Sedimentary Zeolite Deposits in Serbia

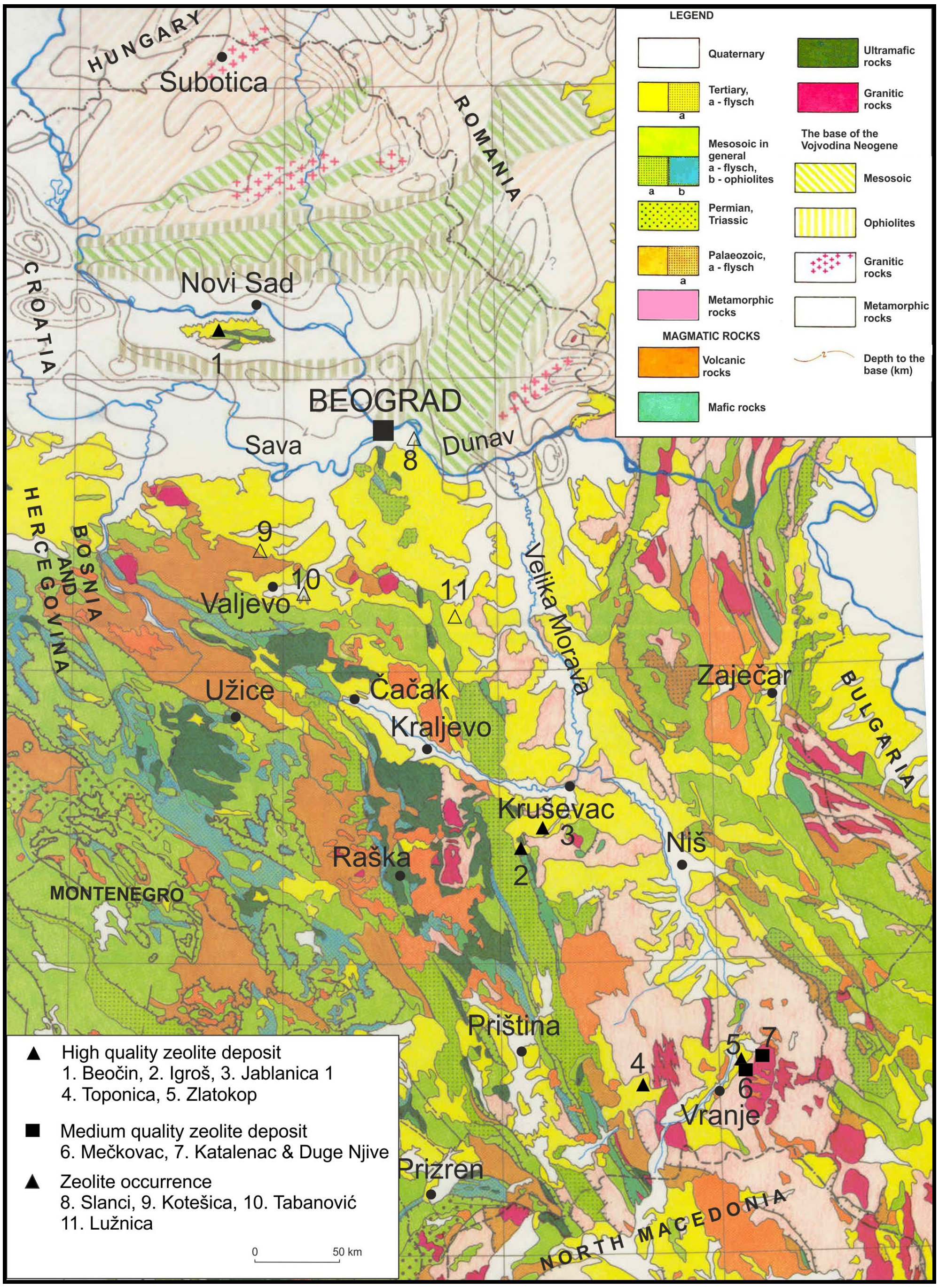

Fig. 1: Zeolite deposits and occurrences in Serbia (Geological Atlas of Serbia, 2002).

Introduction:

Zeolite deposits and occurrences in Serbia are mostly related to subaqueous alteration of ash-fall tuffs of Miocene age, but also to alteration of tuffs of Eocene age. So far, all known commercially interesting zeolite tuff deposits in Serbia are related to Miocene volcanic activity and are of high quality, with zeolite content of over 70 % (Fig. 1). The term "tuff" is here used in a non-genetic sense meaning that it delineates fine-grained material with volcanogenic particles irrespectively of their fragmentation and depositional mechanism.

Due to their industrial significance, zeolite tuffs were the subject of many studies in Serbia (Stojanović, 1968; Nikolić et al., 1975; Obradović, 1977; Gottardi and Obradović, 1978; Obradović, 1988; Simić, 1999; Simić et al., 1999, 2013, 2014; Radosavljević-Mihajlović et al., 2003, 2005, 2013).

The most important parameters of zeolite deposits in Serbia (Table 1) reveal that those deposits are relatively small in term of reserves and/or average thickness. Clinoptilolite is the most abundant zeolite mineral, while mordenite occurs in several deposits in small amounts. There are considerable differences in zeolite tuff quality within the same deposit. Geological and technological knowledge on the Katalenac, Duge Njive, and Mečkovac deposits is rather low because previous exploration campaigns were oriented towards utilisation in the cement industry, so the data on zeolite quality are just indicative, and those potential deposits are not included in this overview.

Table 1. Basic parameters of zeolite deposits in Serbia

Deposit	Age	Over-burden (m)	Rock thickness (m)	Resour-ces (Mt)	Reser-ves (Mt)	CEC (meq/g)	E.M.
Beočin	M	2-28	14.2	2	0.27	1.57	O.P.
					0.15	1.08	
Igroš	M	5-20	1.5	0.1	0.05	1.45	O.P.
Jablanica 1	M	1-15	18.5	2.2	0.20	1.68	O.P.
Zlatokop	M	20-30	2	1.3	0.67	1.64	U.
Katalenac	E	Surface	110	3.4	-	0.70	O.P.
Duge Njive	E	Surface	> 50	1.1	-	-	O.P.
Mečkovac	M	Surface	38	1.0	-	-	O.P.

M = Miocene; E = Eocene || E.M. = Exploitation method; O. P. = Open pit; U = underground

BEOČIN DEPOSIT

Geology:

The Općište deposit is located near the Beočin Monastery, on the northern slopes of Fruška Gora. The zeolitized tuff is interstratified within Badenian marine marlstones. It occurs as one composite layer 8-25 m thick (~ 17 m in average). Overburden is 2-28 m thick, around 5.4 m on average. The whole sedimentary sequence dips to the NNW at 15-25°. Previous exploration revealed three varieties of tuff: zeolite tuff, glass-rich zeolite tuff and glass-rich tuff. Total resources of all tuff varieties are ~2 Mt (Čongradac, 1986). However, in 2006 only a part of the deposit with zeolitized tuff was explored in detail (Mojić and Kovačević, 2007).

Tuff in the Općište deposit is underlain by dark grey marlstone of Badenian age. A typical cross-section through the tuff sequence (bottom to top) is as follows: marly tuff 0.6 m thick at the transition between marlstone and high-quality zeolite tuff (Simić et al., 2013). The economically important tuff sequence starts with medium- to coarse-grained zeolitized tuff 1.0-1.6 m thick, followed by medium-grained zeolite tuff, occasionally with visible biotite flakes, with a thickness of 0.6 to 8.8 m. Fine-grained zeolitized tuff (2.1-8.5 m thick) is the last unit in the zeolite-rich tuffs. The lower quality tuff section starts either (but rarely) with medium-grained glass-rich zeolite tuff 1.3 to 3.0 m thick, or (usually) with fine-grained to very fine-grained glass-rich zeolitized tuff 1.0-16.0 m thick. Fine-grained zeolite-rich tuff (2 m thick) was observed overlying glass-rich zeolitized tuff in one borehole. The tuff section finishes with fragmented marly and clayey tuff. Zeolite-rich and glass-rich zeolitized tuff varieties were distinguished based on CEC values over 130 meq/100g for zeolite-rich tuff (Mojić and Kovačević, 2007).

Mineralogy:

Microscopic study (Simić et al., 2013) revealed that altered volcanic glass (including zeolite minerals) predominantly occurs as glass shards (Fig. 2). Detritus is made up of phenocrysts and, rarely, larger pumice fragments. Quartz occurs as irregular angular grains, rarely in bipyramidal forms. Feldspars, represented mostly by sodic plagioclase, also occur as irregular grains with polysynthetic twinning lamellae. Biotite appears in the form of elongated flakes and is the most abundant mafic mineral, while amphiboles (hornblende) are quite rare. Carbonate minerals occasionally occur. Dimensions of mineral fragments vary according to the variety of tuff and, generally, become smaller upwards, which was also observed at outcrops in the open pit. The studied tuffs have either vitroclastic or vitrocrystalloclastic textures. In medium-grained tuff fossils were detected, indicating sedimentation in aqueous environment. Although the tuff is in general very pure, almost without any allogenic material, large cobbles and boulders of siltstone and fine-grained sandstone indicate a temporary input of terrigenous material into the basin.

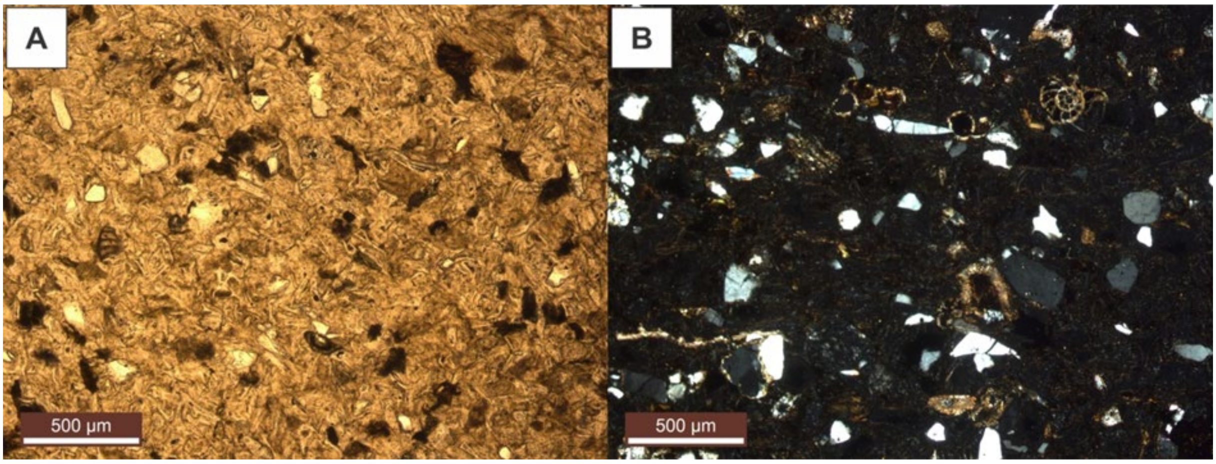

Fig. 2: A-Fine-grained zeolitized tuff – typical texture formed by abundant glass shards in the matrix (Beo-8, ppl); B-medium-grained zeolitized tuff with fragments of quartz and plagioclase phenocrysts and fossils set in glassy matrix (Beo-9, xpl), all from Simić et al. (2013).

According to SEM studies (Simić et al., 2013), clinoptilolite is the dominant zeolite mineral. It differs in shape and grain size, from ~1 µm flakes to ~5x7 µm tabular crystals (Fig. 3). Medium-grained tuff occasionally contains calcite. Glass shard relics are usually visible, displaying evidence of the initial stage of formation of clay and/or zeolite minerals (Fig. 3 C).

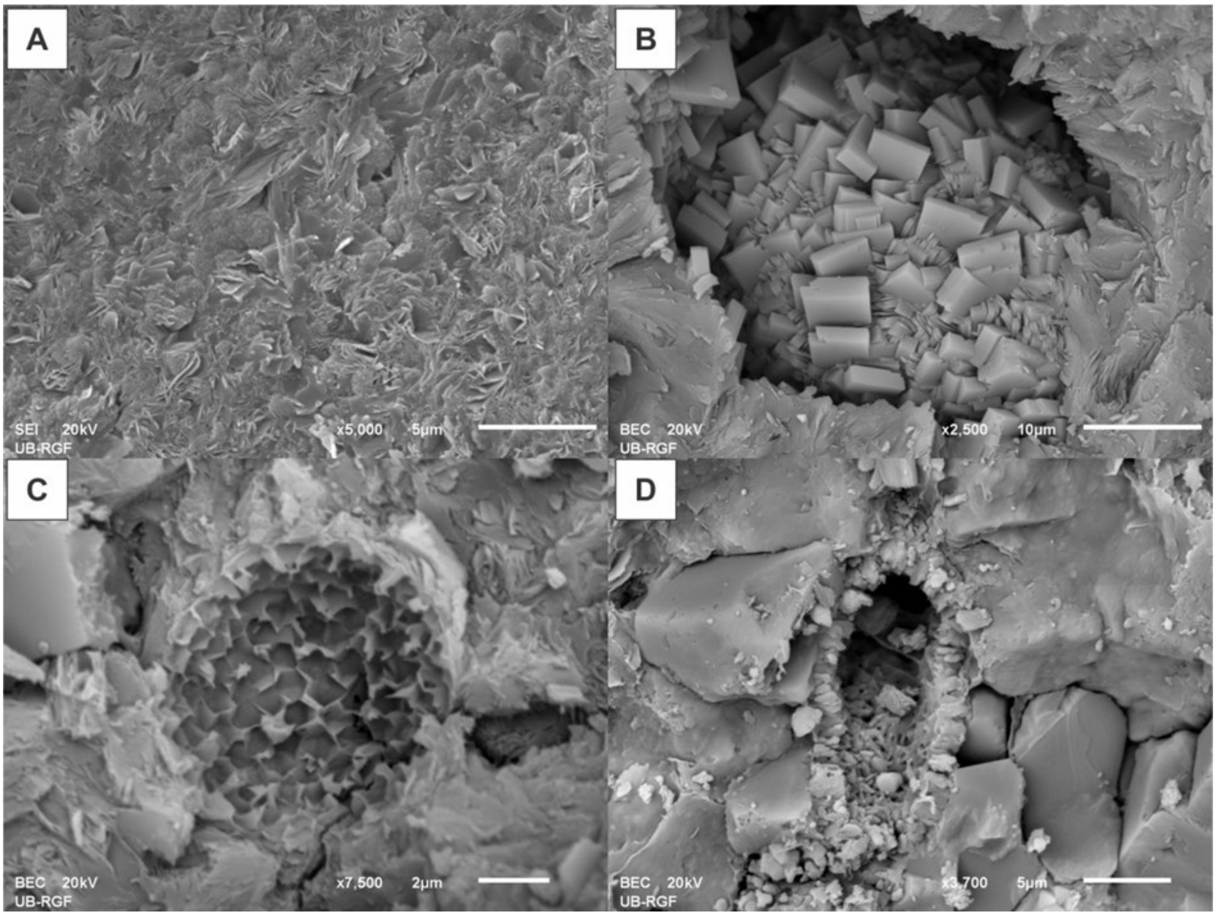

Fig. 3: A –Typical texture of fine-grained clinoptilolite-rich tuff (Beo-6); B – Different grain shape and size of clinoptilolite in fine-grained tuff (Beo-7); C – Clinoptilolite (cpt) formed by alteration of volcanic glass (Beo-6); D – Large calcite crystals in medium-grained tuff; calcite is associated with clinoptilolite (Beo-9), all from Simić et al. (2013).

Chemistry of the rock

The average chemical composition (classical wet method) of high-grade zeolitized tuff (Mojić and Kovačević, 2007) is:

SiO_2	Al_2O_3	Fe_2O_3	FeO	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	$H_2O^{105°C}$	LOI
60.10	15.82	0.87	0.20	3.25	0.86	0.53	2.71	0.27	0.03	4.76	11.07

The average chemical composition (classical wet method) of lower-grade zeolitized tuff (Mojić and Kovačević, 2007) is:

SiO_2	Al_2O_3	Fe_2O_3	FeO	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	$H_2O^{105°C}$	LOI
61.90	11.73	1.02		2.63		1.02	1.96			6.29	10.35

The average content of analysed trace elements of high-grade zeolitized tuff (by different analytical methods, Mojić and Kovačević, 2007) is:

Element	Co	Cr	Pb	Zn	Mn	Sb	Sn	Cd	Cu	Ti	As
Content (mg/kg)	4.49	5.85	16.11	24.36	167.95	21.33	41.25	0.76	2.94	598	0.78

The average cation exchange capacity (Method 9081-EPA) and the estimated zeolite content is (Mojić and Kovačević, 2007):

Zeolite variety	Zeolite (%)	CEC (meq/100 g)
High-grade zeolite tuff	85	154.9
Lower-grade zeolite tuff	60	108.3

Chemistry of the zeolite

Chemistry of clinoptilolite is given as an average from three SEM analyses on a non-polished zeolite tuff sample and is indicative only (Simić, unpublished data).

SiO_2	Al_2O_3	CaO	MgO	K_2O
79.05	13.99	3.45	1.66	1.85

Crystallography

The unit cell parameters of clinoptilolite from Beočin deposit are as follows (Radosavljević-Mihajlović et al., 2013):

a (Å)	b (Å)	c (Å)	ß (º)	V (Å^3)
17.68(4)	17.86(4)	7.41(4)	116.47(3)	2097

Physical properties of the rock

Bulk density: 1.62 tonnes/m^3

Reserves and production

High-grade zeolitized tuff reserves are around 267,000 tonnes and of lower-grade zeolitized tuff around 149,000 t, in total around 416,000 tonnes (Mojić and Kovačević, 2007).

The deposit is not operating, and the estimated average annual production from the open pit mine was 20,000 tonnes (Mojić and Kovačević, 2007).

Main applications

The possible applications of both varieties of zeolitized tuff are as follows (Mojić and Kovačević, 2007):

High-grade zeolite tuff from the Beočin deposit can be used as a feed additive, for remediation of the environment polluted with heavy metals, ammonia, radionuclides and organic contaminants, in agriculture for storage of fruit and vegetable seeds, in fisheries to increase the survival rate of juvenile fish, in all industries (cosmetics industry, beer and wine industry), and for removal of unpleasant odours, filtration of drinking water, and even as a food supplement for the human population.

Lower-grade zeolitized tuff from the Beočin deposit can be used in agriculture to improve the quality of soil (co-fertilizers, reclamation of acid soils, improvement of water-physical properties of soil), in livestock and poultry farming (as a floor litter), as an anti-slaking component in artificial fertilizers and fillers for chemical poisons, and for processing animal and poultry excrement to obtain high-quality fertilizers.

IGROŠ DEPOSIT

Geology:

The Igroš zeolite deposit (Figs. 4 and 5) is located in central part of Serbia, around 250 km from Belgrade. The Igroš village is 8 km away from the nearest town of Brus and is connected by asphalt road to the main road Kruševac-Brus_Kopaonik. The Igroš deposit was studied by Dimitrijević et al (1997), Mojić (1998), Majstorović et al (2010), Kašić et al (2017, 2018)

Detailed geological exploration in the period 1995-2010 revealed that the Igroš deposit consists of two separate bodies, namely Igroš-Vidojevići I and II. Zeolite body I has an area of 170x50 m, with an average thickness of 1.64 m. Zeolite body II has an area of 300x100 m, and an average thickness of 1.82 m.

The zeolite deposit, which originated by diagenesis of ash-fall tuff, contains two varieties of ore – highly zeolitised (~80 % zeolite minerals) light grey to white tuff, and greenish tuff with a lower (40-50 %) zeolite content. Both varieties of tuff occur in ore body II, whereas ore body I is composed of light grey tuff only. The average CEC value in zeolite body I is 167 meq/100 g, whereas in zeolite body II it is 138 meq/100 g.

Fig. 4: General view of the Igroš deposit (photo V. Simić).

Fig. 5: Tuff layer, ore body II (photo V. Simić).

Mineralogy

The mineral composition of both zeolite varieties is similar, with clinoptilolite-heulandite as the main mineral, together with quartz, feldspars, mica, volcanic glass, small amounts of clay, and carbonates. Zircon and apatite are accessory minerals.

Chemistry of the rock

Ore body I

The average chemical composition (classical wet method) of zeolitized tuff ore body I is (Mojić, 2010):

SiO_2	Al_2O_3	Fe_2O_3	FeO	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	$H_2O^{105°C}$	LOI
62.30	12.59	1.20	0.23	4.08	1.94	0.70	0.63	0.22	0.016	4.59	11.06

The average content of analysed trace elements (Mojić, 2010) is:

Element	Co	Cr	Pb	Zn	Mn	Sb	Sn	Cd	Cu	Ti	As
Content (mg/kg)	14.5	2.4	20.2	33.5	149	51	12	1.20	38.4	142.5	0.08

The average cation exchange capacity is (Mojić, 2010):

CEC
167 meq/100 g

Ore body II – average for both varieties of tuff

The average chemical composition (classical wet method) of zeolitized tuff ore body II is (Mojić, 2010):

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	P_2O_5	$H_2O^{105°C}$	LOI
62.23	13.06	2.63	3.53	2.14	1.31	0.68	0.23	0.017	3.89	10.16

The average content of analysed trace elements (Mojić, 2010) is:

Element	Co	Cr	Pb	Zn	Mn	Sb	Sn	Cd	Cu	Ti	As
Content (mg/kg)	13.6	24.2	20.2	43.1	107.8	45	12.5	1.1	24.7	123.9	0.2

The average cation exchange capacity is (Mojić, 2010):

CEC
138 meq/100 g

Ore body II – green tuff only

The average chemical composition (classical wet method) of green zeolitized tuff is (Mojić, 2010):

SiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	$H_2O^{105°C}$	LOI
63.67	12.72	3.38	2.43	1.93	1.27	3.90	8.13

The average cation exchange capacity is (Mojić, 2010):

CEC
99 meq/100 g

Crystallography

The unit cell parameters of clinoptilolite from Igroš deposit are as follows (Radosavljević-Mihajlović et al., 2013):

a (Å)	b (Å)	c (Å)	ß (°)	V ($Å^3$)
17.65(2)	17.94(2)	7.40(9)	116.46(9)	2096

Physical properties of the rock(s)

Bulk density: High-grade tuff 1.40 tonnes/m^3

Bulk density: Lower-grade tuff 1.73 tonnes/m^3

Reserves and production

Total measured resources in both ore bodies are 46,000 tonnes of high grade and 5,000 tonnes of lower grade zeolite. The estimated annual exploitation is 2,000 tonnes (Mojić, 2010).

Main applications

Main applications for both varieties of tuff are presented based on data from the geological report of Mojić (2010). High grade, light grey zeolitized tuff from the Igroš deposit can be used as a feed additive for the production of the mineral adsorber mycotoxin; for remediation of the environment polluted with heavy metals, ammonia, radionuclides and organic contaminants; in agriculture for storage of seeds, fruits and vegetables; in fisheries to increase the survival rate of juvenile fish; in all industries (cosmetics industry, beer and wine industry); in everyday life (removal of unpleasant odours, filtration of drinking water).

Green, lower grade tuff can be used in agriculture to improve the quality of land (co-fertilizers, amelioration of acid soils, improvement of water-physical properties of soil); in livestock and poultry farming (as floor litter); as an anti-caking addition to artificial fertilizers and fillers for chemical poisons; for processing of animal excrement and poultry to obtain high quality fertilizers.

ZLATOKOP DEPOSIT

Geology:

Zeolitic tuffs of the Zlatokop deposit near Vranje (Figs. 6-8) are interstratified in marly rocks of Miocene age (Obradović and Dimitrijević, 1987). The ore body is typically a layer of zeolite-rich dacite tuff. One zeolite-rich layer with an average thickness of 2.09 m, dips towards the NNW at of 5-10°. The deposit was explored over an area of 70 ha.

Tuffs are predominantly pelitic in texture, whitish-yellow and rarely white, with thin limonite films, and low in density (Obradović and Dimitrijević, 1987). They are made up of fine-grained zeolitic volcanic glass which is rather dominant and of rare crystals or fragments of quartz, plagioclase, and biotite, which indicates the dacitic character of the pyroclastic material. Accessory minerals are pyrite, marcasite, gypsum, barite, and limonite.

The tuffs are separated from the overlying laminated marly rocks by a layer of highly silicified tuff with occasional lenses of black chert genetically related to the volcanic glass of the tuffs (Obradović and Dimitrijević, 1987). These silicified tuffs are also laminated with laminates that stand out as alternating fine- to medium-grained chalcedony and quartz. Pyrite and analcime appear in silicified tuffs, especially medium-grained ones.

The overlying marly rocks are built by alternating laminae of carbonate and clay composition with a smaller amount of pyroclastic material, which is less and less common going upwards. The carbonate component, which is usually clearly separated from the clay one, is represented by dolomite which passes upwards into calcite. Smectite is the most common clay minerals, while illite is less abundant.

Fig. 6: General view of the Zlatokop deposit (photo V. Simić).

Fig. 7: Exploitation adit made in zeolite (photo V. Simić).

Mineralogy

Zeolitic tuff from the Zlatokop deposit is made up predominantly of clinoptilolite, while quartz, plagioclase, and biotite are less frequent. Small amounts of mordenite and, occasionally, smectite, have been also detected.

Chemistry of the rock

The average chemical composition (classical wet method) of zeolitic tuff (Matić and Milićević, 2013) is:

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	MnO	$H_2O^{105°C}$	LOI
65.97	12.39	1.62	3.03	1.02	0.92	0.99	0.19	tr.	4.76	10.02

The average content of analysed trace elements (by different analytical methods, Matić and Milićević, 2013) is:

Element	Pb	Cd	Hg	As	Zn	Cu	Se
Content (mg/kg)	6.68	0.05	0.01	26.03	14.54	2.92	19.19

Fig. 8: Zeolite-rich layer 2m thick at the bottom of the bench (photo V. Simić).

The average cation exchange capacity (Method 9081-EPA) is (Matić and Milićević, 2013):

Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	CEC (meq/100 g)
110	21	20	13	164

Chemistry of the zeolite

Composition of clinoptilolite is given as average from three SEM analyses on non-polished zeolite tuff sample and is indicative only.

SiO_2	TiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	Si/Al	SiO_2/Al_2O_3
75.98	0.25	14.40	1.68	3.90	1.63	0.52	1.64	8.97	5.28

Crystallography

The unit cell parameters of clinoptilolite from Zlatokop deposit are as follows (Radosavljević-Mihajlović et al., 2013):

a (Å)	b (Å)	c (Å)	ß (º)	V ($Å^3$)

17.67(5)	17.92(5)	7.41(5)	116.46(4)	2102

Physical properties of the rock(s)

Bulk density 1.58 t/m^3

Average porosity: 34.46 %

Specific pore volume: 0.2386 × 10^{-2} m^3/kg

Reserves and production

Zeolite tuff reserves are around 670,000 tonnes (Matić and Milićević, 2013), and potential resources outside the exploitation permit are around 1.3 Mt.

The exploitation of the deposit started in 1980 (Matić and Milićević, 2013), and the average annual production from underground mine was up to 2,000 tonnes. The operations stopped a few years prior to the COVID crisis.

Main applications

Zeolite tuff from the Zlatopkop deposit was used for animal feed, agriculture, remediation of the environment, and fish farming.

JABLANICA DEPOSIT

Geology

"Jablanica 1" deposit is in the Jablanica village. The deposit covers about 8 ha and consists of a layer of zeolite, with an average thickness of about 18.5 m (Križak et al., 2014). Two varieties of tuff are distinguished in the deposit: massive white tuff and bedded sandy tuff. The bright white tuff makes up the largest part of the deposit and occurs in the upper part of the layer, while the sandy dark grey to grey tuff is in the basal section. The ore body is in the form of layer (Fig. 9) which extends along a NW – SE direction, with a dip of 15-18° (Križak et al., 2014).

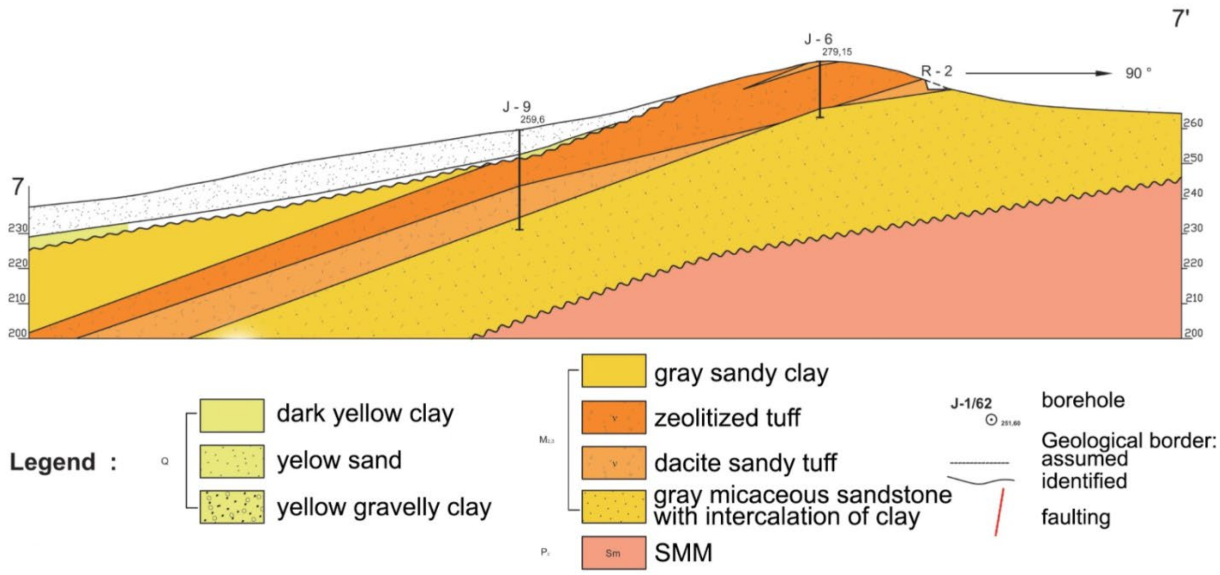

Fig. 9. Geological cross-section trough the Jablanica 1 deposit (Križak et al., 2014).

Mineralogy

Microscopic study revealed that all samples are tuff made up of tiny fragments and ground mass (Križak et al., 2014). They have a massive texture. The ground mass of the tuff, as a major component (70-95%), is completely altered volcanic glass, composed of very fine-grained cryptocristalline, poorly anisotropic aggregates. Fragments include quartz, feldspar and mica. Rare, fractured quartz grains are sometimes bipyramidal, and feldspar grains are fragmented angular-shaped, up to 0.3 mm large and occasionally altered. Fresher plagioclase grains (30 - 35% An) occur as polysynthetic lamellae or have a zonal structure. Among the mafic components, partially chloritized biotite is dominant, while amphiboles and pyroxene are much less abundant. Based on petrology and its chemical composition, the studied rock is defined as fine-grained diagenetic altered dacite tuff, with clinoptilolite as a dominant mineral (Križak et al., 2014).

Chemistry of the rock

The average chemical composition (classical wet method) of zeolitized tuff (Križak et al., 2014) is:

SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	MnO	SO_3	P_2O_5	$H_2O^{105°C}$	LOI
63.58	0.07	13.39	2.58	2.33	3.56	0.91	2.70	0.09	0.13	0.016	4.59	11.06

The average content of analysed trace elements (by different analytical methods, Križak et al., 2014) is:

Element	Pb	Ba	Cd	Cr	As	Zn	Cu	Co	Sr
Content (mg/kg)	51	550	88	38	35	66	10	37	450

The average cation exchange capacity is (Križak et al., 2014):

Ca^{2+}	Mg^{2+}	Na^+	K^+	CEC (meq/100 g)
103	18	4	43	168

Physical properties of the rock(s)

The main physical and mechanical properties of zeolitized tuff (Križak et al., 2014)

Parameter	Mean value
density with natural moisture	1.596 g/cm^3
dry state density	1.243 g/cm^3
specific mass	2.160g/cm^3
porosity	42.45%
water absorbing	28.38%
Mohs' scale of mineral hardness	3-3.5
specific active surface	142 m^2/g
natural radioactivity: ΣU	8.93 Bk/kg
^{226}Ra	8.07
^{232}Th	56.19
^{40}K	258.6
g-index	<1.0
artificial radioactivity: ^{137}Cs	<0.4 Bk/kg

Reserves and production

Zeolitized tuff reserves are around 206,000 tonnes (Križak et al., 2014), and potential resources outside the exploitation permit are around 1.4 Mt.

Main applications

Zeolite from this deposit can be used in pharmacy, cosmetics, water protection, agriculture, and remediation (Križak et al., 2014), although the trace elements content should be carefully checked as some values are quite high (Pb and As).

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