

MINERALOGICAL, PETROGRAPHIC AND CHEMICAL COMPOSITION OF THE GRANODIORITE ROCKS FROM THE KOSOVSKA RIVER LOCALITY IN WESTERN MACEDONIA

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A b s t r a c t: In this paper are shown the results of the mineralogical-petrographic and chemical study of granodiorite rocks from the Kosovska River locality. The mineralogical-petrographic characteristics of samples that were taken, have been determined by use of the polarized light microscope Leitz Vetzlar, Germany. The mineralogical composition was determined by the X-ray diffraction method of the powdered samples (XRD) and using scanning electron microscope. Based on the performed geological study in the field and laboratory study of the particular samples taken from the granodiorite rocks from the Kosovska River locality, were determined the following types of rocks: coarse grained granodiorite and porphyroidal granodiorite.

Key words: granodiorite; mineralogical composition; major minerals; mineral composition; mineral association

INTRODUCTION

The granodiorite of Kosovska River is located about 0.5 km north-west from the Canište village and about 6 km south-east from the village of Kruševica, in the series of gneisses, which has been broken through with granite and granodiorite, as part of the Pelagon metamorphic complex (Figure 1). This area is geographically close to the Selečka Mountain, as an eminent orographic unit in this part of Macedonia. In the past period, up to now, in search of good quality granite, many other regions have been explored in the area of Mariovo on many occasions, but no significant results have been received.

The first brief data from the exploration area and its surroundings were presented in the works of Boue (1891) and Cvijić (1906, 1911). After that, Nikolov (1921, 1924), Tučan (1926), Barić (1940) and Marić (1936, 1940) studied granodiorites, and provided additional data regarding their mineralogical, petrographic and genetic features. Stojanov (1958, 1960, 1968, 1974) studied these terrains and distinguished many varieties of gneisses, micaschists, amphibolite and granitoid rocks. Some of these scientists came up with conclusions regarding the en-

tire Pelagonian, which suggests that at the beginning of the Algonquian orogenic movements, a progressive metamorphosis occurred in the Precambrian complex, and towards the end of the orogenic movements, granodiorite–adamellite masses were initiated. In the period of the preparation of the basic geological map of SFRY (Socialist Federal Republic of Yugoslavia), the authors of the sheet Vitolište (Dumurdžanov and Hristov, 1976) and sheet Prilep (Rakičević, Stojanov and Arsovski, 1965) processed the leave content of the rocks of the sheet Vitolište, where the granodiorite Kosovska River belongs.

Dumurdžanov (1985) explored granitoids in details and concluded that they are mainly represented with granodiorite (70%) and quartzdiorite (20%), and less with quartz monzonites and granites.

The Kosovska River granodiorites have been studied in details by Spasovski et al. (2010); they determined their mineralogical-petrographic composition. Later, granodiorite rocks near this area were investigated by Stojkov and Spasovski (2014), Stojkov et al. (2015) and by Spasovski O., and Spasovski D. (2011, 2015).

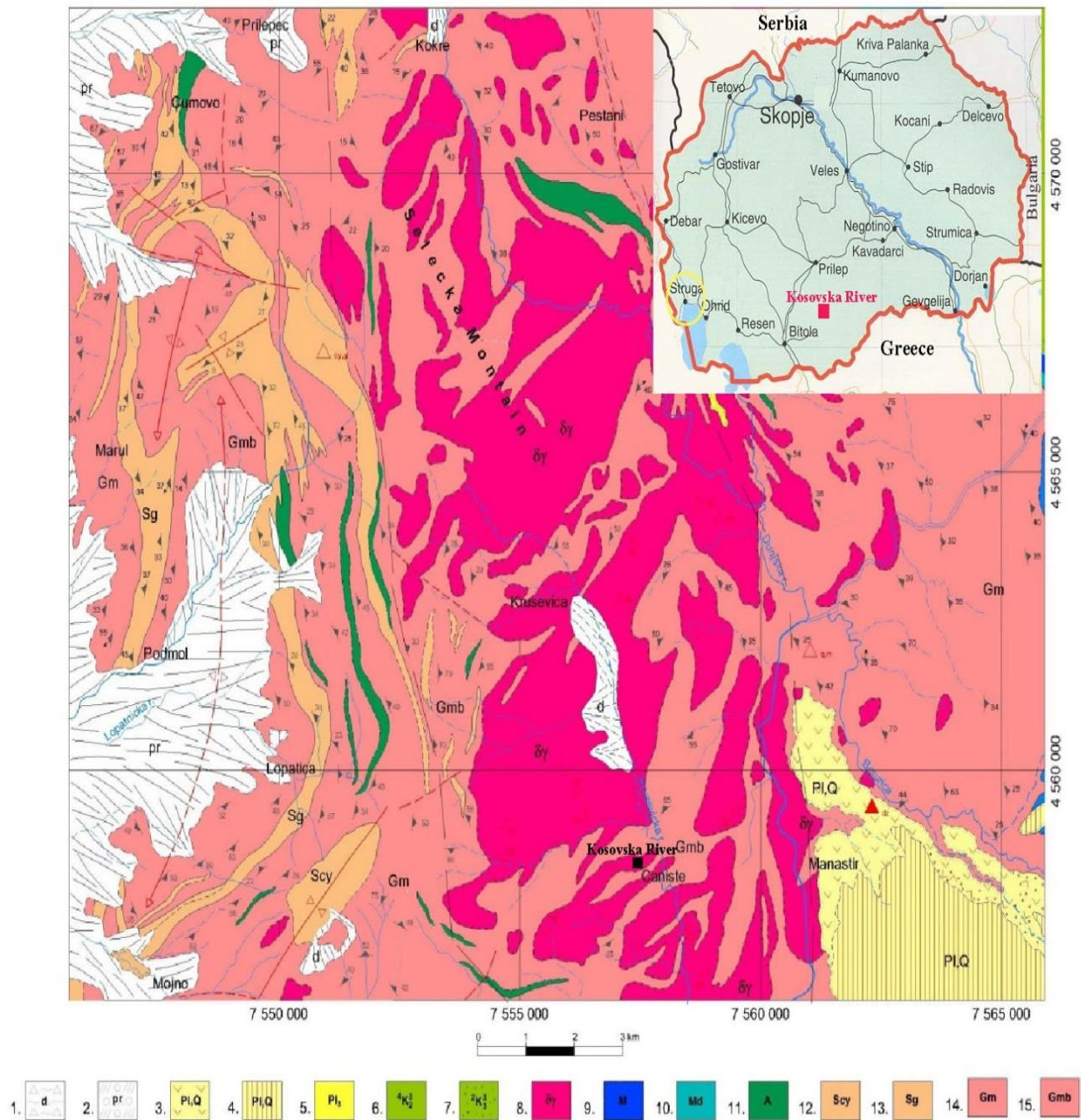


Fig. 1. Geological map of the wider area (according to the Geological Map of the North Macedonia 1 : 100 000)
 1) deluvium; 2) proluvium; 3) volcanogenic sedimentary formations; 4) carbonate-travertine formations; 5) gravels, sands and clays; 6) slabs and banked limestone; 7) sands, marls and conglomerates; 8) granitoides; 9) calcite marbles; 10) gray to gray-white dolomite marbles; 11) amphibolites and amphibolitic shists; 12) granite-kyanite micaschists; 13) granite-staurolite micaschists; 14) strip muscovite gneiss; 15) strip muscovite-biotite gneiss.

METHODOLOGY

The location in Kosovska River was explored using field and laboratory techniques. The field study provided an insight about the terrain, familiarization with its geological and structural-tectonic features, as well as the collection of the representative samples from the granodiorite and definition of

their chemical-mineralogical composition and structural-textural features. The mineralogical-petrographic study was performed at the Faculty of Natural and Technical Sciences in Štip, while the chemical composition of granodiorite was determined in the chemical laboratories at the Faculty of Natural and

Technical Sciences with the inductive coupled plasma atomic emission spectrometer (ICP-AES). The mineralogical and petrographic characteristics of the samples taken, were determined by the use of the polarized light microscope Leitz Wetzlar, Germany. The mineralogical composition of the rocks was determined by the X-ray diffraction (XRD). The powder X-ray analysis was performed on a diffractometer (Shimadzu) XRD-6100, with Cu (1.54060 \AA) radiation operating at 40 kV and 30 mA. The powdered sample was scanned over the $5\text{--}80^\circ$ range with step size of 0.02° and scanning speed of $1.2^\circ/\text{min}$. The chemical compositions of the major mineral phases were determined by scanning electron microscope (SEM). The SEM analyses were recorded on the scanning electron microscopy VEGA3 LMU and INCA Energy 250 Microanalysis System for quantitative analyses of the samples. The

SEM analyses were performed with the SE (Scattered Electrons) detector on 20 kV voltage. First, the samples were cleaned and then a small piece was put on the sample holder with carbon double-adhesive tape on it. The sample's surface was coated with gold on Modular Coater, Quorum Q150R ES and then analyzed in high vacuum mode with more than 0.018 Pa. The sample surface was motorized on 5 axes ($x\text{-}y\text{-}z$, rotation and tilt). For SEM, the VegaTC software was used. The energy-dispersive X-ray (EDX) system for SEM is a fully quantitative SDD with excellent performance at low and high-count rates, which is capable of achieving a resolution better than 125 eV on the MnK α , FK α and CK α peaks. The working distance for X-ray was 15 mm. The detector control and data acquisition were done with INCA software. The SEM-EDS analyses were done on the unpolished surfaces.

GEOLOGIC CHARACTERISTICS

In the geologic structure of the area, that is included in our observation and research, there are two types of rocks: gneiss and granodiorite (Figures 2 and 3). The muscovite gneisses are outspread in the northern and north-eastern part of the researched area. They are characterized by grey colour with glittering radiance from the leaves of muscovite, which can be clearly noticed. They are characterized by slightly distinguished parallel schistose texture. The structure of the gneisses is grano-lepidoblastic with slightly distinguished striped texture. The main minerals in the rock are quartz, feldspar and mica. The participation of salic and femic minerals is approximately equal in quantity, i.e. the salic are slightly more present. The quartz is found in xenomorphic crystals and in feldspars. The feldspar is K-feldspar – orthoclase and plagioclase. The orthoclase is clayed, while the plagioclase is more strongly clayed. The plagioclase is albite to intermediate plagioclase. It is rare to find some larger xenomorphic crystals of orthoclase, as the porphyroblast. The mica is represented with muscovite and biotite, and they are found in not clearly distinguished lines. It is typical for the biotite that it is bleached–baritized; so it has a weak brownish interference. The secondary minerals are the epidote, orthite, rarely garnet and metallic mineral in irregular shapes. The epidote is quite common in long crystals, and it is regularly associated with the mica lines. The apatite and the zircon are accessory minerals.

The granodiorites are most commonly found and mostly constitute the middle part of the researched area (Figure 3). They are characterized by the middle to large-grained content and light grey-pink colour, equally present in the entire sample. The mineral grains are with the size of 5 mm, and rarely slightly bigger ones with 1 cm. With a microscope, it can be spotted that they have hypidiomorphic grain structure. The main minerals are quartz, plagioclase, orthoclase and biotite. The plagioclase is strongly metamorphic, and the products are the epidote and the zoisite, and also a certain zonal allotment of the plagioclase is present (Dumurdžanov, 1985). The separate crystals of the plagioclase have completely turned into epidote with larger crystals of the epidote. The orthoclase is xenomorphic fresh and completely weakly clayed, and regularly poikilitic incorporates smaller crystals in the plagioclase and the biotite. The biotite is found in big square leaves and smaller rectangular leaves outspread, separate and in places grouped in small clumps. The biotite contains idiomorphic spires – microlites of the coesite, and there are also crystals of the epidote and the apatite on the edges. The quartz is found in the interspace with smaller xenomorphic grains. The rock is quite strong, with slight cracks on it, i.e. with slight mechanic deformations, which can be observed with the slightly distinguished undulose darkening of the quartz and the slightly present microcracks at the orthoclase.

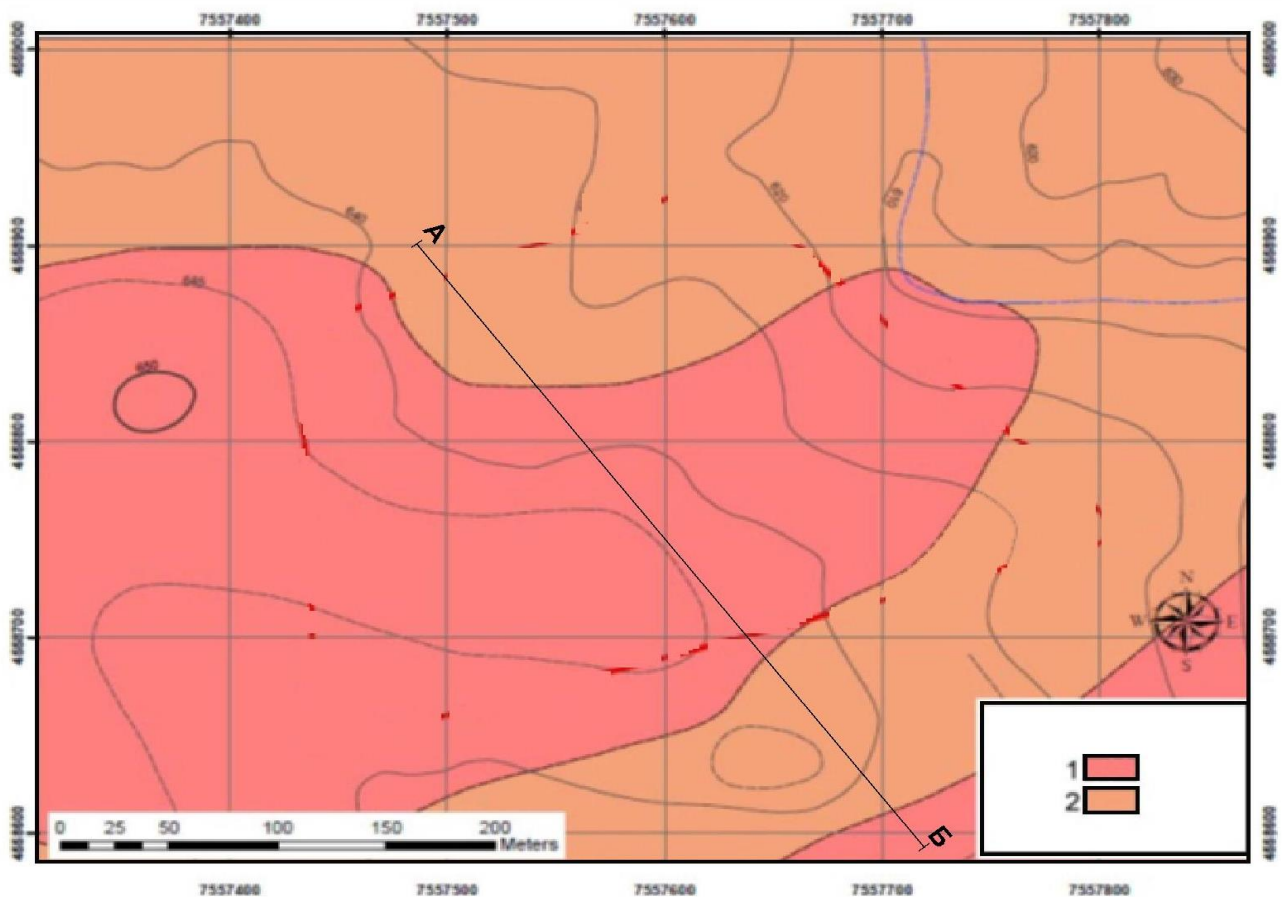


Fig. 2. Geological map of the locality Kosovska River. 1) granodiorite, 2) muscovite gneiss (Spasovski, 2010)

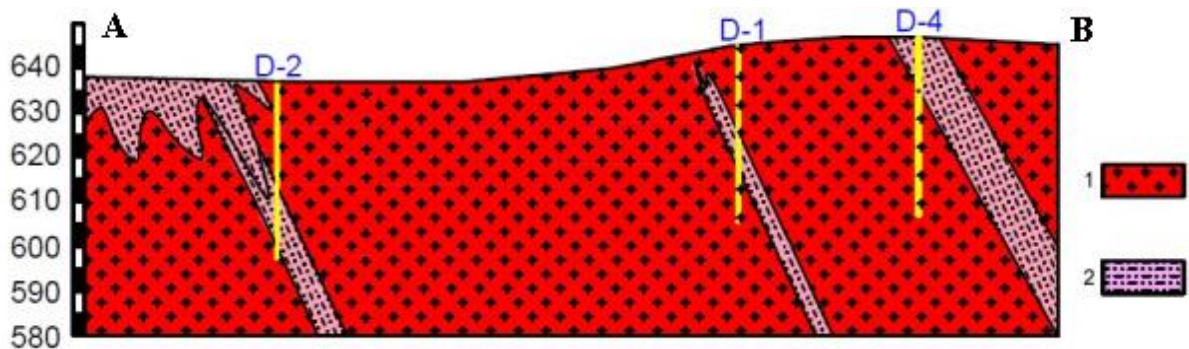


Fig. 3. Geological cross-section of Kosovska River. 1) granodiorite, 2. muscovite gneiss

MINERALOGICAL AND PETROGRAPHIC FEATURES

There were some representative samples from the Kosovska River locality selected for the mineralogical-petrographic study. The mineralogical-petrographic examinations were performed at the Faculty of Natural and Technical Sciences, Institute of Geology, by the authors of the paper. At the Kosovska River locality, two varieties of granodiorite rocks were found: coarse grained granodiorites and

porphyritic granodiorites. The coarse grained granodiorite is mainly grey with dark grey patches of coloured mineral. It consists of coarse granular minerals with a grain diameter of 5–6 mm, which is massive and has hard texture. It has hypidiomorphic granular structure and consists mainly of plagioclase, orthoclase, quartz and biotite. However, plagioclase clearly prevails in the rock.

The **plagioclase** appears as hypidiomorphic crystals. It is significantly altered, and polysynthetic lamellae can be seen very rarely in certain plagioclases. The size of the crystals is in range of 0.3 – 6 mm in diameter or elongation, and the most of them are nearly 2 mm in length. Often, the thin edge of fresh albite can be seen along the marginal parts of the crystals, mainly at the contact parts with plagioclase. The products of the alteration are coesite, epidote and sericite. The crystals of the coesite are long up 0.3 mm (300 microns), and the remaining products are minor constituents (Dumurdžanov, 1986). The products of alterations have chaotic distribution of their crystals. The plagioclase has an intermediary character, type oligoclase-andesine. Often, the major plagioclase is found as the double-twinned crystals.

The **orthoclase** is rare in xenomorphic crystals with a size up to 5 mm in diameter or elongation. Often it fits minor hypidiomorphic and allotriomorphic grains of plagioclase. The crystals of orthoclase are rare, in which some transformation in microcline can be seen; they are slightly micro wedged. The quartz is xenomorphic and it fills the interspace between the crystals; it is cracked and slightly crushed.

The **biotite** is found as the elongated flake-like forms, with the length up to 2 mm. There is a clear pleochroism in brown-slightly green nuance. It appears in thin jets, suppressed in the interspace of the salic minerals. The biotite is very slightly chloritized. The biotite jets are very often associated with fine-grained epidote–ortite and rare garnet crystals, which represent secondary minerals.

Approximately, it is found that the modal composition of the rock – based on the microscopic investigations – consists of plagioclase, 40%; orthoclase, 15%; quartz, 25%; biotite, 13%; secondary minerals, 7%. The porphyritic granodiorite represents rock with grey colour, intersected with white feldspar and dark-grey-green like coloured minerals. The structure is coarse grained with size of the feldspar up to 5–6 mm, where rare major crystals can be seen up to 1.3 cm (most probably K-feldspar). It is hard and with massive texture. Based on the microscopic observation, it could be seen that there is hypidiomorphic texture. It consists of plagioclase, orthoclase, quartz and biotite as main minerals, despite the secondary mineral epidote-ortite and rare sphene.

The **plagioclase** clearly prevails in the rock. It was found as hypidiomorphic crystals, with a length of up to 0.3 – 4 mm. It is significantly altered, and the products are epidote, zoisite and sericite, which

are found as grained and needle-like microcrystals that chaotically fill the plagioclase. Often in plagioclase crystals, integrated by the orthoclase, thin edge of fresh albite can be seen along with the edges of the crystals. The plagioclase was of intermediary character type (oligoclase–andesine–labradorite).

The **orthoclase** is rarer than the plagioclase and can be seen in crystals, larger and as the last product of crystallization, which integrates many crystals of plagioclase and rare thin biotite flakes. The size of these orthoclase crystals is up to 0.8 cm. There are rare crystals of orthoclase also in xenomorphic crystals.

The **quartz** is also less common, slightly cataclased with size of the crystals up to 1 mm. It fills the interspace between the plagioclase in single grains and small lenses.

The **biotite** appears in large masses and elongated thin flakes with size from 0.5 to 2 mm. The biotite is fresh with pleochroism in light brown greenish nuance. The biotite contains rare microcrystals of epidote and needles of coesite. Together with it or separated, the irregular shards shapes (built of minor crystals) and fine granular epidote (thick grouped) are found, as minor thin leaves of biotite – fresh as newly formed biotite. Usually, thin leaves of biotite are around these shard shapes, which could be some coloured mineral fully metamorphosed in secondary products. Some elongated orthogonal shapes could be very slightly seen. The biotite and the jets are suppressed in the interspace of the crystals, so that thin veinlets with limonitization could be found close to them. In particular, in the biotite grains along the margins, the iron component in thin irregular forms and fine grained epidotization were determined.

Approximately, based on the microscopic review, the modal composition of the rock was found as the following: plagioclase, 50%; orthoclase, 13%; quartz, 17%; biotite and secondary minerals, 20%.

In addition to the microscopic observations, the X-ray diffractograms of rocks were produced, which defined very fine mineral grains. For this study, one sample was selected. The mineralogical composition determined with XRD is shown on the diagrams presented in Figure 4.

Based on the position of the peaks, considering the angle 2θ , identification of minerals was performed, and based on intensity of their peak conclusion about their relative quantity in the sample was made.

The quartz is the most common in the studied samples, which suggests that it is represented in great percentage in the matrix of the rock. In the second place comes the microcline. The microcline in

the granodiorite rocks from this particular locality is represented with microcline. In the sample taken from granodiorites of the Kosovska River locality, hornblende and augite were found as well.

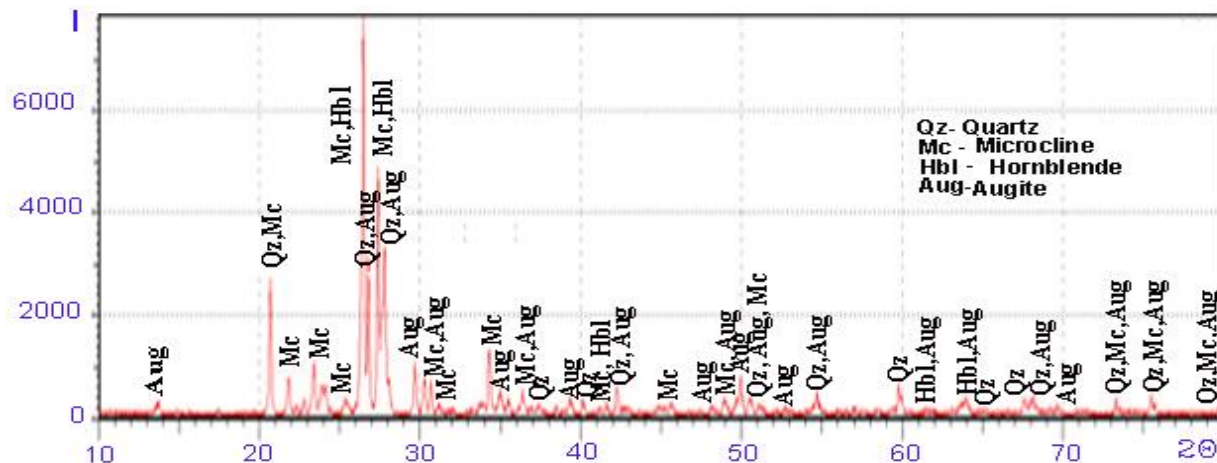


Fig. 4. X-ray diagram of granodiorite

CHEMICAL COMPOSITION OF MINERALS

For the determination of chemical composition of minerals with scanning electron microscopy (SEM), there were separated representative and fresh granodiorites.

The augite is found of the pyroxene with XRD, whose chemical composition is determined with SEM. With the microscopic investigations and the investigations by SEM, pyroxene is determined as augite (Table 1).

The SEM images of augite are given in Figure 5, while Figure 6 shows the EDX spectrum of augite.

The presence of microcline is confirmed of the feldspars; their chemical composition is presented in Table 2, and SEM images of microcline are given in Figure 7, and EDX spectrum of microcline is shown in Figure 9.

The albite is determined of plagioclases with SEM; the corresponding chemical composition is presented in Table 2, the SEM images of albite are given in Figure 8, and EDX spectrum of albite is shown in Figure 10.

The SEM study determined mica (muscovite and biotite), whose chemical composition is presented in Table 3, and respective SEM images of muscovite and biotite are given in Figures 11 and 13, and respective EDX spectrums are shown in Figures 12 and 14.

The quartz – the most common mineral in the granitoid rocks – was confirmed and its chemical composition is given in Table 4, while the SEM images of quartz are given in Figures 15 and 17, and the EDX spectrum of quartz is presented in Figures 16 and 18.

The SEM study also determined epidote, whose chemical composition is presented in Table 5, and respective SEM images are given in Figures 19 and 21, and the corresponding EDX spectrum is shown in Figures 20 and 22.

Table 1

Chemical composition of augite from the Kosovska River locality (%)

Element (K)	Weight	Atomic
O	58.77	73.44
Al	11.51	8.53
Si	15.52	11.04
Ca	10.62	5.31
Na	0.80	0.69
Fe	2.78	0.99
Total	100.00	

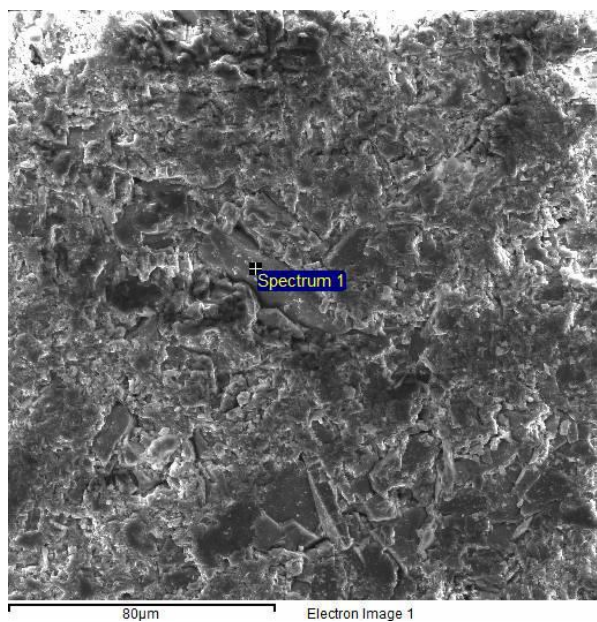


Fig. 5. SEM image of augite, sample 1

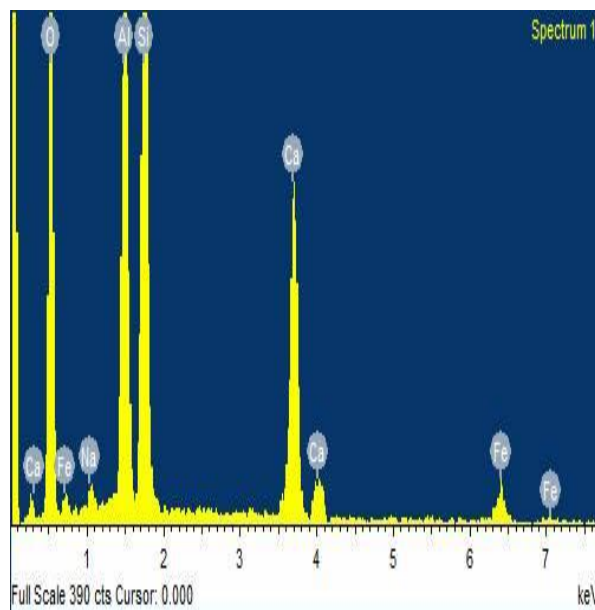


Fig. 6. EDX spectrum of augite, sample 1

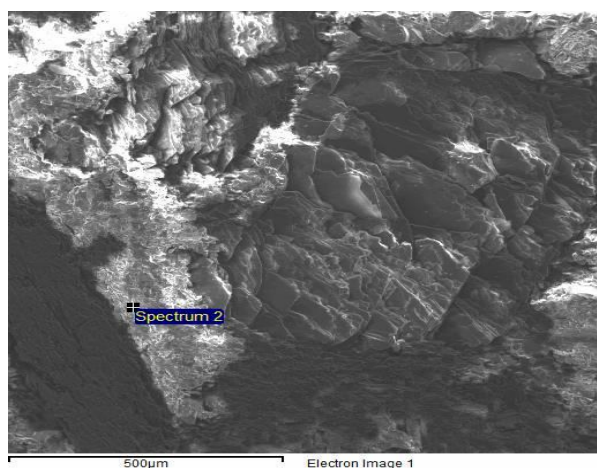


Fig. 7. SEM image of microcline

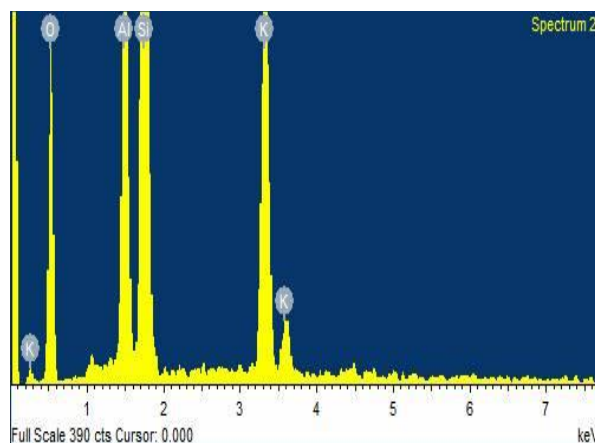


Fig. 8. EDX spectrum of microcline

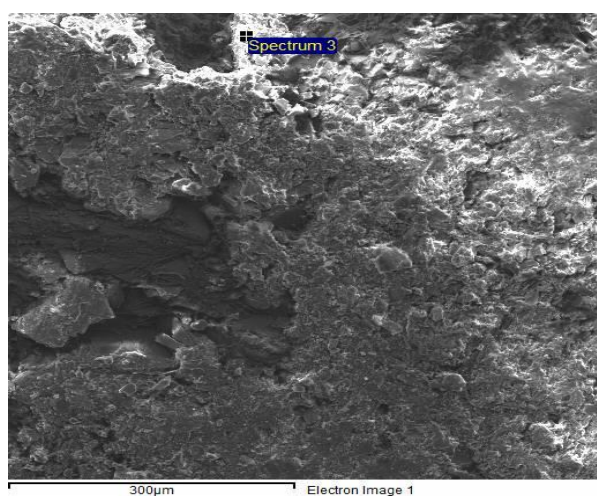


Fig. 9. SEM image of albite

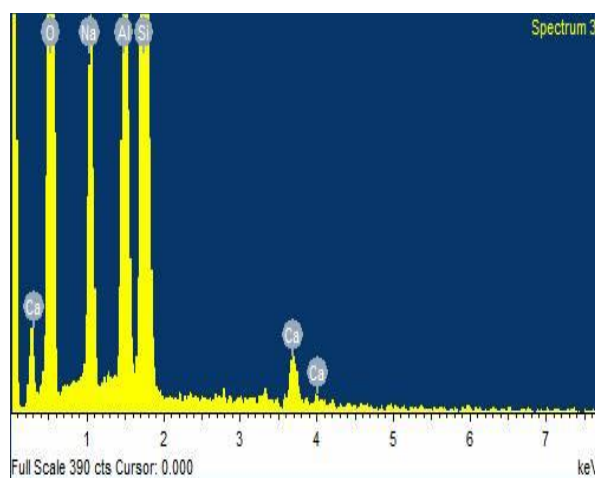


Fig. 10. EDX spectrum of albite

Table 2

Chemical composition of microcline and albite from the Kosovska River locality (%)

Element (K)	Microcline		Element (K)	Albite	
	Weigh	Atomic		Weigh	Atomic
O	44.34	60.16	O	63.05	73.99
Al	8.75	7.04	Na	7.98	6.67
Si	31.01	23.97	Al	8.98	5.95
K	15.91	8.83	Si	19.99	13.39
Total	100.00		Total	100.00	

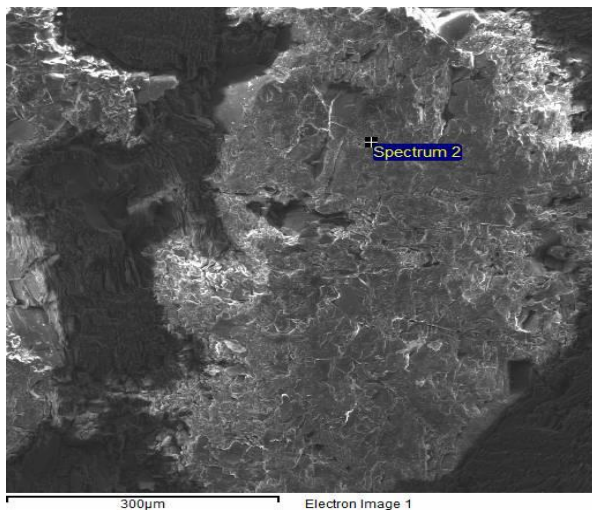


Fig. 11. SEM image of muscovite

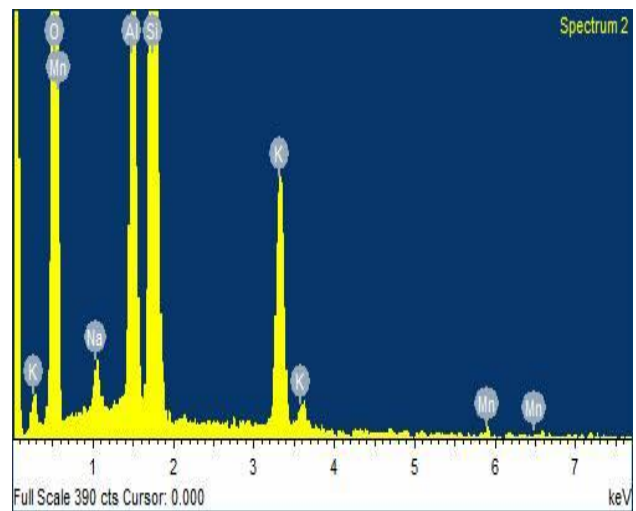


Fig. 12. EDS spectrum of muscovite

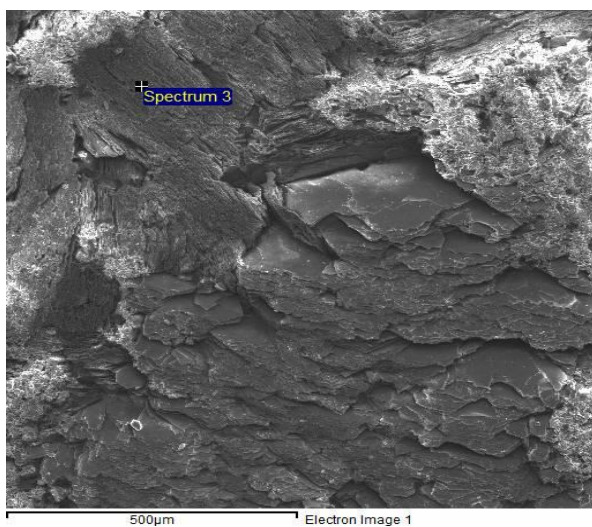


Fig. 13. SEM image of biotite

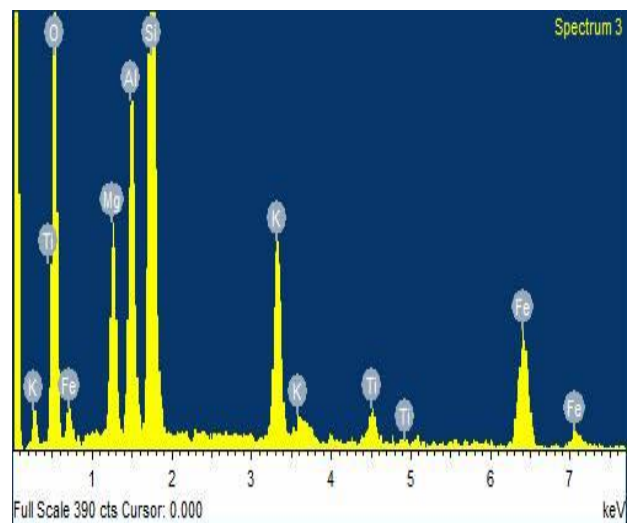


Fig. 14. EDX spectrum of biotite

Table 3

Chemical composition of muscovite and biotite from the Kosovska River locality (%)

Element (K)	Muscovite		Element (K)	Biotite	
	Weight	Atomic		Weight	Atomic
O	64.13	76.45	O	53.91	69.98
Na	1.1	0.91	Mg	6.28	5.37
Al	7.63	5.40	Al	7.64	5.88
Si	21.2	14.39	Si	15.64	11.57
K	5.55	2.71	K	6.15	3.27
Mn	0.39	0.14	Ti	1.36	0.59
Total	100.00		Fe	9.02	3.34
			Total	100.00	

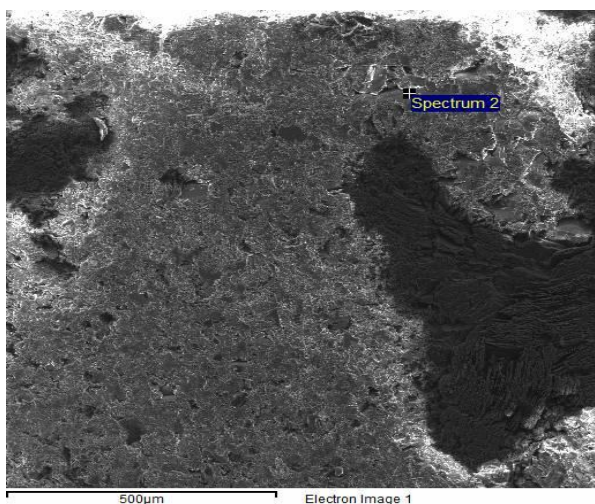


Fig. 15. SEM image of quartz, sample 1

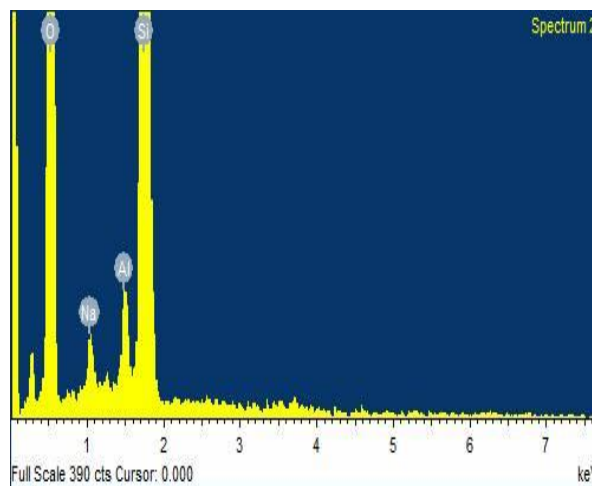


Fig. 16. EDX spectrum of quartz, sample 1

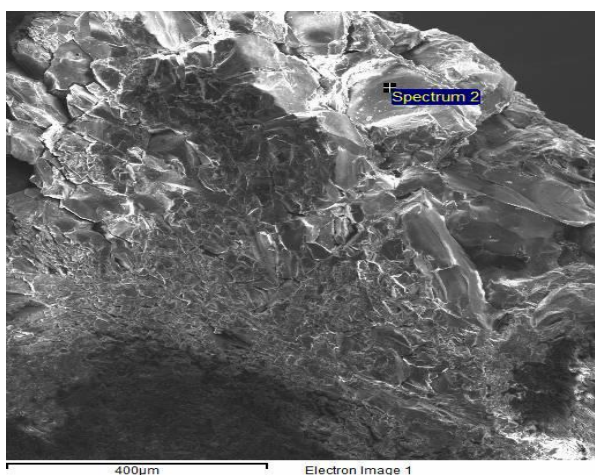


Fig. 17. SEM image of quartz, sample 2

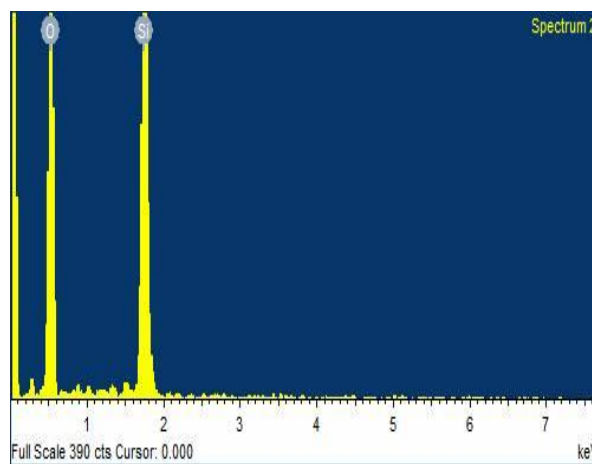


Fig. 18. EDX spectrum of quartz, sample 2

Table 4

Chemical composition of quartz from the Kosovska River locality (%)

Element (K)	Sample 1		Element (K)	Sample 2	
	Weight	Atomic		Weight	Atomic
O	67.57	78.39	O	73.89	83.24
Na	1.04	0.84	Si	26.11	16.76
Al	1.03	0.71	Total	100.00	
Si	30.36	20.06			
Total	100.00				

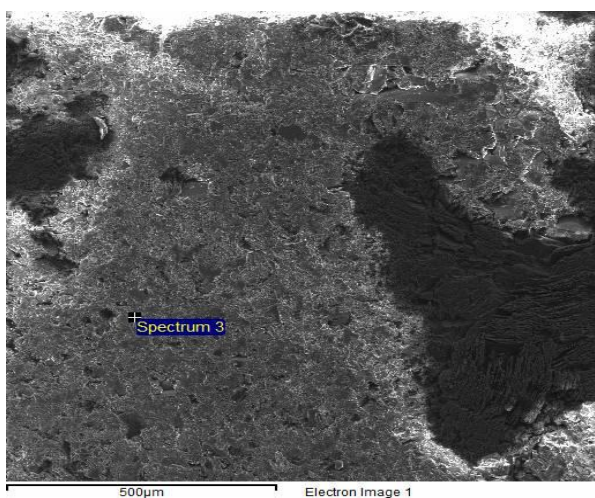


Fig. 19. SEM image of epidote, sample 1

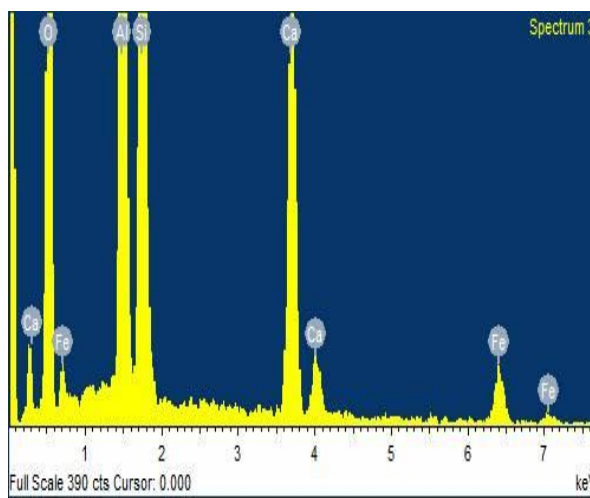


Fig. 20. EDX spectrum of epidote, sample 1



Fig. 21. SEM image of epidote, sample 2

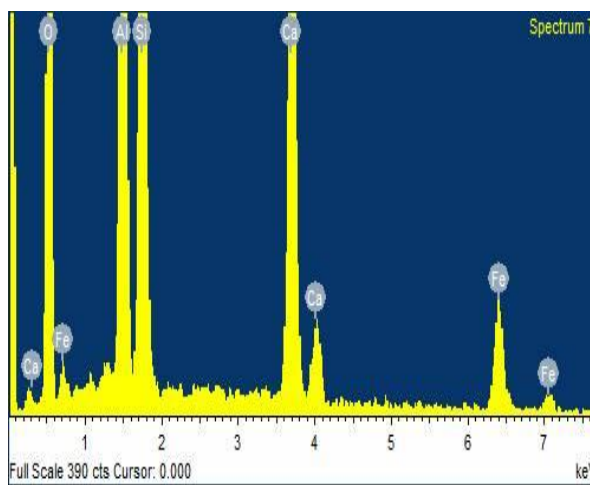


Fig. 22. EDX spectrum of epidote, sample 2

Table 5

Chemical composition of epidote from the Kosovska River locality (%)

Element (K)	Epidote sample 1		Element (K)	Epidote sample 2	
	Weight	Atomic		Weight	Atomic
O	58.84	73.64	O	53.22	69.72
Al	12.11	8.98	Al	11.76	9.14
Si	15.56	11.09	Si	16.50	12.31
Ca	10.27	5.13	Ca	12.75	6.67
Fe	3.22	1.16	Fe	5.77	2.16
Total	100.00		Total	100.00	

CHEMICAL ANALYSES OF ROCKS

The chemical analyses of the granodiorites from the Kosovska River locality, represent a contribution into the broadening of the knowledge for this massive on the territory of the Republic of North Macedonia. Granodiorites characterized with homogeneous – solid to compact texture, which locally turns to porphyroidal. With such arrangement and intergrowth of the mineral components, beige to greenish basic colour spotted with biotite of black colour is formed in the mineral aggregate. For more detailed presentation of the chemical composition of the granodiorites from the Kosovska River locality, four representative samples were taken from the granodiorites. The chemical composition of the analyzed samples is presented in Table 6.

From the chemical analyses (Table 6), it can be stated that the analyzed samples are characterized by a constant chemical composition, which can be seen in the content of SiO₂ (65.71–69.14% SiO₂).

In the granodiorites from the Kosovska River locality the content of P₂O₅ is low and varies in close interval, which is from 0.2 to 0.8 %. The content of the total iron (FeO*) systematically decreases with the increase of SiO₂ and is between 2.39 and 3.92%.

The content of MgO in granitoides from the Kosovska River locality ranges from 0.07 to 0.08%, content of CaO was from 3.41 to 4.35%, while the content of MnO is within the range of 0.69 to 1.27%. The content of Na₂O is from 3.73 to 4.82%, while the content of K₂O ranges from 2.79 to 4.85%, and the content of TiO₂ is from 0.42 to 0.60%.

The variation of the mass participation of the oxides of the major elements in function of the mass participation of SiO₂ is shown of the Harker's diagrams (Figure 23). Correlational trends in Harker's diagrams point on different magmatic processes that

happen in the series of the connected magmas (Prohić, 1998). The geochemical connection between the main elements is used to point on if there are one or more basic processes that will explain the connection between the main elements (Rollinson, 1993).

The variation of the main elements, compared with the increase of the content of SiO₂, indicates that the crystal's functioning is a very important process in the control of the chemistry of the granitoid rocks. However, the geochemical variations of the elements in shreds with the SiO₂ indicate the conclusion that the rocks are incurred during the combined processes. The chemistry of the rock affects the processes of alteration in different degree.

The correlation ratio between the main elements and SiO₂ in the investigated samples of the Kosovska River locality is shown on the variation diagrams in Figure 23.

In Figure 23, a very slight correlation can be noticed between the SiO₂ and Al₂O₃. With the increase of SiO₂ content, the Al₂O₃ decreased. The correlation ratio between SiO₂ and FeO, shown in Figure 23, proves that with increase of SiO₂, the content of FeO decreases. Also, it can be observed that, as the content of the SiO₂ increases, the content of MnO decreases. The correlation ratio between SiO₂ and MgO is shown as well, where an increase of the SiO₂ content is associated with an increase of the MgO content. The correlation between SiO₂ and CaO, shown in Figure 23, confirms that the CaO decreases with an increase of SiO₂ content. The correlation ratio between SiO₂ and Na₂O is shown in Figure 23. There is an increase of SiO₂, which resulted in the decrease of Na₂O. Also, an increase of the SiO₂ results in the decrease of K₂O, which is repeated for SiO₂ and TiO₂ as well as for SiO₂ and P₂O₅ (Figure 23).

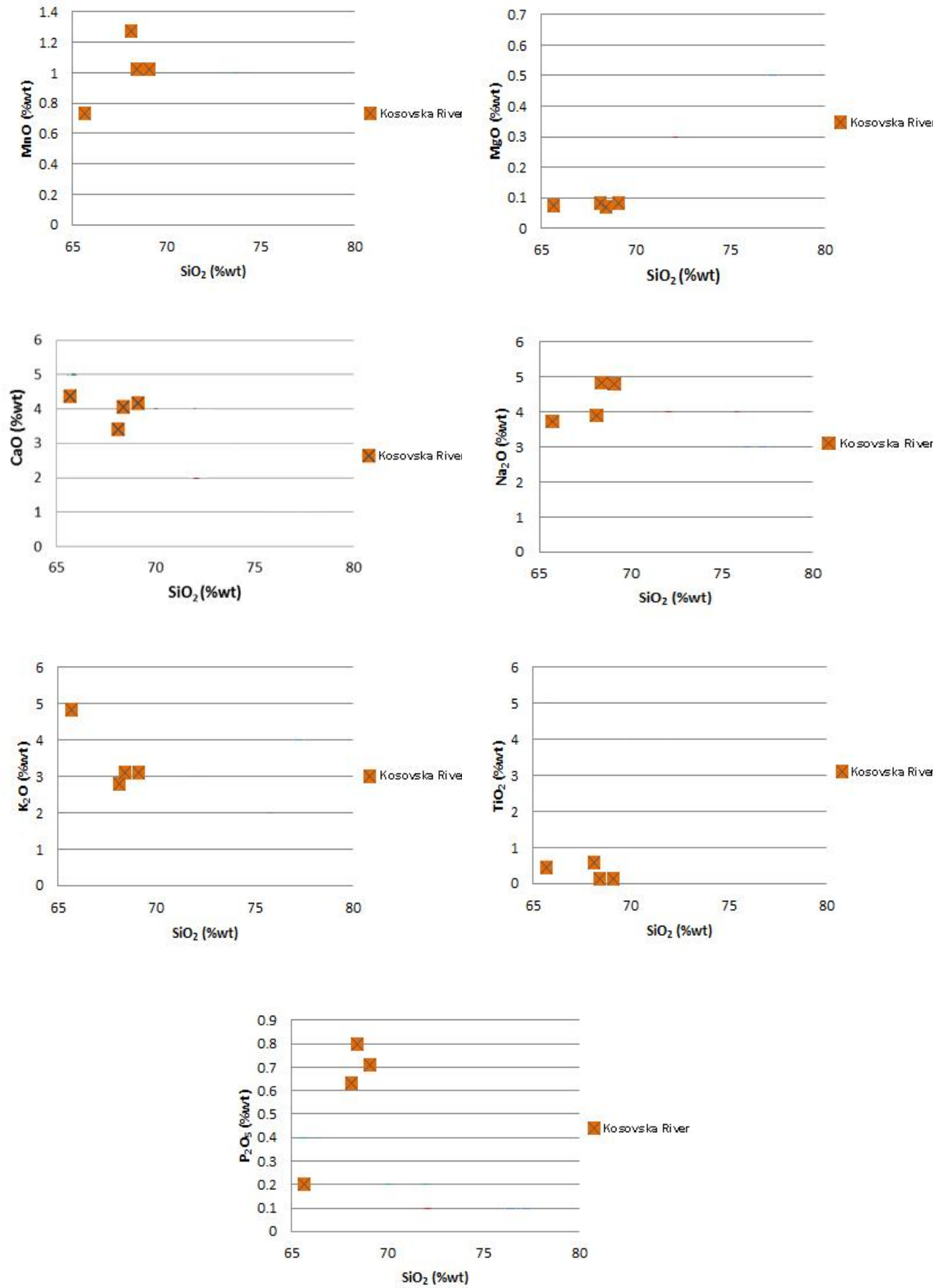


Fig. 23. Variation diagrams

Table 6

Chemical composition of the granodiorites from the Kosovska River locality (%)

Components	Ks-1	Ks-2	Ks-3	Ks-4
SiO ₂	65.71	68.14	68.42	69.14
Al ₂ O ₃	16.37	14.60	12.94	12.80
FeO	2.39	3.92	3.20	3.10
MnO	0.69	1.27	1.18	1.18
MgO	0.07	0.08	0.07	0.08
CaO	4.35	3.41	4.05	4.15
Na ₂ O	3.73	3.89	4.82	4.79
K ₂ O	4.85	2.79	3.10	3.11
TiO ₂	0.43	0.60	0.44	0.42
P ₂ O ₅	0.20	0.63	0.80	0.71
LOI	1.21	0.67	0.98	0.52
Total	100.00	100.00	100.00	100.00

Note: FeO in total Fe, Ks = Kosovska River sample 1 – 4,

CONCLUSION

Based on the structural-textural characteristics and mineralogical composition, there are two basic varieties within the granodiorites, including porphyritic and massive to coarse granular granodiorites. Based on the microscopic observations, significant varieties within the mineralogical composition of the granodiorites are observed, especially within the massive type.

The chemical characteristics of the granodiorite rocks from the Kosovska River locality, represent a contribution to the broadening of the knowledge for this massifs on the territory of North Macedonia. This massif is evidently different from the

surrounding rocks by its content, structural-tectonic features, colour and the manner of its origin. Basically, granodiorite is characterized with homogeneous, solid to compact texture, which locally turns into porphyroidal.

The analyzed samples are characterized with a constant chemical content, which can be seen in the content of SiO₂, in range from 65.71 to 69.14%.

Approximately, based on the microscopic observations was found the modal composition of rock: plagioclase 50%; orthoclase 13%; quartz 17%; biotite and secondary minerals 20%.

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Резиме

МИНЕРАЛОШКИ, ПЕТРОГРАФСКИ И ХЕМИСКИ СОСТАВ НА ГРАНИТОИДНИТЕ КАРПИ ОД ЛОКАЛИТЕТОТ КОСОВСКА РЕКА, ЗАПАДНА МАКЕДОНИЈА

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Клучни зборови: гранодиорит; минералoшки состав; главни минерали; состав на минералите, минерална асоцијација.

Во трудот се прикажани резултатите од минералошко-петрографските и хемиските испитувања на гранитоидните карпи од локалитетот Kosovska Reka. Минералошко-петрографските карактеристики на земените примероци од карпите се одредувани со поларизационен оптички микроскоп Leitz Vetzlar, Germany. Минералошкиот состав е одредуван и со методот на рендгенска дифракција на

примероци во прав (XRD) и со помош на сканирачки електронски микроскоп. Врз основа на теренските и лабораториските испитувања на земените примероци од гранитоидните карпи од локалитетот Kosovska Reka, се утврдени следните типови карпи: крупнозрнест гранодиорит и порфириоден гранодиорит.