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Original scientific paper

MINERALOGICAL CHARACTERIZATION OF BRUCITE ($Mg(OH)_2$) FROM 'RŽANOVO, NORTH MACEDONIA

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A b s t r a c t: The present study was performed to describe an occurrence of very big crystals of brucite and its associated minerals in locality of 'Ržanovo. It was also intended to describe the conditions of crystallization of such a big crystal. Within highly altered peridotite host rock in 'Ržanovo region, acicular, up to 80 cm in size, colorless crystals of brucite were found. It has been also found as small, and in some places large lenses filled with acicular brucite. Crystals were found in serpentinized ultrabasic rocks, within veins. Needles are perpendicular to vein walls. Chemical composition is determinated with ICP-MS. MgO (66.39–69.13), Al_2O_3 (0.343–0.601), FeO (1.956–4.025), MnO_2 (0.239–0.360), TiO_2 (0.003–0.009), LOI (25.1–27.4). Its unit cell is: a = 3.1458 (7), c = 4.766 (2) Å (rhombohedral a = 2,4130 Å, α = 81.36°), V = 40.84 (2) Å^3, Z = 1.

Key words: brucite; 'Ržanovo; big crystals

INTRODUCTION

Brucite is a hydroxide mineral that belongs to the hexagonal crystal system and has the formula of $Mg(OH)_2$ (Mottana et al., 1983). Brucite can be transparent, green, blue or grey, and manganese-rich varieties (substitution of Mn^{2+} for Mg) are deep brown or brownish red. Brucite has a hardness of 2½ and appears in foliated, scaly or finely granular masses (Anthony et al., 2001–2005). Brucite is a metamorphic mineral and is found in carbonate rocks in low-temperature hydrothermal veins where thermal waters could percolate through the carbonate. It is also found in chlorite schists, talc schists and low-temperature serpentinized rocks (Mottana et al., 1983, Magnusson, 1925). The tectonic blocks of peridotites, which in subduction processes are brought into the parts of the continental crust as ophiolitic complexes, are unstable near H_2O under conditions of different temperatures and pressures, during which a process of serpentinization and formation of serpentines occurs. Serpentites are rocks in which there are numerous minerals (a group of serpentine minerals, such as chrysotile, lizardite, antigorite, as well as the presence of brucite, magnetite, talc, chlorite and tremolite). Brucite is widely distributed in ultramafic rocks (Khan et al., 1971; Hora, 1998). It is also found in a variety of exotic settings such as kimberlites (Malkov, 1974) and carbonatites (Lee et al., 2000). The fibrous variety of brucite, nemalite, is common in ultra-mafic rocks, where it coexists with chrysotile (Ross and Nolan, 2003; Khan et al., 1971). Brucite is probably one of the enigmatic minerals that forms during the process of serpentinization (Klein et al., 2013), during the introduction of water into subduction zones (Kawahara et al., 2016; Peters et al., 2020). As a rule, oceanic serpentinites have a lower amount of MgO and have higher concentrations of SiO_2 than on the continental serpentinites (Snow and Dick, 1995), where the brucite is formed as a result of the decomposition of olivine.

Brucite occurrences have recently been recognized in new geological settings, such as in accumulations on the sea floor (Kelley et al., 2001; Früh-Green et al., 2003), and in the future brucite deposits belonging to this category may become of economic interest.

The investigated area 'Ržanovo is a part of the western ophiolite of the Vardar zone and essentially consists of several lithostratigraphic units with distinct, mineralogy, petrology and history of development. Geological position and characteristics of the 'Ržanovo depositwas described in many papers (Ivanov, 1959, 1960; Grafenauer and Strnole, 1966; Maksimović, 1981; Boev, 1982; Boev and Stojanov, 1985; Boev et al., 1992; Boev and Lepitkova, 1994a, 1994b; Boev and Serafimovski, 1995).

The result of many faults and deformations in 'Ržanovo is very complex composition with serpentinized harzburgite, rare dunite, gabbropegmatite and rodingite, cretaceous sediments and sometimes with Triassic sediments. Triassic sediments are represented with two types of slightly metamorphozed rocks; -quartz-sericite-clay schists sometimes with talc and limestones partly recrystallized.

The main units which comprise the geologic setting are followed (Figures 1 and 2):

– Tertiaty volcanics rocks and pyroclastics;
– limestones of Albian-Cenomanian age;
– lateritic Fe-Ni ores of the Cretaceous age;
– ultrabasic complexes (serpentinites) with gabropegmatites and rodingites;
– schistose series of Paleozoik age (phyllites, margillites and quarzites).

The 'Ržanovo zone is a system of parallel thrust sheets composed of altered serpentinite, schist, and marble (Boev and Janković, 1996; Boev et al., 2009; Serafimovski et al, 2013). Several large dislocations pass through the host serpentinized ultrabasic masses.

Based of the chemical composition of olivine in ultrabasic rocks it is clear that they are Alpine type of ultramafics rocks.

Dunites and harzburgites are the main members of the Alpine type ultramafics in the 'Ržanovo-Studena Voda zone. Harzburgites distiguish themselves by the presence of pyroxene (enstatite) 1 cm in size. Serpentinized harzburgites generally occur as blocks which are compact compared to other rocks. Serpentinite occurs as net-like serpentinite which originate from pyroxene and individual flakes and fibres of serpentinite minerals are parallel to the another and possess primary mineral cleavage.

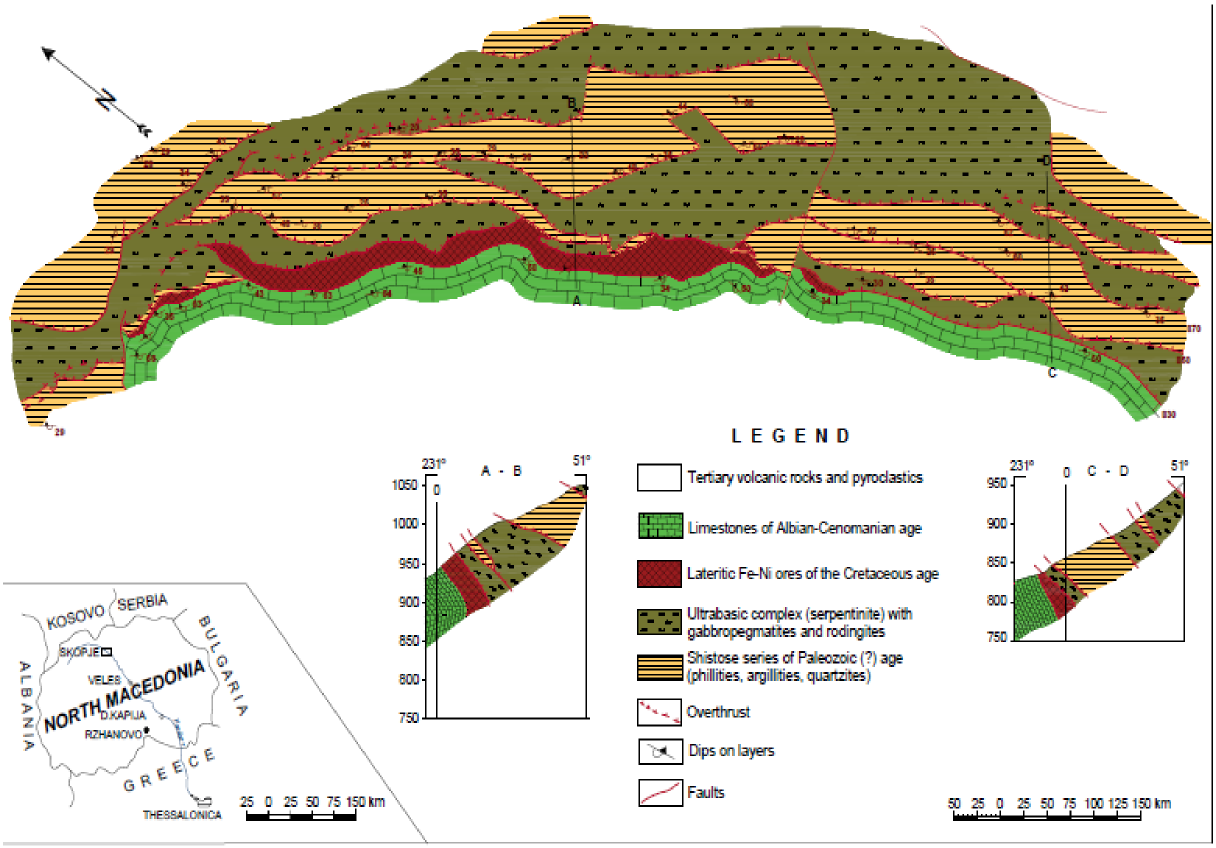
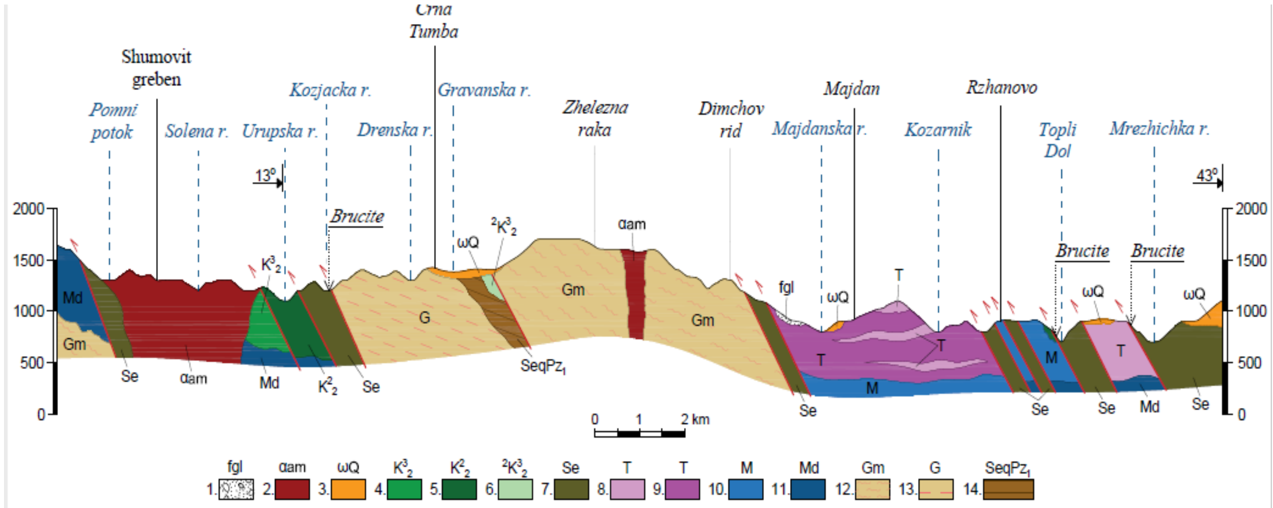


Fig. 1. Geological map with cross sections through the 'Ržanovo deposit



1. fgl – Glaciofluvial sediment; 2. αam – Andensite (Horblende and sanidine) and trachyandesite; 3. ωQ – Pyroclastics aglomeratic and brecia tiff; 4. K^3_2 – Platy and thyckbedded limestons; 5. K^2_2 – Conglomerates and sandstones; 6. $^2K^3_2$ – Shales and conglomerates; 7. Se – Serpentinite; 8. T – Marbly limestones and dolomites; 9. T – Slates and sandstones; 10. M – Marbles; 11. Md – Dolomitic marbles; 12. Gm – Banded muscovite gneisses; 13. G – Porphyroblastic gneiss; 14. $SeqPz_1$ – Quartz-sericite schist.

Fig. 2. Geological cross section of the 'Ržanovo deposit

Serpentinized dunites, unlike serpentinites are due to serpentinization of a harzburgite mas and occur as a large block of compact bodies. Microscopic studies indicate that serpentinite originates from olivine. It is net-like and individual flakes or fibres of serpentinite minerals are arranged along irregular cracks within the primary olivine grains (the first genereation of serpentinite) and as ring radial aggregates replacing individual parts of olivine grains (the second generation of serpentinite).

Gabbropegmatite-rodingites are elongated and cross-cuts the blocks of serpentinites. They are small in size and can be easily recognized on the field. Fresh gabbropegmatites which are usually alterated and affected by rodingitization processes are rarely found. They are characterized by a large grained texture. Microscopic investigations show that they mainly consisit of basic plagioclase and monoclinic pyroxene. Pyroxene alteration is seen as chloritization and prenitization. Rodingite rocks are mainly located in the marginal parts of the serpentinite mass as elongate blocks of variable thickness. They are formed from gabbros and gabbropegmatites during a postmagmatic phase of replacement due to calcium metasomatism. The contacts with surrounding serpentinites are always sharp and clear and are grey and grey-greenish to white depending of the garnet and chlorite contents.

The tectonic blocks of peridotites, which in subduction processes are brought into the parts of the continental crust as ophiolitic complexes, are unstable near H_2O under conditions of different temperatures and pressures, during which a process of serpentinization and the formation of serpentines occurs. Serpentites are rocks in which there are numerous minerals (a group of serpentine minerals, such as chrysotile, lizardite, antigorite, as well as the presence of brucite, magnetite, talc, chlorite, and tremolite).

SAMPLING AND METHODS

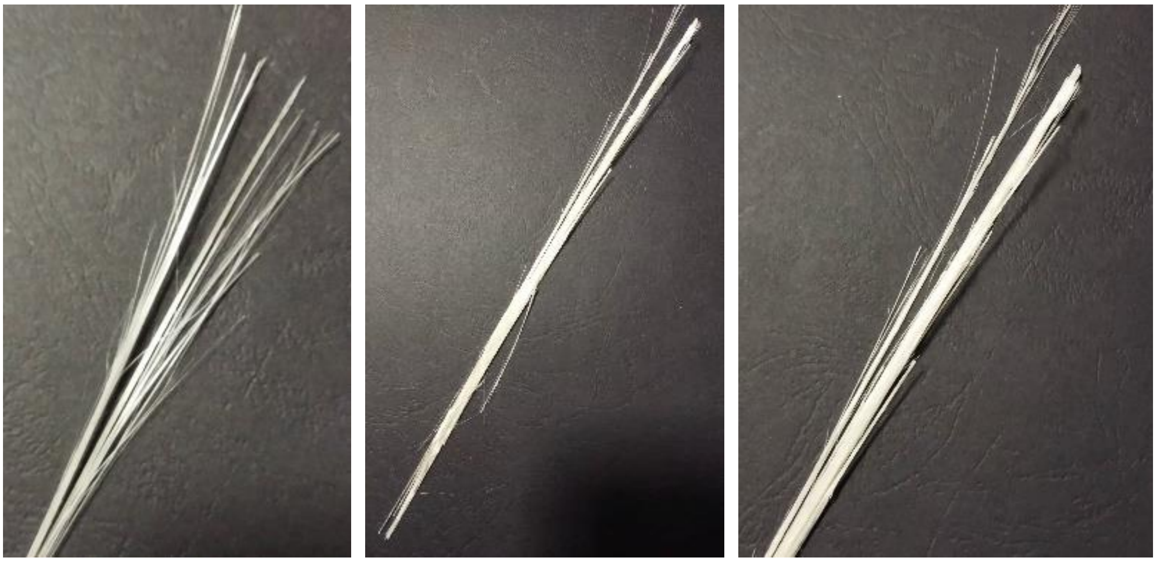
Several samples of brucite were taken from 'Ržanovo locality. Three of them were selected for our investigation. Samples of lizardite and dolomite occurring together with brucite were also analyzed. For reliable characterization of the mineral species in our research following analytical methods were used: ICP-MS and XRD.

ICP-MS. Chemical composition is determinated with ICP-MS. This method provides a rapid and precise means of monitoring up to 50 elements simultaneously for minor- and trace-levels. The ICP-MS technique is widely regarded as the most versatile analytical technique in the chemistry laboratory. When the sample solution is introduced into the spectrometer, it becomes atomized into a mist-like cloud. This mist is carried into the argon plasma with a stream of argon gas. The plasma (ionized argon) produces temperatures close to 7.000°C, which thermally excites the outershell electrons of the elements in the sample.

XRD analyses were carried out by conventional X-ray diffraction (XRD) techniques on samples used a (Shimadzu) XRD-6100 diffractometer with Cu (1.54060 Å) radiation operating at 40 kV and 30 mA. The powdered sample was scanned over the 5 – 80° range with step size of 0.02° and scanning speed of 1.2°/min. The analyzed material is finely ground, homogenized, and average bulk composition is determined. The most intense registered maxima in the studied powder diagrams were compared with the corresponding diagrams from PDF-2 software. Unit Cell software (Tim Holland & Simon Redfern, 1997, new version Apr 2006) was used for calculation on unit cell data.

RESULTS AND DISCUSSION

Macroscopic features on brucite from 'Ržanovo are given in Figure 3. Brucite appears in big acicular, colorless crystals. Cleavage is perfect {0001}. Crystals up to 80 cm in size were found in serpentinized ultrabasic rocks, within veins. Needles are perpendicular to vein walls. Brucite is relatively soft (2½ on the Mohs scale) and has a low density (2.38–2.40 g/cm^3). It is soluble in hydrochloric acid but has no effervescence. Weathering transforms waxy, fresh brucite into a chalk-like material.



Sample 1 Sample 2 Sample 1

Fig. 3. Crystals of brucite

Thin sections of this mineral show low birefringence and significant optical axial angle. It is also uncommon that needles of brucite show biaxial optical figure, with high optical axial angle. It is normally, as hexagonal mineral, uniaxial, but this anomaly could be explained due to the pressure acting to the extremely long crystals. Structural anomalies are not expected. It is also possible that needles of brucite are mixed with much less needles of serpentine (Bermanec et al., 1999). Chemical composition of brucite is given in Table 1.

From Table 1 it can be seen that the concentration of oxides is: MgO 66.39–69.13, Al_2O_3 0.343–0.601, FeO 1.956–4.025, MnO_2 0.239–0.360, TiO_2 0.003–0,009, LOI 25.1–27.4. Among the measured trace elements, just is elevated Ni 401–1910 ppm and all other elements are insignificant (Cr, Zn, Co, Cu, V, Sr, Ce, La, Nb, Y, Sr, and Ba).

It is not likely that P, Si, AI, Ca, K, and Na are really incorporated in crystal structure of brucite, but there was no single solid inclusion visible in the optical microscope. This would explain small percentage of Si and AI in the analyses. There is no high Mn content in the brucite from 'Ržanovo, what was explanation for acicular growth of this mineral in many other occurrences (Bermanec et al., 1999).

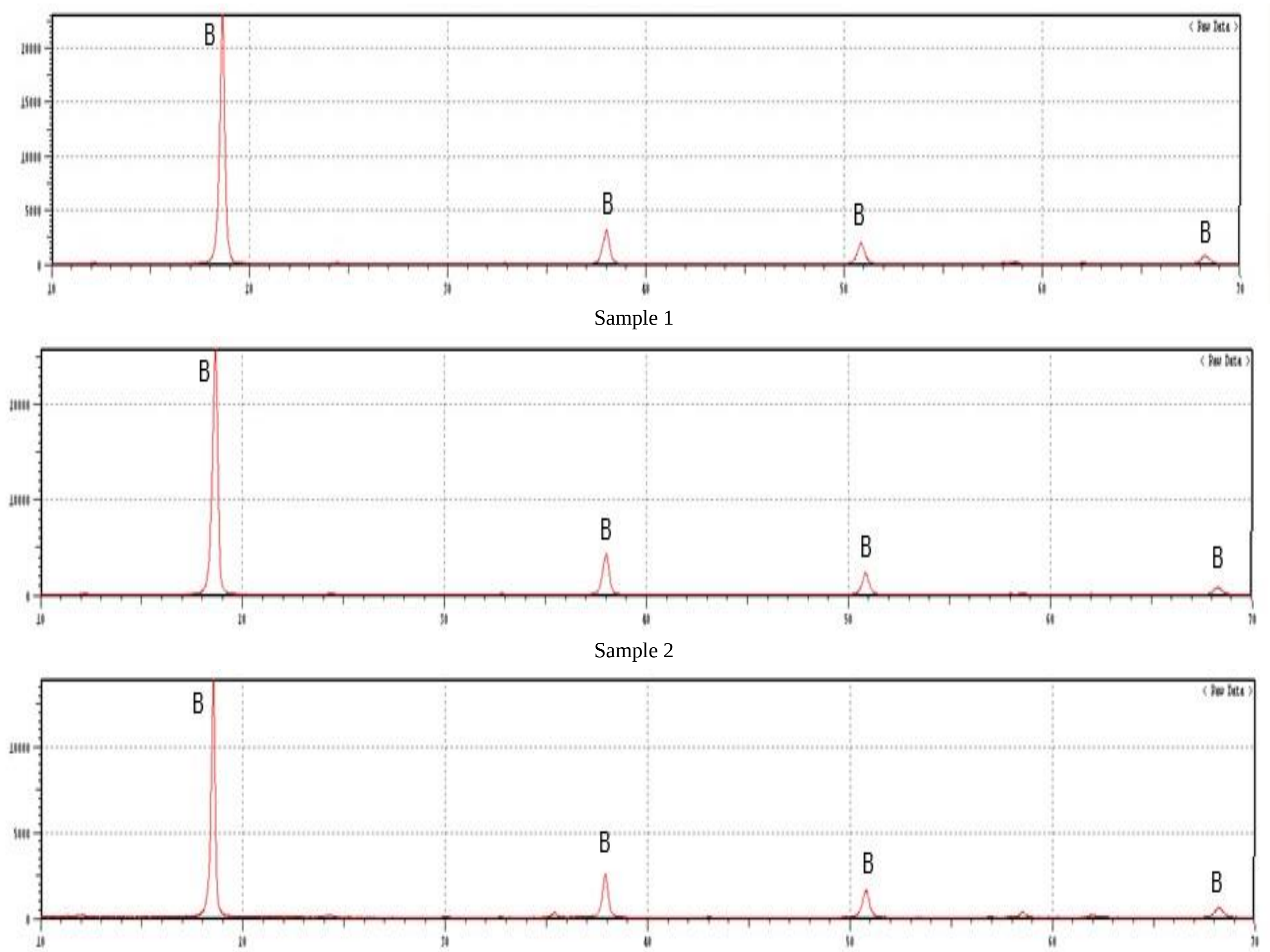
XRD patterns on investigated samples are given in Figure 4. The most intense registered maxima in the studied powder pattern are compared with the corresponding maxima of brucite ICDD 00 007 0239.

Table 1

Chemical composition of brucite from 'Ržanovo

	Sample 1		Sample 2		Sample 3	
	Top	Middle	Top	Middle	Top	Middle
Oxides (%)						
SiO_2	<1	<1	<1	<1	<1	<1
MgO	68.05	68.25	66.39	66.81	68.51	69.13
Al_2O_3	0.359	0.601	0.331	0.343	0.462	0.381
CaO	<0.050	<0.050	<0.050	<0.050	<0.050	<0.050
K_2O	<0.050	<0.050	<0.050	<0.050	<0.050	<0.050
Na_2O	<0.050	<0.050	<0.050	<0.050	<0.050	<0.050
FeO	2.810	1.956	2.255	3.956	2.102	4.025
MnO_2	0.318	0.287	0.342	0.239	0.249	0.360
TiO_2	0.003	0.008	0.003	0.009	0.003	0.003
BaO	0.004	<0.001	0.003	<0.001	0.030	0.009
P_2O_5	0.004	0.004	0.010	0.016	<0.001	0.009
S	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
LOI	26.7	26.2	27.2	27.4	25.1	25.2
Elements (mg/kg)						
Ni	1723	1591	401	456	1858	1910
Co	142	133	118	94	171	180
Cr	11.4	10.4	3.9	2.2	67.4	49.5
Cu	9.90	1.30	9.10	1.50	<1.0	<1.0
Zn	26.8	18.4	22.7	11.7	20.2	17.7
Li	<5	<5	<5	<5	<5	<5
Be	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
B	<10	<10	<10	<10	<10	<10
V	8.6	6.9	6.8	4.6	6.3	13.9
Ge	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
As	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Se	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Rb	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Sr	5.87	2.12	<1.0	1.35	12.07	2.63
Mo	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Pd	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Ag	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Cd	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Sn	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Sb	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Cs	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Ce	4.60	27.80	3.57	14.50	1.28	4.40
W	2.71	2.95	3.21	3.02	2.47	3.41
Tl	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Pb	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Bi	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Th	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
U	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Dy	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Er	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Eu	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Hf	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Ho	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
In*	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
La	2.35	<10	1.26	<1.0	<1.0	1.73
Lu	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0

	Sample 1		Sample 2		Sample 3	
	Top	Middle	Top	Middle	Top	Middle
Nb	3.21	1.02	3.54	1.38	1.12	1.43
Nd	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Sc*	2.12	1.88	<1.0	<1.0	1.20	1.39
Sm	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Tm	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Y	3.86	2.25	2.26	2.01	2.35	2.39
Tb	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Gd	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0



Sample 1

Sample 2

Sample 3

Fig. 4. XRD pattern of the samples 1, 2, 3

The unit cell parameters obtained using the main reflection lines of X-ray diffraction on sample is: a = 3.1458(7), c = 4.766(2)Å (rhombohedral a = 2.4130 Å, α = 81.36°), V = 40.84(2) Å^3, Z = 1.

Samples of lizardite and dolomite occur together with brucite. Lizardite was recognized as a distinct mineral species by Whittaker & Zussman (1956), who found that it possesses a one-layer orthogonal unit-cell and flat layers rather than cylindrical layers as in chrysotile or corrugated layers as in antigorite. Lizardite is green in color that can be seen on Figure 5(a,b). Cleavage is perfect on {0001}. Hardness = 2½, density 2,57 g/cm^3.

Dolomite occurs as lenses (Figure 5(c)). It is white in color. Chemically is very pure. Among the measured trace elements, just Sr – 432 ppm, and Pb – 382 ppm is are elevated and all other elements are insignificant. Chemical compositions of lizardite and dolomite are given in Table 2.

a

b)

c)

Fig. 5. a, b – lizardite, c – white dolomite

T a b l e 2

Chemical compositions of lizardite and dolomite from ‘Ržanovo

	Lizardite	Lizardite	Dolomite
Oxides (%)			
SiO_2	44.0	41.6	–
MgO	38.55	41.00	24.26
Al_2O_3	<200	0.323	<200
CaO	0.361	0.537	30.840
K_2O	<0.050	<0.50	<0.50
Na_2O	<0.050	<0.50	<0.50
FeO	3.527	3.787	1.089
MnO_2	0.081	0.181	0.620
TiO_2	<0.001	0.0017	0.0040
BaO	<0.001	<0.001	0.0064
P_2O_5	0.0035	0.0061	0.0035
S	<0.05	<0.05	<0.05
LOI	11,4	12,3	43.18
Elements (mg/kg)			
Ni	215	486	7.3
Co	14	19	1
Cr	18.8	8.3	<1.0
Cu	<1.0	11.0	2.00
Zn	11.3	13.6	<1.0
Li	19.80	12.70	<5
Be	<1.0	<1.0	<1.0
B	<10	<10	<10
V	5.4	11.1	1.9
Ge	<1.0	<1.0	<1.0
As	<1.0	<1.0	<1.0
Se	<1.0	<1.0	<1.0
Rb	<1.0	<1.0	<1.0
Sr	9.84	10.8	432
Mo	<1.0	<1.0	<1.0
Pd	<1.0	<1.0	<1.0
Ag	<1.0	<1.0	<1.0
Cd	<1.0	<1.0	<1.0
Sn	<1.0	<1.0	<1.0
Sb	<1.0	<1.0	<1.0
Cs	<1.0	<1.0	<1.0
Ce	<1.0	1.09	<1.0
W	<1.0	1.08	<1.0
Tl	<1.0	<1.0	<1.0
Pb	<1.0	<1.0	382
Bi	<1.0	<1.0	<1.0
Th	<1.0	<1.0	<1.0
U	<1.0	<1.0	<1.0
Dy	<1.0	<1.0	<1.0
Er	<0.1	<1.0	<1.0
Eu	<0.2	<0.2	<0.2
Hf	<1.0	<1.0	<1.0
Ho	0.2	0.23	0.39
In*	<1.0	<1.0	<1.0
La	1.53	<1.0	<1.0
Lu	<1.0	<1.0	<1.0
Nb	2.32	2.41	2.25
Nd	<1.0	<1.0	<1.0
Sc*	1.83	<1.0	<1.0
Sm	<1.0	<1.0	<1.0
Tm	<1.0	<1.0	<1.0
Y	2.17	2.60	1.46
Tb	<1.0	<1.0	<1.0
Gd	<1.0	<1.0	<1.0

XRD patterns on investigated samples (lizardite and dolomite) are given in Figures 6 and 7. The most intense registered maxima in the studied powder patterns are compared with the corresponding maxima of lizardite ICDD 00 010 0382 and dolomite ICDD 00 034 0517.

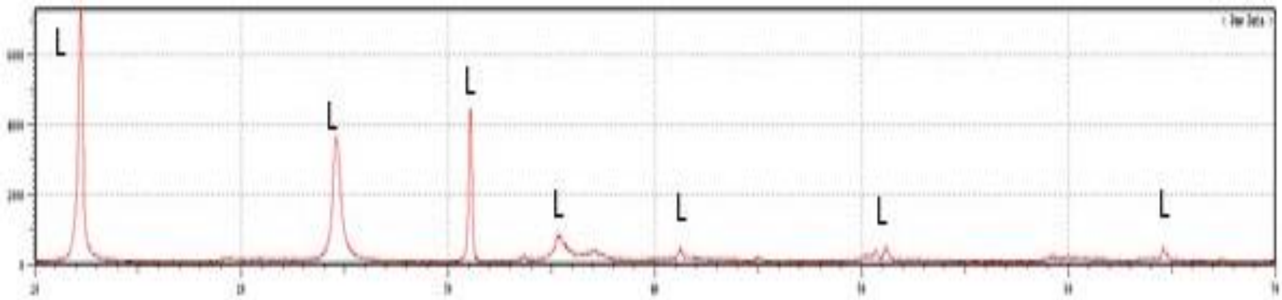


Fig. 6. XRD pattern of the lizardite

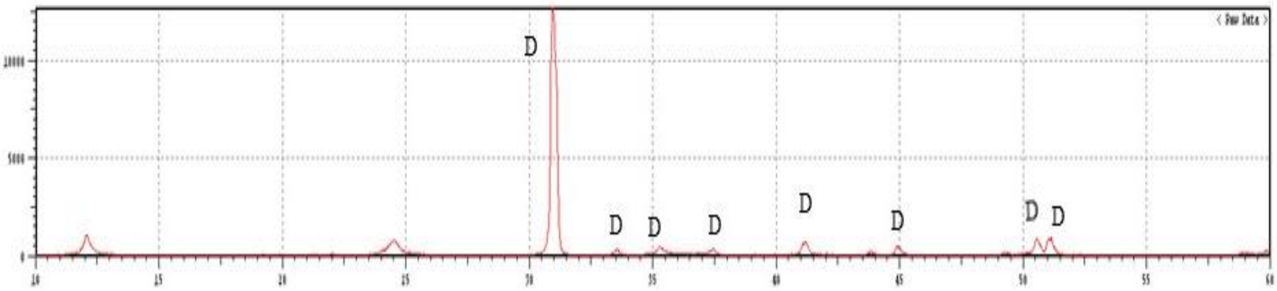


Fig. 7. XRD pattern of the dolomite

CONCLUSION

Brucite is a magnesium hydroxide, $Mg(OH)_2$. Brucite from ‘Ržanovo is the example of the biggest known crystal of these mineral in the world. Crystals up to 80 cm in size were found in serpentinized ultrabasic rocks, within veins. Needles are perpendicular to vein walls. It is a product of hydrothermal alteration of peridotitic host rock. In this process dolomite is also produced. The tectonic blocks of peridotites, which in the process of subduction were brought into parts of the continental crust as ophiolitic complexes, are stabilized in the presence of H_2O under conditions of different temperatures and pressures, during which the process of serpentinization and serpentinite formation occurs. Serpentinites are rocks in which numerous minerals are present (a group of serpentine minerals, such as chrysotile, lizardite, antigorite, as well as the presence of brucite, magnetite, talc, chlorite and tremolite. Brucite is probably one of the enigmatic minerals that forms during the serpentinization process. Brucite is relatively soft (2½ on the Mohs scale) and has a low density (2.38–2.40 g/cm^3). The concentration of oxides is: MgO 66.39–69.13, Al_2O_3 0.343–0.601, FeO 1.956–4.025, MnO_2 0.239–0.360, TiO_2 0.003–0.009, LOI 25.1–27.4. Among the measured trace elements, just is elevated Ni (401–1910 ppm) and all other elements are insignificant (Cr, Zn, Co, Cu, V, Sr, Ce, La, Nb, Y, Sr, and Ba).

Its unit cell is: a = 3.1458(7), c = 4.766(2) Å (rhombohedral a = 2.4130 Å, α = 81.36°), V = 40.84(2) Å^3, Z = 1.

In our research on the occurrence of brucite in the ‘Ržanovo-Studena Voda ophiolitic zone, we are talking about the continental serpentinites that formed in the subduction processes in the zone.

Probable reason for the formation of such big crystals is circulation of hydrothermal solutions through the fractures in the rocks whose thickness increased due to relaxation of the rock during uplift. It is a product of hydrothermal alteration of peridotitic host rock. In this process dolomite is also produced. Brucite has a higher magnesium content than any other raw material, commonly used or considered as ore.

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Р е з и м е

МИНЕРАЛОШКА КАРАКТЕРИЗАЦИЈА НА БРУЦИТ $Mg(OH)_2$ ОД ‘РЖАНОВО, СЕВЕРНА МАКЕДОНИЈА

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Бруцитот е $Mg(OH)_2$. Во ‘Ржаново се најдени најголемите кристали на бруцит во светот. Кристали со големина до 80 cm беа пронајдени во серпентинизираните ултрабазични карпи во вид на вени. Игличестите кристали на бруцит се поставени нормално на ѕидовите на вените. Тие се настанати со хидротермална промена на карпата домаќин, перидотит. Во овој процес настанува и доломит. Тектонските блокови на перидотити, кои во процесите на субдукција се внесени во деловите на континенталната кора како офиолитски комплекси, се нестабилни во присуство на H_2O во услови на различни температури и притисоци, при што доаѓа до процес на серпентинизација и формирање на серпентинити. Серпентинитите се карпи во кои има присуство на бројни минерали (група на серпентински минерали како што се хризотил, лизардит, антигорит, како и присусутво на бруцит, магнетит, талк, хлорит и тремолит. Бруцитот е веројатно еден од енигматични минерали формирани за време на процесот на серпентинизација. Концентрацијата на оксидите во испитуваниот бруцит е: MgO 66,39–69,13, Al_2O_3 0,343–0,601, FeO 1,956–4,025, MnO_2 0,239–0,360, TiO_2 0,003–0,009, LOI 25,1–27,4. Меѓу измерените елементи во траги покачена вредност има само никелот Ni, 401–1910 ppm, а сите други елементи се незначителни Cr, Zn, Co, Cu, V, Sr, Ce, La, Nb, Y, Sr и Ba. Со добиените податоци од рендгенските дифракциони испитувања се пресметани димензиите на елементарната ќелија. Добиени се следниве резултати: a = 3,1458 (7), c = 4.766 (2) Å (ромбоиден a = 2,4130 Å, α = 81,36°), V = 40,84 (2) $Å^3$, Z = 1. Веројатна причина за формирање на толку големи кристали е циркулацијата на хидротермалните раствори низ пукнатините во карпата чија дебелина се зголемила поради релаксација на карпата при издигнување.