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Listvenite as gemstone: the Antina Čuka occurrence (Eastern Serbia)

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Abstract
Antina Čuka listvenite originated by hydrothermal alteration of small ophiolite mass caused by Paleogene magmatic activity. Mineralogical and petrological examination of the listvenite revealed serpentine-rich, silica-rich, and carbonate-rich varieties of the listvenite. Typical minerals are serpentine (group), carbonates (calcite, dolomite, and magnesite), pyrite, gersdorffite, Cr-spinel, barite, limonite, and native Ag. Gemological analysis revealed all serpentine varieties from Antina Čuka (serpentine-rich listvenite, serpentine-rich listvenite with magnetite and fresh serpentinite) have values of both refractive index and specific gravity in the range for serpentine group minerals. A Refractive index value of 1.54 for silica-rich variety confirms the presence of quartz. The results of lapidary processing have proven that both listvenite from Antina Čuka and serpentinite host rock are attractive gemstones. The adequate types of processing of Antina Čuka listvenite are plain cut (different-shaped cabochons) and glyptography/carving.

Keywords
listvenite, mineralogy, petrology, gemological analyses, gemstone, economic potential.

Introduction

The majority of listvenite occurrences in Serbia are related to Jurassic ophiolites of the Vardar zone and Dinarides, but only a few to the Paleozoic ophiolites in Eastern Serbia, such as the Antina Čuka listvenite occurrence located in the eastern part of Serbia, around 200 km ESE of Belgrade (Figure 1a). In the last several decades, exploration of listvenite in Serbia was mainly focused on potential ore mineralization, while minor attention was related to studying the gemstone varieties within listvenite and listvenite as gemstone itself.

Gemstone minerals in Serbia never attracted much interest in Serbia despite the fact that more than 150 deposits and occurrences have been recorded, and some of them have been preliminarily explored in the last 50 years (Miladinović et al., 2016). Silica minerals, belonging to gemstone mineral resources of lower market value, are by far the most abundant (Miladinović et al., 2016; Miladinović et al., 2010; Ilić et al., 2006; Ilić et al., 1998; Malešević et al., 1985). The gemstones as potential raw materials in Serbia were previously studied by Vakanjac (1978) and Ilić (2002, 2006). The gemstone industry in Serbia is at the very beginning – there are just a few people dealing with gemstone processing techniques. There are no official quarries/mines of gemstones due to unsupportive legislation (Miladinović et al., 2016).

Listvenites are uncommon rocks with very complex genesis and have been defined by many authors (for instance, Rose, 1893; Sazonov, 1975; Halls and Zhao, 1995; Hansen et al., 2005; Akbulut et al., 2006; Zharikov et al., 2007, and references therein). Generally, they can be described as hydrothermal and metasomatic rocks originating from ultramafic and mafic protoliths (so-called "true listvenite" - Halls and Zhao, 1995) or silicic igneous and sedimentary protoliths (Zharikov et al., 2007). They are composed of heterogeneous mineral association: quartz, Cr-muscovite (fuchsite), Fe-Mg rich carbonates, followed by the presence of pyrite, spinel, serpentine, talc, Cr-rich chlorite, actinolite and many other mineral phases. In this paper, we use the term listvenite in a broad sense – hydrothermally altered ultramafic rock with both silica and carbonate minerals formed. An overview of listvenite occurrences in central parts of the Balkan peninsula was prepared by Antonović and Vasković (1992), and the possibilities of using selected listvenites in Serbia as a resource of gemstone and decorative stone were recapitulated by Simić et al. (2013b).

Listvenite was historically used as decorative stone predominantly in the former USSR, and even technical instructions were developed – minimal dimension for I and II grade 150 x 150 x 100 mm, and white and grey veinlets and spots up to 10 % and 40 %, respectively (Romanovich, 1986).

In this paper, we aimed to illustrate the potential utilization of the Antina Čuka listvenite as a gemstone and its correlation with mineralogical and petrological characteristics of raw material. We also presented that even small occurrences of gemstone raw material, such as listvenite from Antina Čuka (Eastern Serbia), can be used as a gemstone with economically reasonable interest.

Geological settings

Palaeozoic ophiolites of Eastern Serbia, as host rocks which were altered by hydrothermal fluids genetically related to Paleogene volcanic activity, have been historically studied but mostly from the petrological and geotectonic point of view (Terzić-Petković 1960; Ilić et al., 1967; Grubić 1989; Savov et al., 2001; Zakariadze et al., 2012; Milovanović et al., 2014), without any special attention to listvenite. The continuation of this zone can be followed towards the north to the Tisovita–Iuti Ophiolite in Romania, where listvenite was also found and studied in detail from a genetic point of view (Plissart et al., 2009).

The Antina Čuka area has been occasionally explored since 1979 as the potential zone for Pb-Zn mineralization, including exploration of diamond drilling. Due to that complex exploration programme, it was observed that Pb-Zn-Cu mineralization is of dissemination type, with low metal content (Janković, 1990). It was also discovered that the Antina Čuka area had been hydrothermally altered with several outcrops of listvenite. In 1987 a new exploration project started, targeting listvenite potential as a gemstone and decorative stone. In the Antina Čuka location, six diamond boreholes were drilled, and in one of them, silicified rock that was named aventurine by Petković (1992) was discovered. One bench was also created in 1991 in order to take enough samples for preliminary processing (Petković, 1992). Problems related to the exploration and exploitation of this occurrence were presented by Petković and Kondžulović (1993). A preliminary study of listvenite from Antina Čuka as a potential gemstone has been conducted by Simić et al. (2013a).

The general geology of the studied area (Figure 1b) was summarised after Kalenić et al. (1973). The oldest rocks in the Antina Čuka area are Proterozoic migmatic gneiss, two-mica gneiss, leptinite gneiss, leptinite and micaceous schist, locally with thin lenses of amphibolite. Upper Proterozoic and Paleozoic formations are volcano-sedimentary rocks metamorphosed to the green schist facies. Ultramafic and mafic rocks (gabbros and serpentinite) are of Early Devonian in age (Zakariadze et al., 2012). In the Antina Čuka area, serpentinite is in tectonic contact with other rocks (Grubić, 1989). According to Petković (1992), it is derived from dunite. Permian units are represented by granite and red sandstone. The Mesozoic units consist predominantly of carbonate rocks of the Cretaceous and Jurassic ages (Kalenić et al., 1973).

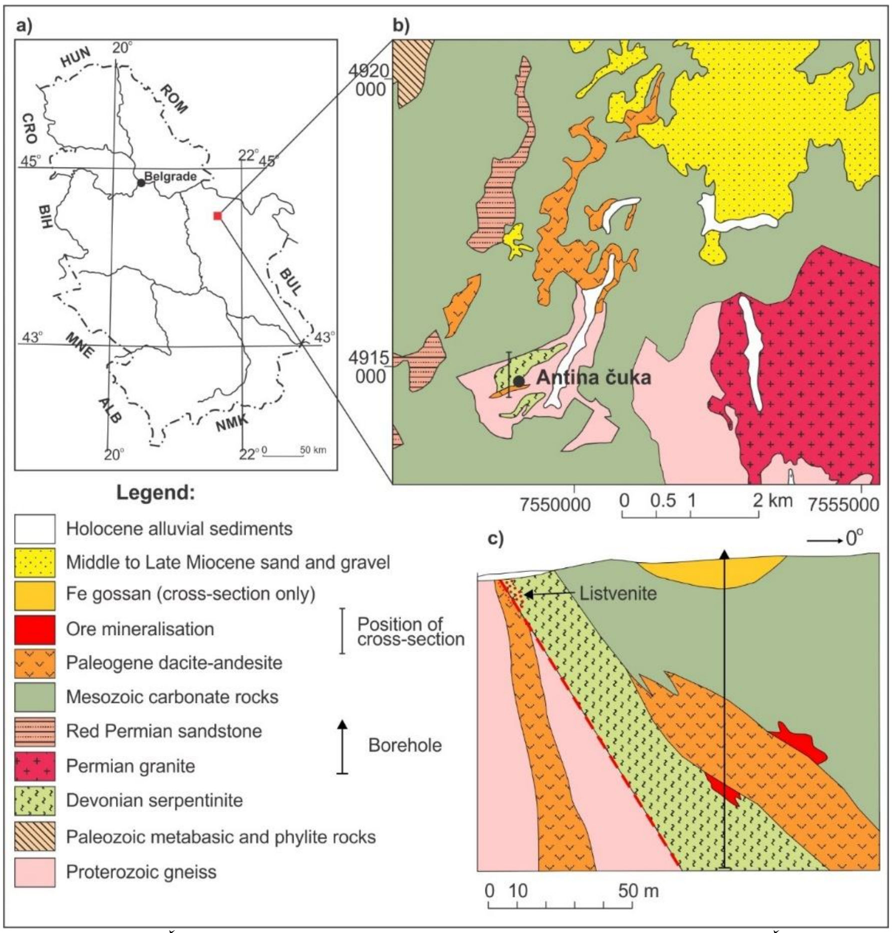

Figure 1: a) Position of the Antina Čuka listvenite occurrence, b) simplified geological map of the Antina Čuka area (after Kalenić et al., 1973) and c) schematic cross-section of the Antina Čuka area with the position of identified listvenite (modified after Janković, 1990).

During Paleogene in the Antina Čuka area, dacite-andesite intrusions into different rocks resulted in the formation of different metasomatic and metamorphic rocks - hornfels, skarn, marble and listvenite, occasionally with small ore bodies (Figure 1c). Middle to Late Miocene sediments consists of sand and gravel, occasionally with shale. Alluvial deposits of the Holocene age are mostly made of sand and gravel.

Based on the results of six diamond boreholes and exploration trench and bench, beneath saprock and alluvial loose sandy clay, up to 9.6 m thick, one lens of decorative stone was drilled along the contact of andesite and serpentinite, with the estimated thickness of up to 5 m (Figure 2).

The stone was described as light green with a transition to light or dark brown. We found the same rock in the field and had one sample from the collection of the University of Belgrade - Faculty of Mining and Geology, and they were defined in this study as listvenite.

Materials and methods

Samples of listvenite were obtained from the old trench near Antina Čuka and selected in such a way as to represent all macroscopically different varieties. One sample of fresh serpentinite was taken from the outcrop.

Petrographic analyses were done macroscopically using a binocular lens and diluted HCl, and under he polarized microscope with transmitted light (type Leica DMLSP). Thin sections were made using Canada balsam in which rock slices were embedded.

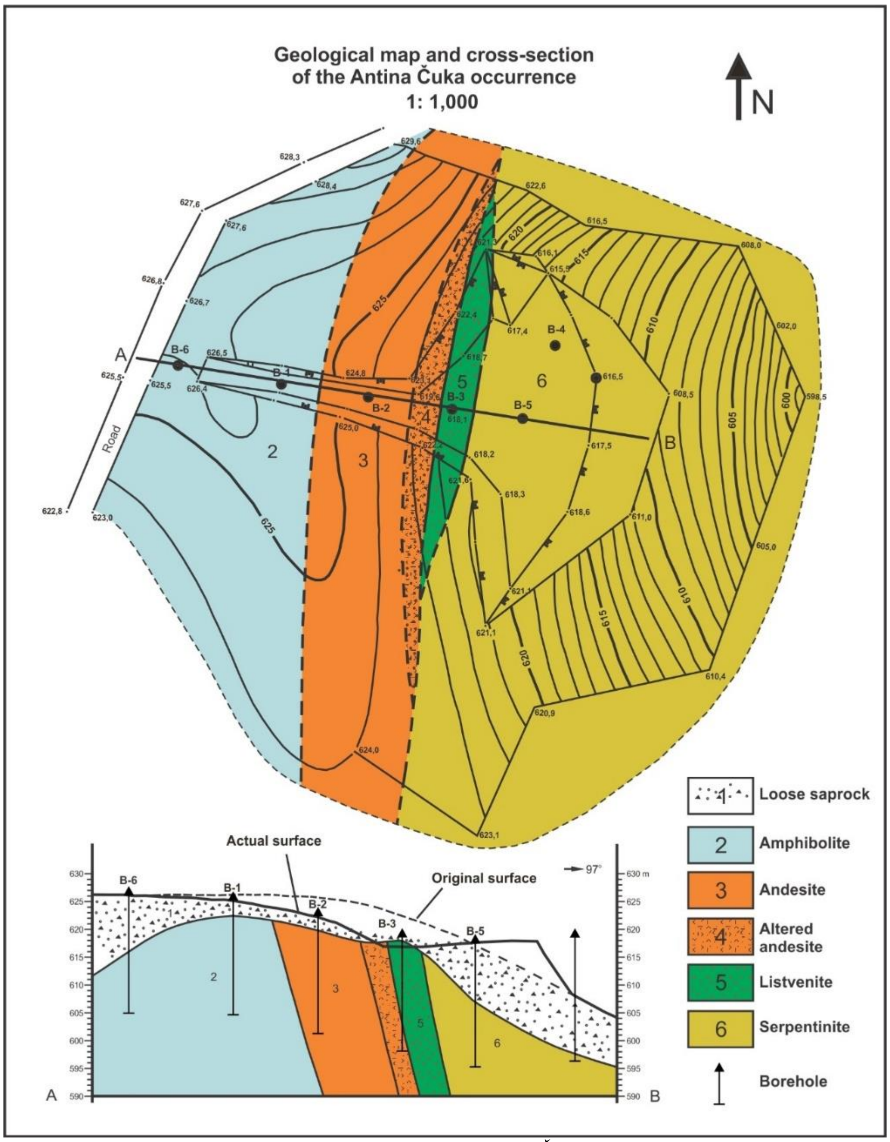

Figure 2: Geological map and cross-section of the Antina Čuka listvenite (after Petković, 1992)

Chemical compositions of minerals were obtained at the University of Belgrade – Faculty of Mining and Geology using a JEOL JSM–6610LV scanning electron microscope (SEM) coupled with an energy-dispersive X-Max Large Area Analytical Silicon Drifted spectrometer (Oxford). The samples were covered by carbon using a sputter machine type BALTEC-SCD-005. The analyses were done under an acceleration voltage of 20 kV, a beam current of 20 nA and a spot size of 1 µm. Tungsten filament was used as a beam source. Appropriate internal and external standards were used for the analyses. The detection limit for most elements was ~0.1 %.

Samples were also subjected to gemological investigations (index of refraction and specific gravity), as well as lapidary processing.

Samples for determination of refractive index (R.I.) and specific gravity were flat polished in order to determine the adequate refractive indices. A method for determining the refractive index of a sample with a flat surface was performed according to Matlins and Bonanno (2007) with a standard refractometer using 1.81 R.I. contact liquid. The specific gravity of specimens was calculated according to the hydrostatic method (Liddicoat, 1977).

Five different raw listvenite samples were subjected to lapidary processing. Dimensions of rough samples ranged from about 4x5x7 cm to about 7x8x12 cm, except in the green quartz sample (Fig. 5D), whose dimensions were slightly larger – about 8x10x20 cm. All samples were cut into 6 to 8 mm thick slabs, presenting the starting material for further processing (trimming, grinding and polishing). The slab of green quartz was obtained by cutting along the quartz vein in predominantly calcite mass. Lapidary cutting methods applied on Antina Čuka listvenite specimens included cabochon cutting and carving. Cabochon cutting was performed on a Lortone TS-8 diamond saw using sintered diamond blades. The cabochons' grinding, sanding and polishing were done on a homemade grinder/polisher similar to Lortone Beaver, equipped with a diamond grinding wheel and expanding drum with diamond resin belts for sanding and polishing. The carving was performed on Foredom CT flexible shaft with diamond electroplated grinding tips and hard felt tips with diamond paste. Applied cabochon cut and gemstone carving ("carving in the round") techniques are explained in Introduction to Lapidary (Kraus, 1987). Grinding and polishing of flat samples used for gemstone identification (R.I.) were done on the Ming Xing LZM-2E faceting machine using electroplated diamond laps.

Results and Discussion

Mineralogical and petrographic characteristics

Mineralogical and petrographic analyses were conducted in such a way as to point out the features important for the lapidary processing of this gemstone.

Basic mineralogical and petrographic studies of the Antina Čuka listvenites have already been reported by Simić et al. (2013a). The authors have emphasized that the investigated rocks are very heterogeneous in composition and fabrics, even at a distance of only a few centimetres (Figure 3a). The most obvious macroscopic characteristics are banding, the presence of veins, angular fragments, lenses, and cumulates of different mineral phases. Heterogeneity in the composition is reflected by the developing of different zones and parts with serpentine domination (Figure 3b), carbonate domination (Figure 3c) and a zone of quartz and calcite alternation (Figure 3d), all of them characterized by a relatively uniform distribution of opaque minerals and other accessory minerals.

The serpentine-rich zones contain around 80-85 % vol. of serpentine. Calcite of different degrees of crystallinity is developed within the veins and cracks. A small amount of quartz and opaque minerals, such as pyrite, spinel, magnetite and limonite, were also detected. Magnetite content can occasionally be much higher, giving added value to the such specimen after lapidary processing.

Varieties with calcite domination contain around 85 % vol. of this phase. The most characteristic feature is the significant discrepancy in grain size of calcite - from cryptocrystalline in the groundmass to crystalline and coarse-grained within some veins or small parts of the groundmass. A small amount of serpentine was also detected. Opaque minerals occur as dusty aggregates formed within the veins and cracks, as patches of limonitic crust or as individual grains, sometimes with limonitic aureoles.

The studied zones, characterized by the alternation of green and white bands and lenses, are made of quartz and carbonates, respectively. The groundmass, composed of fine-grained anisotropic quartz and locally concentrated limonite, is intersected by fractures and cracks filled with coarse- to fine-grained quartz and carbonates – dolomite and magnesite.

SEM-EDS analyses (Figure 4, Table 1) confirmed more than ten different accessory phases within the studied samples. Most of them are metallic minerals – pyrite, Cr-spinel, limonite, gersdorffite, native silver, but also clacite, dolomite, magnesite and barite. Compared to the results of Ferenc et al. (2016), we did not find so many different minerals, which might indicate a lower level of hydrothermal influence.

Table 1. Selected chemical analyses of minerals from the Antina Čuka listvenite performed by SEM-EDS method. The composition is given in wt.% and normalized to 100 %, except for carbonates.

Anality-cal spot	Sample/ site	Mineral	S	Fe	Co	Ni	As	Sb	Total									
1	464/2-1	pyrite	56.05	43.95					100.00									
2	464/2-1	pyrite	55.85	42.81	0.41	0.93			100.00									
3	464/2-1	pyrite	56.28	42.90		0.82			100.00									
4	464/4-1	pyrite	56.73	41.64		1.05	0.57		100.00									
5	464/4-7	pyrite	54.15	40.72	2.55	1.70	0.87		100.00									
6	464/4-1	gersdorffite	24.31	2.55		32.50	39.76	0.88	100.00									
7	464/4-1	gersdorffite	21.89	0.75		32.22	44.69	0.46	100.00									
Anality-cal spot	Sample/ site	Mineral	O	Mg	Al	Si	S	Ca	Ti	Cr	Mn	Fe	Ni	As	Zn	Ba	Ag	Total
8	464/2-4	Cr-spinel	27.53	0.42	0.59	0.90		0.29	0.49	31.19	0.62	33.28			4.69			100.00
9	464/2-4	Cr-spinel	26.88	0.21		0.30			0.49	31.20	1.18	38.39			1.35			100.00
10	464/2-4	dolomite	16.30	11.26		0.22		19.93					2.52					50.24
11	464/4-7	limonite	42.08			3.03		2.05		0.25	0.24	44.80	0.46	7.10				100.00
12	464/4-7	barite	21.96				8.73	1.41				2.45				65.45		100.00
13	464/2-6	limonite	31.99		1.19	0.53		1.18			6.30	52.27	4.92	1.62				100.00
14	464/2-6	calcite	15.69	2.62		0.21		34.14				0.39						53.05
15	464/2-6	magnesite	16.88	21.22		0.52			1.07			5.64						45.32
16	464/2-9	native Ag	6.93	0.52		2.82						0.51					89.22	100.00
17	464/2-9	native Ag	13.18			2.78						0.68					83.36	100.00

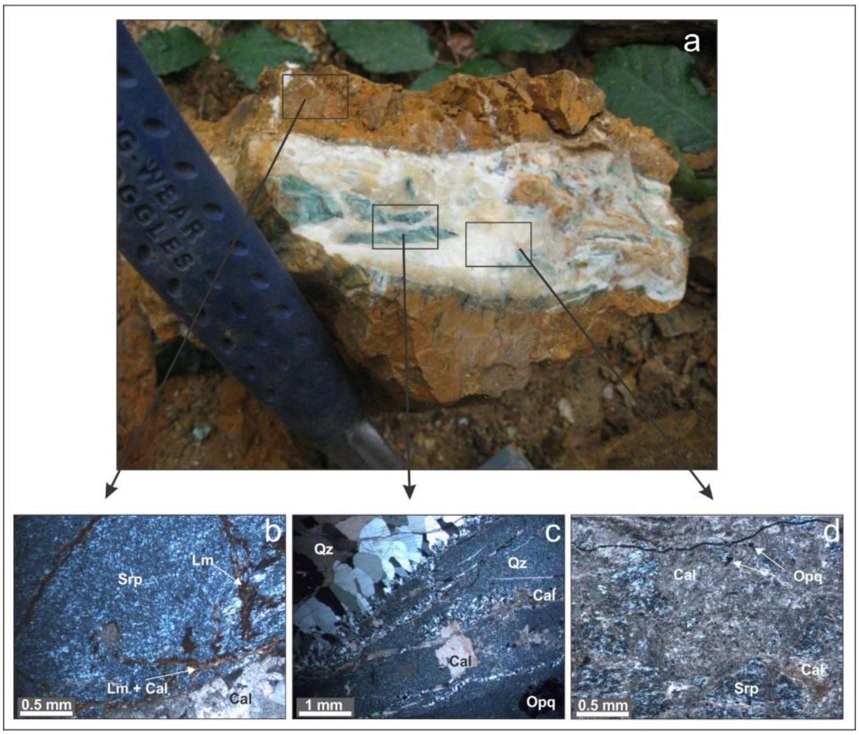

Figure 3. Petrographic characteristics of listvenites: a - macroscopically visible heterogeneity represented by bands, lenses and fragments differing in colour and mineral composition (Simić et al. 2013); b - zone with the domination of serpentine (Srp) intersected by veins of limonite (Lm) and calcite (Cal); c - zone with quartz (Qz) as a groundmass mineral; this part is characterized by the existence of microcrystalline quartz and calcite (Cal) within the veins and cracks, and dissemination of opaque mineral phases (Opq); d - zone with groundmass predominantly composed of calcite with serpentine (srp) and dissemination of opaque minerals (Opq).

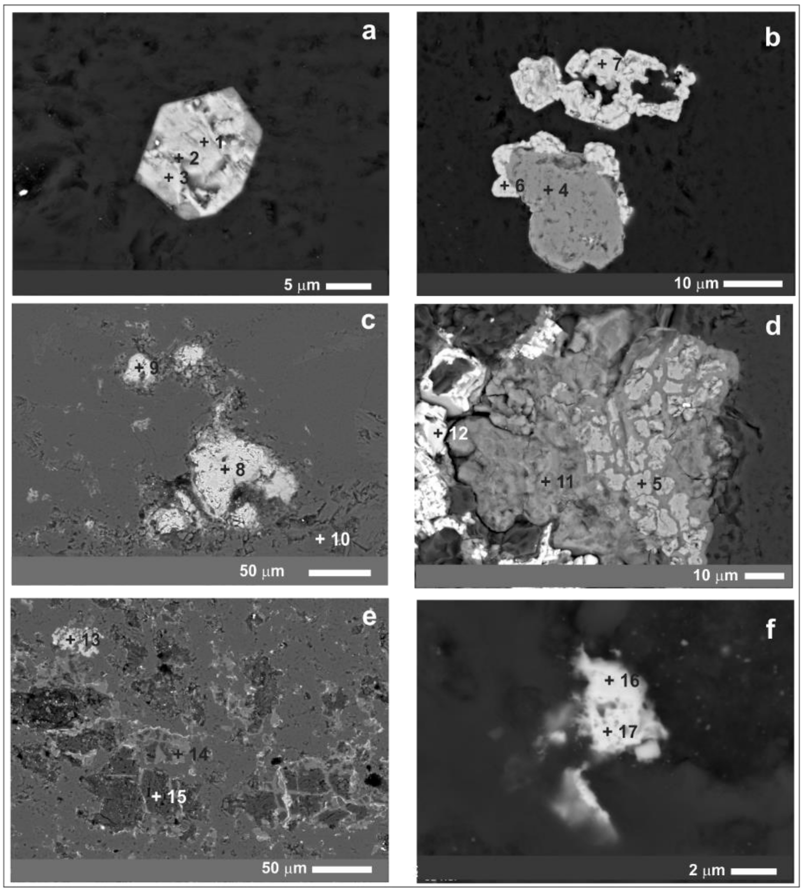

Figure 4. Backscattered electron images illustrate the distribution of different minerals with numbers indicating the positions of analyzed mineral phases. a. pyrite (1-3), b. pyrite (4) with gersdorffite (6-7), Cr-spinel (8-9) and calcite (10), d. pyrite (5) and limonite (11) with barite (12), e. limonite (13) with calcite (14) and magnesite (15), f. grain of native silver (16-17). Chemical analyses are given in Table 1.

Gemological analysis

Gemological methods are actually applied geological laboratory methods which are non-destructive to already processed (cut) gemstones. Obtaining those values (for instance, refractive index and specific gravity) are very useful at a moment when a gemstone from a deposit starts to be present in the market as finished products (cabochons, carvings, jewellery).

The heterogeneity of mineral composition was expected to be a limiting factor for the application of these methods, but measured values showed a high level of accordance with petrographic and chemical analysis. Gemological analysis was applied to already processed specimens (presented in Figure 5) to obtain their refracting index values. In addition to samples representing the final products of lapidary processing (Figure 5), a sample obtained from a white (carbonate) vein was also subjected to this type of analysis. These carbonate veins are not regarded as a specific gemstone variety from Antina Čuka gemstone occurrence but significantly contribute to the aesthetic values of those which are (Figure 5A and 5B).

Table 2. Results of gemological analysis

Specimen	Index of refraction (RI)	Specific gravity (SG)
5A – Serpentine rich listvenite	1.57	2.50
5B – Serpentine rich listvenite with magnetite	1.56	2.70
5C – Fresh serpentinite	1.55	2.45
5D – Green quartz	1.54	2.61
5E – Silica rich listvenite	1.55	2.59
Carbonate vein	1.50-1.68	2.88

For serpentine group minerals, standard refractive index values are from 1.560 to 1.571, while specific gravity values are between 2.44 and 2.66 (Schumann, 2011). All serpentine varieties from Antina Čuka (serpentine-rich listvenite, serpentine-rich listvenite with magnetite and fresh serpentinite) have values (both RI and SG) in the range for serpentine group minerals. Serpentine-rich listvenite with magnetite has slightly higher SG, probably due to the greater content of magnetite.

A very attractive green silica variety (Figure 5D) that occurs in veins (Figure 3A) is identified as quartz. According to the index of refraction, silica is represented only in crystalline form. There is no detectable presence of cryptocrystalline (chalcedony) or amorphous silica.

Carbonate-rich variety subjected to gemological analysis has a high value of birefringence – 0.180, and with a refractive index of 1.500 – 1.680 and specific gravity of 2.88, corresponds to dolomite.

One of the most important issues for the estimation of whether Antina Čuka listvenite could be used as a gemstone is to analyze the properties that turn a mineral or a rock into a gemstone class, such as beauty, durability and rarity.

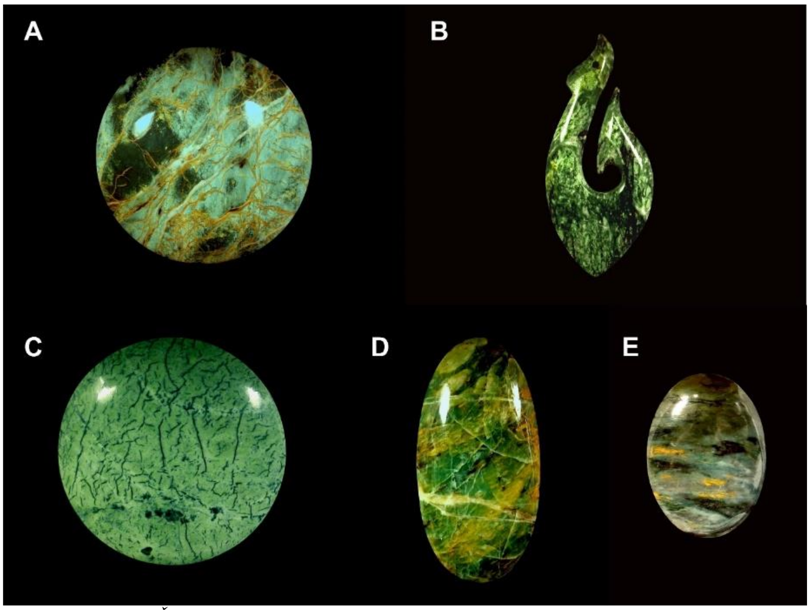

Figure 5. Lapidary processed Antina Čuka listvenite varieties. A. Serpentine-rich listvenite (38 mm), B. Serpentine-rich listvenite with magnetite (57x25mm), C. Fresh serpentinite (38 mm), D. Green quartz (40x20 mm), E. Silica-rich listvenite (26x18mm)

Beauty, as the most important gemstone property, is best represented through its optical characteristics: colour, transparency, lustre, pattern, a play of light, etc. Durability is a group of physical and chemical properties that makes a mineral or a rock susceptible to gemstone processing (cutting) and, of course, its subsequent stability over time. Rarity mostly influences the commercial value of a gemstone. However, this third property comes as a result of the previous two. If beauty and durability are more amplified, the gemstone is rarer. Gemstone cutting is the process that aims to enhance good natural gemstone's characteristics and eliminate bad ones or conceal them. Therefore, we considered that the best possible way to assess the quality of Antina Čuka listvenite is to subject it to adequate gemstone cutting.

There are three main ways to cut a gemstone (Schumann, 2011) – by faceting, cabochon cutting or engraving (carving). Faceting is not aplicable on Antina Čuka listvenite since it lacks transparency. The most prominent aesthetic characteristics of Antina Čuka listvenite varieties are their colour and pattern. The best way to enhance those aesthetic properties is by applying two other main types of cut: cabochon cut and engraving (carving).

Green quartz variety is maybe the most beautiful variety because of its intense green colour with weak transparency. It also obtains very good lustre after polishing. This variety produced attractive cabochons (Figure 5D). An issue with green quartz is that it usually occurs in relatively thin veins, sometimes brecciated, which makes it inadequate for carving due to its small size.

Silica-rich listvenite consists of minerals with different hardness, and the presence of small cracks makes it also inadequate for carving. Those cracks are easily avoided with proper sawing, and high-quality cabochons can be produced (Fig 5E). On the other hand, fresh serpentinite (Fig. 5C) and serpentine-rich listvenite varieties have a rather homogenous hardness composition, and that makes them suitable for both carving and cabochon cutting (Figures 5A and 5B).

According to its aesthetic properties and utilization possibilities, the listvenite varieties from the Antina Čuka deposit are comparable to the antique verde serpentinite (5A), lizardite serpentinite (5C), nephrite (5B), aventurine (5D) and green jasper (5E). We believe that this material could be most adequately used for making silver jewellery (sterling silver .925).

Conclusion

Antina Čuka listvenite is a small lens-like ore body formed at the contact of andesite and serpentinite. It was subdivided into serpentine-rich, silica-rich and silica- and carbonate-rich varieties, all of which can be used as attractive gemstones.

The potential of the Antina Čuka listvenite as a resource of dimension stone is restricted due to the small volume of the deposit. On the other hand, the good aesthetic and polishing properties of the listvenite make this deposit a potential resource of decorative stone, but for the small products and small total production.

The valid economic potential of the occurrence can be determined only after more detailed geological exploration and studies.

Antina Čuka listvenite varieties fall into the broad inexpensive group of coloured gemstones. Since the gemstone-cutting process greatly increases rough gemstone value, it would be the most economically appropriate for a considerable part of Antina Čuka listvenite to be processed in a local lapidary workshop that needs to be opened nearby. Only smaller quantities of Antina Čuka listvenite could be sold on the market as a bulk (uncut) gemstone. Antina Čuka gemstone occurrence could be eventually utilized as a geo-touristic attraction in addition to two nearby caves – Ravništarka and Ceremošnja.

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