MINERALS FROM THE REPUBLIC OF MACEDONIA
WITH AN INTRODUCTION TO MINERALOGY

by

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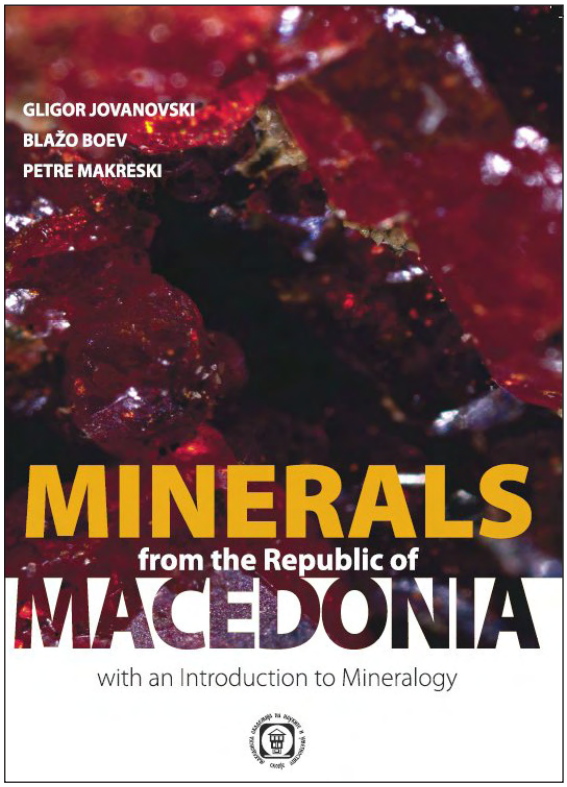
with continued contributions from Branko Kaitner, Trajče Stafilov and Bojan Šoptrajanov



The reviewed monograph is the first extended work in which data on the minerals of the Republic of Macedonia have been described. This book is written in English to make the information on the mineralogy of the Republic of Macedonia accessible to a worldwide audience. The monograph contains a general introduction to mineralogy, which makes it useful for people starting their education in this science, i.e. students of geology, amateur mineralogists and mineral collectors in the Republic of Macedonia. Thus, the monograph is a dual-purpose book.

The first part of the book is an introduction to mineralogy with the necessary connections to the related sciences of physics, chemistry and crystallography. The starting point is the keystone: what a mineral is, what mineralogy is and how minerals are formed. Furthermore, we see material on the crystallographic basis of mineralogy and a chapter devoted to the crystalline state of substances. The next point is on the chemistry of minerals. This significant chapter explains modern conceptions on the constitution of solids and the types of chemical bonds. An important part of the basis of mineralogy is the theory of isomorphism and solid solutions, which is also reflected in the monograph. The physical properties of minerals and an introduction to optical mineralogy are found in the next chapter. A long chapter is devoted to modern methods of mineralogical research, including physical assays such as X-ray diffraction and various spectroscopic methods as well as chemical analysis. The final chapter of the first part of the book presents the principles of the classification of minerals and some examples.

The second main part of the monograph contains collected information on the minerals of the Republic of Macedonia and their localities. A chapter is devoted to the general geological characteristics of the Republic of Macedonia. The four major geotectonic units of the territory, namely the West-Macedonian zone, the Pelagonian massif, the Vardar zone and the Serbo-Macedonian massif are described, focusing on mineragenetic aspects.

The next chapter seems especially valuable, as it includes a description of the most interesting mineral localities of the Republic of Macedonia (with a geographical map showing their distribution). This information was almost unknown to the worldwide community of both professional and amateur mineralogists and the monograph mainly fills in this blank. Among others, the descriptions of localities of global mineralogical significance are given. First, there is the famous Allchar, an unusual low-temperature hydrothermal Au-Sb-As deposit with incredibly rich and diverse thallium mineralization, a source of many new minerals and the location of a unique project on solar neutrinos. Alinci is a very

interesting location, as it is a group of alkali-syenite related occurrences: a new mineral, macedonite, or $PbTiO_3$, was first discovered here and several rare species have been found. Nežilovo, a belt of metamorphic and metasomatic rocks with unusual mineralogy and geochemistry, similar to the famous Långban in Sweden, is also described, focusing on the distribution of exotic complex oxygen compounds with Pb, Ba, Zn, Mn, Sb, Ti, As, *etc*., with endogene origin. Other outstanding objects characterized in the book are an ultrabasic complex with nickel deposits at Ržanovo, a polymetallic deposit related to manganese-rich skarns at Sasa, as well as the occurrence of nice, big crystals of pink corundum and even ruby in the marbles at Sivec. From the monograph, we can learn about remarkable finds of giant crystals at localities in the Republic of Macedonia such as epidote (up to 1 m), titanite, gypsum, *etc*.

The next chapter comprises of mineral descriptions where all mineral species and varieties known in the Republic of Macedonia are listed. The minerals are divided in two groups: the first, much larger group, is related to the studied and characterised minerals by the authors of the monograph, whereas the second group contains information concerning the non-collected minerals (in the monograph termed as *other minerals*). For each mineral, general data (chemical formula, symmetry, physical properties, occurrence, origin of the name, *etc.*) and information on its distribution in the Republic of Macedonia are given. Furthermore, for the studied minerals, quantitative chemical composition analysis, X-ray powder diffraction data with determined unit cell parameters, optical data and infrared and/or Raman spectrum are given. These characteristics were obtained from mineral samples collected in the Republic of Macedonia.

The last chapter includes a list of publications of the authors on the mineralogy of the Republic of Macedonia and indices of minerals and localities mentioned in the monograph. It is necessary to note that each chapter of the book contains a detailed list of references that is valuable in itself.

An important part of the monograph is the illustrations. Besides common figures necessary to describe basic concepts (especially in the first part, the introduction to mineralogy), we can enjoy many color photographs of minerals from the Republic of Macedonia, including nice full-page pictures. This seems valuable because the mineral specimens from this country are not well-known and not widespread in foreign museums and private collections, and readers will acquire considerable new information and impressions from these pictures. The high printing quality of the book merits praise.

Thus, I can recommend this monograph to a wide audience, from professional mineralogists and geologists to people who are taking their first steps in this field.

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Publisher: Macedonian Academy of Sciences and Arts, Skopje, Macedonia

Co-publisher: Skenpoint, Skopje, Macedonia

http://mineralsfrommacedonia.blogspot.mk/

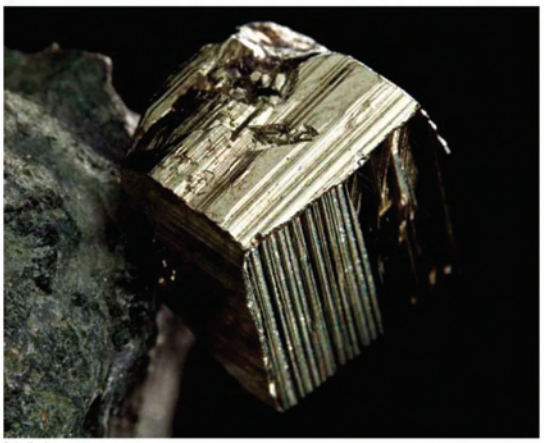
CONTENTS

1.3. Crystalline State and the Chemical Composition of Minerals

Both (primary and secondary) mineral formation processes result in the creation of various crystal forms with perfect geometrical shapes and beautiful colours that have continuously fascinated people throughout human history. Before the processes of mineral formation were scientifically considered and understood, various religious myths regarding minerals were created based on their apparent magical powers. The profound scientific consideration of the Earth and consequently of its rocks and crystal mineral forms started about 200 years ago [1].

The term **crystal** originates from the Greek word *κρυστάλλος* (*krystallos*), meaning *clear ice* [2, 8]. It has been shown that crystals can be formed by almost any solid substance. The word *crystal* is applied to the appropriate mineral sample of a regular shape surrounded by various flat surfaces with different forms [8].

As mentioned, minerals have definite but not necessarily fixed chemical compositions. This means that any mineral has its own chemical formula [3, 4]. For instance, the definite and fixed chemical formula of the mineral pyrite, FeS_2, means that it is



Pyrite (Sasa), 14×14×13.

Fluorite (Sasa), NMHH collection.

1.5.3. Chemical Bonds

In general, there are three main ways of linking or bonding of the atoms where the valence electrons are involved. These are known as: *ionic bond*, *covalent bond* and *metallic bond*. Other types of bonds where no valence electrons are included are *hydrogen bond* and *van der Waals bonds*. The latter two types are so-called interactions between closed-shell molecules.

Ionic Bond

The commonest type of bonding in minerals is the ionic bond. In its simplest form, it is formed when one of the atoms loses one or more electrons, thereby becoming a cation (A^{1+}, M^{2+}, etc.), whereas the second one accepts the corresponding number of electrons and transforms into an anion (A^{-}, A^{2-}, etc.). The resulting ions cohere by the electrostatic Coulombic interaction [3, 8, 14]. Therefore, both constituents of the ionic bond tend to acquire the electron configuration 8 in their outermost shell (characteristic configuration for the energetically most stable elements – noble gasses). In mineral halite ($NaCl$), for example, the outermost electron of the sodium atom ($3s^1$) also transfers to the outermost shell of the chlorine atom ($3p^5$). The result of this transfer is the formation of the sodium cation Na^+ with an electron configuration of neon (Ne) and the chlorine anion Cl^- with an electron configuration of argon (Ar). These two ion types with stable electron configurations but opposite charges attract each other and form ionic crystals of $NaCl$. This interionic attraction is balanced by a corresponding repulsion, keeping the ions at the so-called *equilibrium distance*. The product of this bonding is the formation of a *chemical compound* represented by a *chemical formula* (e.g. $NaCl$).

In addition to the elements that form ions with equal but opposite valency, the ionic bond can be also formed between the elements forming ions (including complex ions) with different oxidation states (e.g. Mg^{2+} and $2Cl^-$ in $MgCl_2$; $2Mg^{2+}$ and SiO_4^{4-} in Mg_2SiO_4). It is evident that the ions bond in a ratio that ensures a balance between the positive and negative charges, thereby forming an electrically neutral crystalline solid. The ionic bonding results in the formation of various types of surroundings of cations by anions and vice versa. This number of nearest neighbours of opposite charges is called the coordination number (CN) and depends on the corresponding ionic radii.

For example, the CN for Na^+ and Cl^- in the ionic structure of $NaCl$ is 6, which means that each sodium cation is surrounded by six chlorine anions and vice versa. Depending on the particular CN value, various types of so-called coordination polyhedra are

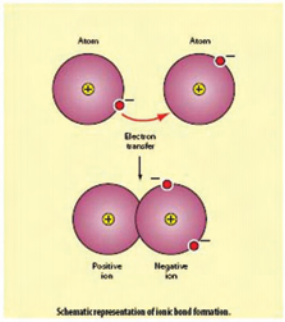
formed, the most common being triangle (CN = 3) (see Table 4), tetrahedron (CN = 4), octahedron (CN = 6) and hexahedron or cube (CN = 8). Their packing together builds up the crystal of the corresponding mineral [8]. The cation/anion radius ratio for the most common regular coordination polyhedra are given in Table 4 [3].

Table 4. The cation/anion radius ratio for the most common regular coordination polyhedra

CN	Polyhedron	Cation/anion radius ratio
2	Line*	< 0.155
3	Triangle*	0.155–0.225
4	Tetrahedron	0.225–0.414
6	Octahedron	0.414–0.732
8	Cube	0.732–1
12	Cubo-octahedron	~1

*"Line" and "triangle" are in fact tritious polyhedra that refer to the linear and triangular coordination of the central atom by two and three atoms, respectively.

In general, the ionic solids (minerals) are brittle, hard and have high melting temperatures.

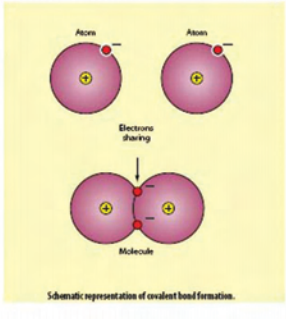


Schematic representation of ionic bond formation.

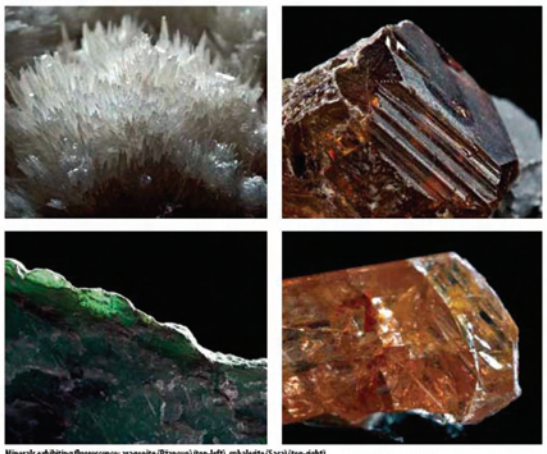
Covalent Bond

Electron pair sharing between the atoms is the main characteristic of the covalent bond. The equal sharing of the electron pair results in the formation of the so-called *non-polar* covalent bond between the analogue atoms (as in H_2, N_2, O_2, Cl_2, etc.). By contrast, the unequal distribution of the electron cloud along the bond is typical for forming a *polar* covalent bond between the various atoms (as in CO, HCl, etc.) [14]. The molecules with polar covalent bond possess an *electric dipole moment*. Similar to the ionic bond, the formation of the stable noble gas electron configuration 8 in outermost shells is also particular for the electron pair sharing atoms coming close to each other and forming a covalent bond [3, 8].

The strength of the covalent bond depends on the degree of the overlapping of the adjacent atom orbitals. In general, three different types of molecular bonds can be obtained as a result of the overlapping of various atomic orbitals: sigma bond, σ; pi bond, π; and delta bond, δ. To explain the geometric arrangement of various structural constituents in the substances (minerals) where the covalently bonded atoms appear, the so-called *hybrid orbitals* are introduced.



Schematic representation of covalent bond formation.



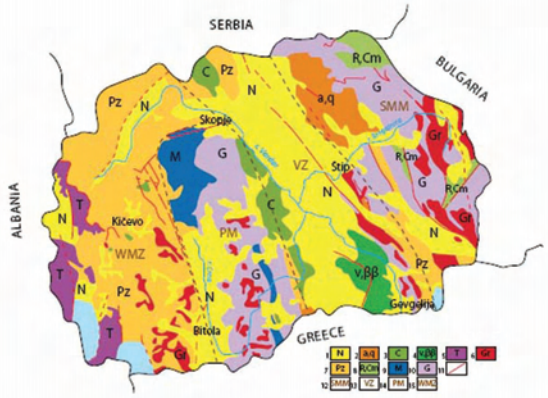
Minerals exhibiting fluorescence: aragonite (Rzanovo) (top-left), sphalerite (Sasa) (top-right), talc (Rzanovo) (bottom-left) and zircon (Canlite) (bottom-right).

Minerals exhibiting phosphorescence: calcite (Mrzen) (left) and fluorite (Sivec) (right).

Chalcanthite (Bučim) (previous page).

2.1. Geological Characteristics of the Republic of Macedonia

In general, four geotectonic regions (units) are present on the territory of the Republic of Macedonia: West-Macedonian zone (WMZ); Pelagonian massif (PM); Vardar zone (VZ); Serbo-Macedonian massif (SMM) [1–17].



The geological map of the Republic of Macedonia: (1) Neogene; (2) Volcanics; (3) Cretaceous; (4) Gabbros-diabases; (5) Triassic; (6) Granitoids; (7) Palaeozoic; (8) Riphean-Cambrian; (9) Marbles; (10) Gneisses; (11) Faults; (12) SMM; (13) VZ; (14) PM; (15) WMZ.

Quartz (Sasa) (previous page).

2.2. Deposits of the Collected Minerals of the Republic of Macedonia

The mineral localities within the territory of R. Macedonia are given in the map below.

Map of the deposits in the Republic of Macedonia where the minerals were collected from: (1) Alinci, (2) Alšar, (3) Bebrtče, (4) Beliište, (5) Bogoslovec, (6) Bonče, (7) Brest, (8) Bučim, (9) Budinarci, (10) Bukovik, (11) Crni Vrv, (12) Čanište, (13) Češinovo, (14) Čemovo, (15) Dumjan, (16) Debar, (17) Drenovci, (18) Dunje, (19) Gimovi, (20) Ilovica, (21) Kobilino Pole, (22) Kokre, (23) Kočino, (24) Kozjak, (25) Krastov Kamen, (26) Krstov Dol, (27) Lagovo, (28) Lojane, (29) Mrežičko, (30) Mzren, (31) Nebregovo, (32) Negilovo, (33) Nistrovo, (34) Niževpole, (35) Orsojnica, (36) Pčinja, (37) Pehčevo, (38) Pekliište, (39) Pelagon, (40) Petrova Reka, (41) Plavica, (42) Plešenci, (43) Prilepec, (44) Rabrovo, (45) Radnja, (46) Ramna Niva, (47) Rzanovo, (48) Sasa, (49) Sazdevo, (50) Selečka Planina, (51) Sivec, (52) Stavo Bonče, (53) Streški, (54) Sopur, (55) Stavica, (56) Tajmište, (57) Toranica, (58) Trojaci, (59) Vasev Grad, (60) Veprčani, (61) Veselčani, (62) Vitolište, (63) Vodno, (64) Vrbske, (65) Zletovo, (66) Zvegor.

Quartz (Budinarci), 92×74×67 (next page).

2.2.9. Budinarci

The village of Budinarci is situated about 12 km north-west of Berovo. The surrounding rocks include micaschists, gneisses and granites. Very wide granite and quartz veins have been found there [2]. Gneisses are irregularly transected by pegmatite quartz veins of variable sizes. The larger pegmatite veins are made up of quartz, which appears in the form of beautiful crystal aggregates or individual crystals.

Quartz (Budinarci), 80×53×32, MMRK collection.

Fluorite (Sivec), 5×5×3.

Augen two-mica amygdaloidal gneisses are connected to the contact parts of the granitoid intrusions and gradually grade into two-mica biotite-muscovite gneisses.

In the lower parts, the gneisses are massive and stratified pseudolayered and schistose owing to the presence of mica. On the surface, they erode into various irregular, oval shapes or sporadically there are 0.5 m thick gneiss arenite. Gneisses are light red, so the portions that contain more quartz and feldspar-albite are brighter and those with more mica are grey-greenish. As a rule, at the lower parts of gneisses (presented as biotite muscovite gneisses) microcline prevails, whereas in the upper parts albite and microcline are the most abundant. Porphyroblastic gneisses are closely related to the contact parts of the granites or the gneiss-granites in the valley of the Babuna river (at Teovoi). The thickness of the gneiss series in the northern part of the Pelagon is estimated to be between 5000 and 7000 m.

The series of micaschists is less widespread than the gneiss series. It was discovered at Plavenski Rid, Osoj and Silegarnik, Begovi Virovi and Kadino Pole, Ubava, north-west of the Kadina Reka basin and mt. Kitka. To the east of Prilep they were distinguished in mt. Prisoj and mt. Vicrila as well as other places. As a whole, the series of micaschists is present as micaschists, quartzites and graphite schists, which facially grade one into another, both laterally and vertically. Unlike gneisses, micaschists possess a clearly pronounced dark brown to dark schistose texture depending on the presence of the graphite material. In addition to quartz, micaschists also contain micas (muscovite, biotite), chlorites and garnets (of up to 30% in some parts), kyanite, staurolite, albite, amphibole and tourmaline. There are also garnet micaschists rich in garnet with crystals reaching several centimetres in size. They are abundant in mt. Vicrila, north of Kozjak. Large kyanite crystals of 10 cm have been found in the micaschists in this area.

The mixed series in the northern part of the Pelagon occupies a sequence of metamorphic rocks of variable petrographic compositions: augen schists, feldspathic-chlorite schists, clipolines, talc schists and marbles where rocks grade one into another both laterally and vertically. Gneisses (mainly albite) prevail (especially in the areas of Markova Reka, Kitka, Ruen and the village of Izvori among all lithological varieties, whereas the presence of metarhyolites is also evidenced.

Fluorite, CaF_2

Fluorite (Sivec), 32×31×29.

Name origin:	After its composition containing fluorine. Latin *fluere* means to flow
Colour:	White, yellow, green, red, blue
Hardness:	4
Density:	3.13 (exp.); 3.18 (calc.)
Cleavage:	{111} perfect
Optical characteristics:	Isotropic, $n = 1.433$
Localities in the R. Macedonia:	Sivec

Fluorite (Sasa), 197×116×111, MNHN collection (next page).

Fluorite most commonly forms cubes, and rarely octahedra and rhombododecahedra. It can also occur in granular, compact and massive habits. It is one of the minerals with the largest varieties of colours, ranging from white to black (purple, green, yellow, pink, red, blue, brown, colourless). Fluorite is transparent to transluscent with a vitreous lustre and white streak. It occurs in hydrothermal mineral veins. Fluorite is associated with quartz, calcite, dolomite, barite, galena, sphalerite, gypsum, tourmaline and other minerals. This mineral is the main source of fluorine used in the chemical industry. Fluorite is also used in steelmaking and other metallurgical processes, in special optics and as an additive in toothpastes.

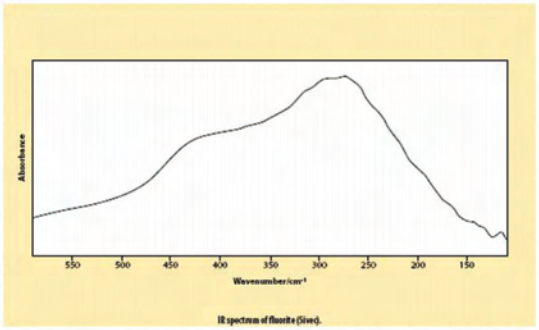
Fluorite on dolomite (Sivec), 20×14×12.

Fluorite is not a common mineral in the territory of the Republic of Macedonia. It has been identified in the dolomite marbles at the Sivec deposit. It occurs in the form of the nice cubic monocrystals of pinkish-bluish colour. Sometimes twinned crystals occur. Most frequently it is associated with quartz, calcite and dolomite.

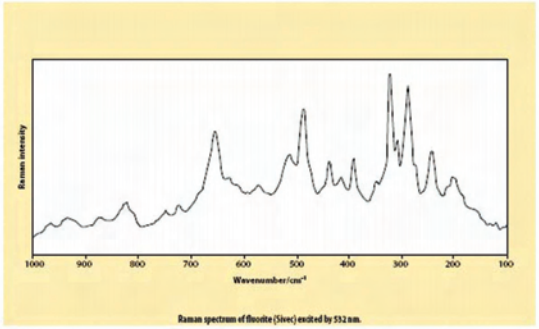
X-ray microprobe chemical analysis of fluorite (Sivec)

Element or element oxides	Content/%			
	Point 1	Point 2	Point 3	Theoretical
Al_2O_3	0.18	<0.01	<0.01	
SiO_2	<0.01	<0.01	<0.01	
CaO	70.89	71.02	72.15	71.83
F*	49.06	49.18	48.44	48.67
FeO	0.09	0.07	<0.01	
Cl	0.09	<0.01	<0.01	
Total	99.66	99.57	100.20	100

*Fluorine content is expressed as oxygen (F) to adjust the total content to 100%.



IR spectrum of fluorite (Sivec).



Raman spectrum of fluorite (Sivec) excited by 532 nm.

XRPD data for fluorite (Sivec)

h	k	l	d_{obs}	d_{cal}	d_{obs}
1	1	1	3.1480	3.1565	-0.0085
2	2	0	1.9331	1.9329	0.0002
3	1	1	1.6486	1.6484	0.0002
4	0	0	1.3671	1.3668	0.0003
3	3	1	1.2544	1.2542	0.0002
4	2	2	1.1161	1.1160	0.0001

Unit Cell Par.[obs]
$a = 5.4630$ Å
$V = 163.035$ Å³

Unit Cell Par.[a]
$a = 5.4639$ Å
$V = 163.12$ Å³
$Z = 4$
Cubic ($Fm3m$)

[a] S. Speziale, T. S. Duffy, Single-crystal Elastic Constants of Fluorite (CaF_2) to 9.3 GPa, *Physics and Chemistry of Minerals*, 29, 465–472 (2002).

Fluorite on dolomite (Sivec).

Fluorite (Sasa), MMNH collection.

2.4. Published Papers by the Members of the Research Group Regarding the Studies of Minerals from the Republic of Macedonia

1987

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1994

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