The Average Structural Density of Barite Crystals of Different Habit Types

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Abstract

Eight different habit types of barite crystals which could be classified into 4 essentially different groups were determined by morphological analysis: E_b group - barite crystals elongated in the [010] direction; E_a group - barite crystals elongated in the [100] direction; T_c group - barite crystals tabular on the basal pinacoid {001}; E_c group - barite crystals elongated in the [001] direction.

By determination and comparison of the average structural densities of different habit types of barite crystals, it was possible to establish the spans of their values from 276.1 to 495.8, and overlapping of values for E_b group with E_a group and T_c group with E_c group. The sequence of habit types, with regard to the decreasing average structural density, i.e. the priority sequence of developing of barite crystal types, was determined: E_b group - E_a group - T_c group - E_c group.

Sažetak

Morfološkom analizom određeno je 8 različitih tipova habitusa baritnih kristala koji se mogu svrstati u 4 bitno različite skupine: E_b skupina - kristali barita izduljeni smjerom [010]; E_a skupina - kristali barita izduljeni smjerom [100]; T_c skupina - kristali barita pločasti po baznom pinakoidu {001}; E_c skupina - kristali barita izduljeni smjerom [001].

Određivanjem i usporedbom srednjih srtukturnih gustoća različitih tipova habitusa baritnih kristala utvrđeni su rasponi njihovih vrijednosti od 276.1 do 495.8, te su uočena preklapanja vrijednosti za E_b i E_a skupine, kao i za T_c i E_c skupine. Utvrđen je redosljed tipova habitusa s obzirom na smanjenje srednje strukturne gustoće kristala, tj. prioritetni slijed razvoja baritnih kristala: E_b skupina - E_a skupina - T_c skupina - E_c skupina.

1. INTRODUCTION

Crystals of the same mineral species can appear in different forms, i.e. habit types. Besides numerous researchers, KOSTOV (1966, 1969, 1970, 1975) has investigated the changes of mineral habit during the crystallization of a particular mineral species. He pointed to the influence of crystalline structure (unit cell dimensions, nature of bonds in the structure) and external conditions during growth (chemical composition of solution, supersaturation degree of solution, type of ingredients) on the change of crystal habit of the same mineral species. BRAVAIS (1851) observed the evident dependence between reticular density and probability of appearance of a particular crystal form. Priority in crystal growth has the crystal form with maximal reticular density (BRAVAIS, 1851). In consideration of this fact, it is possible to determine the development sequence of barite crystal types by introduction of the term "average structural density of crystal" (KRIVOVIČEV, 1971) which defines the total contribution of reticular density of all crystal form faces, and by calculation of the values of average structural densities for different habit types of barite crystals.

Barite is one of the mineral species, which could have extremely different habit types. BRAUN (1932)

has determined seven different habit types of barite crystals on the basis of detailed investigation of relationships between morphological characteristics and mineral association. Some specimens of barite from Westmorland and County Durham in the north of England, show up to six changes of habit during growth, from the crystals elongated in the [100] direction, through the crystals tabular on the basal pinacoid {001}, to crystals elongated in the [010] direction (SEAGER & DAVIDSON, 1952). A similar sequence is described for barite crystals from the Tjuja-Mujun mine (SOLODOVNIKOVA, 1927). KRIVOVIČEV (1971) separated three generations of different habit types of barite crystals from the Belorečensko locality, and determined the development sequence from the prismatic crystals elongated in the [010] direction, through the thick-tabular crystals elongated in the [010] direction, to crystals tabular on the basal pinacoid. This sequence is in accordance with decreasing average structural density of the crystals (KRIVOVIČEV, 1971). An inverse development sequence was determined on barite crystals from Krapinske Toplice (ZEBEC, 1976). A sequence from crystals tabular on the basal pinacoid, through crystals elongated in the [100] direction, to crystals elongated in the [010] direction, was accompanied with a change of genesis temperature and decrease of $SrSO_4$ content (ZEBEC, 1976). Observations of phantoms and inclusions in crystals of barite from Muddy Creek, Colorado, suggest at least five episodes of crystal growth, with

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changes in morphology with respect to time (TRUEBE, 1981). Recently, researchers attempted to determine the dependence of barite morphology on different controlled conditions during experimental crystal growth (FERNANDEZ-DIAZ et al., 1990; PRIETO et al., 1992).

Numerous barite specimens with different crystal habits are stored at the collection of Department of Mineralogy and Petrography of the Croatian Natural History Museum in Zagreb. Systematic researchs of barite morphology and the reticular density of the crystal form faces was undertaken in order to try to define the priority sequence of developing of barite crystals types, on the base of calculated values of average structural density of crystal.

This paper is the result of investigations performed as part of a Master's Thesis (RADANOVIĆ-GUŽVI-CA, 1995).

2. MATERIAL AND METHODS

The crystals of barite from the Mrzle Vodice locality in Gorski Kotar region, Sivac on mountain Petrova gora and Ričice in Lika region from Croatia, Žune in north-western Bosnia and Herzegovina, the valley of the river Krivaja, Mačkara district, Veovača and the environs of Kreševo in the central part of Bosnia and Herzegovina, and Prača in eastern Bosnia and Herzegovina, Šuplja Stena on Avala hill and Veliki Majdan in Serbia, Yugoslavia, and Stari Trg (Trepča) in Kosovo, Serbia, Yugoslavia, are elaborated in this paper.

Under the research programme, the morphological, as well as relevant crystallographic analyses of the barite crystals, were performed.

Morphological analysis included goniometric measurement of the crystals, performed for the most part (62 crystals) on a Goldschmidt type two-circle reflecting goniometer, and for a smaller number (5 crystals) on a contact goniometer. On the basis of faces determination from a gnomon projection, and by the help of data for barite by GOLDSCHMIDT (1897) and PALACHE et al. (1951), construction of the parallel perspective drawings of measured crystals was undertaken using the method described by TERPSTRA & CODD (1961), as well as the determination of the different habit types of the barite crystals.

Crystallographic research was performed by the X-ray powder method on the Philips vertical x-ray goniometer with Cu K radiation and graphite monochromator. Standard conditions of recording were: 40 kV, 30 mA, 1° 2 /min for the counter, and 1 cm/min for the recording paper, in the angular region from 9° to 62° 2 . The barite X-ray diffraction pattern was indexed conformably to JCPDS card no. 24-1035. The unit cell dimensions are the result of refinement by the method of least squares performed by the GITTER computer program (HUMMEL, 1982) on the base of 28 to 32 indexed diffraction lines.

The reticular density of a particular crystal form is the ratio of the number of all atoms in the section of a crystal structure, defined by the elementary parallelogram S_{hkl} and interplanar spacing d_{hkl}. The number of all atoms in a certain section was performed by the ATOMS computer program (DOWTY, 1991), which gave us the display of structure based on the data (coordinate system, unit cell parameters, space-group symmetry, atom coordinates). The data according to HILL (1977) are used for the display of barite structure. The surface of the elementary parallelogram was calculated according to the formula:

$$S^2_{hkl} = h^2 S^2_{100} + k^2 S^2_{010} + l^2 S^2_{001} + 2(hk S_{100} S_{010} \cos + kl S_{010} S_{001} \cos + lh S_{001} S_{100} \cos \mu)$$

where: S_{hkl} - surface of the elementary parallelogram of the face (hkl) in [$Å^2$]; $S_{100} = b_0 c_0 \sin$, $S_{010} = c_0 a_0 \sin$, $S_{001} = a_0 b_0 \sin$ (a_0, b_0, c_0 - unit cell dimension in [Å]; , , - angles between crystal axes); = (100) ((010); = (010) (001); μ = (001) (100)) (PHILLIPS, 1946). This formula can be simplified since barite is orthorhombic:

$$S^2_{hkl} = h^2 b^2_0 c^2_0 + k^2 c^2_0 a^2_0 + l^2 a^2_0 b^2_0$$

According to KRIVOVIČEV (1971) the average structural density of crystal "E_{CR}" was calculated according to the formula:

$$E_{CR} = (M \times E_{hkl})$$

where: M is the morphological persistence of a particular crystal form (FRANK-KAMENECKII, 1951), E_{hkl} is the reticular density of a crystal form in number of atoms in $Å^3$, and the product "$M \times E_{hkl}$" defines the contribution of reticular density of a given crystal form in the total structural density of crystal. Morphological persistence is the ratio of the total surface of all faces of a certain crystal form and the surface of the whole crystal, and was expressed in percent. "M" is calculated thus: face surfaces of emphasized crystal forms were measured on several crystals of a certain habit type, and the average value of these surfaces for a respective crystal form was calculated.

The studied barite specimens are stored in the collection of the Department of Mineralogy and Petrography of the Croatian Natural History Museum in Zagreb, and are marked by the inventory numbers: 600:ZAG;3310:MP1, 600:ZAG;3602:MP1, 600:ZAG;3641:MP1, 600:ZAG;5183:MP1, 600:ZAG;5677:MP1, 600:ZAG;5689:MP1, 600:ZAG;5718:MP1, 600:ZAG;5780:MP1, 600:ZAG;6688:MP1, 600:ZAG;6691:MP1, 600:ZAG;6703:MP1, 600:ZAG;8541:MP1, 600:ZAG;8542:MP1, 600:ZAG;8546:MP1, 600:ZAG;9013:MP1, 600:ZAG;9014:MP1, 600:ZAG;9016:MP1, 600:ZAG;9017:MP1, 600:ZAG;9018:MP1, 600:ZAG;9021:MP1, 600:ZAG;9022:MP1, 600:ZAG;9023:MP1, 600:ZAG;9025:MP1, 600:ZAG;9026:MP1, 600:ZAG;9027:MP1, 600:ZAG;
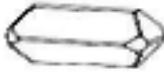
HABIT TYPE	LOCALITY	FORM	M (%)	E_{hkl} (atoms/Å³)	M x E_{hkl}	$E_{OR} = \Sigma$ M x E_{hkl}
	Šuplja Stena	101	63.4	5.572	353.3	495.8
		011	17.6	4.335	76.3	
		001	11.1	3.578	39.7	
		210	2.8	3.443	9.6	
		100	2.3	4.441	10.2	
		213	1.3	1.960	2.5	
		010	1.1	2.725	3.0	
		211	0.4	3.102	1.2	
	Veliki Majdan	101	31.9	5.561	177.4	419.5
		001	21.9	3.570	78.2	
		210	20.8	3.437	71.5	
		201	12.6	3.787	47.5	
		011	7.4	4.326	32.0	
		103	4.3	2.298	9.9	
		010	1.1	2.719	3.0	

Table 1 The average structural density of barite crystals elongated in the [010] direction (E_b group).

9029:MP1, 600:ZAG;9030:MP1, 600:ZAG;9031:MP1 and 600:ZAG;9039:MP1.

3. RESULTS

3.1. MORPHOLOGICAL ANALYSIS

From morphological analysis of barite crystals from 12 localities, 8 different habit types were determined. These habit types could be classified into 4 essentially different groups:

The E_b group of barite crystals (Table 1) is represented by 2 habit types elongated in the [010] direction. One habit type was observed only in Šuplja Stena, and had been described before (BARIĆ, 1949). These colu-

mnar crystals have prominent {101} and somewhat smaller {011} faces. According to Braun's classification, this habit type is intermediate between ore - IIIb type - P^b_{001}[010] and carbonate - IVa type - A_b (BRAUN, 1932). Another habit type was observed in Veliki Majdan. These columnar crystals have well-developed {001} and {101} faces, and somewhat smaller {011} and {210} faces, and correspond to the carbonate - IVa type - A_b (BRAUN, 1932).

The E_a group of barite crystals (Table 2) is represented by 2 habit types elongated in the [100] direction. One habit type was noticed in Ričice and Kreševo (Raštelica, Dubrave, Dusina). These thick-tabular crystals have prominent {001}, {011} and {101} faces, and correspond to the rectangularly-tabular - I type -

HABIT TYPE	LOCALITY	FORM	M (%)	E_{hkl} (atoms/Å³)	M x E_{hkl}	$E_{OR} = \Sigma$ M x E_{hkl}
	Ričice	011	45.3	4.330	196.1	431.3
		001	34.6	3.569	123.5	
		101	20.1	5.557	111.7	
	Dubrave, Kreševo	011	52.5	4.320	226.8	429.2
		101	20.3	5.539	112.4	
		001	18.0	3.559	64.1	
		010	4.5	2.718	12.2	
		210	3.1	3.425	10.6	
		213	1.6	1.950	3.1	
	Mačkara	011	47.1	4.327	203.8	415.8
		001	25.4	3.566	90.6	
		101	13.8	5.553	76.6	
		210	10.6	3.433	36.4	
		310	1.4	2.593	3.6	
		010	0.6	2.721	1.6	
		230	0.6	1.678	1.0	
		100	0.5	4.424	2.2	

Table 2 The average structural density of barite crystals elongated in the [100] direction (E_a group).

HABIT TYPE	LOCALITY	FORM	M (%)	E_{hkl} (atoms/Å²)	$M \times E_{hkl}$	$E_{CR} = \Sigma M \times E_{hkl}$
	Žune	001	66.3	3.581	237.4	
		011	13.5	4.338	58.6	
		101	13.5	5.574	75.2	
		010	2.5	2.726	6.8	394.1
		210	1.8	3.442	6.2	
		100	1.3	4.438	5.8	
		201	1.1	3.772	4.1	
	Mrzle Vodice	001	57.3	3.584	205.4	
		210	32.3	3.440	111.1	
		101	6.7	5.575	37.4	
		211	2.4	3.101	7.4	365.3
		010	1.0	2.724	2.7	
		011	0.3	4.337	1.3	
	Veovača	001	64.1	3.571	228.9	
		210	24.7	3.434	84.8	
		610	5.1	1.425	7.3	
		011	3.2	4.328	13.8	
		201	0.8	3.763	3.0	346.3
		100	0.7	4.428	3.1	
		101	0.7	5.559	3.9	
		230	0.4	1.678	0.7	
		010	0.3	2.721	0.8	
	Kreševo	001	55.2	3.575	197.3	
		210	30.6	3.437	105.2	
		101	11.0	5.564	61.2	
		011	2.8	4.332	12.1	377.2
		100	0.3	4.431	1.3	
		610	0.1	1.426	0.1	
	Prača	001	51.4	3.579	184.0	
		210	39.9	3.442	137.3	
		102	5.1	3.319	16.9	
		103	2.7	2.304	6.2	349.1
		101	0.7	5.571	3.9	
		201	0.2	3.771	0.8	
	Veliki Majdan	001	39.1	3.576	139.8	
		210	21.7	3.438	74.6	
		211	14.3	3.097	44.3	
		201	13.0	3.768	49.0	
		111	8.7	3.890	33.8	354.6
		101	1.1	5.566	6.1	
		102	1.1	3.316	3.6	
		010	0.6	2.720	1.6	
		100	0.4	4.433	1.8	
	Stari Trg (Trepča)	001	57.6	3.580	206.2	
		210	25.8	3.441	88.8	
		211	9.0	3.101	27.9	
		101	5.3	5.571	29.5	361.4
		011	1.3	4.338	5.6	
		010	0.6	2.727	1.6	
		100	0.4	4.435	1.8	

Table 3 The average structural density of barite crystals tabular on the basal pinacoid {001} (T_c group).

P^a_{001}[100], [010] (BRAUN, 1932). Another habit type was observed only in the Mačkara district. These columnar crystals have well-developed {001}, {011}, {101} and {210} faces, and correspond to the carbonate - IVb type - A_a (BRAUN, 1932).

The T_c group of barite crystals (Table 3) is represented by 2 habit types tabular on the basal pinacoid {001}. One habit type was noticed only in Žune. These tabular crystals have well-developed faces from the [100] and [010] crystallographic axe's zones, specially

HABIT TYPE	LOCALITY	FORM	M (%)	E_{hkl} (atoms/Å²)	$M \times E_{hkl}$	$E_{cr} = \Sigma M \times E_{hkl}$
	Sivac	410	51.3	2.050	105.2	276.1
		210	13.3	3.434	45.7	
		100	12.2	4.426	54.0	
		001	11.6	3.564	41.3	
		110	4.2	2.319	9.7	
		211	3.1	3.101	9.8	
		230	1.7	1.679	2.9	
		213	1.3	1.954	2.5	
		010	0.7	2.722	1.9	
		101	0.6	5.552	3.3	
	Krivaja	210	64.4	3.443	221.7	349.4
		001	20.9	3.574	74.7	
		211	11.7	3.102	36.3	
		101	3.0	5.568	16.7	

Table 4 The average structural density of barite crystals elongated in the [001] direction (E_c group).

{011} and {101} faces. Another habit type was the most frequent type, and discovered at the following localities: Mrzle Vodice, Žune, Veovača, Kreševo, Prača, Veliki Majdan and Stari Trg (Trepča). These tabular crystals have marked {210} faces, and correspond to the silicate - VI type - P^b_{001}[010] (BRAUN, 1932).

The E_c group of barite crystals (Table 4) is represented by 2 habit types elongated in the [001] direction. One habit type was observed only in Sivac. These crystals have well-developed {410} and {100} faces and somewhat smaller {001} and {210} faces, and correspond to the Wolnyn - VII type - A_c (BRAUN, 1932). Another habit type was noticed only in Krivaja. These isometric to elongated crystals have prominent {210} faces and well-developed {001}, {101} and {211} faces, and correspond to the cubic-pyramidal - V type - $I_{001,210}$[120] (BRAUN, 1932).

A detailed review of the results of morphological analysis is given in RADANOVIĆ-GUŽVICA et al. (in press).

3.2. AVERAGE STRUCTURAL DENSITY OF BARITE CRYSTALS

The average structural density of crystal was calculated according to aforesaid procedure for 14 barite samples, e.g. 8 different habit types of crystals (Tables 1-4).

Values of the average structural density of a crystal are greatest in the case of barite crystals elongated in the [010] direction, and amounts vary from 419.5 to 495.8 (Table 1). Barite crystals elongated in the [100] direction have values of average structural density in a relatively small range from 415.8 to 431.3 (Table 2). Values of the average structural density of barite crystals tabular on the basal pinacoid {001} are 346.3 to 394.1 (Table 3). Values of the average structural density of a crystal are smallest in the case of barite crystals elongated in the [001] direction, and range from 276.1 to 349.4 (Table 4).

The ranges of values of the average structural densities of the four main groups of barite crystal habit types are shown in Fig. 1. Overlapping of the E_b group (barite crystals elongated in the [010] direction) with the E_a group (barite crystals elongated in the [100] direction) was observed, as well as the T_c group (barite crystals tabular on the basal pinacoid {001}) with the E_c group (barite crystals elongated in the [001] direction). Besides, it is necessary to highlight that the value of the average structural density of columnar barite crystals,
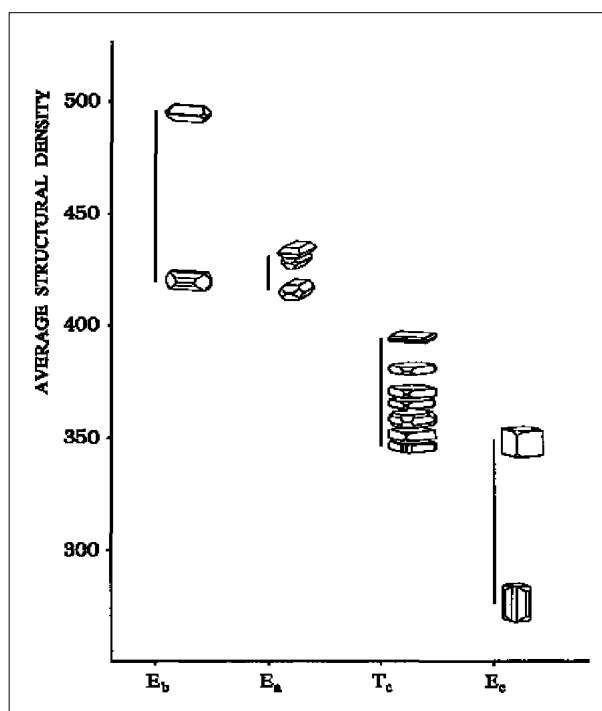

Fig. 1 The ranges of values of average structural densities of four main groups of barite crystal habit types: E_b group - barite crystals elongated in the [010] direction; E_a group - barite crystals elongated in the [100] direction; T_c group - barite crystals tabular on the basal pinacoid {001}; E_c group - barite crystals elongated in the [001] direction.

elongated in the [010] direction, with well-formed {101} and {001} faces, as well as with somewhat smaller {210}, {201} and {011} form faces, which were noticed in Veliki Majdan mine is very close to the value of average structural density of columnar crystals elongated in the [100] direction, with well-formed {011} and {001}, as well as with somewhat smaller {101} and {210} form faces, from Mačkara district. According to Braun's classification, both habit types correspond to the carbonate type, i.e. crystals elongated in the [010] direction are carbonate - IVa type - A_b, and the crystals elongated in the [100] direction are carbonate - IVb type - A_a (BRAUN, 1932).

4. DISCUSSION AND CONCLUSIONS

By comparison of the values of average structural densities of different habit types of barite crystals, the development sequence of barite crystal types in consideration to a decrease of average structural density, was determined. In general, the priority sequence was: barite crystals elongated in the [010] direction (E_b group) - barite crystals elongated in the [100] direction (E_a group) - barite crystals tabular on the basal pinacoid {001} (T_c group) - barite crystals elongated in the [001] direction (E_c group) (Fig. 1).

From similar previous researches (SOLODOVNIKOVA, 1927; SEAGER & DAVIDSON, 1952; KRIVOVIČEV, 1971; ZEBEC, 1976), together with the one performed here, it is possible to conclude that the sequence of developing of barite crystal types is not strictly defined, and is changeable (alternate) from case to case. Besides the density of the atomic arrangement, which defined the development sequence of crystal habit types, there are many other factors, which are able to cause a change in the sequence, or a change of direction.

It would be of great interest to know something about the mineral associations in which particular habit types of barite crystals occur, since the morphology of barite crystals together with information on mineral association, can help identify conditions under which some mineral assemblages were formed. However, research on mineral associations, as well as comparison with Braun's group was beyond the scope of this paper and is left for future investigation.

Acknowledgements

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