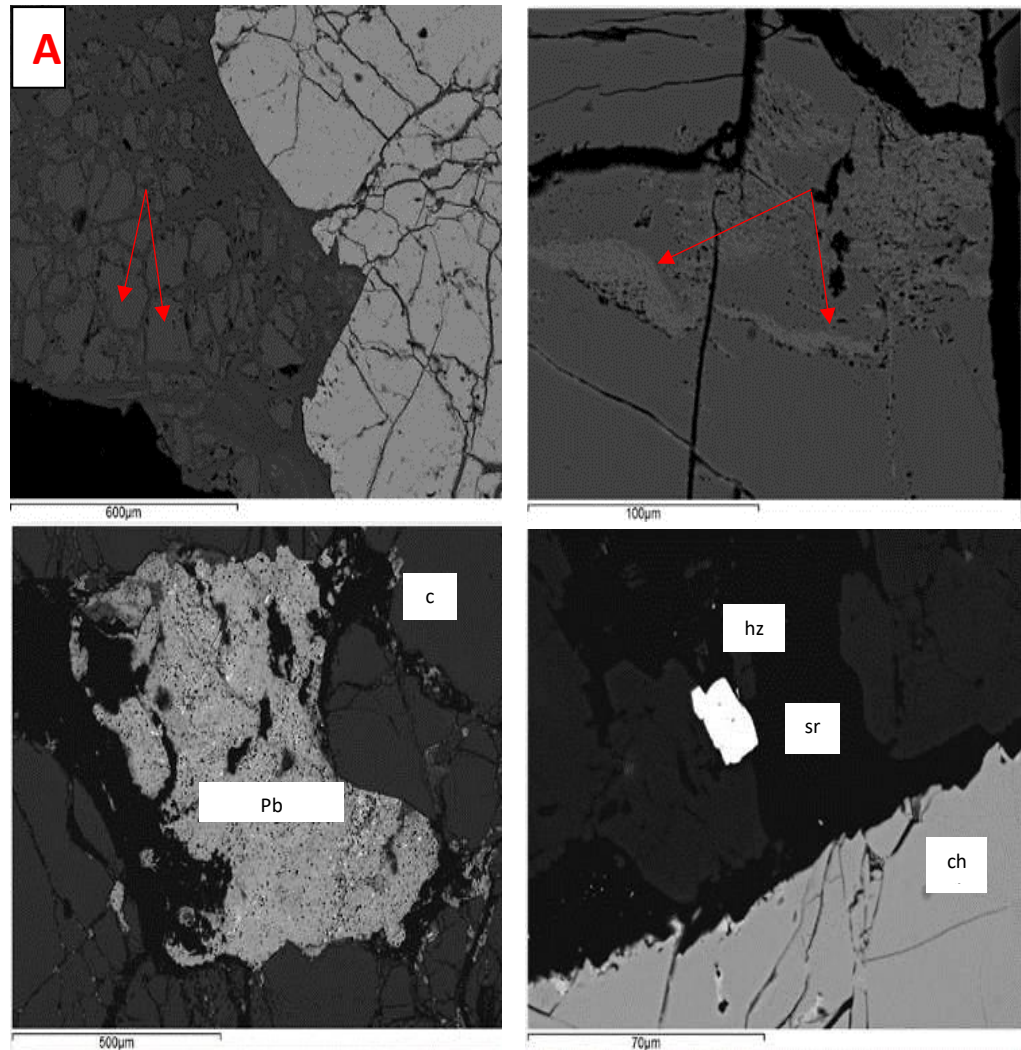


Fig. 3: SEM images of chromite crystals (A) in contact with the serpentinized mass and the presence of relict crystals, (B) the presence of ferrochromite at the peripheral parts of the chromite crystals, (C) Pb-antimonites that appear to occupy the crystals of chromite and (D) the presence of heazlewoodite in the form of small euhedral crystals embedded in the serpentine groundmass. Abbreviations according to Whitney & Evans (2010).

4.3 MINERAL CHEMISTRY OF TINOS CHROMITITES

Representative microanalyses of chromites are reported in Table 1 and plotted in Figure 4. On a Cr-Fe³⁺-Al by Stevens (1994) the Tinos chromites plot in the field of Al-chromite with only a few representatives in the field of Fe-chromite (Fig.4A). In the binary classification diagram for spinels (fields for spinel - hercynite - Mg-chromite - chromite) the Tinos samples extend in the fields of Mg-chromite and chromite sensu strictu (Fig. 4B). According to the mineral chemistry, the chromite crystals have values of Cr number [Cr#: Cr/(Cr+Al)] higher than 0.69, whereas the Mg number [Mg#: Mg/(Mg+Fe²⁺)] is higher than 0.15. In the Al₂O₃ vs Cr₂O₃ plot (Fig. 4C), the analyzed samples display a slightly upward trend, occupying a wide range area of the plot. The chromite crystals have values of Al₂O₃ up to 15 wt. % whereas the Cr₂O₃ ranges from 55 to 70 wt. %. The chromitites from the ophiolite complex in Tsiknias, represent crystallized products, in a mantle environment.

In the semi-log TiO₂ versus Al₂O₃ plot (Fig. 4D) the chromitites have a composition similar to that of the peridotites from subduction zones, whereas only some of the analyzed samples fall in the field of back-arc basin peridotites.

5. DISCUSSION

The Tsiknias ophiolite complex in Tinos Island is a succession of partially dismembered and structurally repeated ultramafic and gabbroic rocks, whereas the upper crustal lithologies are not present (Lamont et al. 2019). Vakondios (1997) described that the host rock of Tinos chromitites are mainly the serpentized ultramafic rocks, the protolith of which consists of harzburgites and to a lesser degree dunites. According to the abundance of their mineralogical components, they fall into the field of harzburgites, similar to the ophiolites of Vourinos complex, with increased Cr₂O₃ content (schlieren) which shows a high degree of partial melting (Vakondios, 1997). The studied lithotypes included serpentinites, meta-dunites and meta-peridotites, which are also the host rocks of the chromite mineralization. The meta-peridotites have a composition similar to typical harzburgites, with porphyroblastic and often mylonitic texture. Olivine crystals are anhedral, with boxwork texture, pyroxenes are characterized by tectonic twinning and kink-band textures and chromite crystals are often brecciated. Serpentine and chlorite are the main alteration products of olivine and orthopyroxene. Serpentine shows hourglass and often bastitic texture,

due to replacement against orthopyroxene. The meta-dunites are holocrystalline with porphyroblastic texture. Olivine shows signs of deformation (twinning, and elongation of the crystals) and alteration (serpentine, chlorite, and magnesite). Chromite is euhedral to subhedral, containing olivine inclusions. In the serpentinites, the relict minerals are rarely visible, except for chromite which occurs with subhedral crystals and shows brecciated texture.

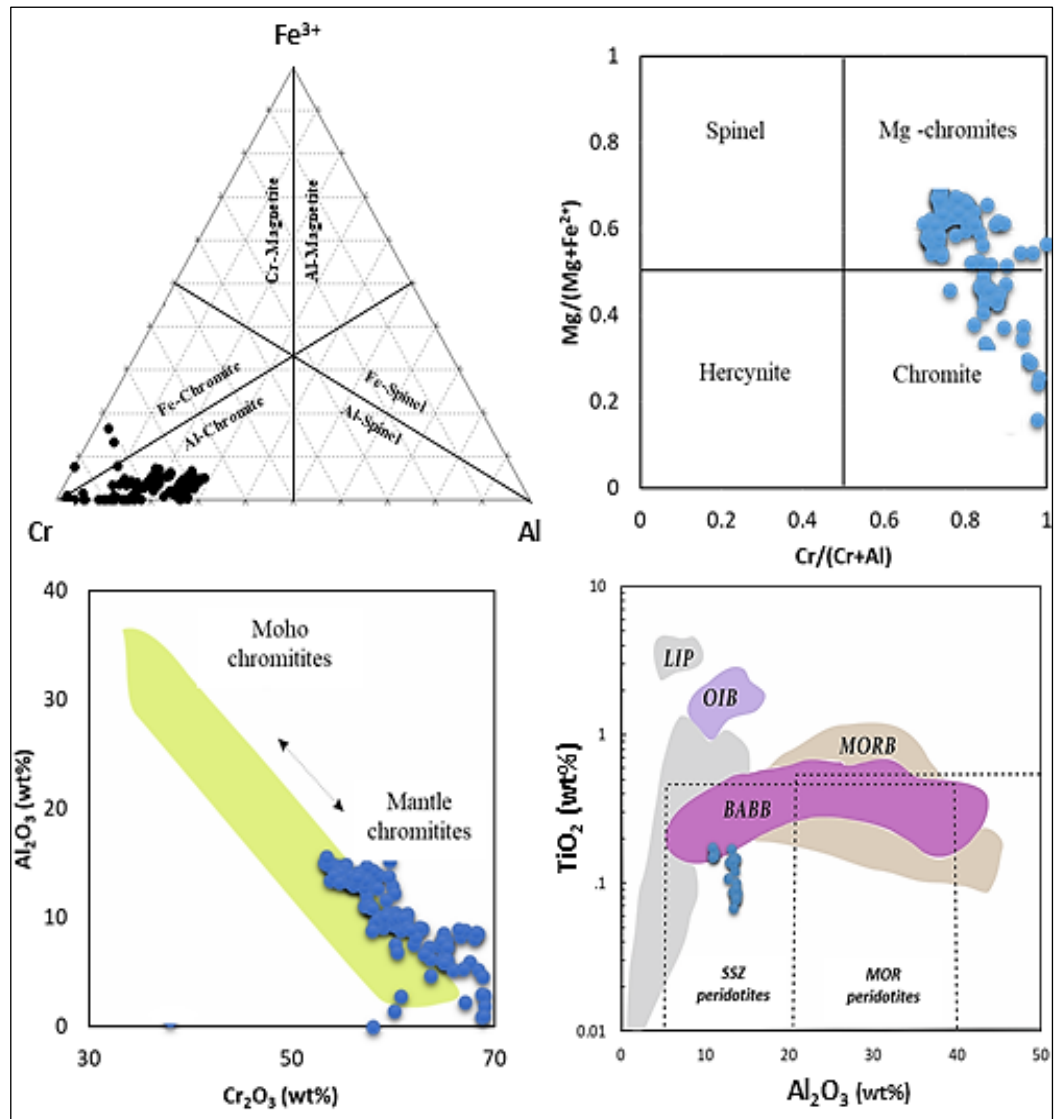


Fig. 4: Analyzed chromites from the chromitites of the ophiolite complex of Mount Tsiknias (A) Cr – Fe³⁺ – Al (Stevens, 1944), (B) Al₂O₃ vs Cr₂O₃ (Bonavia et al., 1993), (C) Mg / (Mg + Fe²⁺) vs. Cr / (Cr + Al) diagram. (D) TiO₂ vs Al₂O₃. Fields: LIP – Large Igneous Provinces, OIB – Ocean Island Basalts, MORB – Mid Ocean Ridge Basalts, BABB – Back Arc Basin Basalts (Kamenetsky et al., 2001).

Table 1. Representative Electron Microprobe (*) and Microscope analyses of Mg-chromites, chromites and Fe-chromites from the chromitites of the ophiolite complex of Mount Tsiknias (-: below detection limit).

Sample	C213_2*	C134-5*	C101-8	C219a-3	C117_2	C219b-3	C219b-1	C219b-3	C134-9	C113-4	C113-6
	Mineral		Mg-Chromite			Chromite			Fe-Chromite		
ZnO	0.03	0.02	-	-	-	-	-	-	-	-	-
SiO₂	0.01	0.02	-	-	-	-	-	-	-	-	-
MgO	14.30	13.92	12.38	12.87	13.25	12.28	8.22	8.97	8.49	4.51	4.44
Al₂O₃	13.41	13.68	5.87	11.06	13.27	14.67	7.50	6.67	6.13	0.85	0.95
TiO₂	0.13	0.09	-	-	-	-	-	-	-	-	-
FeO(t)	15.60	15.47	16.71	18.86	14.71	15.75	20.77	22.03	20.55	24.82	25.68
V₂O₅	0.10	0.18	-	-	-	-	-	-	-	-	-
Cr₂O₃	56.05	56.44	65.05	57.21	57.74	57.37	62.03	63.04	64.83	68.75	68.93
MnO	0.26	0.26	-	-	-	-	-	-	-	1.08	-
Total	99.89	100.08	100.01	100.00	98.97	100.07	98.52	100.71	100.00	100.01	100.00
Cations to 32 Oxygens											
Zn	0.01	0.01	-	-	-	-	-	-	-	-	-
Si	0.00	0.00	-	-	-	-	-	-	-	-	-
Mg	5.45	5.29	4.92	5.02	5.10	4.67	3.37	3.62	3.45	1.93	1.90
Al	4.04	4.11	1.84	3.41	4.03	4.41	2.43	2.12	1.97	0.29	0.32
Ti	0.03	0.02	-	-	-	-	-	-	-	-	-
Fe²⁺	2.48	2.60	3.03	3.00	2.82	3.21	4.47	4.28	4.40	5.61	5.88
Fe³⁺	0.96	0.77	0.77	1.26	0.40	0.17	0.33	0.78	0.31	0.39	0.31
V	0.02	0.04	-	-	-	-	-	-	-	-	-
Cr	11.32	11.37	13.71	11.82	11.77	11.56	13.46	13.46	13.94	15.59	15.62
Mn	0.06	0.06	-	-	-	-	-	-	-	0.26	-
Cations	24.26	24.20	24.20	24.38	24.08	24.00	24.03	24.18	24.04	24.03	24.00
Mg#	0.69	0.67	0.62	0.63	0.64	0.59	0.43	0.46	0.44	0.26	0.24
Cr#	0.74	0.73	0.88	0.78	0.74	0.72	0.85	0.86	0.88	0.98	0.98

The secondary mineralogy includes serpentine, chlorite, magnesite, iddigsite and metallic minerals. Serpentine forms hourglass and bastitic texture, which indicates a possible peridotite protolith. In addition, secondary development of serpentine veins which crosscut the initial formed serpentine are probably associated with subsequent

deformation or metasomatism related to the emplacement of Tinos pluton (Andreani et al., 2004; Mellini et al., 2005).

The chromitites are characterized by brecciated, poikiloblastic texture due to serpentine and chlorite inclusions in the chromite crystals, resulted from the substitution of olivine. In some cases, the alteration of chromite is quite advanced so that transition zones of Fe-chromite, are formed to the margins of the crystals, where the presence of magnetite is sometimes observed locally. The modification of the chromite composition occurs due to post-magmatic processes such as alteration (Onyeagocha, 1974), regional metamorphism and/or metasomatism. According to Bliss and McLean (1975), the formation of ferrochromite is attributed to serpentinization in which magnetite is initially formed and replaced then by ferrochromite, during higher conditions of metamorphism, up to the lower amphibolitic facies ($T \geq 500^\circ \text{C}$). The interstitial groundmass to the chromite crystals consists of serpentine, which creates a network with irregular dimensions and orientation around, or inclusions within the chromite crystals.

On a Cr-Fe³⁺-Al by Stevens (1994) the Tinos chromites plot mainly in the field of Al-chromite, whereas the Fe-chromite compositions were obtained from the chromite rims and are attributed to metamorphism, metasomatism and supergene oxidation. In the binary classification diagram for spinels, the analyzed chromite crystals fall in the field of the typical chromite and Mg-chromite, and in the Al₂O₃ vs Cr₂O₃ plot they fall in the field of “mantle chromitites”. The low contents of Al₂O₃ and TiO₂ in the chromites and their high MgO, and especially Cr₂O₃ contents suggest that they initially originated from melts with boninite composition. According to a number of researchers (Bai et al., 2000; Ahmed and Arai, 2002; Edwards et al., 2002; Matsumoto and Tomurtogoo, 2003; Ahmed et al., 2005; Rollinson, 2008; Tzamos et al., 2017) chromitites with high Cr# and a relatively low content of TiO₂ can be derived from boninitic melts that are formed in Supra-Subduction Zone (SSZ), i.e. in a depleted mantle wedge (Tzamos et al., 2017). However, only some of the analyzed samples fall in the field of back-arc basin peridotite. Boninitic melts are formed from hydration of the accretionary prism over subduction zones, followed by partial melting of the residual mantle in a forearc setting (Tzamos et al., 2017).

Base metal (Fe, Ni and Cu) sulfides, oxides and alloys as well as incompatible element phases (Pb, Sb, Mo, Sn and Zn) e.g. oxides, sulfides and antimonides, were

also identified, either dispersed in the serpentine groundmass or adhered at the chromite crystals. The origin of these various incompatible metallic phases i.e. galena, sphalerite, wurtzite, antimonites, as well as Fe-Pb oxides, requires further study, suggesting possible metasomatic and/or hydrothermal processes related to the emplacement of the neighboring Tinos composite granodiorite-leucogranite.

6. CONCLUSIONS

Based on petrography and mineral chemistry of Tinos chromitites we can highlight the following conclusions:

- The Tsiknias ophiolite complex, comprises of serpentized-tectonized ultramafic rocks. The studied lithotypes are serpentinites, meta-dunites and meta-peridotites, which are the host rocks of the chromite mineralization. The ultramafic host rocks show characteristics typical of a depleted mantle.
- Chromites from Tinos chromitites have compositions which fall mainly in the field of Mg-chromite and chromite *sensu strictu*. Their MgO and Cr₂O₃ contents show that they originated from melt with a boninitic composition, typical of a Supra-Subduction Zone.
- The mineralogical assemblages identified in the chromites (base metal as well as incompatible element phases), are influenced by other processes i.e. serpentization, and are probably linked to the metasomatic and/or hydrothermal processes, related to the emplacement of the neighbouring Tinos composite granodiorite-leucogranite. The chromitites of the Tsiknias ophiolite complex, seem to have been influenced by post-magmatic processes, intriguing the interest for further research.

7. AUTHOR CONTRIBUTIONS

M.K. performed all the petrographic analyses. M.K. and E.K. conducted the SEM experiments and wrote the manuscript. S.F.T. contributed to manuscript writing. All authors contributed to data analysis and commented on the manuscript. K.St.S. supervised the project.

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