

## **Preiswerkite, Ca-(Mg,Fe)-paragonite and Ca-“ephesite” in metadiorite from the HP Kechros Metamorphic Complex in Eastern Rhodope, NE Greece**

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**Abstract.** Preiswerkite, Ca-(Mg,Fe)-paragonite and Ca-“ephesite”, three unusual micas, have been found in a metadiorite within an antigorite serpentinite from the Kechros *HP* Metamorphic Complex in Eastern Rhodope, Greece. The preiswerkite displays a solid solution towards Na-phlogopite up to 18 mol%, towards margarite up to 2.4 mol% and towards phlogopite up to 6 mol%. The Ca-(Mg,Fe)-paragonites and Ca-“ephesites” form solid solutions between paragonite, Mica-L, margarite, “ephesite” and muscovite. In Ca-(Mg,Fe)-paragonite, the paragonite component ranges from 49 to 66 mol%, the Mica-L component from 29 to 41 mol%, the margarite component from 2 to 10 mol% and the “ephesite” component from 0 to 13 mol%. In Ca-“ephesite”, the “ephesite” component ranges from 29 to 53 mol%, the paragonite component from 22 to 46 mol%, the margarite component from 14 to 26 mol% and the Mica-L component from 0 to 18 mol%.

The preiswerkite coexists with oligoclase and Ca-“ephesite”. They replace Ca-(Mg,Fe)-paragonite and magnesiotalcarnite during decompression to amphibolite facies conditions. The Ca-(Mg,Fe)-paragonite represents relict phase formed at the *HP* metamorphic stage.

*Key words:* preiswerkite, Ca-“ephesite”, *HP* metamorphism, Greek Rhodope

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## **Еврипидис Боскос, Мариа Пераки. Прайсверкит, Са-(Mg,Fe)-парагонит и Са-„ефесит” в метадiorити от високобаричния Кехроски метаморфен комплекс в Източните Родопи, СИ Гърция**

**Резюме.** Прайсверкит, Са-(Mg,Fe)-парагонит и Са-„ефесит”, три необичайни слюди са намерни в метадiorити всред антигоритови серпентинити от високобаричния Кехроски метаморфен комплекс в Източните Родопи, СИ Гърция. Прайсверкитът образува твърди разтвори с Na-флогопит до 18% mol, с маргарита до 2,4% mol и с флогопита до 6% mol. Са-(Mg,Fe)-парагонит и Са-„ефесит” образуват твърди разтвори с парагонита, слюда-L, маргарита, „ефесита” и мусковита. В Са-(Mg,Fe)-парагонит парагонитовият компонент е между 49 и 66% mol; слюда-L компонентът – между 29 и 53% mol; маргаритовият компонент – от 14 до 26% mol и „ефеситовият” компонент – от 0 до 13% mol. В Са-„ефесита” „ефеситовият” компонент е между 29 и 53% mol; парагонитовият компонент – между 22 и 46% mol, маргаритовият компонент – от 14 до 26% mol и слюда-L компонентът – от 0 до 18% mol.

Прайсверкитът съществува с олигоклаз и Са-„ефесит”. Те заместват Са-(Mg,Fe)-парагонит и Mg-тарамит при декомпресия до условията на амфиболитов фациес. Са-(Mg,Fe)-парагонит представлява реликтова фаза, образувана при високобаричния метаморфен етап.

## Introduction

Preiswerkite a sodic trioctahedral mica, with the ideal formula  $\text{Na}_2 \text{Mg}_4^{\text{VI}} \text{Al}_2 (\text{Si}_4^{\text{IV}} \text{Al}_4) \text{O}_{20} (\text{OH})_4$ , is very rare in nature. It was first described by Keusen and Peters (1980) from a metarodingite in the Geisspfad ultramafic complex in the Swiss Penninic Alps. Since then, it has been found in Allalinhorn, Switzerland (Meyer 1983), Liset, Norway (Smith & Kechid 1983; Kechid 1983; Smith 1988), in La Copointrie, Vedée, France (Godard & Smith 1984; Godard 1988; Godard and Smith 1999), in Koralpe and Saualpe, Austria (Miller 1990), in Rio La Palmilla, Guatemala (Harlow 1994), in Blengsvatn, Bamble, Norway (Visser et al. 1999), and in the Dabie UHP metamorphic belt, China (Wang et al. 2000). Most of these occurrences have in common a polymetamorphic history involving the retrogression of high-pressure (*HP*) rocks (Table 1).

Ideal end-member aluminium silicon preiswerkite was synthesized by Franz and Althaus (1976) who called it “sodium-brittle mica”, and by Líu (1989). They examined the solid solution between sodium end-member phlogopite and preiswerkite (corresponding to the tschermakite substitution  $^{\text{VI}}\text{Mg}+\text{Si}=^{\text{VI}}\text{Al}+^{\text{IV}}\text{Al}$ ) at different *P* and *T* conditions.

Three of the above mentioned preiswerkite occurrences (La Compointrie; Liset; Koralpe/Saualpe) consist of *HP* eclogites, in which preiswerkite appeared during retrogression as coronae or symplectites around kyanite. In the same kyanite eclogites, also linked to the breakdown of kyanite, there also exists another very rare mica, Na-(Mg,Fe)-margarite. This mica is described as being a solid solution between margarite ( $\text{Ca}_2\text{□}_2^{\text{VI}}\text{Al}_4[\text{Si}_4^{\text{IV}}\text{Al}_4]\text{O}_{20}(\text{OH})_4$ ), paragonite ( $\text{Na}_2^{\text{VI}}\text{Al}_4[\text{Si}_6^{\text{IV}}\text{Al}_2]\text{O}_{20}(\text{OH})_4$ ) and a hypothetical 2.5-octahedral end-member with the ideal formula  $\text{Na}_2\text{□}_1(\text{Mg,Fe})_1^{\text{VI}}\text{Al}_4[\text{Si}_4^{\text{IV}}\text{Al}_4]\text{O}_{20}(\text{OH})_4$  (“Mica L”, Smith 1988; Godard & Smith 1999). The last one can be considered as the (Mg,Fe) equivalent of the trioctahedral Li-mica ephesite  $\text{Na}_2\text{Li}_2^{\text{VI}}\text{Al}_4[\text{Si}_4^{\text{IV}}\text{Al}_4]\text{O}_{20}(\text{OH})_4$ .

Margarite, Mica L and ephesite are related by the cation exchanges:  $\text{Ca}_2\text{□}_2=\text{Na}_2\text{□}_1(\text{Mg,Fe})_1=\text{Na}_2\text{Li}_2$  (Godard & Smith 1999).

Ephesite occurs in nature as pure phase of near end-member composition. It is also known to form extensive solid solution with margarite, with Na substituting for Ca and Li for vacancy (Schaller et al. 1967). Ephesite has been synthesized by Warhus and Chatterjee (1984) at temperatures between 300 and 700°C and pressures up to 3.5 GPa. Franke et al. (1982) reported the synthesis of a lithium-free “ephesite-like” sodium mica, whose composition was given as  $\text{Na}_2\text{Al}_{4.66}[\text{Al}_4\text{Si}_4]\text{O}_{20}(\text{OH})_4$ .

These micas are of particular interest due to their extreme rarity, which might be related to very specific metamorphic conditions in pressure, temperature, nature of the fluid, and/or chemical potentials.

In this article, we describe the petrology of a new occurrence of a preiswerkite, Ca-(Mg,Fe)-paragonite and Ca-“ephesite” association in a metadiorite lens within serpentized peridotite from the Kechros *HP* metamorphic complex in Eastern Rhodope and discuss the chemical properties, in particular the possible solid solutions, and the metamorphic conditions of the formation of these unusual micas. This may contribute to a better understanding of their genesis. Ca-(Mg,Fe)-paragonite and Ca-“ephesite” are described as being solid solutions between paragonite, Mica L, “ephesite” and margarite. Paragonite and Mica L end-members dominate in Ca-(Mg,Fe)-paragonite and “ephesite” plus margarite and members in Ca-“ephesite”.

## Geological setting

In Eastern Rhodope, a discrete tectonic contact separates the Kechros complex from the overlying Kimi complex (Fig. 1). The Kechros complex consists of orthogneisses, reworked migmatites containing muscovite metapelite lenses, pelitic gneisses, high-alumina metapelites and scarce occurrences of marbles. Large bodies of antigorite serpentinites are tectonically intercalated. These ultramafic

Table1. Occurrences of preiswerkite and Na-(Mg,Fe)-margarite and Ca-“ephesite” associated with preiswerkite from the literature (From Godard and Smith 1999, data about Dabie UHP from Wang et al. 2000)

Preiswerkite					
Localities	Host rocks	Petrography	Metamorphic conditions	XMg	References
Geisspfad, Switzerland	Metarodngite	Mica + zoisite nodules	Greenschist to amphibolite facies	0.93	Keusen and Peters (1980)
Allalinhorn, Valais, Switzerland	Eclogitized gabbro	Symplectite	Depressurisation		Meyer (1983)
Liset, Vestlandet, Norway	Kyanite eclogite	Symplectite around kyanite	Depressurisation	0.76	Smith and Kechid (1983), Kechid (1984), Smith (1988), Godard and Smith (1999)
La Compointrie, Vendee, France	Kyanite eclogite	Coronae around kyanite	Depressurisation	0.96	Godard and Smith (1984), Godard (1988), Godard and Smith (1999)
Vumba, Botswana	Serpentinite	Symplectite around spinel and hoegbomite	Retrogression	0.73	Rammlmair et al. (1988)
Koralpe and Saualpe, Austria	Kyanite eclogite	Symplectite around kyanite	Depressurisation	0.90	Miller (1990)
Rio La Palmilla, Guatemala	Jadeitite	Mostly with alteration products	Albitisation	0.75-0.86	Harlow (1994, 1995)
Blengsvatn, Bamble, Norway	Scapolitite	Tourmaline + biotite + scapolite±corundum±plagioclase+...	Greenschist to amphibolite facies	0.81-0.85	Visser et al. (1999)
Dabie Shan, China	Quartz jadeitite	Plagioclase+magnete+taramite	Retrogression, High-grade greenschist facies	0.84	Wang et al. 2000
Kechros Complex, Rhodope Metamorphic Province, Greece	Metadiorite			0.73-0.83	This study

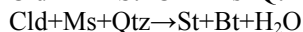
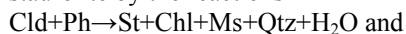
bodies contain boudins of gabbroic pegmatites, diabase dykes, rodingitic gabbros and trontjemites subsequently affected by *HP* metamorphism (Mposkos et al. 1997; Iliadis 2006). Boudins of eclogites, eclogite-amphibolites and

amphibolites occur within orthogneisses, serpentinites and between serpentinites and under- and overlying metapelites (Mposkos et al. 2011). *P-T* conditions of 1.7-2.2 GPa and 570-620°C are constrained from the eclogites

Table1. (continuation)

Na-(Mg,Fe)-Margarite associated with preiswerkite					
Localities	Host rocks	Petrography	Metamorphic conditions	Mica L mol%	References
Liset, Vestlandet, Norway	Kyanite eclogite	Symplectite after kyanite	Depressurisation	27	Smith and Kechid (1983), Kechid (1984), Smith (1988), Godard and Smith (1999)
La Compointrie, Vendée, France	Kyanite eclogite	Coronae around kyanite	Depressurisation	30	Godard and Smith (1984), Godard (1988), Godard and Smith (1999)
Koralpe and Saualpe, Austria	Kyanite eclogite	Symplectite after kyanite	Depressurisation	13	Miller (1990)
Kechros Complex, Rhodope Metamorphic Province, Greece	Metadiorite				This study

(Mposkos et al. 2011). In the orthogneisses and metapelites the *HP* event is documented by the presence of phengitic white K-mica reaching 3.50 and 3.43 Si *apfu*, respectively (Mposkos 1989). In metapelites, the formation of staurolite by the reactions



(abbreviations after Whitney and Evans, 2010, and Prw=preiswekrite, Eph=ephesite) suggests nearly isothermal decompression from maximum pressures of 2.2 GPa to 0.4 GPa (Mposkos 1989; Mposkos & Liati 1993), implying rapid uplift. At a later stage, at pressures below 0.3 GPa andalusite is replacing chloritoid and green biotite is replacing garnet and muscovite.

Orthogneisses and metapegmatites have Variscan protolith ages, as is indicated by Rb-Sr age of 334 Ma of magmatic muscovite from metapegmatite; (Mposkos & Wawrzenitz 1995) and U-Pb ages between 326-299 Ma of magmatic zircons from orthogneisses (Peytcheva & von Quadt 1995; Cornelius 2008; Liati et al. 2011). An U-Pb SHRIMP age of magmatic zircons from a kyanite-eclogite

yielding 255 Ma is interpreted as the age of the gabbroic protolith (Liati et al. 2011) indicating that the subsequent *HP* metamorphism is Alpine in age. The minimum age of the *HP* metamorphism is limited by Rb-Sr and Ar-Ar white mica ages of 36-42 Ma from mylonitic orthogneisses (Wawrzenitz & Mposkos 1997; Lips et al. 2000). Emplacement of the Kechros complex to shallow crustal levels and juxtaposition against the Kimi complex by extensional deformation between 36 and 32 Ma is constrained by Rb-Sr biotite ages of granodiorites and subvolcanic rocks that intruded the Kechros complex and the overlying Kimi complex (Del Moro et al. 1988).

### Petrography

The preiswerkite-, Ca-(Mg,Fe)-paragonite- and Ca-“ephesite”- bearing rock described in this paper is found in the Flambouro area east of the Organi village within foliated serpentinite (Fig. 1). It forms a small boulder with visible dimensions 3x5m. In hand specimen, it is fine grained and green in colour. No deformation



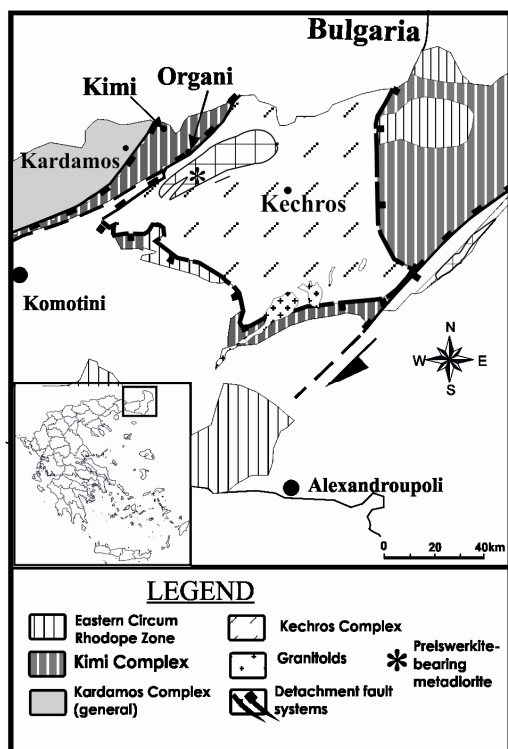


Fig. 1. Simplified geological map of the Kechros complex in eastern Rhodope (from Mposkos, 1990). Star marks the locality of the metadiorite of the present work within the antigorite serpentinite (coordinates 41°15' 37"N, 25° 43' 05"E)

textures can be observed, suggesting that deformation was concentrated in the surrounding serpentinite. The bulk and CIPW norm composition of this rock is that of Na-Al rich leucodiorite with norm olivine 0.95 wt.% and norm plagioclase 87.4 wt.% (Table 2). The rock is relative enriched in Li (278.6 ppm), suggesting that small amounts of lithium may be present probably in the crystal lattice of the micas and amphiboles. The Al-rich Na-micas designated in this work as Ca-(Mg,Fe)-paragonite and Ca-“ephesite”, and the preiswerkite could be considered as potential Li-bearing phases in this rock (see below mineral chemistry).

Based on thin section and SEM observations the total mineral assemblage in the Na-Al-rich micas bearing metadiorite: Amp+Bt+Ab+Pl+Pg+“CaEph”+Prw+Czo+Ph+Chl+Rt+Ttn+Apt. In the SEM images Ca-(Mg,Fe)-paragonite and Ca-“ephesite” could not be distinguished from each other. They are distinguished only on the basis of their chemical composition (see below).

Preiswerkite is commonly associated with Ca-“ephesite”, amphibole, plagioclase and K-feldspar. Textural relationships show preiswerkite, plagioclase and K-feldspar to form at the expense of Ca-(Mg,Fe)-paragonite and amphibole (plus phengite) (Fig. 2 a, b, c). Preiswerkite, Ca-“ephesite” and plagioclase commonly show textures like “graphic intergrowths” (Fig. 2 b, c, d) replacing pseudomorphically preexisting Na-micas or amphiboles. Ca-(Mg,Fe)-paragonite occurs as inclusions in amphibole (Fig. 2 a, b). Matrix Ca-(Mg,Fe)-paragonite, and amphibole are replaced by preiswerkite, Ca-“ephesite”, oligoclase and K-feldspar. The corroded edges of the amphibole grains indicate that amphibole participates in the preiswerkite-forming reaction. The small size, the symplectitic intergrowth of preiswerkite with Ca-“ephesite” and plagioclase (Fig. 2 b, d, e), and the irregular distribution prevent an accurate modal estimation of preiswerkite and Ca-“ephesite” in the rock (both participate with <5% in volume).

Table 2. Chemical composition and CIPW norm of the preiswerkites-, Ca-(Mg,Fe)-paragonite- and Ca-“ephesite”-bearing metadiorite from the Kechros HP metamorphic complex

SiO <sub>2</sub>	59.10	MgO	2.65	Or	6.21	Il	0.68
TiO <sub>3</sub>	0.35	CaO	5.20	Ab	57.80	Mt	0.92
Al <sub>2</sub> O <sub>3</sub>	20.70	Na <sub>2</sub> O	6.75	An	23.39		
Fe <sub>2</sub> O <sub>3</sub>	0.63	K <sub>2</sub> O	1.04	Di	5.79		
FeO	1.50	LOI	0.87	Hy	4.68		
MnO	0.04	Li	2.78	OI	0.95		
		(ppm)					

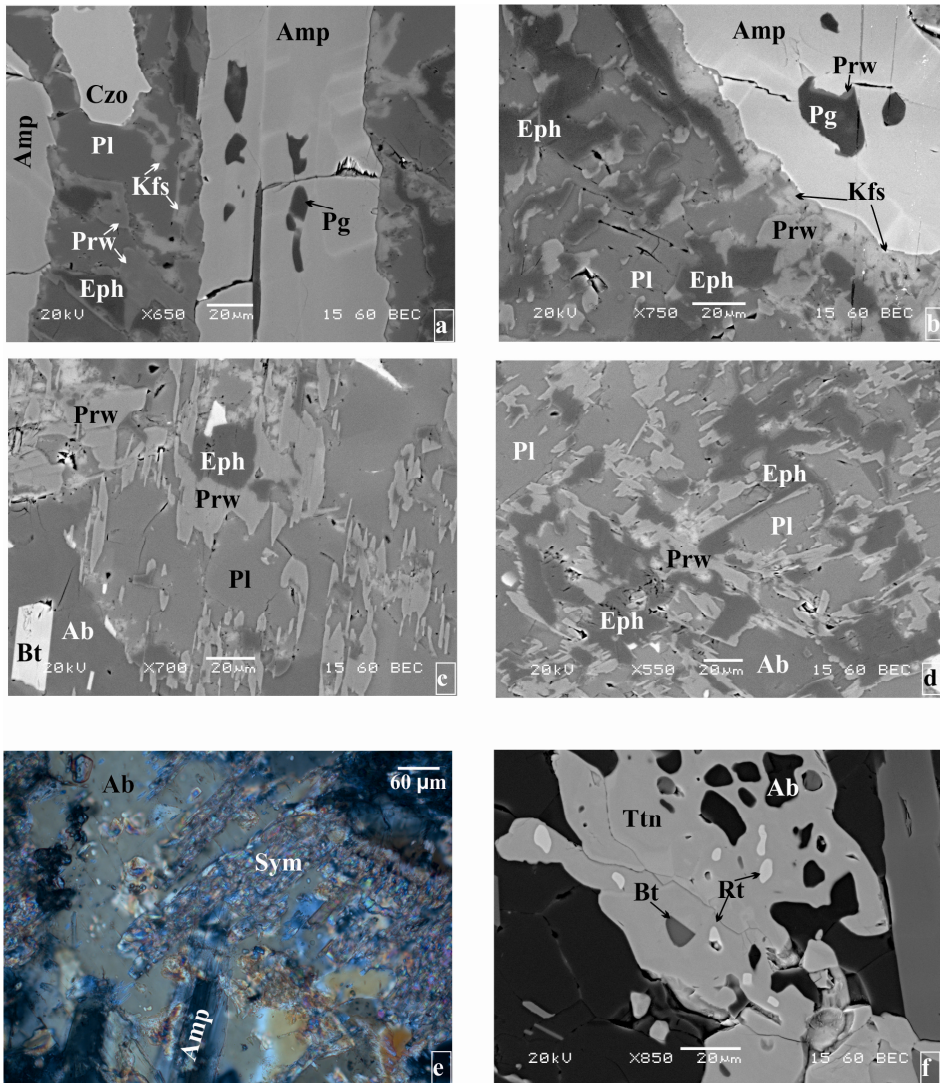


Fig. 2. a) Preiswerkite (Prw) is associated with oligoclase (Pl), Ca-“ephesite” (Eph), cloudy K-feldspar (Kfs), amphibole (Amp) and clinozoisite (Czo). Amphibole and clinozoisite show corroded edges. Amphibole contains inclusions of Ca-(Mg,Fe)-paragonite (Pg), BSE b) Amphibole with inclusions of Ca-(Mg,Fe)-paragonite (Pg). Between paragonite inclusion and amphibole host preiswerkite is formed. The amphibole shows resorbed edges in contact with preiswerkite and K-feldspar. Matrix: preiswerkite and Ca-“ephesite” form “graphic”-like intergrowths within oligoclase. The lines on the right of the picture are artifacts, BSE c) Preiswerkite flakes enclosed in oligoclase (Pl). The thin prismatic to needle shaped preiswerkite crystals replaced pseudomorphically previous, thin, prismatic amphiboles. Eph=Ca-“ephesite”, Ab=albite, Bt=biotite, BSE. d) Oriented intergrowths of preiswerkite, Ca-“ephesite” and oligoclase replaced Ca-(Mg,Fe)-paragonite, BSE. e) Symplectitic intergrowths of preiswerkite and plagioclase (Sym) replacing pseudomorphically Ca-(Mg,Fe)-paragonite inclusions in albite (Ab), Amp =amphibole, CPL. f) Titanite (Ttn) with inclusions of rutile (Rt), biotite and albite, BSE

Albite and oligoclase are the dominant phases ( $\approx 70\%$  in volume). Albite forms porphyroblasts up to 1 mm in size and is rich in inclusions of amphibole, biotite, clinozoisite, Ca-“ephesite”, rutile and titanite. Oligoclase is associated with albite, preiswerkite, Ca-“ephesite” and K-feldspar.

Amphibole is a major phase ( $\approx 15\%$  in volume). It forms thin prismatic to fibrolitic crystals 20-50x200-1000 $\mu\text{m}$  in size, not oriented in the rock matrix. Amphibole contains inclusions of Ca-(Mg,Fe)-paragonite, albite, biotite, and titanite. In figure 2b is shown a Ca-(Mg,Fe)-paragonite inclusion in amphibole replaced by preiswerkite at the contact to the amphibole host. Inclusions of Ca-“ephesite” in amphibole are not observed, suggesting that Ca-“ephesite” which in the rock matrix always is associated with preiswerkite is formed in a subsequent stage of metamorphism than the amphibole and the Ca-(Mg,Fe)-paragonite. Biotite ( $\approx 8\%$  in volume) occurs in oriented flakes in albite/oligoclase replacing pseudomorphically former phengites. Phengite is observed only as inclusion in albite. Clinozoisite is minor ( $\approx 2\%$  in volume). The corroded edges of clinozoisite (Fig. 2a) suggest that clinozoisite participated in plagioclase forming reactions.

Apatite is common but minor. It contains inclusions of biotite, albite, oligoclase and clinozoisite. Small (20-50  $\mu\text{m}$ ) two-phase (gas-liquid) fluid inclusions occur in apatite grains. Fluid inclusions are elongated parallel to the apatite *c*-axis. Titanite contains inclusions of rutile, albite, and biotite (Fig. 2f). The rutile inclusions in titanite record a preceded probably *HP* stage of metamorphism in this rock.

## Analytical methods

Major element composition and Li content of the rock were determined by AA.  $\text{Fe}_2\text{O}_3$  is determined titrimetric with  $\text{K}_2\text{Cr}_2\text{O}_7$ . Loss on ignition (LOI) was determined from the total weight change after heating the powdered sample at 1000  $^\circ\text{C}$  for 1 h. Microprobe analyses were performed using a JEOL 6380 LV SEM

equipped with an EDS INCAx-Sight 7388 at the School of Mining and Metallurgical Engineering, National Technical University of Athens. Amphibole nomenclature is after Leake et al. (2004). Representative mineral analyses are given in Tables 3, 4 and 5.

## Mineral chemistry

*Preiswerkite:* The preiswerkite has compositions with  $X_{\text{Mg}}$  ratios [ $X_{\text{Mg}} = \text{Mg}/(\text{Mg} + \text{Mn} + \text{Fe})$ ] ranging from 0.73-0.83, similar to  $X_{\text{Mg}}$  ratios shown in amphibole which is replaced by the preiswerkite ( $X_{\text{Mg}}^{\text{Amp}} = 0.68-0.78$ ) (Tables 3 and 5). The Kechros preiswerkite displays a solid solution towards Na-phlogopite up to 18 mol%, (the Si *apfu* are more than 4 and the Al atoms less than 6, Table 3). Complete solid solution has been demonstrated experimentally at 0.2 GPa, 600 $^\circ\text{C}$  (Liu 1989; Tlili 1990). Since the analyzed preiswerkites from the Kechros metadiorite show lower values of Mg+Fe+Mn+2Ti atoms than those of the Si atoms, it seems possible that the deficit in octahedral occupancies is compensated by Li atoms suggesting minor solid solution of ephesite component. The margarite solid solution in preiswerkite ranges from 0 to 2.4 mol% and the phlogopite from 0 to 6 mol%.

*Ca-“ephesite”/Ca-(Mg,Fe)-paragonite:* Ca-bearing dioctahedral Na-micas in nature have been interpreted as being solid solutions between margarite and paragonite (e.g. Ackermann & Morteani 1973; Höck 1974; Guidotti et al. 1979; Baltatzis & Katagas 1981; Frey et al. 1982; Feenstra 1996). The solid solutions and miscibility gap between margarite and paragonite are well known in nature and have been investigated experimentally (Franz et al. 1977; Tlili et al. 1990). Na-margarites also contain significant amounts of Mg and Fe. These Na-(Mg,Fe)-margarites are complex solid solutions of margarite –“Mica L”-paragonite with margarite as major component (Godard & Smith 1999). A solid solution between the Li-bearing trioctahedral Na-mica ephesite and margarite is

Table 3. Chemical compositions of preiswerkite from the metadiorite of the Kechros HP metamorphic complex

	1	2	3	4	5	6	7	8
SiO <sub>2</sub>	30.73	29.94	30.42	30.21	30.44	29.98	31.28	31.40
TiO <sub>2</sub>	-	-	-	-	-	0.17	-	0.24
Al <sub>2</sub> O <sub>3</sub>	34.65	34.51	34.85	34.95	34.04	34.22	34.57	33.60
FeO	8.45	8.19	8.46	8.01	8.66	7.40	7.69	7.27
MnO	0.43	0.20	0.21	0.30	-	0.01	-	0.14
MgO	13.99	14.18	13.64	13.45	14.09	15.18	14.37	15.04
CaO	0.26	-	0.29	0.30	0.19	0.09	-	0.16
Na <sub>2</sub> O	6.86	6.74	6.77	6.55	6.67	6.98	7.20	6.85
K <sub>2</sub> O	0.33	0.45	0.20	0.35	0.54	0.32	-	0.46
Total	95.40	94.21	94.44	94.12	94.63	94.35	95.11	95.16
Si	4.275	4.226	4.282	4.257	4.284	4.211	4.342	4.364
Al <sup>VI</sup>	3.725	3.774	3.718	3.743	3.716	3.789	3.658	3.636
	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al <sup>VI</sup>	1.956	1.967	1.993	2.062	1.930	1.876	1.997	1.869
Ti	-	-	-	-	-	0.018	-	0.025
Fe	0.983	0.967	0.996	0.945	1.019	0.870	0.893	0.845
Mn	0.051	0.024	0.025	0.035	-	0.001	-	0.016
Mg	2.901	2.983	2.862	2.826	2.955	3.178	2.973	3.116
	5.891	5.941	5.870	5.868	5.904	5.943	5.863	5.871
Ca	0.039	-	0.044	0.045	0.029	0.014	-	0.024
Na	1.850	1.845	1.848	1.789	1.821	1.902	1.938	1.846
K	0.059	0.081	0.036	0.063	0.097	0.057	-	0.082
	1.948	1.926	1.928	1.897	1.947	1.973	1.938	1.952
XMg	0.74	0.75	0.74	0.74	0.74	0.78	0.77	0.78
Mg+Fe+ Mn+2Ti	3.935	3.974	3.864	3.806	3.974	4.085	3.866	3.977
Al <sub>tot</sub>	5.681	5.741	5.711	5.805	5.646	5.665	5.655	5.505

also reported (e.g. Schaller et al. 1967; Velde 1971; Chopin et al. 1991).

The Ca-“ephesites” from Kechros, and the Ca-(Mg,Fe)-paragonites have unusual compositions reported according to our knowledge for the first time (Table 4). Since the ferric/ferrous iron ratios are not known from microprobe data the analyses are calculated assuming total iron as Fe<sup>2+</sup>. Li cannot be analyzed with the means of microprobe. Therefore we calculated the chemical formula of the microprobe analyses on a Li-free basis (Table 4, Fig. 3). From table 4 is shown that the <sup>VI</sup>Al occupancies in the chemical formula are more than 4 atoms indicating that the Ca-“ephesite” and Ca-(Mg,Fe)-paragonite from Kechros contain

either “ephesite-like” mica component (Na<sub>2</sub><sup>VI</sup>Al<sub>4.66</sub> [Al<sub>4</sub>Si<sub>4</sub>]O<sub>20</sub>(OH)<sub>4</sub>) or if Li is present, ephesite component. The Ca-“ephesite” and Ca-(Mg,Fe)-paragonite can be considered as solid solution of “ephesite” (referred in this work to true ephesite or/and “ephesite-like” sodium mica), paragonite, “Mica L” Na<sub>2</sub>(Mg,Fe)<sub>1</sub><sup>VI</sup>Al<sub>4</sub>[Si<sub>4</sub><sup>IV</sup>Al<sub>4</sub>]O<sub>20</sub>(OH)<sub>4</sub>, margarite and muscovite.

In the Ca-(Mg,Fe)-paragonite the margarite component ranges from 2 to 10 mol%, the “ephesite” from 0 to 13 mol%, the “Mica L” from 29 to 41 mol%, the paragonite from 49 to 66 mol% and the muscovite from 0 to 2.8 mol%. The Na-mica inclusions in amphiboles show the highest paragonite and “Mica L”

Table 4. Representative chemical compositions of Ca-(Mg,Fe)-paragonite and Ca-“ephesite” from the metadiorite from Kechros HP metamorphic complex

	Ca-(Mg, Fe) - Paragonite						Ca-“ephesite”						
	1	2	3	4	5	6	7	8	9	10	11	12	13
SiO <sub>2</sub>	39.02	39.60	41.02	39.16	38.15	38.79	36.16	33.24	33.98	33.93	33.69	34.68	33.74
TiO <sub>2</sub>	-	-	-	-	-	-	0.28	-	-	0.15	-	-	-
Al <sub>2</sub> O <sub>3</sub>	45.02	44.85	44.08	44.72	45.66	45.09	48.24	51.65	50.51	50.93	51.20	49.70	50.53
FeO	0.59	0.80	0.73	0.71	0.92	0.80	0.83	0.55	0.26	0.90	-	0.67	0.58
MnO	-	0.15	-	-	0.07	0.11	-	-	-	-	-	-	-
MgO	1.68	1.05	0.94	1.71	1.55	1.10	-	0.44	0.35	-	-	0.53	0.38
CaO	0.92	0.83	0.43	0.47	0.72	1.39	2.27	2.72	3.22	2.94	3.77	2.85	3.26
Na <sub>2</sub> O	7.38	7.13	7.36	7.49	7.26	6.87	6.44	6.03	6.05	6.02	5.77	6.08	5.94
K <sub>2</sub> O	-	-	-	0.26	0.06	0.34	0.03	-	-	-	-	-	-
Total	94.61	94.41	94.57	94.46	94.36	94.51	94.11	94.63	94.36	94.87	94.42	94.52	94.45
Si	5.086	5.166	5.321	5.116	4.996	5.077	4.752	4.362	4.468	4.447	4.424	4.554	4.441
Ti	-	-	-	-	-	-	0.028	-	-	0.015	-	-	-
Al	6.917	6.895	6.740	6.886	7.049	6.956	7.471	7.987	7.828	7.867	7.924	7.693	7.840
Fe	0.065	0.087	0.080	0.078	0.101	0.088	0.091	0.060	0.029	0.098	-	0.074	0.064
Mn	-	0.017	-	-	0.007	0.012	-	-	-	-	-	-	-
Mg	0.326	0.204	0.182	0.333	0.303	0.214	-	0.086	0.068	-	-	0.103	0.075
Ca	0.128	0.115	0.060	0.066	0.102	0.196	0.320	0.382	0.453	0.412	0.530	0.401	0.459
Na	1.866	1.804	1.851	1.880	1.832	1.744	1.642	1.535	1.542	1.530	1.468	1.549	1.516
K	-	-	-	0.042	0.009	0.057	0.005	-	-	-	-	-	-
Mrg	0.064	0.060	0.031	0.033	0.052	0.095	0.163	0.200	0.226	0.211	0.265	0.210	0.232
Pg	0.540	0.583	0.660	0.537	0.494	0.522	0.374	0.180	0.234	0.220	0.205	0.277	0.220
Mica L	0.390	0.308	0.262	0.411	0.411	0.315	0.091	0.146	0.097	0.098	-	0.177	0.135
“Eph”	0.006	0.045	0.047	-	0.038	0.040	0.370	0.474	0.443	0.471	0.530	0.336	0.409
Ms	-	-	-	0.021	0.005	0.028	0.002	-	-	-	-	-	-

components. In Ca-“ephesite” the “ephesite” component ranges from 29 to 53 mol%, the paragonite from 22 to 46 mol%, the margarite from 14 to 26 mol% and the “Mica L” from 0 to 18 mol%. Potassium is below the detection limit in the analysed Ca-“ephesites”. Deviation in the amounts of the end-member components is expected if Li is included in the calculation of the chemical formula. As the Li<sub>2</sub>O content is not known in the analyzed Ca-“ephesites” we recalculated the chemical formulae adding as Li<sub>2</sub>O is required to reduce <sup>VI</sup>Al atoms to 4. The Ca-“ephesite” analyses need 2.18 to 2.54 wt.% Li<sub>2</sub>O to reduce <sup>VI</sup>Al atoms to be 4, and the Ca-(Mg,Fe)- paragonite analyses need 0.2 to 0.78 wt.% Li<sub>2</sub>O. The total Li<sub>2</sub>O content in the rock is 0.0278 wt.% and the Ca-“ephesite”

participates in the rock with ≈2 vol%. Assuming that the total Li content of the rock is bound only in the Ca-“ephesite”, then the calculated “ephesite” component in the Ca-“ephesite” analyses comprises mainly true ephesite- and less “ephesite-like” Na-mica component.

The relationship between paragonite-margarite-“ephesite”, paragonite-Mica L-“ephesite” and paragonite-Mica L-margarite is shown in figures 3a, 3b and 3c respectively. Figure 3d shows the relation between paragonite, Mica L and “ephesite” plus margarite. Based on the paragonite, “Mica L”, “ephesite” and margarite components the two compositional groups are well distinguished: (a) the Ca-(Mg,Fe)-paragonite with low contents

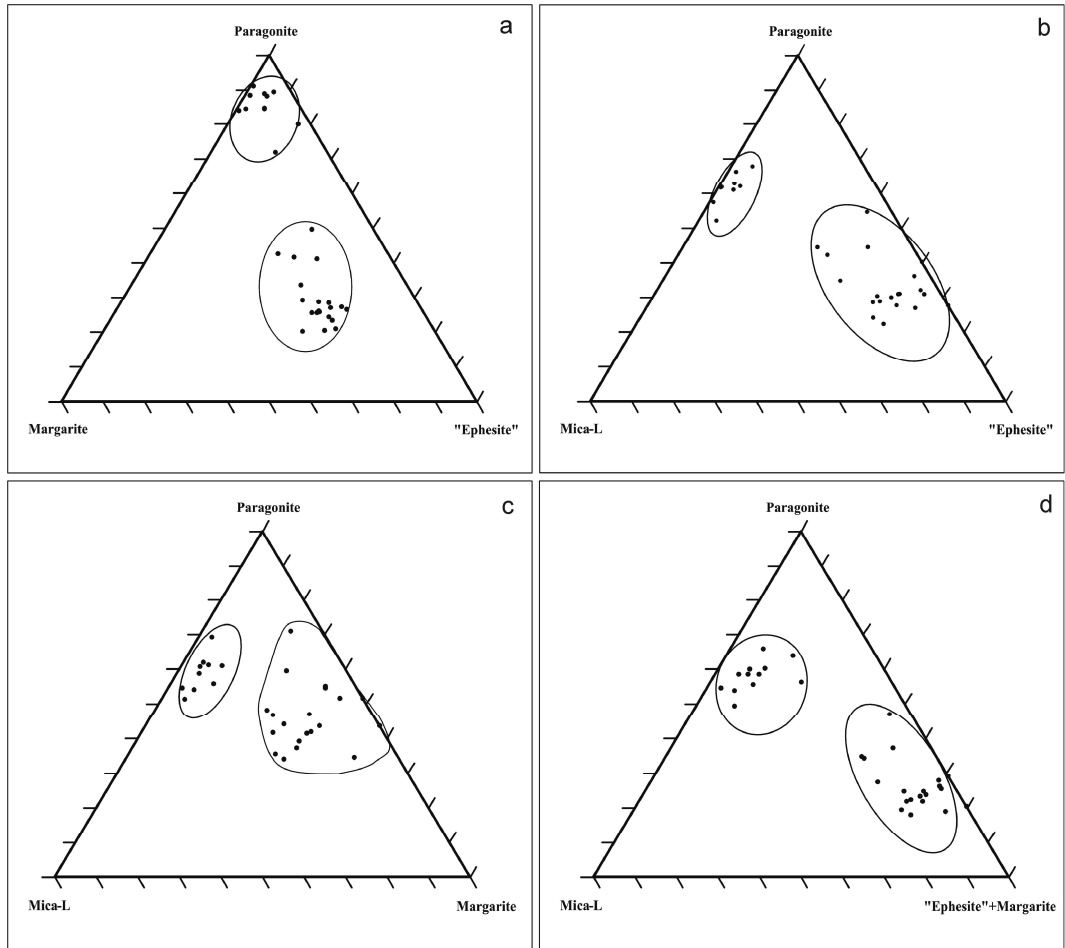


Fig. 3. Ca-(Mg,Fe)-paragonite and Ca-“ephesite” compositions from the Kechros metadiorite. End-member proportions are calculated with respect to the system Pg-Mrg- Mus-“Mica L”-“Eph”. “Eph” comprises in this work the calculated on a Li-free basin “ephesite-like” Na-mica. The calculation sequence is as follows:  $X_{Mus}=X_K$ ;  $X_{Mrg}=X_{Ca}$ ;  $X_{Pg}=[Si-(SixX_{Mar}+SixX_{Mus}):1-(X_{Mar}+X_{Mus})-4]:2x1-(X_{Mar}+X_{Mus})$   
 $X_{MicaL}=(Mg+Mn+Fe)$ ,  $X^{“Eph”}=1-(X_{Mus}+X_{Mrg}+X_{Pg}+X_{mical})$

in margarite and “ephesite” component and high contents in Mica-L component in solid solution; (b) the Ca-“ephesite” with lower Mica L and paragonite and higher margarite component than the Ca-(Mg,Fe)-paragonite (Fig. 3 b, c, d). The Ca-(Mg,Fe)-paragonite and Ca-“ephesite” from Kechros show considerable solid solution towards Mica L (Fig. 3 b, d). The Ca-(Mg,Fe)-paragonite displays the greatest

Mica L proportion ever reported for paragonite. Similar paragonite and Mica L compositions to those of the Kechros Ca-“ephesite” show Na-margarites associated with preiserwerkite from the Liset retrogressed kyanite eclogite (Kechid 1984; Godard & Smith 1999).

*Amphibole:* Amphibole grains are commonly homogeneous in composition. Small compositional zoning with increasing FeO

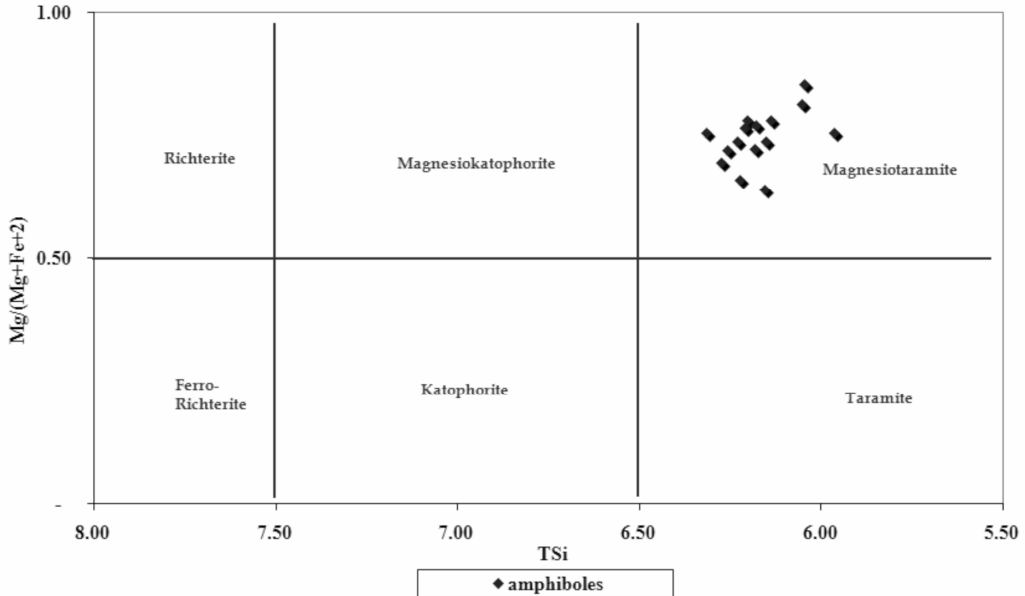


Fig. 4. Chemical composition of the Na-Ca-amphiboles in terms of  $\text{Si}:\text{Mg}/(\text{Mg}+\text{Fe}^{2+})$

content toward the rim is observed in some amphibole grains. Representative compositions are given in table 5. The amphiboles are Al-rich Na-Ca-amphiboles with 6.054 to 6.259 Si *apfu*, 1.076 to 1.543 Na *apfu*, 0.623 to 0.715 Na<sub>B</sub> *apfu* and  $\text{Mg}/(\text{Mg}+\text{Fe}^{2+})$  ratio from 0.70 to 0.77. They plot in the compositional field of magnesiotaramite (Fig. 4).

*Biotite* is rich in Mg; the  $\text{Mg}/(\text{Mg}+\text{Fe})$  ratio ranges from 0.69-0.80. Only one *phengite* flake was analyzed (Table 5); it is found as inclusion in albite. The Si content in *phengite* is 6.785 *apfu*. The anorthite component in *albite* ranges from 1 to 6 mol% and in associated *oligoclase* from 13 to 20 mol%. The  $\text{Fe}_2\text{O}_3$  content in *clinozoisite* ranges from 5.92-7.07 wt.%.

## Discussion

Preiswerkite is still a rare mineral of mica group. Since its discovery in 1980, preiswerkite has been found only in eight localities worldwide (Table 1). It is found for the first time in the Rhodope *HP* metamorphic area.

The most known localities of preiswerkite involve metabasic, metaultramafic and metaacidic rocks, in which preiswerkite formed during retrogression following eclogite- or amphibolite facies metamorphism, or during a late to retrograde stage greenschist to amphibolite facies overprint (Table 1). In three localities (Liset, La Compointrie, and Koralpe/Sauvalpe) preiswerkite occurs in retrogressed eclogites and is associated with Na-(Mg,Fe)-margarite. In the Kechros complex the studied preiswerkite occurs in a metadiorite included within antigorite serpentinite, and is associated with Ca-bearing Al-rich Na-mica named in this work as Ca-“ephesite”. The mineral assemblage  $\text{Ab}+\text{Pl}+\text{Amp}+\text{Pg}+\text{Bt}+\text{Ca-Eph}+\text{Prw}+\text{Czo}+\text{Ttn}$  in the metadiorite is diagnostic for the transition zone of the albite-epidote-amphibolite facies to amphibolite facies metamorphic conditions. Typical *HP* mineral phases, like Na-rich clinopyroxene, are not found in the metadiorite. Only the *phengite* inclusion in albite and the rutile inclusions in titanite suggest that a *HP* event preceded the

Table 5. Representative chemical compositions of Na-Ca-amphiboles, biotite, phengite, plagioclase and clinozoisite from the metadiorite from the Kechros HP complex

	Na-Ca-Amp				Bt		Ph	Pl		Czo
	1	2	3	4	5	6	7	8	9	10
SiO <sub>2</sub>	42.88	43.26	42.52	42.98	38.93	38.35	49.95	64.80	63.43	38.94
TiO <sub>2</sub>	0.47	0.45	0.67	0.59	1.67	1.01	-	-	-	-
Al <sub>2</sub> O <sub>3</sub>	19.24	19.86	20.03	19.04	18.57	20.37	26.17	22.10	22.88	29.25
FeO <sub>t</sub>	7.85	8.75	8.83	7.75	9.11	7.01	3.82	-	-	Fe <sub>2</sub> O <sub>3</sub> =5.22
MnO	0.51	0.24	0.27	-	-	-	-	-	-	-
MgO	11.58	11.47	11.44	12.34	16.60	18.21	3.76	-	-	-
CaO	8.54	8.55	8.33	8.52	-	-	-	3.34	4.02	24.20
Na <sub>2</sub> O	5.52	3.87	4.37	5.41	0.91	1.81	-	9.92	9.44	-
K <sub>2</sub> O	0.52	0.86	0.74	0.55	8.79	7.82	10.97	-	-	-
Total	97.12	97.32	97.20	97.21	94.60	94.58	94.68	100.16	99.78	98.32
Si	6.181	6.203	6.121	6.173	5.631	5.479	6.785	2.850	2.807	3.000
Ti	0.051	0.049	0.072	0.064	0.183	0.109	-	-	-	-
Al	3.268	3.356	3.399	3.224	3.166	3.430	4.190	1.146	1.193	2.657
Fe	0.946	1.050	1.063	0.931	1.102	0.837	0.434	-	-	Fe <sup>3+</sup> =0.343
Mn	0.062	0.029	0.033	-	-	-	-	-	-	-
Mg	2.488	2.453	2.455	2.642	3.579	3.879	0.761	-	-	-
Ca	1.319	1.314	1.285	1.312	-	-	-	0.157	0.191	1.998
Na	1.543	1.076	1.220	1.507	0.256	0.500	-	0.846	0.810	-
K	0.096	0.158	0.136	0.101	1.622	1.425	1.901	-	-	-
X <sub>Mg</sub>	0.72	0.70	0.70	0.74	0.76	0.82	-	-	-	-

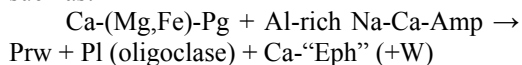
albite-epidote-amphibolite facies and amphibolite facies metamorphism. Minimum pressure of  $\approx 1.2$  GPa is constrained from the Si content in phengite (Si=6.785 *apfu*, Table 5) for assuming temperature of  $\approx 550^\circ\text{C}$  (Massonne & Szpurka 1997). However, eclogites and partially or completely amphibolitized eclogites occur within the antigorite serpentinite as well as in the neighboring crustal rocks indicating that the whole Kechros complex underwent eclogite facies metamorphism. Pressures at 1.7 GPa are constrained from the neighboring eclogites (Mposkos et al. 2011). Decompression was isothermal up to a pressure of  $\approx 0.4$  GPa within the narrow stability field of chloritoid-staurolite-chlorite constrained from the associated metapelites (Mposkos 1989; Mposkos & Liati 1993). The mineral assemblages in the metadiorite probably record various stages of metamorphic overprint

following decompression. The chemical composition of the Na-Ca-amphiboles indicate amphibole formation at moderate high pressures (Laird & Albee 1981), within the albite-epidote-amphibolite facies field as indicate the albite inclusions in the amphiboles and the coexistence of magnesiotaramite with albite in the matrix. The Ca-(Mg,Fe)-paragonite inclusions in the Na-Ca-amphibole may represent relicts of the *HP* stage as paragonite is stable at the peak *P-T* conditions (1.7 GPa /  $570^\circ\text{C}$ ) constrained from the neighboring eclogites.

Textural relationships show that preiswerkite is associated with oligoclase and Ca-“ephesite” replacing Ca-(Mg,Fe)-paragonite and Na-Ca-amphibole (Fig. 2 a, b, c, e). Figure 2b shows preiswerkite replacing Ca-(Mg,Fe)-paragonite inclusion in amphibole. These textures indicate that preiswerkite is formed consuming Ca-(Mg,Fe)-paragonite and



amphibole probably according to a reaction such as:



The “graphic intergrowth” like textures shown in figures 2b and d represent former Ca-(Mg,Fe)-paragonite flakes replaced pseudomorphically by preiswerkite, Ca-“ephesite” and plagioclase by the above mentioned reaction. The reaction was more complex since minor K-feldspar is commonly associated with preiswerkite, oligoclase and Ca-“ephesite”. It is formed consuming potassium from the amphibole and the Ca-(Mg,Fe)-paragonite. However, participation of phengite in the formation of K-feldspar is not excluded.

Geothermobarometry cannot be performed on preiswerkite or Ca-“ephesite”, as the thermodynamic properties of these micas are unknown. However, textural relationships indicate that the Kechros preiswerkite coexists with oligoclase (Fig. 2 a, b, c, d) and is probably formed at the transition from the albite-epidote-amphibolite to amphibolite facies conditions, during the isothermal decompression. In the Liset, La Compointrie and Koralpe/Saualpe eclogites preiswerkite is formed by water consuming reactions during retrogression (Godard & Smith 1999). In these localities preiswerkite coexists with Na-(Mg,Fe)-margarite and both phases are formed consuming kyanite and amphibole. The preiswerkite from Kechros metadiorite coexists with Ca-“ephesite” and is formed by a reaction consuming Al-rich Na-Ca-amphibole, Ca-(Mg,Fe)-paragonite (plus phengite and clinozoisite) and producing preiswerkite, Ca-“ephesite”, oligoclase, K-feldspar and probably water.

Preiswerkite and other Na-micas close to the Na-phlogopite-preiswerkite solid solution were synthesized by Carman (1974), Hewitt and Wones (1975), Franz and Althaus (1976), Koons (1982), Líu (1989) and Tlili (1990), at *P-T* conditions indicating stability of preiswerkite, corresponding to upper greenschist- and amphibolite facies. In nature, preiswerkite appears mainly in former *HP* rocks but its genesis is favored by a drop in pressure

(Godard & Smith 1999; Table 1). As stated by Godard and Smith (1999), preiswerkite’s rarity is not related to extreme or unusual *P-T* conditions but to unusual chemical compositions. Preiswerkite is a trioctahedral Na-mica with the maximum tschermakitic substitution ( $^{\text{VI}}\text{Al}^{\text{IV}}\text{Al}=\text{Mg Si}$ ) i.e. it is Al-rich and Si-poor. “Mica L”, margarite and ephesite contain even more Al than preiswerkite from which they are derived by the substitutions ( $3\text{Mg}=2^{\text{VI}}\text{Al}$ ), ( $\text{Ca}^{\text{VI}}\text{Al}=\text{Na}2\text{Mg}$ ) and ( $4\text{Mg}=2\text{Li}2^{\text{VI}}\text{Al}$ ). Instead of the common paragonite-margarite solid solution, the more aluminous solid solution “ephesite”-margarite is observed (Table 4, Fig. 3).

These facts indicate that preiswerkite and Ca-“ephesite” appear in Na-Al rich systems, like that of the Kechros metadiorite, at *P-T* conditions of the transition zone between albite-epidote amphibolites and amphibolite facies. The increase in paragonite component seems to stabilize the Al-rich sodic micas at higher pressures. Paragonite richer components show the sodic mica inclusions in the Na-Ca-amphiboles and are formed at the *HP* stage of metamorphism whereas those in the matrix, which are associated with preiswerkite and are formed during decompression show much higher margarite and “ephesite” components (Table 4, Fig. 3).

## Conclusions

The studied preiswerkite from the Kechros metadiorite has compositions with  $X_{\text{Mg}}$  ranging from 0.73 to 0.83, similar to  $X_{\text{Mg}}$  shown in amphibole ( $X_{\text{Mg}} \text{ Amp}=0.68-0.78$ ) which was consumed by the preiswerkite formation. The preiswerkite displays a solid solution towards Na-phlogopite up to 18 mol%, towards margarite up to 2.4 mol% and towards phlogopite up to 6 mol%.

The studied Ca-(Mg,Fe)-paragonites and Ca-“ephesites” can be considered as solid solutions between paragonite, Mica-L, margarite, “ephesite” and muscovite. The Ca-(Mg,Fe)-paragonite is characterized by high “Mica L” contents and low margarite and “ephesite” contents. The paragonite component ranges

from 49 to 66 mol%, the Mica-L component from 29 to 41 mol%, the margarite component from 2 to 10 mol% and the “ephesite” component from 0 to 13 mol%. In Ca-“ephesite” the “ephesite” component ranges from 29 to 53 mol%, the paragonite component from 22 to 46 mol%, the margarite component from 14 to 26 mol% and “Mica-L” component from 0 to 18 mol%.

As indicated by experimental and natural occurrences, the stabilities of preiswerkite and Ca-“ephesite” are not restricted to extreme or unusual *P-T* conditions. The rarity of these micas in nature seems to be due to unusual chemical compositions; they appear in H<sub>2</sub>O-saturated Na-Al-rich Si-poor compositions principally at greenschist- or amphibolites facies *P-T* conditions (see also Godard and Smith 1999). Such conditions are realized in the quartz-free metadiorite from the Kechros high-*P* metamorphic complex during decompression. The preiswerkite from the Kechros metadiorite coexists with oligoclase and Ca-“ephesite” and the three minerals are formed replacing Ca-(Mg,Fe)-paragonite and Al-rich Na-Ca-amphibole at the transition from the albite-epidote-amphibolite facies to amphibolite facies conditions during decompression. The Ca-(Mg,Fe)-paragonite represents relict phase formed at the *HP* metamorphic stage.

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