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PHYSICAL PROPERTIES AND TEMPERATURES OF CRYSTALLIZATION OF COEXISTING GARNETS AND BIOTITES FROM PARAGNEISSES OF THE LITTLE CARPATHIANS

(Fig. 1—3, Tab 1—4)



Abstract: The evaluation of measured and theoretical physical properties of biotites ($V_m = 148,01 - 150,66 \text{ cm}^3/\text{mol}$; $V = 491,50 - 500,29 \times 10^{-10} \text{ m}^3$; $n_z = 1,633 - 1,645$; $\rho = 2,91 - 3,09 \text{ g/cm}^3$) and coexisting garnets ($V_m = 115,61 - 116,39 \text{ cm}^3/\text{mol}$; $a_o = 11,560 \times 10^{-10} \text{ m}$; $n = 1,804 - 1,810$; $\rho = 4,15 - 4,19$) has made possible indication of recrystallization temperature of these minerals. These temperatures were compared with temperatures determined thermodynamically ($450 - 610^\circ\text{C}$).

Резюме: Оценка измеренных и теоретических физических свойств биотитов (V_m , V , n_z , ρ) и сосуществующих гранатов (V_m , a_o , n , ρ) представило возможность индикации температуры рекристаллизации этих минералов, что и было сравнено с температурами определенными термодинамическим путем ($450 - 610^\circ\text{C}$).

Introduction

The occurrence of index minerals in metamorphosed rocks is often used for determination of the degree of recrystallization alteration of rocks. Besides the methods determining the intensity of metamorphism on the basis of mineralogical valuation of paragenesis, the investigation of metamorphism of valid geothermal and geobarometric data leads to reconstruction from thermodynamical standpoint comes more to the foreground. Treatment of temperature and depth conditions of the recrystallization process. Approximation of temperature and pressure isolines is obtained in the studied geotectonic area, making possible to trace the thermal field of the area, immediately linked with deep structures. These lines also determine erosional downcutting of tectonic structures and indicate the position of their axes. For determination of thermodynamic parameters of metamorphism those minerals are suitable, in which solid solutions play part and easily reach the equilibrium conditions. The form of mineral occurrence permits to decide whether it is possible to understand the total mineral association as equilibrium on the basis of microscopic structural properties of rocks.

With equilibrium distribution of elements among coexisting phases, which undergoes Nernst's distribution law, continual change of chemical composition of paragenesis forming mineral takes place with changes of the grade of metamorphism.

The distribution of Mg^{2+} and Fe^{2+} in garnet and biotite, garnet and staurolite, biotite and cordierite..... is mostly elaborated from these viewpoints. The isomorphism of these elements in garnet and biotite causes at the same time the change of their physical properties.

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Refraction properties of magnesium and iron are very different, ($R_{Mg} = 0,36$; $R_{Fe^{2+}} = 2,29$; $R_{Fe^{+3}} = 5,90$) and therefore in the work it is set out from the assumption that the mutual substitution of these elements will have a different contribution to summary refraction of the matter. Individual Mg and Fe concentrations ratios are at given temperature indicated with distribution coefficient (K_D). The investigation of relation between temperature and refractive index, density, volume of unit cell and molar volume of co-existing garnets and biotites was carried out with comparing of the theoretical and measured physical properties of these minerals.

Methodical approach

The choice of the working method was preceded by microscopical study of metapelitic rocks of the Little Carpathians. The garnets are found in the studied rocks mostly as idiomorphic porphyroblasts without inclusions, phenomena of rotation and microscopical inhomogeneities. Garnets equally as biotites are a product of the post-tectonic recrystallization stage of selected samples.

The analysed material was taken from a piece of rock, the second part of which was available for chemical analyses and thin section material. After crushing of rock in the jaw mill (2 kg), sieving and decantation, heavy mineral fractions of rocks were obtained. As dividing liquid bromoform was used. The hand-separated amount of minerals was sufficient for preparation of samples for X-ray diffraction, electron microanalysis, determination of refractive indices and density.

Observation of garnet and biotite with separation permits to conclude that the analysis can be rendered more difficult also with these mistakes. The concentrates usually contain impurities (mainly samples from lower metamorphic degree, which are „badly“ crystallized and contain inclusions), which are difficult to remove by consequential manual separation. The mistakes are also resulting from the presence of grains of different chemical composition (e.g. two generations of mineral, zonality of mineral alternation...). In these cases, indicating chemical unequilibrium, the analyses of mineral are of no significance for interpretation. Removal of these doubts should be preceded by detailed study by microscopical structural and macroscopic features of the rock or by detailed observation of the chosen physical property of minerals.

The lattice parameter of garnets (a) was determined by the method of X-ray diffraction on powder of sample with application of CuK_α radiation. Tracing of slow records was performed with evaluation of diffraction intensities in the range $6 - 60^\circ 2\theta$.

For determination of parameters of biotite unit cells (Tab. 1) the methods of powder diffraction were applied. In the first approach θ and „d“ values were generated, compared with the measured values. The lines were indexed on the basis of the assumption that biotite of samples is polytype 1M with symmetry $C2/m$. The number of reflexions for getting more precise lattice parameters varied 7–10. They were the following reflexions: 001, 020, 110, 112, 003, 131, 023, 004, 132, 331.

Table 1
Unit cell parameters of biotites from Little Carpathian paragneisses*

	a	b	c	β	V
6.	5,346(2)	9,224(9)	10,248(9)	100,24	497,37
7.	5,334(3)	9,288(8)	10,240(8)	99,85	499,88
8.	5,349(3)	9,206(7)	10,277(9)	101,29	496,32
9.	5,331(8)	9,172(8)	10,243(5)	101,14	491,50
11.	5,343(2)	9,242(9)	10,247(9)	100,19	498,11
12.	5,325(2)	9,282(7)	10,185(9)	98,90	497,42
14.	5,349(2)	9,238(9)	10,267(9)	100,50	498,96
17.	5,339(2)	9,255(8)	10,244(9)	99,57	499,23
18.	5,345(2)	9,234(3)	10,223(8)	100,10	496,83
KB-1	5,337(3)	9,206(8)	10,191(9)	100,30	493,06
KB-2	5,335(1)	9,226(5)	10,238(7)	100,40	495,70
KB-3	5,342(2)	9,221(9)	10,217(9)	99,98	495,76
KB-4	5,342(2)	9,278(8)	10,234(9)	99,51	500,29
KB-5	5,337(2)	9,215(7)	10,264(9)	101,04	495,57

* In dimensions: $\times 10^{-10}\text{m}$.

The structural studies of natural and synthetic biotites have shown (G. Donay et al., 1964; R. Hazen — C. Burnham, 1973), that polytype *1M* is often found in biotites. On the other hand, it is not possible to distinguish polytypes *1M* and *3T* in routine way by powder diffraction, moreover, polytypism in natural biotites seems to be complex (M. Ross et al., 1966). For these reasons biotites were traced as polytype *1M*. With regard to this way of interpretation satisfactory accordance between the theoretical and measured values resulted. The chosen way also provides the possibility to compare the obtained data with values already published by other authors. (D. Wones, 1963; R. Crowley — R. Roy, 1964; M. Rutherford, 1973; D. Hewit — D. Wones, 1975), who also interpreted biotite with polytypism *1M*.

Measurements of refractive indices were performed by immersion method. Garnets were measured in methyleniodide saturated with sulphur. Sodium light was used. Density values obtained by micropiknometry were reproducible in the range 0,01-0,02.

Results and discussion

In determination of physical properties I calculated first refractive index of biotite n_z on empirical basis. For this approach the value d_{331} was used, obtained by X-ray diffraction. With introduction of this value in the equation.

$$n_z = - 3,9002 + 3,5848 d_{331}$$

refractive indices n_z for the observed biotite were obtained. This equation resulted from treatment of values d_{331} and n_z for synthetic biotites, published by D. Hewit and D. Wones (1975).

Comparing these refractive indices with the measured ones [Tab. 2], no satisfactory accordance resulted. This follows from the difference of synthetic and natural biotites.

The extent of measured and calculated refractive indices is close with regard to all the biotite series. This fact reduces correlation between the measured and calculated value of refractive indices ($r = 0,36$).

There may be several causes of this discordance. The peak d_{-331} can be overlapped with other reflexions, which may cause unprecise location of the peak and incorrect assignment of the value θ .

Reflexion 060, which may have approximately half the intensity of peak d_{-331} , mostly contributes to the mistake from this standpoint. With reduced intensity and shift to the peak d_{-331} , the mentioned unexactness can so occur. The presence of elements Ti, Mn, Ca, Na, causing the shift of reflection position and not included in the system, studied by D. Hewit and D. Wones (1975), can also have disturbing effect on this relation.

The refractive index is of diagnostic importance in subdivision of the biotite series into the individual members. The density, however, does not occur as diagnostic feature. In the studied biotites relation between n and ρ has not been found, although in isostructural phases closer relationship between refractive index and density is supposed.

The causes of this low correlation are necessary to search in the presence of elements, having various contribution to refraction with nearly the same atomic weight. Although with their increasing contents Fe^{2+} and Fe^{3+} have close contribution to increasing density, their contribution to total refraction is different. Fe^{3+} has essentially greater atomic refraction than Fe^{2+} and influences more the refractive index value.

Substitution of Si by aluminium in tetrahedral positions, which may lead to 30 % substitution of silicon positions by aluminium in biotites, also is of considerable influence on the refractive index value. The larger Al ion radius, which substitutes Si in tetrahedral positions, lower the refractive index approximately by 2 %. It means, that with a little change of biotite density a change of the refractive index on the second decimal place occurs. Substitution of Fe^{2+} and Mg by aluminium in octahedral positions is also important. The refractive index in biotites is thus a complex function not only of chemical composition but also essentially influenced by distribution of atoms in individual structure positions.

The density (ρ), molar volume (V_m) and volume of elementary cell (V) of biotites, however, display equal complex dependence on chemical composition and structural properties as the refractive index. Mutual relations between these physical properties are summarized in Tab. 3.

In the studied samples of biotites no correlation between the refractive index and density, volume of unit cell and molar volume has been found. In garnets relation between the refractive index and molar volume; density and volume of unit cell; and density and molar volume has not been found.

The properties of garnets are usually delimited by endmembers pyrope, almandine, spessartine, grossular and andradite. In garnets (in simple case) ideal formation of solid solutions is considered. The physical properties of given member of the garnet group result then from the physical nature of the end-members.

Table 2
Physical properties of coexisting garnets and biotites from Little Carpathian paragneisses

	n	ρ	a_o	V_m	$n^{calc.}_z$	n_z	ρ	V	V_m	ToC
6.	1,809	4,17	11,549	115,97	1,638	1,634	2,97	497,37	149,78	540
7.	1,806	4,16	11,548	115,94	1,638	1,640	2,98	499,88	150,53	560
8.	1,810	4,19	11,544	115,82	1,636	1,639	3,00	496,32	149,46	460
9.	1,809	4,16	11,554	116,12	1,618	1,637	2,89	491,50	148,01	460
10.	1,808	4,18	11,549	115,97		1,637	2,96			520
11.	1,808	4,18	11,547	115,91	1,638	1,636	3,04	498,11	150,00	450
12.	1,804	4,15	11,539	115,67	1,630	1,643	3,00	497,42	149,79	610
14.	1,808	4,18	11,553	116,09	1,638	1,634	3,02	498,96	150,26	510
17.	1,807	4,17	11,537	115,61	1,637	1,633	2,98	499,23	150,34	510
18.	1,807	4,18	11,563	116,39	1,639	1,641	2,92	496,83	149,62	460
KB-1	1,807	4,18	11,560	116,30	1,639	1,643	3,05	493,06	148,48	570
KB-2	1,806	4,16	11,539	115,67	1,630	1,633	2,98	495,70	149,28	570
KB-3	1,807	4,17	11,549	115,97	1,635	1,635	2,99	495,76	149,29	560
KB-4	1,810	4,19		115,98	1,642	1,645	2,99	500,29	150,66	570
KB-5	1,807	4,18	11,551	116,00	1,632	1,636	2,91	495,57	149,24	590

Table 3

Relations between physical properties of biotites {a} and garnets {b} from Little Carjatian paragneisses

a) biotite		b) garnet	
$n_z - n_z^{calc.}$	$r = 0,36$	$n - \rho$	$a = 0,68$
$\rho - V$	$r = 0,38$	$n - a_0$	$r = -0,73$
$\rho - V_m$	$r = 0,38$	$V_m - a_0$	$r = 0,99$
$V - V_m$	$r = 0,99$		
$V_m = -0,5794 + 0,3022 V$		$\rho = -2,8434 + 3,8822 n$	
		$a_0 = 12,0296 - 0,2661 n$ $a_0 = 7,6787 + 0,0333 V_m$	

n, n_z — index of refraction

ρ — density [g/cm³]

a_0 — unit cell edge

V — unit cell volume ($\times 10^{-10}$ m)

V_m — molar volume (cm³/mol)

Garnets occur in small amount in the studied rocks and so do not essentially determine chemical composition of metamorphosed rocks.

In metamorphic reaction

{Mg+Fe} biotite + {Fe+Mg} garnet = {Fe+Mg} biotite + {Mg+Fe} garnet, in which substitution phenomena take place, chemical composition of biotite and garnet is influenced by temperature, pressure and chemical composition of rocks.

With the course of these reaction, which are conditioned by higher metamorphic grade, the changes of physical properties of coexisting garnet and biotite, which can be schematically expressed by following model reactions, are connected:

Flog75Ann25	+	Pyr16Alm84	=	Flog58Ann42	+	Pyr33Alm67
$n = 1,609$		1,791		1,628		1,811
$\rho = 2,91$		4,075		3,00		4,20
$V = 501,1$		11,503		504,0		11,515
$V_m = 151,13$		114,62		151,95		114,96
$\ln K_D =$	2,70				1,29	
$T(^{\circ}\text{C}) =$	420				680	

Flog ₉₁ Ann ₉	+	Pyr ₄₁ Alm ₅₉	=	Flog ₇₅ Ann ₂₅	+	Pyr ₅₀ Alm ₅₀
n = 1,592		1,782		1,609		1,772
ρ = 2,82		4,01		2,91		3,95
V = 498,4		11,498		501,1		11,492
V _m = 150,34		114,46		151,13		114,28
lnK _D =	2,73				1,09	
T(°C) =	400				700	

Flog ₈₃ Ann ₁₇	+	Pyr ₅₀ Alm ₅₀	=	Flog ₇₅ Ann ₂₅	+	Pyr ₅₈ Alm ₄₂
n = 1,600		1,772		1,609		1,762
ρ = 2,87		3,95		2,91		3,89
V = 499,8		11,492		501,1		11,487
V _m = 150,73		114,28		151,13		114,12
lnK _D =	1,60				0,76	
T(°C) =	600				800	

The position of these reaction is designated in Fig. 1, with numbers I, II, III. The temperatures in the models are given according to the work of L. Perčuk (1973). The values of the end-members pyrope, almadite, phlogopite, annite were taken from the works: B. Skinner (1956), D. Hewitt — D. Wones (1975), R. Robie — D. Waldbaum (1968).

In the applied model as end-members phlogopite and annite were accepted only. For descriptive and thermodynamic purposes algebraically suitable end-member oxibiotite $K\text{Fe}^{3+}_3\text{AlSi}_3\text{O}_{12}(\text{H}_{-1})$ was not taken into account for simplified approach although. D. Wones and H. Eugster (1965) have shown that biotites in the phlogopite-annite series also contain oxibiotite component.

This theoretical approach does not lead to the supposition that garnets and biotites of some physical properties represent higher grades of metamorphism. This statement applies to those properties, which are essentially influenced by distribution of magnesium, iron and original chemical composition of rocks. The influence of pressure and structure order-disorder phenomena in these minerals were not taken into account in the calculated models.

The physical properties of biotites and garnets coexisting in paragneisses of the Little Carpathians are summarized in Tab. 2. The calculated model Pyr-Alm/Flog-Ann does not illustrate sufficiently all the chemical variability of coexistence of biotite and garnet and causes disagreement, mainly in evaluation of the unit cell volume and the molar volume of coexisting garnets

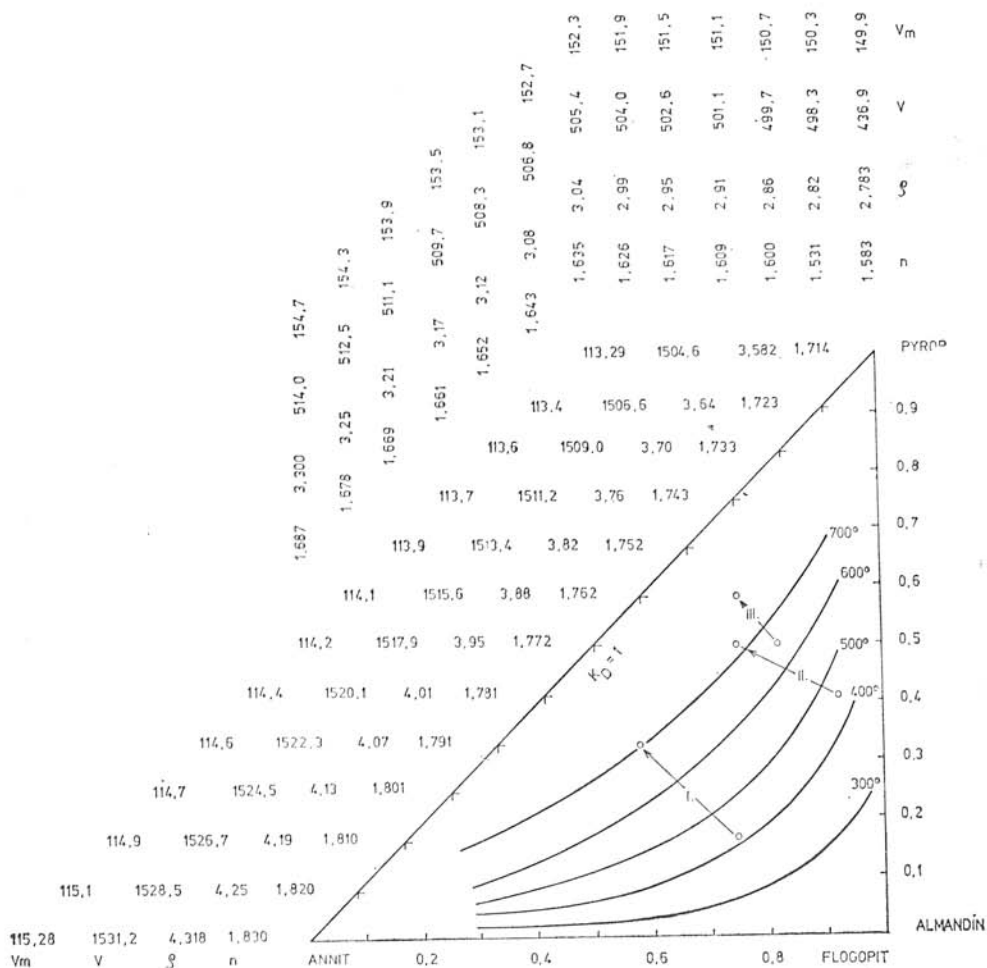


Fig. 1. Model of changes of physical properties of garnets (in the Pyr-Alm series) and biotites (in Flog-Ann series) at various temperature of coexistence. The scale of temperature is adopted from the work by L. Perčuk (1973).

and biotites. V and V_m probably more sensitively reflect the changes of chemical composition. The presence of elements not included in the model causes deviation from the model approach to such an extent that comparison can be unactual from this standpoint.

From evaluation of densities $\rho_{Bio-Gar}$ and refractive indices $n_{Bio-Gar}$ partial accordance between temperatures indicated by these physical properties and temperatures of recrystallization results, determined for these rocks by thermodynamic approach (M. Dyda, 1977).

Table 4

Temperatures of coexistence of biotites and garnets in Little Carpathian paragneisses obtained on the basis of $K_D^{Mg^{Fe}}$ and temperatures indicated by $\rho^{Bio-Gar}$

Sample	$\log K_D$	Temperature(°C)	$\rho^{Bio-Gar}$	Indicated temperature(°C)
6.	0,63	540	2,97/4,17	550
7.	0,59	560	2,98/4,16	570
8.	0,73	460	3,00/4,19	550
9.	0,71	460	2,89/4,16	450
10.	0,65	520	2,96/4,18	510
11.	0,74	450	3,04/4,18	610
12.	0,45	610	3,00/4,15	610
14.	0,65	510	3,02/4,18	580
17.	0,65	510	2,98/4,17	550
18.	0,76	460	2,92/4,18	450
KB-1.	0,54	570	3,05/4,18	620
KB-2.	0,55	570	2,98/4,16	570
KB-3.	0,56	560	2,99/4,17	570
KB-4.	0,55	570	2,99/4,19	540
KB-5.	0,49	590	2,91/4,18	440

From this standpoint, the evaluation of densites of coexisting biotites and garnets appear to be most purposeful (Tab. 4., Fig. 2 and 3).

Garnets from metapelites have delimited chemical composition and physical properties to a narrow extent. Therefore coexisting biotite with higher variability of properties essentially influences the position of samples in the given model. From the generalization of theoretical data is also evident that garnets with lower V cannot unambiguously be attributed by a higher grade of metamorphism, also when this relation is generally evident in metapelites (see e.g. K. Nandi, 1967). Biotites are from this standpoint less suitable in the studied rocks as they are more influenced by the chemical composition of rocks.

The temperature in Tab. 4 are obtained on the basis of data by L. Perčuk [1973]. Calculation of recrystallization temperature according to S. Saxena [1973], taking into regard also distribution of elements in the individual structural positions, does not result in the same temperature values. Recrystallization temperatures determined for the studied samples according to A. Thompson [1976 a, b] are also different ($T^{\circ}C_{Perčuk}$ v. $T^{\circ}C_{Thompson}$ $r = 0,73$).

Conclusion

Mutual coherency of properties of the mineral association is a function of the metamorphic process in the whole range of P-T-X conditions determining also physical properties of coexisting phases.

It is evident from evaluation of molar volumes, unit cells volumes, refractive indices and densities of coexisting biotites and garnets in Little

Carpathian paragneisses (Tab. 2) that a suitably chosen physical property can be an indicator of the grade of metamorphism. The degree of suitability is given by the choice of model, which should take into account such components that no essential difference are caused between model system and real sample.

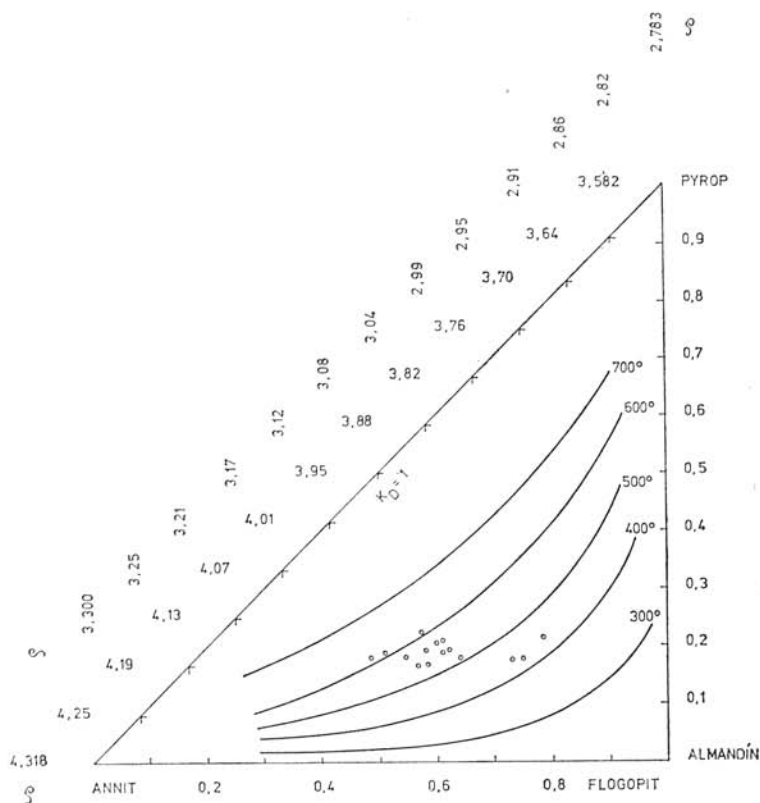


Fig. 2. Densities of coexisting garnets and biotites of the Little Carpathian paragneisses plotted in the calculated model.

The recrystallization temperatures of Little Carpathian paragneisses, obtained by thermodynamic calculation (450-610 °C) and evaluation of physical properties of coexisting biotites and garnets (Tab. 4), indicate actuality of the applied approach in approximation of metamorphic temperatures.

Acknowledgment

For making possible investigations, their thematic direction, introduction in the field, interest and remarks to the work I express many thanks to the

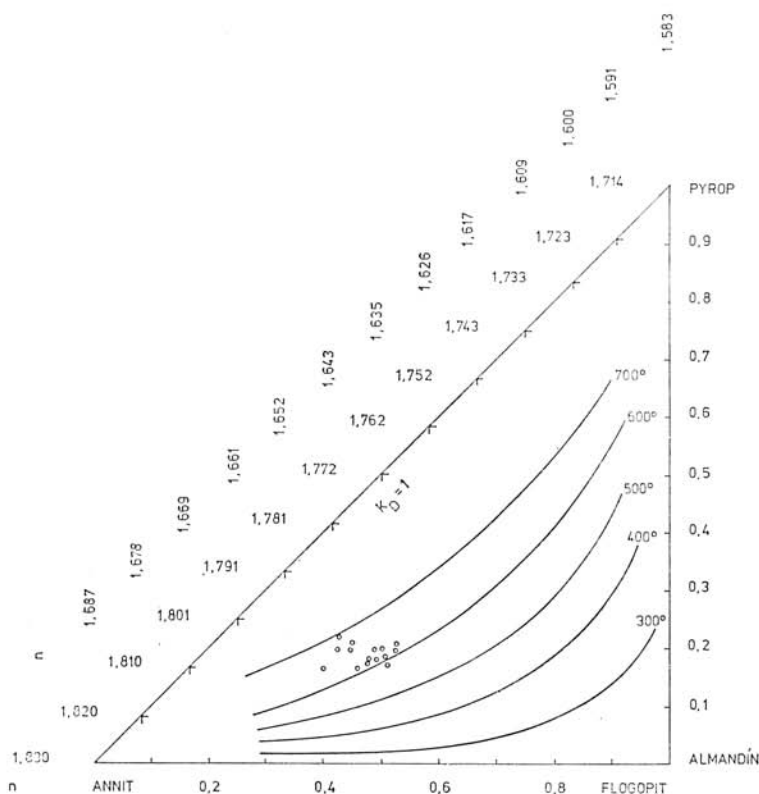


Fig. 3. Refractive indices of coexisting garnets and biotites of Little Carpathian paragneisses plotted in the calculated model.

director of the Geological Institute of the Slovak Academy of Sciences, Academician B. Cambel. To Ing. M. Handlovic CSc. and RNDr. J. Král. I am indebted for the remarks to the manuscript of the work.

Geological position and location of samples

The Little Carpathians are the westernmost and most external core mountains of the West Carpathian arc. Postkinematic granitoid rocks of intrusive character mostly occur here, which penetrated into the higher parts of the sedimentary mantle. The sedimentary envelope was affected by epizonal metamorphism before intrusion and the periplutonic metamorphic process, caused by the intrusion, became the predominant metamorphism in many places. This process was taking place with terminal folding of the Variscan mountain system (T. Buday, B. Cambel, M. Maheľ, 1962), B. Cambel (1976).

The fundamental rocks of the Little Carpathian metamorphic crystalline are biotite phyllites, biotite paragneisses with garnet, staurolite and accessory andalusite and fibrolite sillimanite.

Spreading of these rocks and suitability of their mineral associations for the study of the delimited mineralogical questions have made possible more detailed observation of the individual mineral groups.

The study of samples with its thematical direction continues from the works by B. Cambel (1954, 1958), M. Čajka — E. Šamajová (1961), B. Cambel — J. Macek (1972) and is part of complex investigation of the Little Carpathian crystalline complex.

In definition of the nomenclature, grain-size classes and structures of the observed rocks I set out from the works by: B. Hejtmán (1962), A. Dudek — F. Fedíuk — M. Palivcová (1962), J. Kamenický — D. Hovorka (1972).

- Sample N° 6y.: biotite-quartz paragneiss (p), fine-grained; Bratislava, Dúbravka, near the waterworks.
- Sample N° 7y.: garnet-sillimanite-biotite paragneiss (p), fine-grained. Limbach, Slnéčné údolie valley, elev. p. 306.
- Sample N° 8y.: biotitic paragneiss (p), fine-grained. Baba, road Pernek-Baba, ~ 500 m before the hotel.
- Sample N° 9y.: garnet-staurolite-biotitic paragneiss (p), fine-grained. Baba, near the parking-place.
- Sample N° 11y.: staurolite-biotitic paragneiss (p), fine-grained. Limbach, Slnéčné údolie valley, adit near the forester's house.
- Sample N° 12y.: arctic migmatite, fine-grained. Jur near Bratislava, road NW from the community Jur, deviation near the outcrop of migmatites.
- Sample N° 14y.: garnet-staurolite-biotitic paragneiss (p), fine-grained. Road Baba-Pernek, ~ 300 m of the cottage Baba.
- Sample N° 17y.: biotitic paragneiss (p), fine-grained. Road Bratislava—Devín, Lištie diery, mouth of Modrý Jarok brook.
- Sample N° 18y.: biotitic paragneiss (p), fine-grained. Road Bratislava—Devín, ~ 300 m S of the big quarry.
- Sample N° KB—1y.: sillimanite-garnet-biotitic paragneiss (p), fine-grained. Bratislava, Železná studnička, ~ 100 m of the concrete bridge, on the left side of the brook.
- Sample N° KB—2y.: garnet-biotitic paragneiss (p), fine-grained. Bratislava, Železná studnička, Road of Youth, ~ 150 m of the houses on the left side of the brook.
- Sample N° KB—3y.: biotitic paragneiss (p), fine-grained. Bratislava, Železná studnička, ~ 100 m above the restaurant Klepáč.
- Sample N° KB—4y.: biotitic paragneiss (p), fine-grained. Bratislava, Železná studnička, ~ 200 m above the restaurant on the way Železná studnička—Kamzík.
- Sample N° KB—5y.: garnet-sillimanite-biotitic paragneiss (p), fine-grained. Bratislava—Lamač, cottage area ~ 200 m of the last cottage in the narrowed valley of brook.

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