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Original scientific paper

VUČKOVICA CARBONATE-SILICA GEMSTONE DEPOSIT (CENTRAL SERBIA): GEOLOGIC PROPERTIES, GENETIC PROCESSES AND DEPOSITION AGE

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A b s t r a c t: Carbonate-silica veins and masses in theVučkovica locality in central Serbia, classified as Kraubath type deposit, despite their thickness, are not economically significant as magnesite deposit due to silica content well above 0.3%. What makes this mineralization economically insignificant as magnesite deposit – heavy silification of magnesite – is what enables its use as a gemstone. The regional hydrothermal activity along deep faults of the Sava-Vardar zone of the central Serbia had caused the alteration of serpentinite and formation of carbonate-silica mineralization. Unlike other deposits of the same type in this ophiolite belt, here the depth of the erosional level provides an opportunity to explore previously unknown features of these mineralizations, such as veins' trending, dip direction, dip angle and thickness, and somewhat different relationships between carbonate and silica constituents – namely predominance of magnesite with minor content of green dolomite and chalcedony. X-ray diffraction analyses have shown that carbonate minerals present are predominantly magnesite with minor dolomite, and that silica is fully crystallized despite its colloform structure. Energy-dispersive X-ray fluorescent (EDXRF) analysis indicated the presence of significant content of nickel, which can cause the green colour of dolomite. Optical microscopy has shown that the precipitation process is carried out continuously between cryptocrystalline magnesite and crystalline dolomite, and that silica crystallite (fibre) size varies between cryptocrystalline and microcrystalline. As a gemstone deposit, Vučkovica can be considered small and with complicated internal setting, but with significant depth span, as is typical for Kraubath type deposits. Present gemstone types are considered "semi-precious", of low to moderate economic value, but with very pleasing aesthetic properties.

Key words: hydrothermal veins; silica; carbonat; altered serpentinite

INTRODUCTION

Vučkovica gemstone deposit is situated 12 km west of Kragujevac in central Serbia (Figure 1). Gemstone mineralization is cropping out as a steep section over Vučkovačka River and beneath the local hill top of Lipa (444 m). The deposit has been discovered in 20th century, during the field examinations for the needs of basic geologic map of the area. The systematized geological data on this area were first given by Marković et al. (1968). Company "Magnohrom" from Kraljevo has conducted the examination of magnesite veins in this deposit in 1968 to define their potential for use as a raw material for refractive industry (Pejčić et al., 1985) with negative results due to heavy silification of magnesite. During the time period between 2003 and 2005, the deposit has been the subject of interest in research of gemstone raw materials (Marčeta, 2005). Gemstone types in this deposit are presented by chalcedony varieties with carbonates – magnesite and dolomite.

Fig. 1. Geographic position of the Vučkovica deposit.
Inset: Position in Serbia.

GEOLOGIC SETTING

Vučkovica gemstone deposit is located within the highly tectonically deformed serpentinite belt marking the Central regional fault of the Vardar zone (Vukašinović, 1976) or "Šumadija dislocation" (Anđelković, 1967 and references therein). Serpentinite is a part of ophiolitic sequence found within the accretionary wedge formed in subduction processes that eventually led to Vardar Ocean closure (Toljić et al., 2018). The NNW continuation of this serpentinite belt is hosting other, previously examined genetically related gemstone deposits (Marčeta, 2005; Kurešević & Dević, 2014, 2015; Kurešević et al., 2015, 2017). Toljić et al. (2019) define this belt as a part of the Central Vardar zone, where Cretaceous flysch and Palaeogene molasses mark the postion of the Sava-Vardar suture. It belongs to Stragari-Vučkovica gemstone district as defined in Marčeta (2005).

Serpentinite host rock is given Upper Jurassic age. It is in tectonic contact with Cretaceous sediments towards east: Barremian-Aptian flysch (sandstone, shale, conglomerate) and Albian-Cenomanian sediments (sandstone, sandy marlstone and limestone, Marković et al., 1968). Their contact is partly covered by Miocene sediments of Gruža lake: sand, sandstone, gravel, limestone, marlstone, clay with coal of Pannonian age (Marković et al., 1968). At the field surface, serpentinite body has small outcrops which in some parts contain carbonate-silica mineralization. These are best developed and observable in the Vučkovica deposit. A very small gemstone occurrence is also found in Donja Vučkovica (Marčeta, 2005), one km to the south, with no economic interest (Figures 2 and 3).

In tectonic fabric of this area, the most notable faults are trending NW-SE, marking tectonic contact between serpentinite and Cretaceous sediments, and perpendicular faults trending NE-SW in Cretaceous sediments. Their accompanying local faults and joint systems enabled the circulation of generative hydrothermal fluids.

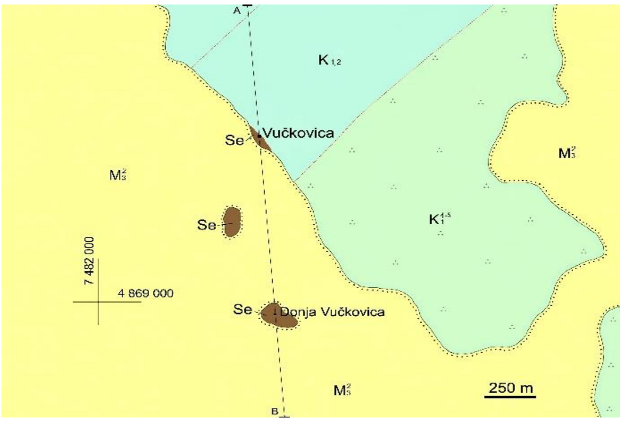

Fig. 2. Simplified geologic map of area between Vučkovica and Donja Vučkovica (modified from Pejčić et al., 1985, and Marković et al., 1968).
Key: K_1^{4-5} Barremian-Aptian flysch; $K_{1,2}$ Albian-Cenomanian sediments; M_3^2 Pannonian sediments (details in text).

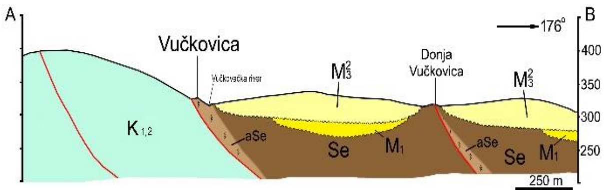

Fig. 3. Cross-section through Vučkovica (along the line A–B in Figure 2). Deposit extension into the depth is hypothesised. Markings as in Figure 2; aSe – altered serpentinite; M_1 – assumed presence of Lower Miocene sediments, as marked on the Basic Ggeologic Map (Marković et al., 1968). Vertical scale is stretched for visibility

DEPOSIT GEOLOGY AND GEMSTONE VARIETIES

Beneath of the Lipa hill crest, serpentinite lenticular body is cropping out beneath Miocene Lake sediments, horizontally extending 25 m at the surface, trending approximately north-south. The open section profile is up to 7 m high. Hydrothermal alteration has transformed serpentinite along faults into opal-CT and magnesite mass, wherein introduced carbonate-silica masses (magnesite, dolomite, chalcedony, agate, jasper) have precipitated along cm-dm joints as stockworks and veins and as much thicker, approximately planar veins. The deposit shape is determined by the shape of serpentinite body altered along the fault as a stock. If it does indeed extend into the depth as presented in Figure 3, then it can be considered a sheet. Within it, the gemstone types are distributed as stockworks, irregularly shaped masses and veins.

Predominant gemstone type in the Vučkovica deposit – a paragenetic sequence of carbonate (magnesite and dolomite) and silica (chalcedony, agate, rarely jasper and opalised serpentinite) is named "carbonate-silica onyx", due to its banded structure. This is geologically a misnomer which serves as a gemmological designation of the three main components – magnesite, dolomite and chalcedony, occurring layer upon layer as linings or encrustations in the most beautiful pieces of the gemstone (Figures 4 and 5), giving it the appearance of an agate. Their spheroidal, colloform structures testify of their formation by coagulation from a colloidal dispersive system.

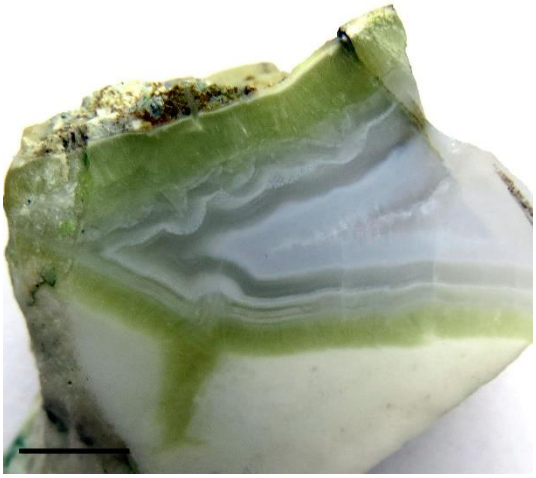

Fig. 4. Carbonate-silica "onyx": magnesite (white), dolomite (green), chalcedony (centre, bluish to colourless). Detailed explanation in text. Scale bar 1 cm

Fig. 5. Magnesite nodule with silica and Fe-minerals enriched rims (SiFe) encrusted with dolomite aggregates of perpendicular, platy, flattened crystals forming rosettes. Scale bar 12 mm

"Kraubath" deposit type is namely considered an economic type of magnesite deposit and is formed in a near-surface epithermal system. Its description given in Pohl (1990) perfectly fits Vučkovica, with the exception of having higher silica content. The gemstone mineralization appears in two main ore body forms:

1. Planar, subvertical interparallel veins trending nearly east-west (dip direction 340–354º, dip angle 74–85º). The thickest observed vein, visible at the outcrop section (called "main vein") is ~120 cm thick. Some 15 m further northward, there is another one parallel to it, 10–12 cm thick. The thickness of each vein is varying along its vertical extent. These interparallel veins have occupied the extensional fault and fracture system. Pohl (1990) describes this action as syndeformational filling of tensional structures. Although planar and apparently simple veins, their internal setting is not uniform (Figure 6). The main vein is composed of magnesite nodules coalesced together, with dolomite veinlets up to 2–3 mm thick filling up the interspaces (Figure 6, marked M+D). The rims of the nodules in places have higher contents of silica and Fe-minerals giving them rusty pigmentation (Figures 5 ahd 6, marked SiFe). These enrichments in some nodules reach the point of forming jasper, and in others it is just silicified, iron-stained magnesite. Silification of magnesite nodules is the most intense and conspicuous along the rims of the main vein. From the zone of altered serpentinite (Figure 6, marked aSe), the vein is separated along both selvages, by chalcedony-dolomite vein (Figure 6, marked SiV). In it, chalcedony forms its typical double parallel wall-liners that are generally central axis symmetric. Small magnesite nodules can be present (Figure 7). The thickness of chalcedony veins changes along their vertical extent up to the point of pinch out in places (such as in the eastern selvage shown in Figure 6), usually 2 mm to 6 cm, branching into several thin veinlets and again merging into one thicker vein. Within the main magnesite vein, the mineralization shows jointing subparallel to vein selvages (Figure 6, marked fr), probably due to tectonic activity affecting the brittle mineralization.

2. Numerous irregularly-shaped masses and semi-planar veins of carbonate-silica "onyx" in irregular patterns, forming stockworks within the altered serpentinite. They consist of snow-white magnesite, green dolomite and colourless to bluish chalcedony, juxtaposed in curved banded structures of differing shapes (Figures 4, 5, 7), sometimes with botryoidal surfaces. As in the "main vein", magnesite nodules form the main mass of the mineralization. Nodules are irregular in form, with gnarled surfaces and (when not lined with younger minerals) spheroidal globules visible (Figure 8, red arrows), or with rims containing the previously described type of silica and Fe-enrichment (Figure 5, "SiFe"). In some places, gemstone mineralization includes opalized serpentinite irregular (brecciated) mm-cm pieces. Flattened platy crystals of dolomite, perpendicular to the surfaces of magnesite nodules, form an encrustation layer over magnesite, 2.5–7 mm thick (Figure 5, "D"). Within the SE continuation of

the Neotethyan ophiolite belt within the Sistan suture zone in the eastern Iran, the genetically related deposits are called silica-carbonate listvenites and this type of dolomite is defined as "sparry crystalline dolomite" (Boskabadi et al., 2020). Where subsequent precipitation of chalcedony is absent, botryoidal surfaces of magnesite nodules covered with dolomite end with net-like surface formed by intersecting, blade-like rhombohedron crystal shape points resembling rosette or "desert rose" aggregates (Figure 5, "D" on the right side of the photo). These aggregates look identical to nickeloan dolomite from Mount Keith mine in Western Australia [1]. In some places, dolomite crusts lie directly on altered serpentinite as the only visible precipitate in fracture wall linings. Surfaces of magnesite nodules or parts thereof without dolomite encrusting reveal the presence of globules (Figure 8). These (or the rugged nodule surfaces) are called "cauliflower texture" and testify of derivation via colloid-sol-gel sequence, and have probably precipitated first as nesquehonite (Pohl, 2020), or rather hydromagnesite $Mg_5(CO_3)_4(OH)_2 \cdot 4H_2O$ or similar minerals that tend to form spherulites.

Chalcedony occurs as translucent to transparent, colourless or bluish, as a) part of "onyx" type mineralization, where it coats dolomite layer or fills up the small remaining voids, usually forming agate with absent colour variations between the bands (as in Figure 4) or b) in separate veins, where it formed as approximately planar fracture fillings (as in Figure 6a – left-side selvage, Figure 6b – SiV, or Figure 7), with botryoidal surfaces or giving way to drusy quartz crystals, most often up to 1 mm long, lining central void walls. Within the excavation digs (an adit with short lateral hallways) dating from 1968, it can be seen that chalcedony veins cut through older carbonate veinlets grown sideways from the oldest and thickest magnesite-dolomite veins. The older carbonate vein and younger chalcedony vein usually grow few to ten cm apart, with identical trending.

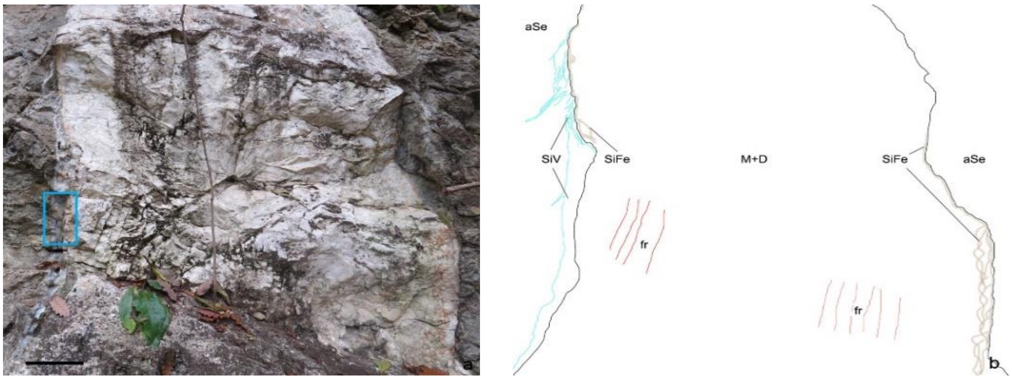

Fig. 6. Zoning in the "main vein": **a)**. photograph (size bar 15 cm), **b)** drawing
(Key: aSe – altered serpentinite; SiV – vein with predominant chalcedony; SiFe – magnesite zones enriched with silica and iron minerals; M+D – main vein mass made up of magnesite and dolomite; fr – fractures parallel with vein walls; detailed explanations given in text.)

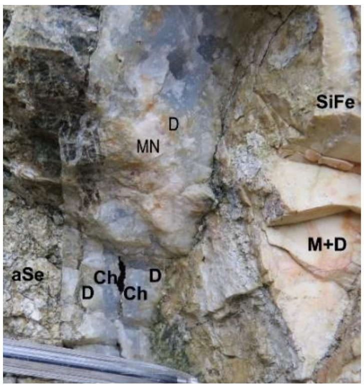

Fig. 7. Detail of a chalcedony vein marked with rectangle in Figure 6a. (Key: aSe – altered serpentinite; D – dolomite; Ch – chalcedony; MN – magnesite nodule; M+D – magnesite with dolomite veinlets; SiFe – magnesite enriched with silica and Fe-minerals within main vein selvage.
Scale pen diameter 8 mm.

Fig. 8. Magnesite (±silica) globules on a surface of magnesite nodule with dolomite encrustation.
The largest arrow pointing the one 2.35 mm in diameter.
Size bar 5 mm.

Common opal hosting these veins is in fact hydrothermally altered serpentinite (Kurešević & Vušović, 2012), however, not all altered serpentinite is opalized. Its colour is mostly green, in several interchanging hues in each piece. Unlike other related gemstone deposits in this serpentinite belt, Vučkovica contains negligible percentage of common opal.

LABORATORY EXAMINATIONS

X-ray diffraction analysis

X-ray diffraction analysis is performed in laboratory of the Institute for Technical Sciences of the Serbian Academy of Science and Art in Vinča. Patterns are collected using a Philips PW 1050 powder diffractometer with Ni-filtered CuKα radiation (α = 1.541874 Å) and a scintillation detector. Measurements were done at room temperature over the 2θ range from 9.810º to 79.810º, with a scanning step width 0.05° and –0.19° 2θ correction. Number of points 1401. Peak matching done via Match! Crystal impact software. The method only gives approximate quantitative relationships of the constituents. Results are shown in Figures 9 and 10. The samples examined are a) "carbonate-silica onyx" (Figure 9) – the complete vein shown in Figure 4, and b) "monomineral" layer of pale green dolomite (Figure 10).

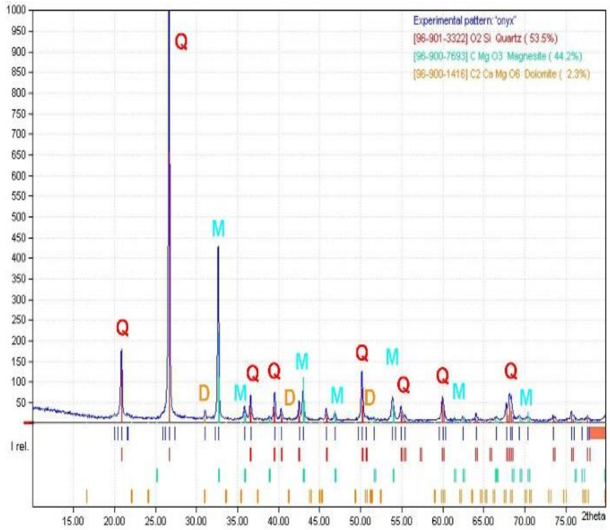

Fig. 9. XRD pattern of "onyx" sample.
Key: Q – quartz, M – magnesite, D – dolomite.
Upper right – semiquantitative results

XRD analysis results of "onyx" sample confirm the earlier examinations of Pejčić et al. (1985) – that "onyx" consists of silica and carbonates. Silica, macroscopically classified as chalcedony (agate variety) gives the reflection of completely crystalline quartz, with no traces of opal-A, -C, -T or -CT, even though colloform structures give off its formation from a gel. Quartz peaks are the most conspicuous. Also, semiquantitavely, silica is a predominant constituent (53.5%), even though macroscopically magnesite appears to predominate. This is another proof of magnesite being heavily silicified. Dolomite peaks are weak, and its content is 2.3%.

XRD analysis results of dolomite, however, have shown some surprising data. Even though the sample preparation included a careful separation of pure dolomite layer, the results show content of magnesite of 10.4%. The remaining 89.6% is in fact dolomite. Also, the results show complete absence of silica in this layer.

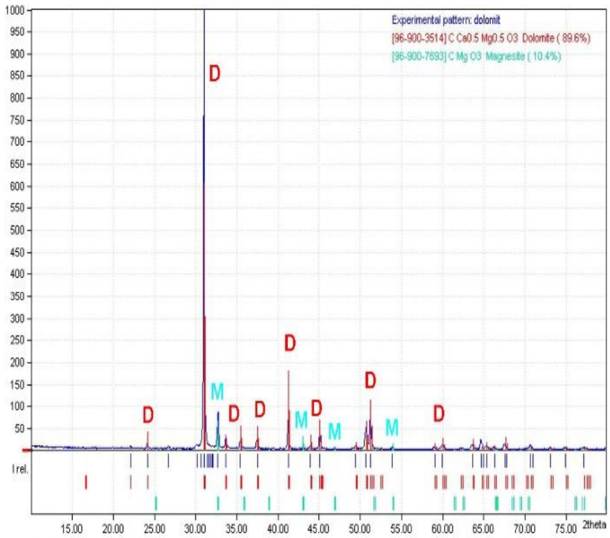

Fig. 10. XRD pattern of "monomineral" dolomite sample.
Key: D – dolomite; M – magnesite.
Upper right – semiquantitative results

Microscopic examination

Microscopic examinations and photomicrograph capturing are performed in Stone and Aggregate Laboratory of the IMS Institute in Belgrade, on microscopic thin sections with polarizing microscope Ernst Leitz, model RP 48 with digital camera Samsung SDC HY2307. "Onyx" variety is examined aiming to establish its internal setting and depositional relationships (Figures. 11 and 12).

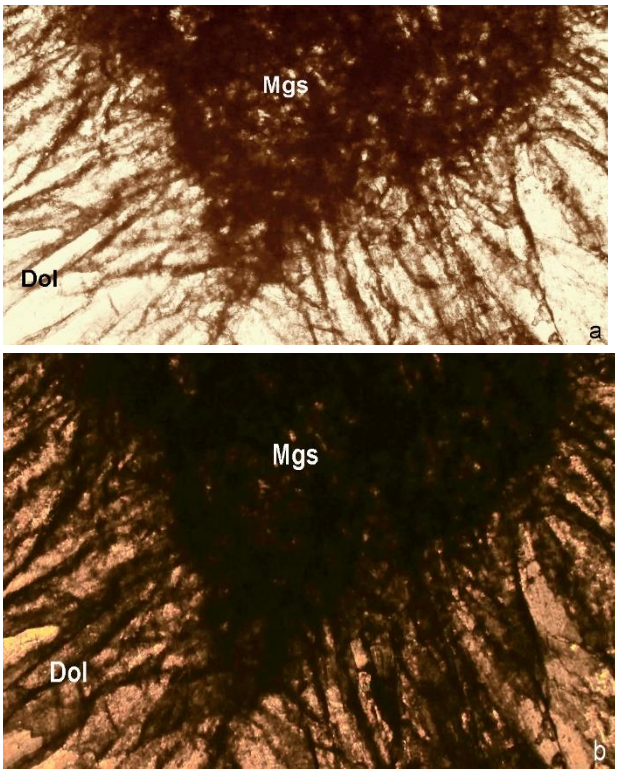

Fig. 11. Transition between magnesite (Mgs) and dolomite (Dol): **a)** PPL and **b)** XPL. FOW 3 mm

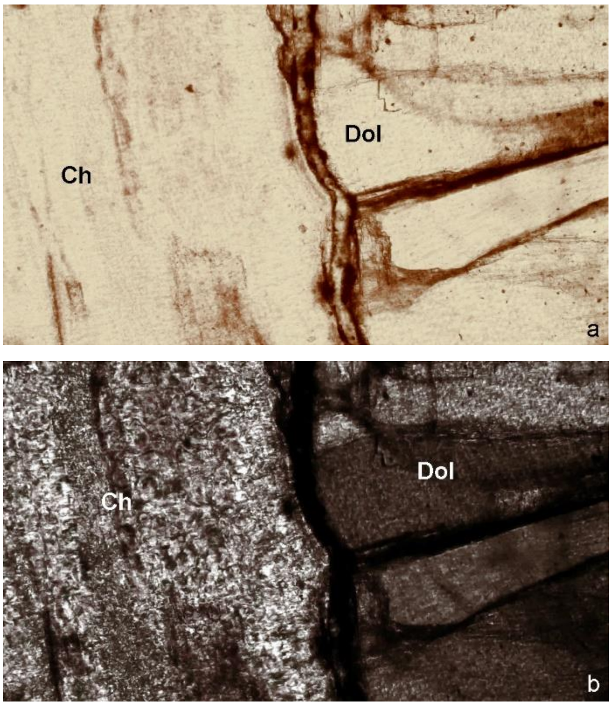

Fig. 12. Transition between dolomite (Dol) and chalcedony (Ch): **a)** PPL and **b)** XPL. FOW 3 mm.

Figure. 11 shows that magnesite is cryptocrystalline and its precipitation hasn't been completely finished at the time when dolomite crystals started to form. Their co-precipitation lasted for a certain time period before solely dolomite precipitated, providing an explanation for the occurrence of ~10% magnesite in XRD analysis results of macroscopically monomineral dolomite vein. Figure 12 shows that silica appears in various crystallite sizes from cryptocrystalline to microcrystalline, with sporadic occurrences of Fe-oxyhydroxide staining that follows certain "layers" of silica. Unlike magnesite-dolomite gradual transition, the dolomite-chalcedony boundary is very sharp and clear.

Energy-dispersive X-ray fluorescence analysis

The aim of this analysis was to detect the possible causes of green colour of dolomite crystals. The analysis is performed at the cement, chemistry and mortars laboratory of the IMS Institute in Belgrade. The preparation of samples involves drying until constant mass at 105 ± 5°C, milling in Herzog-Siemens mill type Simatic C7-621, pressing into pellets under 25 t in Specac T-40 hydraulic press to produce stable pellets. Quantitative and qualitative analysis is performed with the Spectro Xepos Energy-dispersive XRF (EDXRF) instrument with a binary cobalt/palladium alloy thick-target anode X-ray tube (50 W/60 kV) and combined polarized / direct excitation. A silicon drift detector (SDD) design was used as a detector of Spectro Xepos, and air as a cooler. Control software is Spectro XRF Analyzer Pro. The most relevant results (analytes present in concentrations above their detection limit) are presented in Table 1.

Vučkovica is the first gemstone deposit in the explored serpentinite belt with macroscopically visible dolomite content, therefore we can not compare the obtained results with other related deposits in this area. Compared to available data on dolomite (Table 1), it can be seen that sodium-oxide content is below detection limit in both dolomites, magnesium-oxide content is higher in Vučkovica (24.16% compared to 22.18% in Sivec dolomite) probably due to magnesite contents indicated by XRD and microscopic analyses; silica content is 4.788% compared to only 0.23% in Sivec. This, along with significantly higher content of iron(III) oxide (1.401% in Vučkovica, compared to 0.03% in Sivec can also be partly ascribed to magnesite content, since it is enriched with both silica and iron-oxyhydroxide. Calcium-oxide content in Vučkovica of 24.52% is lower than 30.52% in Sivec. Compared to Clarke number values for nickel given in Taylor (1964) – crustal average: 75 ppm, basalt average 150 ppm it is clear that dolomite from Vučkovica reflects the high Ni contents of host ultrabasic rocks.

Table 1

Most relevant results of EDXRF analysis of Vučkovica green dolomite (1), compared to data on available dolomitic marble (2) from Sivec (N. Macedonia) tested under the same conditions, and green dolomite (3) formed at very similar conditions in Malentrata in Ligurian ophiolites (Boschi et al., 2009)

Analyte	Contents (1)	Contents (2)	Contents (3)	Analyte	Contents (1)	Contents (2)
Na_2O	<0.0027 %	<0.0027 %	0.13 %	Cu	2.5 ppm	–
MgO	24.16 %	22.18 %	21.29 %	Zn	1.9 ppm	2.0 ppm
Al_2O_3	0.2821 %	0.06 %	0.06 %	Ga	0.6 ppm	*
SiO_2	4.788 %	0.23 %	bdl	As	1.7 ppm	*
K_2O	0.0179 %	0.01 %	bdl	Rb	0.5 ppm	*
CaO	24.52 %	30.52 %	26.66	Y	0.9 ppm	*
Fe_2O_3	1.401 %	0.03 %	3.32 %	Nb	0.6 ppm	*
LOI	44.66 %	46.64%	48.74 %	Cd	1.1 ppm	*
Mn	470.1 ppm	16.4 ppm	542.1 ppm	Ba	18.1 ppm	*
Co	13.00 ppm	5.29 ppm	*	V	42.6 ppm	*
Ni	319.8 ppm	18.3 ppm	bdl	Ti	22.1 ppm	*
Cr	127.5 ppm	7.20 ppm	410.5 ppm	W	48.2 ppm	*
Sr	472.7 ppm	85.8 ppm	*	U	1.0 ppm	*

Note: Total iron content is automatically recalculated and presented as Fe_2O.
*data unavailable; bdl – below detection limit

GEMSTONE POTENTIAL

Even though magnesite is the predominant mineral phase in the Vučkovica deposit, its silica content being well over 0.3% (Pohl, 2020), excludes the possibility of its economic use as magnesium raw material. The intense silification of magnesite, however, is what gives it favourable properties for use as a gemstone – retaining the given form, the possibility of taking a perfect polish and resistivity to scratching. Gemstone colours and patterns are pleasant. Presence of green dolomite and bluish agate is what gives this material its prized aesthetic properties. The colour combination white-green-bluish is unique. At the market, these gemstone varieties are considered to be of low to moderate economic value, named "semi-precious".

Opalized serpentinite can have beautiful green hues and be transparent when wet (with natural moisture present), but as it dries, it becomes partly opaque, brittle and it cracks easily. Although its aesthetic properties are good, it can not be used as a gemstone due to its brittleness.

The hardness values of "onyx" variety are medium to high. As a gemstone raw material, magnesite is usually used dyed as an imitation of other gemstone types, notably turquoise. However, magnesite from Vučkovica is naturally silicified, and can therefore be processed and used as some harder materials. Chalcedony alone is of high quality and can be processed and finished in automated process. Its Mohs' hardness is between 6 and 7. It can be cut and polished for cabochon production. The processing of "onyx", due to its partial brittleness, demands some adjustments. Despite the hardness differences among "onyx" constituents (3.5 for dolomite to 7 for chalcedony; hardness of magnesite varies depending on silica content, can be up to 7), this gemstone type can be cut and polished into various decorative items, however, the combination of automated processing and hand finishing must be used (Pejčić et al., 1985).

Chalcedony and opalized serpentinite have conchoidal fracture with smooth surfaces, whereas magnesite has semiconchoidal and irregular fracture with rough, pseudo-grainy surfaces (the feature uncommon for Kraubath type magnesite).

Pejčić et al. (1985) have performed explorative drilling to the depth of 70 m, based on which they estimated the indicated resources of carbonate-silica "onyx" to be 22868 t, and chalcedony in separate veins 147 t. These numbers seem exaggerated and are much lower today due to intense hand-picking from surface and the available exploration digs, and using the gemstones for experimental processing. Mentioned explorers have classified this deposit into the third category of the geoeconomic classification of deposit types by Ilić (2003) – deposits with complicated geologic structure, intense variability of mineralization properties and irregular distribution of useful mineral raw material, where detailed exploration aiming at specifying the proved and probable reserves (A and B category according to the classification used in Serbia) is not economically feasible. In any case, even though Kraubath type veins can be traced into depth up to several hundred meters, this deposit, as gemstone and not commercial magnesite deposit, is considered small, since its market pricing does not allow deep digging excavation.

The absence of pronounced pull-apart structures, tearing, moving, sliding and subsequent healing indicates that tectonic action during generation process or after its ending hasn't been intense. Minor fracturing of the gemstone mineralization and the presence of genetically related carbonate-silica mineralization within the lacustrine sedimentary packages of the adjacent Miocene lakes testify that the host rock mass hasn't been moved far, if at all, from where precipitation took place, during or after the generative episode. The exploration digs (an adit with side hallways) driven into the side of Lipa hill and partly through the "main vein", dug out in 1968, aiming to serve as a survey room and for sampling of raw material, still stand without any support amidst a still active seismic zone, thus testifying of carbonated and silicified serpentinite in Vučkovica being a stable mining environment, at least at the ground level.

GENETIC PROCESSES AND ORDER OF FORMATION

As in all other examined genetically related gemstone deposits in this zone (Marčeta, 2005; Kurešević & Dević, 2014, 2015; Kurešević et al., 2015, 2017), two main phases of precipitation are present. In the first phase, the first formed mineral is magnesite, precipitated as irregularly shaped nodules (as described in detail by Pohl, 1990) from a colloidal dispersive system, with Fe-minerals ± silica in some places along the nodule rims. This depends on local variations of silica and Fe-minerals concentrations, but appears to be especially pronounced where a lateral fracture comes to an end and fluid circulation becomes stagnant. These are the locations where brown jasper forms. Oversaturation of the generative fluid with respect to magnesite and its precipitation may be enhanced by fluid evaporation and degassing (Ulrich et al., 2014). It is now well established that CO_2-rich fluids, originating from devolatilization of deeply subducted oceanic slabs, ascend through major fault lines and perform alteration upon present ophiolite sequences (Peng et al., 2020). Dissolution of serpentinite bodies due to CO_2-rich fluid action results in their transformation into carbonate-silica masses while progressively increasing the environmental pH (Ulrich et al., 2014). At pH > 9 magnesite precipitates, while silica at this pH is usually solubilised. However, Williams & Crerar (1985) found that a) amorphous silica also precipitates from dense colloids in supersaturate alkaline aqueous solutions; b) even in undersaturated solutions, silica co-precipitates with insoluble metal hydroxides – namely Mg- and Fe-hydroxides (which is clearly apparent in all genetically related gemstone deposits in this serpentinite belt); and c) carbonates enhance precipitation of silica. These are the most obvious reasons why magnesite and part of the present silica co-precipitate, and there is observed paragenetic connection between magnesite, silica and Fe-minerals.

Where iron-coloured nodule rim is absent, dolomite directly encrusts the white magnesite nodules, also filling up the gel-contraction cracks where present. Obradović et al. (1992) explain this precipitation order to be caused by low solubility of magnesite in the carbonate solution and passing into dolomite due to decrease in magnesium concentration. When colloidal dispersive system got depleted in carbonate, in some places silica free from observable iron impurities (but not completely, as indicated by microscopic analyses) precipitated in leftover free spaces. Where chalcedony is absent, dolomite "desert rose" crystal aggregates forming encrustation around magnesite nodules remain visible (Figure. 5). In other related deposits this phase is represented by irregular magnesite and jasper veins (Kurešević & Dević, 2015, Figures 1–4; Kurešević et al., 2015, Figures 2–8; Ibid., 2017, Figures 7 amd 8). The first phase products appear as semi-planar veins and stockworks, formed by open space filling, since such veins exhibit a colloform habit (Augustithis, 1995).

The second phase is in other related deposits represented by chalcedony veins filling up newly opened joints or tiny contraction cracks in the first-phase minerals while they were still unsolidified; in Vučkovica it is represented by veins containing similar paragenesis as the first phase of precipitation, but in different volume relationships – chalcedony is predominant, containing dolomite along selvages and a small content of magnesite nodules encrusted with dolomite. Chalcedony surfaces are lined with minute drusy quartz crystals in some places where enough space had remained at the moment when the generic fluid had turned from silica oversaturated to undersaturated. This happens when circulation conduits get sealed during silica precipitation, so the dispersive system can not continue being oversaturated in silica, and thus the colloid passes into a molecular solution of monomeric silicic acid, precipitating quartz crystals (Xu et al., 1998; Williams & Crerar, 1985).

Based on the field and laboratory examinations of Vučkovica mineralization, the deposit profile, as described in Lefebure et al. (1995), is "I17 cryptocrystalline ultramafic-hosted magnesite veins" (the synonym being "Kraubath-type"), related to faults cutting ultramafic rocks, in our case serpentinite, formed from hypogene low-temperature CO_2-rich fluids that caused the metasomatism of ultramafic rocks with veins emplaced along steep faults. The description of "Kraubath near-surface epithermal system" given in Pohl (1990) fits Vučkovica down to the smallest detail, with difference in silica content defining Vučkovica as gemstone and not magnesite deposit in the geoeconomic sense. This description even explains the formation of the near-by contemporaneous hydrothermal-sedimentary deposits in the Miocene lakes, common in many locations in central Serbia (Anđelković, 1986; Krstić et al., 2003).

The absence of prominent spherulites, namely in silica, which are so typical of other related deposits formed in shallower terrain levels (Marčeta, 2005; Kurešević & Dević, 2014, 2015; Kurešević et al., 2015, 2017), might be a signal that the sol-gel medium has not been abruptly neutralized by the introduced ions, and that the gradual decrease in ambient temperature had caused the precipitation of the phases present, according to their (in)solubility. That insolubility line is as follows: magnesite (±silica±iron-minerals), dolomite, silica (± iron-oxyhydroxides).

Absence of sulphide minerals, along with rusty pigmentation of jasper testifies of the oxidative conditions prevailing during the precipitation in the near-surface environment. Iron freed from primary mafic minerals, now forming Fe-oxyhydroxides such as limonite, serves as pigment for chalcedony. However, as jasper is a minor component in this deposit, it can be concluded that Fe-oxyhydroxides were present in significantly lower quantities than in other related deposits. This conclusion is in agreement with previously stated on slower precipitation.

AGE OF DEPOSITION

The fact that the mineralization has occupied planar fractures as planes of extension within serpentinite, whose dip direction and dip angle it is possible to measure, provides an opportunity to relate its genetic process with a certain tectonic event and thus attempt at giving it an age span, as no dating of these gemstone mineralizations along the Central regional fault of the Vardar zone has been undertaken so far. Also, the mineralizsation types have been observed and examined in Vučkovica that have not been found in other examined deposits in this serpentinite belt – namely the magnesite-dolomite-silica paragenesis. These have revealed the genetic connection with hydrothermal-sedimentary deposits of the same composition (Ilić, 1966/67) in Lower Miocene lake sedimentary basins of central Serbia (Anđelković, 1986; Obradović et al., 1992), providing another unique opportunity to contribute to genetic model of this deposit type.

According to available data, lacustrine hydrothermal-sedimentary magnesite-dolomite-silica masses have common inner texture and composition as mineralization observed in Vučkovica. They form concordant and syngenetic layers and pods within the lacustrine sedimentary sequences of Lower Miocene age (Ilić, 1968). This deposit type is named "lacustrine" or "Bela Stena" type (Pohl, 2020). The age of mineralization in Vučkovica can be narrowed down – if not set precisely – exactly via the mentioned sedimentary sequences, since Kraubath and Bela Stena deposit types are noted to often grade into each other (Pohl, 2020). Krstić et al. (2003) found that Lower Miocene lakes were so heavily mineralized by regional magmatic and hydrothermal activity that they can not be considered strictly fresh-water lakes. Due to the elevation of Vučkovica terrain, Lower Miocene sediments are not present by the exact location but crop out beneath Pannonian sediments some 4 km to the west. However, Vučkovica deposit is located near the edge of the Miocene Gruža lake, implying that the faults that enabled the subsidence and lake basin formation have also enabled the circulation of the same hydrothermal fluids that formed mineralization within the lacustrine sequence to precipitate it also within the fault and fracture systems within serpentinite along the basin rim. In many if not all central Serbia Miocene lakes, syngenetic magnesite-dolomite-silica layers are also present along lake bays (Ilić, 1966/67), but only in Lower Miocene sediments (Anđelković, 1986).

During the second Sava tectonic phase in Lower Miocene (Eggenburgian-Ottnangian), the Gruža lacustrine basin (whose eastern boundary is situated right along Vučkovica outcrop) and Gornji Milanovac lacustrine basin (in its northern continuation) became interconnected and had coeval sedimentation (Anđelković, 1986). The sedimentary package in the Gornji Milanovac basin named Nevade series of this age contains three magnesite layers. They have not yet been located in Gruža lacustrine sediments due to the overlay of thick younger Miocene sediments that are absent in the Gornji Milanovac basin (Figures 2 and 3).

This sedimentation cycle has begun after decompressional tectonic movements (Sava II), namely subsidence, which produced both the sedimentary basins (and left Vučkovica hill made up of thrust serpentinite as a conspicuous peak by the lake rim) and NE-SW to east-west tectonic structures (Anđelković & Anđelković, 1996), occupied by precipitated veins in Vučkovica.

Based on all stated data, we infer the coeval formation of magnesite-dolomite-silica mineralizations, both in Lower Miocene lacustrine sedimentary basins and in all hydrothermal vein gemstone deposits within the ophiolite sequence of the Central regional fault of the Vardar zone in Serbia, described in our investigations so far (Marčeta, 2005; Kurešević & Dević, 2014, 2015; Kurešević et al., 2015, 2017), including Vučkovica. We propose this age to be within upper Eggenburgian to lower or middle Ottnangian time span. The deposits show indications of formation through more than one hydrothermal pulsation, but these are as close to one another in time as it takes for the partial gel contraction to take place and synaeresis cracks to start appearing (these are more obvious in other explored deposits than in Vučkovica). Syngenetic magnesite-dolomite-silica mineralizations in central Serbia Lower Miocene lacustrine sediments are also present in two or sometimes three layers (Ilić, 1966/67) and their NiO content is up to 0.02% (Pavlović & Radukić, 1959).

DISCUSSION

Scientific importance of this deposit lies in the opportunity to observe the deeper parts of this genetic type of gemstone deposits and new data it provides. Other gemstone deposits of the same type in this serpentinite belt are not as deeply cut and their outcrops are represented by much poorer mineralizations, present at the field surface almost exclusively as altered serpentinite eluvium containing pieces of poor silica-magnesite mineralization. At the level of erosion of the Vučkovica deposit magnesite predominates and dolomite is also present. Pohl (1990) explains this separation in precipitation products to be due to migration of iron and silica towards the field surface, while magnesite tends to precipitate first and rather abruptly, via sol-gel phase. Intense weathering and formation of eluvium in other deposits made it impossible to find any ore bodies *in situ*, while in the Vučkovica deposit deeper levels of the same mineralization are cut open and retained *in situ*, which enables observation of its geologic properties, and even measure its dip direction and angle, without the need to deduct its original features and position; the phases of precipitation provide a clearer picture of the geologic succession of tectonic movements and hydrothermal precipitation. Vučkovica is the first deposit of this type where it is possible.

Ilić (1952) made a revolutionary discovery while examining magnesite deposits in Neogene lacustrine basins, concluding that they precipitated from hydrothermal solutions formed due to magmatic activity, rich in juvenile CO_2, that had leached Mg from peridotite-serpentinite basement and arriving into lacustrine sediments through the same faults that enabled the formation of lacustrine basins. He named this type of deposits hydrothermal-sedimentary, lately named Bela reka type, after his most important discovery in Serbia. The circulation of the same hydrothermal fluids generated the Kraubath type veins in surrounding serpentinite masses present along lake rims. The authors before his time had considered carbonate-silica mineralizations in spatial association with serpentinites and accompanying rocks to be of lateritic origin. Although Ilić's hypothesis can be corroborated by presence of large hidden magmatic body indicated by aeromagnetometry (Vukašinović, 1970) extending from Rudnik Mt. at north to Gledićke Mts. (immediately south of Vučkovica) at south, contemporary explorations have added the processes of metamorphism of deeply subducted oceanic plates along with accompanying ocean floor sediments as the source of CO_2 participating in genetic processes of these mineralizations. Devolatilization of subducted sediments leads to formation of hydrothermal fluids with CO_2 that enact alteration of ophiolites, first causing the leaching of silica and Mg, and later on, in the near-surface zones of ophiolites, deposition of silica and carbonate minerals (Ague & Nicolescu, 2014; Peng et al., 2020; and references therein), defined as listvenitesation s. lat. Serpentinite belt in which the gemstone mineralizations in question are located belongs exactly to the area west of former Vardar ocean forearc and within its suture zone, i.e. accretionary wedge (Toljić et al., 2018, 2019). However, according to Halls & Zhao (1995), as these mineralizations do not contain fuchsite, they are not considered real listvenite, and according to Figure 2 (op. cit.), real listvenite with gold can be present in deeper ground levels, as Vučkovica and related gemstone deposits are formed at lower temperatures with lower intensity of alteration. Cited authors state listvenite to form at sub-greenschist facies conditions, at temperatures of 200 – 300ºC and pressures around 1.5 Kb, while Vučkovica and related gemstone deposits are formed at lower P-T conditions in near-surface terrain levels.

The process of magnesium leaching from ultramafic rocks via reaction between them and CO_2-rich fluids, given in Pohl (2020), is accomplished in deeper levels, although magnesite is hardly the product being primarily formed and transported in the fluid, but rather some of its hydrated relatives. These "magnesite precursor colloids" (Pohl, 2020) moved upward through available fault-fracture-fissure systems. In this process, magnesite as less soluble precipitated earlier than silica, although both are present in all related deposits, and often intermixed. As these fluids reached Miocene lacustrine environments, the separation between magnesite and silica has been more efficient than in Vučkovica or other examples of Kraubath type deposits in the area.

The absence of chromite grains in magnesite-dolomite-silica mineralization testifies that these are not formed by *in situ* alteration of serpentinite, but are precipitated from the introduced fluid (Pohl, 2020). On the other hand, host rock along veins' walls is made up of opalized serpentinite with abundant magnetite grains formed by hydrothermal alteration of chromite (Pejčić et al., 1985; Kurešević & Vušović, 2012). Stable Carbon and Oxygen isotope analyses in these mineralizations have shown that carbon-dioxide came from different sources, including meteoric and ascending waters of magmatic, diagenetic, metamorphic and mantle derivation. The process of mixing of juvenile fluids with cooler meteoric waters is the most probable cause of physico-chemical debalancing of the fluid and precipitation, just as is the case with the Bela Stena type deposits (Pohl, 2020).

CONCLUSION

The Vučkovica gemstone deposit provides scientific value by yielding a view of *in situ* mineralization unlike any other deposit of the Kraubath type in the same serpentinite belt. The minerals that are otherwise present as thin irregular veins in eluvium are found here in a 15–20 m wide zone of altered serpentinite, forming thick veins and stockworks that can be observed *in situ*. Also, in other deposits, silica varieties predominate, whereas in this one the main, predominant component is magnesite. We hypothesise that this type of mineralization is present in all other occurrences in this belt, but as it is located in lower ground levels, it is not yet cropping out at surface.

The conspicuous subvertical veins have a common east-west trending. Other ore body types appear randomly distributed, just like in other gemstone deposits of the same type in this serpentinite belt, with no conclusive pattern or preferred direction, typical thickness, certain stable and unchangeable mutual mass relations and percentages within the ore bodies.

The presence of roughly spherical bodies (in our case magnesite nodules) and colloform structures they form, testifies of their precipitation from a colloidal dispersive system evolved into a complex gel (Augustithis, 1995).

Magnesite had precipitated the first, containing variable contents of silica. Magnesite nodules are lined with dolomite. Where nodules are so densely packed that they appear to have coalesced, dolomite linings look like thin, contorted veinlets inside magnesite mass. In some places, as shown by microscopic analyses, there is a gradual transition between magnesite and dolomite precipitation, while in others, magnesite nodules precipitated and even had time to form gel contraction joints before the onset of dolomite precipitation. Dolomite also precipitated in newly opened fractures, some of which are completely filled by it.

According to Toljić et al. (2019, Figure 1 op. cit.), location of Vučkovica is directly within the basal thrust of the major tectonic unit – reverse thrust fault along which the thrusting of the Eastern Vardar zone (fore-arc domain) has been carried out over the Central Vardar zone (suture domain). This is also the reason why ophiolites are cropping precisely in this elongated belt, why they have been intensely deformed, and why most gemstone veins are intensely fractured and rarely ever found *in situ*. From the observed relations, we conclude that along with precipitation of carbonate-silica mineralization, the extensional tectonic activity continued after a short pause, isodirectional to the predepositional one, and of low intensity, barely enough to open some new, isodirectional fractures within the brittle serpentinite host rock, at or near the selvages of already formed carbonate veins with minor silica. These were in places – where thinner – completely filled by dolomite, and in others, precipitation continued with chalcedony over dolomite-lined walls. Also, the position of related lacustrine mineralizations, points to the absence of intense horizontal tectonic movements at the time of their generation or afterwards in this area. This contributed immensely to the quality of the Vučkovica mineralization and the possibility of its use as a gemstone.

There is an ongoing juxtaposition of genetic models for this type of deposits – lateritic (Ulrich et al., 2014; Jurković et al., 2012 etc.) and hydrothermal (Pohl, 1990; Marčeta, 2005 etc.). Both could be actuated in appropriate times and settings, however, in our case, the lateritic model can not explain why there is a narrow zone of altered serpentinite with vein-stockwork precipitation of carbonates and silica exclusively within and in a narrow zone around the deep, regional faults which serve as feeder conduits for introduction of devolatilization products of deeply subducted slabs with accompanying oceanic sediments, and nowhere further from them, as unaltered serpentinite masses spread over large areas of the Balkan Peninsula. Therefore, we classify Vučkovica according to the deposit profiles of Lefebure et al. (1995) as I17 cryptocrystalline ultramafic-hosted magnesite veins, i.e. Kraubath hydrothermal deposit type, the heavily silicified variant.

Finding an absolute match of Kraubath type deposit (described by Pohl, 1990) in Vučkovica, down to the smallest detail, and macroscopic properties of gemstone to parallel those of previously examined, it can now be confirmed that either all related gemstone deposits explored so far (Marčeta, 2005; Kurešević & Dević, 2014, 2015; Kurešević et al., 2015, 2017) continue at greater depth into Kraubath magnesite type deposits or they form under approximately the same conditions, within the same geologic environment, apparently randomly distributed within the serpentinite belt along the Central regional fault of the Vardar zone as a conduit for generative fluids.

Hydrothermal alteration of serpentinite into carbonate-silica masses (apparently, exclusively opal-CT and magnesite) along tectonic lines, concurrently with deposition of the introduced silica (chalcedony, jasper, agate) and carbonates (magnesite, dolomite) in available tectonically opened spaces is a commonplace for all genetically related gemstone deposits along this serpentinite belt. The introduced precipitation products originate from the same alteration processes performed in deeper serpentinite zones, as described in detail by Ulrich et al. (2014) and Boschi et al. (2009). The precipitation process probably started abruptly, by deposition of mixed magnesite-silica amorphous to cryptocrystalline mass. The ongoing precipitation of magnesite continued for some time along with the onset of dolomite precipitation in the form of lath-like crystals of green colour with high Ni content (~320 ppm). As XRD analysis hasn't indicated the presence of takovite $Ni_6Al_2(OH)_{16}[CO_3]\cdot 4H_2O$, népouite $(Ni, Mg)_3(Si_2O_5)(OH)_4$, theophrastite $Ni(OH)_2$, willemseite $Ni_3Si_4O_{10}(OH)_2$ or other green minerals typical of precipitates found in serpentinites (Maksimović, 1969), the nickel content is likely tied to ion substitution within the crystal lattice due to comparable ion radii (Mg^{2+} 86 pm; Ni^{2+} 83 pm). A more in-depth study might reveal why green-col-ouring agent – presumably nickel – in some places colours magnesite (e.g. Laverton, Western Australia [2]), in some deposits dolomite (as in Vučkovica), and in some chalcedony (e.g. deposits in Karaganda region in Kazakhstan [3].

Where circulation paths were not infilled and clogged by carbonate precipitation, relatively pure silica formed chalcedony cryptocrystalline to microcrystalline aggregates in the available spaces as the last precipitate.

Two-phase deposition is a common feature of this gemstone deposit type, however, from the observed relationships in Vučkovica, the impression is that the process of deeply-seated subducted slab devolatilization has been continuous, and also the formation of CO_2-rich fluids derived in it, which caused the *in situ* alteration of host serpentinites into carbonate-silica masses along circulation paths, but the generative process (vein precipitation in free spaces) carried out within the upper Eggenburgian to lower or middle Ottnangian time span has been somewhat disturbed or interrupted by the onset of syndepositional tectonic activity of unchanged direction and of low intensity.

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НАОЃАЛИШТЕ НА КАРБОНАТНО-СИЛИКАТЕН СКАПОЦЕН КАМЕН ВУЧКОВИЦА (ЦЕНТРАЛНА СРБИЈА) – ГЕОЛОШКИ СВОЈСТВА, ГЕНЕТСКИ ПРОЦЕСИ И СТАРОСТ НА НАОЃАЛИШТЕТО

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Карбонатно-силикатни жици и маси во локалитетот Вучковица во централна Србија, класифицирани како наоѓалиште од типот Краубат, и покрај нивната моќност не се економски значајни како наоѓалиште на магнезит поради содржината на силициум-диоксид над 0,3%. Она што ја прави оваа минерализација економски безначајна како наоѓалиште на магнезит – силната силификација на магнезитот – ја овозможува неговата употреба како скапоцен камен. Регионалната хидротермална активност покрај длабоките раседи на Савско-Вардарската зона во централна Србија предизвикала промена на серпентинитот и формирање на карбонатно-силикатна минерализација. За разлика од другите наоѓалишта од ист тип во овој офиолитски појас, овде длабочината на ерозивното ниво дава можност да се истражат претходно непознатите карактеристики на овие минерализации, на пример правците на жиците, насоката на протегање, аголот на залегнување и моќноста, како и различните односи помеѓу карбонатните и силикатните конституенти – имено, доминација на магнезит со мала содржина на зелен доломит и калцедон. Рендгенските дифракциони анализи покажаа дека присутните карбонатни минерали се претежно магнезит со малку доломит и дека силициум-диоксидот е целосно кристализиран и покрај неговата колоформна структура. EDXRF покажа присуство на значителна содржина на никел, што може да е причина за зелената боја на доломитот. Оптичката микроскопија покажа дека процесот на преципитација се одвивал континуирано помеѓу криптокристалниот магнезит и кристалниот доломит и дека големината на силикатните кристалити (влакна) варира помеѓу криптокристални и микрокристани. Наоѓалиштето на скапоцени камења Вучковица може да се смета за мало и со комплицирана внатрешна градба, но со значаен длабински распон, што е карактеристично за наоѓалиштата од типот Краубат. Сегашните видови скапоцени камења се сметаат за „полускапоцени“, со мала до умерена економска вредност, но со многу пријатни естетски карактеристики.