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Original scientific paper

HYDROTHERMALLY OPALIZED SERPENTINITES IN TETHYAN OPHIOLITE SEQUENCE IN CENTRAL SERBIA

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A b s t r a c t: Intensely fractured serpentinite protrusions in Vardar tectonic zone of central Serbia are a part of accreted Jurassic Tethys ophiolite sequence. Primary ultramafic rocks presenting remnants of oceanic crust were regionally metamorphosed into serpentinites after middle Jurassic. Tertiary magmatic activity of calc-alkaline character occurred along deep faults marking the zone of closure of Tethys ocean trench. Hydrothermal activity driven by magmatic heat, through fracture and fault zones, has caused opalization of ultramafic host and vein deposition. Results of various examinations of these opalized serpentinite are presented: field examinations, chemical, microscopic, X-ray diffraction (XRD) and X-ray fluorescence (XRF) analyses. Opalized serpentinite has been identified as predominating tridymite-cristobalite type opal-CT. Practical significance of these opal-CT occurrences lies in possibility for its use as a gemstone and in prospection of genetically and spatially related hydrothermal deposits (cryptocrystalline magnesite veins and other siliceous gemstone types). Occurrences of opalized serpentinite of this type have been discovered on many locations along Tethys suture zone.

Key words: hydrothermal alterations; serpentinite; opal-CT; Tethys

INTRODUCTION

Opalized serpentinite masses in central Serbia have beed used in the 20th century as fluxer masses without significant examinations. The first examinations were performed by Poharc-Logar & Logar (1998), and our aim was to add more information on further examinations on this interesting occurrence which could be more widely used as an industrial mineral.

Geological history of terrain comprised by territory of Serbia, as a part of East European Alpine belt, is closely tied to evolution of Tethys ocean - its opening, widening and its closure, as well as relative movement between Eurasian and Adriatic plates seen through global plate tectonics. Vardar tectonic zone is a part of Tethys ocean domain in central part of Balkan Peninsula, characterized by significant ophiolite sequence marking the zone of Tethys ocean trench, as a part of Jurassic ocean crust (e.g. Janković, 1990). This suture zone has a more or less continuous spreading through Serbia and further to the southeast over Macedonia, Greece and Turkey (Janković, 1990), where it turns east, to Izmir-Ankara zone (Elmas & Yiğitbaş, 2001). To the northwest, it extends over Pannonian basin and Croatia, Austria - Eastern Alps towards northern Italy (Dimitrijević, 1995), but is mostly covered by thick Cenozoic sediments. Many authors have given their opinion of geotectonic evolution of Vardar zone. One of the most detailed studies of geohistorical evolution for the whole Balkan Peninsula including Vardar zone, has recently been given by Karamata (2006).

There are two main ophiolitic belts in Vardar zone, marking the deep faults indicated also by aeromagnetic studies (Vukašinović, 1976). These regional faults are known as: 1. the Central deep fault and 2. the Western deep fault (Fig. 1). The opening of these deep faults happened during Jurassic, in formation of Tethys trench ocean basin (Janković, 1990). Afterwards, during late Jurassic and Cretaceous, due to regional pressures, Tethys trenches were closed and transformed into suture zones located above eastward subducting Adriatic plate suture zone. Ultramafic rocks from lower parts of the ocean floor have been compressed, protruded and obducted over younger rock formations, and regionally altered to serpentinites in the oxidizing conditions in near-surface and surficial levels of the terrain. The continued regional pressures during Cenozoic have caused partial melting in lower parts of the crust. In syn- to postcollisional extensive settings, intensive magmatic activity of calc-alkaline character (from rhyolite to basalt and from granodiorite to diorite) has occurred along regional fractures and splay faults, which was accompanied and followed by hydrothermal activity. Thus, mostly siliceous hydrothermal veins have been formed (chalcedony, jasper, quartz). There are also different types of hydrothermal alterations present in serpentinites, encircling these veins (Marčeta, 2005).

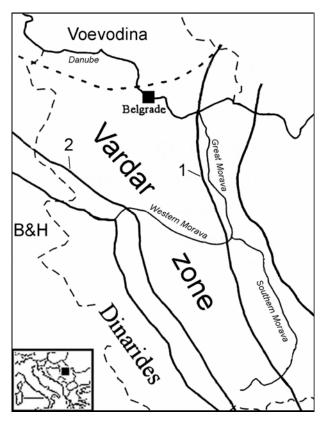


Fig. 1. Generalized tectonic map of central and southern Serbia with main regional fault zones and extension of Vardar tectonic zone. Inset – location in Europe. Key: 1 – Central deep fault; 2 – Western deep fault

Based on field studies and laboratory examinations, it has been found that primary ultrabasic rocks in ophiolitic belts in central Serbia have been predominantly hartzburgites, less often lherzolites, while dunites were rare. In regional serpentinization process, primary minerals (olivine, bronzite, diallage, magnetite, chromite) have been transformed into various serpentine minerals (antigorite, bastite), talc, spinel group minerals, magnesite and other carbonates. Additionally, due to intensive regional pressures and tectonic processing, serpentinites have been intensely crushed and have tectonic contacts with surrounding rocks.

Due to magmatic activity and hydrothermal activity associated with it, regional serpentinization of ultramafic rocks in these ophiolite sequences has been followed by various hydrothermal alterations, occurring sporadically along regional and splay faults, and mineral composition has again been substantially changed. Differences in chemical compositions between primary ultrabasic rocks, serpentinized and hydrothermally opalized are presented in Table 1. Ignition loss on 450 and 1000 °C results have shown high values. According to Flörke et al. (1991), a distinction is made between molecular water (H₂O_{mol}) and silanol groups (H₂O_{SiOH}) in different silica species structures. However, distinction between two subtypes (A isolated molecules and hydroxyl groups trapped in the structure, and B - strongly hydrogen-bonded accumulations of water molecules of hydroxyl groups within the structure or on external and internal surfaces) can be made with infrared and near-infrared spectroscopic studies. Former examinations have shown that ignition loss can be due to capillar water and Fe-hydroxide dispersed in it (Poharc-Logar & Logar, 1998).

Table 1

Chemical analyses results of generally unaltered
to poorly altered peridotite, serpentinized
peridotite and hydrothermally opalized
serpentinized peridotite

Substance	wt%			
Substance	1	2	3	4
SiO ₂	40,69	41,29	94,28	87,68
TiO ₂	0,12	0,11	_	_
Al_2O_3	0,98	0,93	0,02	0,05
Fe ₂ O ₃	4,76	6,48	0,75	3,82
FeO	4,19	2,89	0,14	0,29
MnO	0,10	0,13	_	_
MgO	43,18	35,13	_	_
CaO	1,10	0,68	_	_
Na ₂ O	0,07	0,04	_	_
K ₂ O	_	0,03	0,05	0,01
H_2O	4,78	11,94	4,89	8,15
Ignition loss ^{450°}	/	/	11,60%	5,66%
Ignition loss ^{1000°}	/	14,28%	12,93%	6,28%

1) Peridotite (average for central Serbia); 2) Serpentinite i.e. serpentinized peridotite (central Serbia); 3) Opalized serpentinite ("emeraldgreen opal-CT"); 4) Opalized serpentinite ("variegated opal-CT").

3

Opalization of serpentinite accompanied with silica vein deposition has been observed in 13 localities in central Serbia (Šumadija region) in Vardar zone so far, in the area through which the deep faults (Fig. 1) extend. The formation of opal is caused by hydrothermal activity driven by magmatic heat – in metasomatic interactions of hydrothermal solutions with ultramafic rock, which resulted in partial to complete transformation of the rock into a mixture of silica polymorphs (opal-CT) \pm carbonates and clay minerals.

HYDROTHERMAL ALTERATIONS OF SERPENTINITES – FIELD OBSERVATIONS

The predominating types of alteration of examined serpentinites are: silicification, carbonatization, pyritization (and subsequent limonitization) and opalization. These processes are generally termed listwenitization and are common in ultramafic rocks in zones comprised by former Tethys domain (Antonović & Vasković, 1992). Silicification is tied to limited zones around siliceous veins and thin-veined stockworks. The influx of silica is due to its extraction from the lower zones of serpentinites (Protić et al., 1998; Ilić, 1969) or other rock formations through which hydrothermal solutions have circulated, and its precipitation in shallower environment due to changes in chemical (redox, pH, saturation state) and physical (T, P)properties (Yee et al., 2003; Potapov, 2004). The products of other types of alteration are intimately intermixed and spread through the whole mass of hydrothermally altered serpentinites around various fractures, most often tectonic contacts.

Opalization of serpentinite resulted in formation of opal-CT. The zones of opalized serpentinite can be found around all siliceous \pm carbonate hydrothermal mineralizations in these rocks, and around fractured zones, which were a flow path of hydrothermal fluids, even when no pronounced vein deposition took place.

Almost all former works in Serbia examining these opalized serpentinites (with or without siliceous veins) treated these mineralizations as weathering profiles, until works of Miladinović (2005) and Marčeta (2005) suggested, based on field observations of fault-focused zoned alteration patterns with decreasing alteration intensity away from the fluid pathway and other rock formations pinched in the same fault zones also intensely silicified, and laboratory examinations, that these mineralizations are of hydrothermal origin.

Opalization of serpentinites is due to hydrothermal alteration by CO₂-rich low-temperature water solutions (White et al., 1956; Işik et al., 2005). The degree of opalization is visibly decreasing further from the influx fault and, gradually, opalized serpentinite passes into hydrothermally unchanged, plain serpentinite. Oxygen isotopic composition analyses of specimens from other parts of Tethys suture zone found in very similar geological setting have indicated the temperature of the mineralising fluid of 65 to 80°C (Işik et al., 2005).

The main products of hydrothermal alteration are the various types of silica and carbonate minerals (mainly magnesite, dolomite, ankerite).

Cristobalite-tridymite opal is common in hydrothermal magnesite deposits in serpentinites and adjacent rocks (Ilić et al., 1984; Radovanović S., 1985; Đurić S. et al., 1990), as veinlets and stockworks. However, specimens examined in this study have been chosen from a large opalized serpentinite mass with magnesite veins generally absent at the surface.

The habit of serpentinite in opal-CT is partly preserved, but has obtained a glassy, jointed jellylike habit, with slight or complete change of colour. Its fracture is conchoidal, lustre vitreous to greasy; it is opaque to semitransparent in thinner sections, prevailingly isotropic or with very weak birefringence under microscope. Its density varies from 2.06 to 2.23 g/cm³, and its refractive indices from 1.441 to 1.455 (Poharc-Logar & Logar, 1998). The chemical composition of zones of opalized serpentinite without intensive colouring (with Fe₂O₃ content below 1 wt%), which are predominant in all locations, shows average silica content around 90 wt%. The most conspicuous change in chemical composition during alteration is increase in silica content from about 40 to about 90 wt%, and decrease in magnesia content from around 40 wt% to 0.

THE RESULTS OF X-RAY POWDER DIFFRACTION ANALYSES

The results of powder XRD analyses on specimens of opalized serpentinite have shown

domination of low temperature tridymite in their composition, with lower content of low tempera-

ture cristobalite, amorphous silica (opal) and quartz. Quartz, tridymite and cristobalite have close *d* values, and these formations are generally termed opal-CT (Flörke, 1967). Short descriptions of XRD analyses results for these specimens are given below, and diffractogrames are shown in Figures 2 and 3 (Operating conditions: Philips PW 1710 CuK α 1.54178 Å, voltage 40 kV, current intensity 30 mA, 2 Θ from 5 to 60°, 0.02° step for 0.5 s).

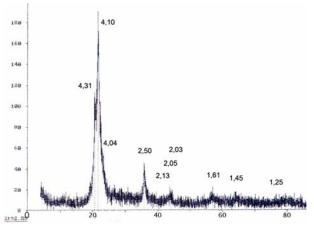


Fig. 2. X-ray diffractograme of opalized serpentinite (variegated opal-CT) from Šumadija

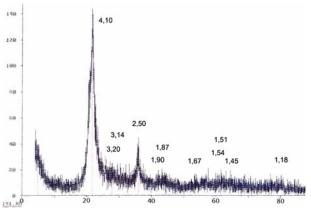


Fig. 3. X-ray diffractograme of opalized serpentinite (emerald-green opal-CT) from Šumadija

Majority of authors examining silica of sedimentary and/or hydrothermal origin have stated that opal-CT is an intermediate product of amorphous silica to quartz transformation process predominantly of low cristobalite type \pm interstratified stacking of tridymite layers in the structure (e.g. Mizutani, 1977). Interestingly, opals formed in lateritic zones, i.e. weathering profiles of ultrabasic rocks (not of hydrothermal origin) of the Vardar zone have been reported to be of predominating cristobalitic type (e.g. Maksimović, 1980), in contrast of examined hydrothermal occurrences.

Specimen 1: variegated opal-CT

The specimen was taken from the zone of the serpentinite around the fault along which the hydrothermal alterations have been most intensive. It has completely opalized matrix, lighter in colour than the parent rock, and sporadic darker zones with larger content of dispersed, tiny metallic mineral grains macroscopically visible.

XRD results show that major components of the specimen are low tridymite (strongest peaks on 4.10, 4.31, 2.50 and 2.05 Å), low cristobalite (smaller peaks on 4.04 and 1.61 Å), quartz (on 1.45 and 1.25 Å) and amorphous silica (few broadened peaks). Because of the similarities in *d* values of low cristobalite and low tridymite, such mineral masses are generally marked as opal-CT, and only the 4.3 Å reflection is ascribed exclusively to tridymite (Flörke 1967; Flörke et al. 1991; Cady et al. 1996).

Specimen 2: green opal-CT

The specimen was taken from the zone of the serpentinite, which makes up the transitional zone from completely opalized to hydrothermally unaltered serpentinites. Its colour is so-called emerald-green, much darker than the previous specimen. It has partly preserved granular texture with conspicuous bastitized pyroxene relicts in opalized matrix.

XRD results show that major component of the specimen is tridymite (low temperature tridymite) whose strongest peaks are on 4.10 and 2.50 Å, and weaker on 1.54 and 1.51 Å. Some weaker peaks belong to quartz (on 1.45 and 1.18 Å), low cristobalite and amorphous silica.

Poharc-Logar & Logar (1998) first found this hydrothermally altered serpentinite to be opal-CT, based on detailed mineralogical laboratory analyses. Based on performed XRD analyses, conclusion was made that few stronger reflections of opal-CT could come from the basic tridymite structure as defined in former works of Flörke (1967), but the intensities are different. Infrared absorption spectrometry of opalized serpentinite has shown presence of bands characteristic for polymorphs of SiO₂ (on 1100 cm⁻¹ and 470 cm⁻¹), while the band on 780 cm⁻¹ characteristic for disordered SiO₂ polymorphs shows asymmetry indicating minor content of quartz, as shown also by XRD analyses. The bands characteristic for ordered cristobalite and opal-C are absent, while present bands could not be ascribed to opal-A due to higher intensities and sharper peaks. Same authors have also undertaken a detailed study of the colour origin and found that it originates from iron oxyhydroxide minerals of varying composition – intertransitions of limonite, hydrohematite and hematite.

THE RESULTS OF X-RAY FLUORESCENCE ANALYSES

X-ray fluorescence (XRF) analyses have been performed in Institute IMS Laboratory in 2007 (Table 2) (Operating conditions: Energy-dispersive x-ray fluorescence, spectrometer Oxford ED 2000 with Ag target anode, Si(Li) detector and SMART digital pulse processor, excitation time 200 s, resolution 170–185 eV, voltage 5–50 kV. Analyses done in Institute IMS Laboratory for materials examination, Belgrade).

XRF analyses results of serpentinite and its hydrothermal alteration products - green and variegated opal show interesting relations. Na, Mg, Cl, Mn, Fe, Co and Cu have been subtracted during alteration process in varying degree. Decrease in Na content from 1400 mg/kg to 0, Mg from 120000 to 400 and 80, and Fe from 67000 to 17000 and 2100 are the most conspicuous and show that the intensity of their subtraction is correlated to intensity of alteration degree increase. Si, P, S, K, Ca, Ti, V, Cr, Ba and W have been introduced during alteration, also in varying degree. The introduction of Ca is readily observed in all examined occurrences, and is due to its leaching from surrounding Cretaceous limestone formations caught up in fracture zones together with serpentinites. Cr, Ni, S, K, Ca, Ti, V and Zn display a varying pattern – an increase in partially opalized and decrease in completely opalized serpentinite. Only Se and Pb have retained their content unchanged during alteration process.

Table	2
	Comparative XRF analyses results

Element		ppm	
Element -	1	2	3
Mg	120000	400	80
Al	9200	9400	2800
Si	244500	380000	430000
Р	20	250	340
S	20	70	60
Cl	77	20	15
K	90	300	270
Ca	2700	4000	900
Ti	170	250	180
V	74	120	11
Ce	10	11	16
Ba	7	13	18
Cr	1830	6000	2000
Mn	150	50	53
Fe	67000	17000	2100
Co	48	30	20
Ni	455	520	40
Zn	10	22	8
Se	1	1	1
Zr	3	4	4
W	0	2	0
Pb	7	7	7
Na	1400	-	_
Cu	1	_	_

1) hydrothermally unaltered serpentinite; 2) partially opalized serpentinite, "green opal"; 3) completely opalized serpentinite, "variegated opal")

THE RESULTS OF MICROSCOPIC EXAMINATION IN TRANSMITTED LIGHT

Variegated opalized serpentinite (green-black, white-black and green-white)

Specimens of serpentinite taken from the zone around the influx fault show intensive opalization. Pseudorelicts of serpentine group minerals can be seen under microscope, and grains of spinels as relicts of primary peridotite minerals. Serpentinite and silica (tridymite, cristobalite, opal) replacing it make up weak birefringence matrix, with grains of bastitized pyroxenes. Veinlets of chalcedony are readily observed. They account for small amounts of quartz registered in XRD analyses (Fig. 2).

Figure 4 shows photomicrographs of intensely opalized serpentinite matrix with chalcedony veinlets.

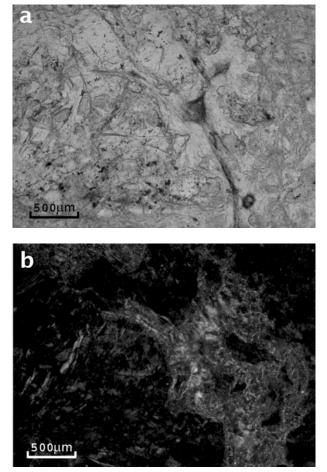


Fig. 4. Photomicrographs of opalized serpentinite a) parallel nicols, b) crossed nicols.

Green-coloured opalized serpentinite (so-called emerald-green)

Specimens of serpentinite taken from the transitional zone towards hydrothermally unaltered serpentinites (away from the fault that served as an influx channel for hydrothermal solutions) display lower degree of opalization. The rock is composed of serpentine minerals (antigorite relicts), bastitized pyroxene relicts and chlorite, all transformed into silicified mass (tridymite, cristobalite, opal) with sporadic primary minerals (spinel group).

Microscopically, appearance of this type of opalized serpentinite is very similar to previously

described, except bastitized and chloritized pyroxenes are more conspicuous, due to lower degree of opalization. Their birefringence is more intensive and discernible in fine-grained opalized serpentine matrix. Intense fracturation of pyroxenes caused by exhumational folding of serpentinite masses is also more conspicuous, because opalization, which has healed the same in more intensely opalized types, was not fully exerted.

Figure 5 shows bastitized pyroxenes in opalized serpentinite matrix (so-called emerald-green opal).

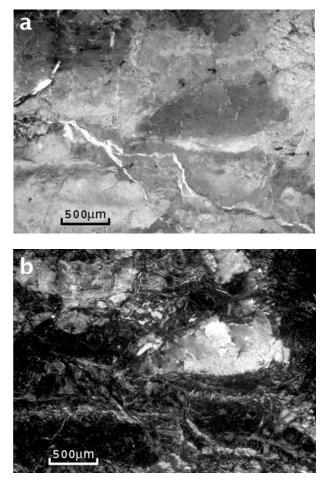


Fig. 5. Photomicrographs of partial opalization of serpentinite a) parallel nicols, b) crossed nicols.

DISCUSSION OF RESULTS

As can be seen from the photomicrographs of opalized serpentinite, the pseudorelicts of serpentinite can be clearly observed, and its texture is mostly preserved. However, powder XRD analyses did not confirm the presence of minerals typical for these rocks, but only silica polymorphs presence; chemical analyses and IR absorption spectrometry also show predomination of silica. This indicates that, even though the habit of serpentinite is partly preserved, its composition is completely or almost (within XRD sensitivity range) completely changed, by pseudomorphic replacement by silica. The material has optically higher relief than serpentine minerals, and is optically pseudoisotropic under crossed Nicol prisms unlike serpentine minerals, due to submicroscopic grain size of silica (Cady et al., 1996).

CONCLUSION

Hydrothermal alteration that lead to opalization of serpentinite was presumably caused by lowtemperature hydrothermal action, but the process of opal genesis by hydrothermal opalization of serpentinite is not yet completely elaborated. According to the model of hydrothermal alteration and deposition by CO₂-rich fluids in serpentinites provided by Isik et al. (2005), the decrease of temperature at shallower depths would cause transformation of deep HCO_3^- fluids into CO_2 -rich, acidic fluids. Under such conditions, Mg²⁺ would be soluble and carried away by fluids, while silica would be immobile, and precipitate. Further from the fluid-channelling fault and near-fault joint systems, due to pressure release and outgassing of CO₂, the precipitation of cryptocrystalline MgCO₃ would occur, which is indeed observed during the field study.

Frondel (1979) has suggested that silica polymorphs can be formed as residues left by leaching serpentine minerals by acids. He also noted that silica residues can inhere "parent" silicate framework, as has been found by microscopic examination of opal-CT from central Serbia. Leaching of antigorite he found decreases its birefringence virtually to zero, which was also found in examined opal-CT.

Metasomatic interactions between serpentinites and hydrothermal solutions resulted in significant silica enrichment and magnesia subtraction, indicated by analyses of chemical composition. Action of hydrothermal solutions containing dissolved carbonic acid initiates in lower parts of terrain, and thus formed silica is transported towards surficial levels by fluids. Magnesia has already been partly removed from near-surface and surficial levels by weathering, and the remaining part was by fluids transported and precipitated at the boundary of the zone of hydrothermal influence, as has been observed in almost all examined localities, and also in other various hydrothermal deposits related to ultramafic rocks (e.g. Hora, 1997). Genetic and spatial relationship between opalized serpentinite and chalcedony veins deposited in them has been noted in all examined occurrences. Chalcedony veins have been formed by silica deposition from the same hydrothermal fluids, which have caused alteration of serpentinite. As has been observed before (Bettermann & Liebau, 1975; Korytkova et al., 2002), type of silica formed depends mostly on primary material; in discussed case – amorphous silica containing various impurities taken over during circulation through joint systems in the first case or opalized serpentinite in the second.

These processes have also been observed in other similar geological environments along Tethys suture zone (e.g. Miladinović, 2005; Işik et al., 2005).

PRACTICAL SIGNIFICANCE

Siliceous mineralizations (plain, blue and purple chalcedony, jasper, onyx, carnelian, sard, agate) occurring spatially and genetically associated with opalized serpentinites are being used as gemstone material. Due to larger area comprised by opalized zones and the possibility of their aeromagnetic prospecting, opalized zones of serpentinite have been successfully used as prospecting

clues in their localization and discoveries of new siliceous gemstone occurrences during special regional prospection (Marčeta, 2005). Opal-CT is also being used as a gemstone. Closely genetically and spatially related to these opal occurrences are hydrothermal deposits of cryptocrystalline magnesite. Large deposits of this type have been discovered in Greece, Turkey and Serbia.

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Резиме

ХИДРОТЕРМАЛНИ **ОПАЛИЗИРАНИ СЕДИМЕНТИ ВО ТЕТИСКАТА ОФИОЛИТСКА СЕКВЕНЦИЈА ВО ЦЕНТРАЛНА СРБИЈА**

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Клучни зборови: хидротермални измени; серпентинит; опал-ЦТ; Тетис

Интензивно фрактурирани серпентински изданија во Вардарската тектонска зона во централна Србија се дел од офиолитската низа на јурскиот Тетис. Примарните ултрабазични карпи кои претставуваат остатоци од океанската земјина кори беа местимично метаморфозирани во серпентинити после средината на јура. Терцијарната магматска активност од бигорно-алкален карактер се случила по должината на длабоки фрактури кои ги обележуваат оните на повлекувањето на Тетисот. Хидротермалната активност, предизвикана од врелата магма, преку фрактури и тектонски зони предизвика опализација на ултрабазичен матрикс и наталожување во пукнатините. Презенти-222 рани се резултати од разни испитувања на овие опализирани серпентинити : теренски испитувања, хемиски, микроскопски анализи, рендгенска дифракција (XRD) и рендгенска флуоресценција (XRF). Опализирани серпентинити се идентификувани главно како тридимит-кристобалит од типот на опал-ЦТ. Практично значење на појавата на опалот-ЦТ се состои во можноста тој да се користи како скапоцен камен и генетски и просторно да се истражуваат хидротермални депозити (криптокристални магнезитни жили и други видови скапоцен силициумски камен). Појави на опализиран серпентинит од овој тип се откриени на многу локации должината на шавната зона на Тетисот.