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## STRASHIMIRITE AND CORNWALLITE (COPPER ARSENATES) FROM RĘDZINY (LOWER SILESIA, POLAND)

**Abstract.** Two rare copper arsenates, strashimirite and cornwallite, have been found in the dolostone quarry in Rędziny. They occur in zones of dispersed ore mineralization, first of all in tyrolite veins, where form aggregates of very fine, intergrown fibrous individuals up to 20  $\mu\text{m}$  long, developed as white-green and light green spherulites. Both minerals have been identified on the basis of X-ray and chemical microprobe analyses. The composition of strashimirite corresponds to:  $(\text{Cu}_{3.90}\text{Mg}_{0.05}\text{Fe}_{0.03}\text{Ca}_{0.02}\text{Zn}_{0.01})[(\text{AsO}_4)_{1.99}](\text{OH})_{2.00} \cdot 2.5 \text{H}_2\text{O}$ ; the mineral reveals typical values of the unit-cell parameters:  $a = 9.719(2) \text{ \AA}$ ,  $b = 18.806(5) \text{ \AA}$ ,  $c = 8.937(3) \text{ \AA}$ ,  $\beta = 97.31(3)^\circ$ . Also the composition of cornwallite does not depart from the theoretical formula, its formal unit is  $(\text{Cu}_{4.92}\text{Mg}_{0.02}\text{Fe}_{0.04}\text{Ca}_{0.03}\text{Zn}_{0.01})[(\text{AsO}_4)_{2.09}](\text{OH})_{3.67}$ , and the unit-cell parameters are:  $a = 4.603(3) \text{ \AA}$ ,  $b = 5.785(3) \text{ \AA}$ ,  $c = 17.344(1) \text{ \AA}$ ,  $\beta = 91.66(5)^\circ$ . Both minerals are the products of reaction of  $\text{Cu}^{2+}$  and  $[\text{H}_2\text{AsO}_4]^-$  ions in the zone of weathering of dispersed sulphide and sulphide-arsenide mineralization, present in the Rędziny dolostones.

**Key-words:** strashimirite, cornwallite, X-ray data, unit-cell parameters, chemical composition, Rędziny, Poland

## INTRODUCTION

Strashimirite,  $\text{Cu}_8(\text{AsO}_4)_4(\text{OH})_4 \cdot 5\text{H}_2\text{O}$ , is monoclinic with a the space group P2/m, P2 or Pm and the unit-cell parameters  $a = 9.71 \text{ \AA}$ ,  $b = 18.85 \text{ \AA}$ ,  $c = 8.94 \text{ \AA}$ ,  $\beta = 97.12^\circ$  (Minčeva-Stefanowa 1968). The mineral was first described from the Zapachitsa copper deposits in western Stara-Planina, Bulgaria, as white greenish, fine-flake aggregates and concentric spherulites about 0,5 mm in size, being pseudomorphs after tyrolite and cornwallite.

Cornwallite,  $\text{Cu}_5(\text{AsO}_4)_2(\text{OH})_4$ , is monoclinic with the space group P21/a and the unit cell parameters  $a = 4.60 \text{ \AA}$ ,  $b = 5.81 \text{ \AA}$ ,  $c = 17.61 \text{ \AA}$ ,  $\beta = 92.25^\circ$  (Berry 1951) or  $a = 4.600(2) \text{ \AA}$ ,  $b = 5.757(3) \text{ \AA}$ ,  $c = 17.380(6) \text{ \AA}$ ,  $\beta = 91.87(3)^\circ$  (Arlt, Armbruster 1999). The mineral was first described from Wheal Gorland, St. Day,

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Cornwall, England. Cornwallite, known also as erinite, is dimorphous with cornubite (Claringbull et al. 1959). Its aggregates are similar to those of strashimirite, only their colour is more intensive.

## GEOLOGICAL SETTING AND MATERIAL

The deposit of dolostones quarried in Rędziny has the form of a lens about 1 km long with a width reaching about 200 m, and strikes NNE-SSW. It lies some 500–800 away of the field of the abandoned mine in Czarnów and about 500 m from the eastern margin of the Karkonosze granite (Teisseyre 1973; Sawicki 1995). Brownish-red zones of dispersed, mainly sulphide-arsenopyrite mineralization and of secondary mineralization with oxidized compounds of Fe–Cu–Zn–Pb–Mn–V are visible on western walls of the pit. More detailed geological description was provided in earlier papers concerning the mineralization in Rędziny (e.g. Gołębiowska, Pieczka 1997).

The mineralized zones are most often developed as small, some centimetres large insets and veinlets as well as small nests in dolostone. They reveal a rich inventory of mineral phases: e.g. from this locality several minerals from the groups of Ca–Cu (conichalcite, tyrolite, clinotyrolite; Gołębiowska et al. 1998) and Ca–Fe arsenates (yukonite; Pieczka et al. 1998), as well as cassiterite (Gołębiowska, Pieczka, 1997) were described. Strashimirite and cornwallite are very rare in Rędziny, and most often they co-occur with other, more frequent arsenates, such as conichalcite, scorodite or tyrolite. The two minerals in question form aggregates of very fine-grained, intergrown, elongated individuals up to 20 µm long. Principally they occur as radial aggregates or concentric spherulites, with the size not exceeding 1 mm. The colour of both minerals is white-green and light green, being more intensive in thicker aggregates.

## METHODS

Strashimirite and cornwallite have been identified applying X-ray and electron microprobe analyses. X-ray measurements were carried out on powdered samples of both minerals, using a Philips X'Pert diffractometer and the following conditions: graphite-monochromatized  $\text{CuK}_{\alpha 1}$  radiation ( $\lambda = 1.540562 \text{ \AA}$ ), operating voltage 34 kV, operating current 30 mA, the range of recording  $4\text{--}60^\circ(2\theta)$ , scanning speed  $0.02^\circ(2\theta)/\text{s}$ , quartz as the internal standard.

The quantitative chemical analyses were conducted at the Institute of Non-ferrous Metals in Gliwice, using a JCSA-733 JEOL electron microanalyser in the energy-dispersion mode. The specimens were covered with the graphite layer ca. 100 Å thick. The following standards were used: wollastonite  $\text{CaSiO}_3$  (Ca, Si), InAs (As), MgO (Mg), GaP (P),  $\text{FeS}_2$  (S),  $\text{Al}_2\text{O}_3(\text{Al})$ , and metallic Cu, Zn, Fe and V. Specimen current was  $2.15 \times 10^{-8} \text{ A}$ , voltage 20 kV, beam

diameter 2 µm, acquisition time for each analysis 20 s. The results were ZAF corrected after subtraction of  $\text{SiO}_2$  considered an admixture, and recalculated on the basis of four (strashimirite) and five (cornwallite) divalent ions.  $\text{H}_2\text{O}(+)$  was stoichiometrically added to compensate for an excess of the positive charge of cations.

## RESULTS AND DISCUSSION

Detailed characterization of copper arsenates is difficult considering their similar chemical compositions that differ sometimes only in the amount of water, and because of the presence of polymorphs. In addition to chemical analyses, the unambiguous identification requires precise phase analyses. As the amount of analytical material is often small, such an identification is in some cases impossible as on the recorded X-ray patterns the strongest reflections belong to more common secondary iron minerals (scorodite, pharmacosiderite).

X-ray powder data of strashimirite from Rędziny

TABLE 1

Strashimirite							
d <sub>hkl</sub>	JCPDS 21-289	I	hkl	d <sub>hkl</sub>	JCPDS 21-289	I	hkl
18.5	18.7	10	010	2.405	2.403	1	$\bar{4}01$
9.52	9.46	8	020	2.317	2.316	4	223
8.96	8.97	9	001	2.254	2.256	2	$\bar{3}23$
8.08	8.15	4	011	2.140	2.144	3	$\bar{4}40$
	7.03	1	$\bar{1}01$	2.086	2.082	1	$\bar{2}81$
6.14	6.21	2	101	2.034	2.038	1	313
5.60	5.62	3	$\bar{1}21$	1.981	1.981	2	$\bar{1}73$
4.784	4.79	8	200	1.949	1.947	1	333
4.657	4.65	2	$\bar{1}31$		1.901	2	263
4.467	4.47	1	$\bar{2}01$	1.871	1.879	2	343
4.208	4.21	8	140		1.839	1	234
4.027	4.01	2	022	1.797	1.794	1	470
	3.82	2	230	1.772	1.773	2	005
3.622	3.62	1	032	1.739	1.740	1	$\bar{3}91$
	3.35	8	240	1.703	1.703	2	210.1
3.134	3.13	9	060	1.666	1.665	1	522
	3.07	1	202	1.647	1.649	1	551
2.972	2.97	5	250	1.623	1.627	2	481
2.869	2.86	10	$\bar{1}61$	1.615	1.612	2	$\bar{5}43$
	2.78	4	321	1.597	1.596	2	$\bar{5}62$
2.697	2.69	1	152	1.584	1.580	4	310.1
2.644	2.65	6	340	1.564	1.560	2	$\bar{5}53$
2.577	2.57	1	$\bar{2}23$	1.534	1.535	4	$\bar{2}11.2$
2.476	2.476	7	261	1.528	1.528	4	$\bar{6}41$



The author faced the same problem with strashimirite and cornwallite — small sizes of aggregates of these minerals and difficulties in their separation made impossible obtaining X-ray patterns with reflections of only these two phases. Several other minerals were recorded alongside, including scorodite and tyrolite. Nevertheless, the presence of strashimirite and cornwallite is well documented and the sets of their  $d_{hkl}$  values (Table 1, 2) correspond well with standards from JCPDS files (cards 21-289, 39-1357). Unit-cell parameters of both minerals studied basically do not depart from the literature data and they are as follows:  $a = 9.719(2) \text{ \AA}$ ,  $b = 18.806(5) \text{ \AA}$ ,  $c = 8.937(3) \text{ \AA}$ ,  $\beta = 97.31(3)^\circ$  for strashimirite and  $a = 4.603(3) \text{ \AA}$ ,  $b = 5.785(3) \text{ \AA}$ ,  $c = 17.344(1) \text{ \AA}$ ,  $\beta = 91.66(5)^\circ$  for cornwallite.

The microprobe analyses of strashimirite and cornwallite from Redziny are presented in Table 3. In the case of strashimirite,  $\text{Cu}_8(\text{AsO}_4)_4(\text{OH})_4 \cdot 5\text{H}_2\text{O}$ , the

TABLE 2  
X-ray powder data of cornwallite from Redziny

Cornwallite							
$d_{hkl}$	JCPDS 21-289	I	hkl	$d_{hkl}$	JCPDS 21-289	I	hkl
8.82	8.68	60	200	2.1401	2.141	3	$\bar{4}21$
	5.48	14	110	2.1118	2.113	11	421
	4.806	40	210	2.068	2.055	3	212
	4.591	25	001	2.0442	2.040	3	620
	3.534	45	$\bar{1}11$	1.9152	1.912	3	130
	3.499	16	111	1.8814	1.877	30	230
	3.200	45	$\bar{4}01$	1.8293	1.825	13	330
3.1167	3.105	30	401	1.7974	1.793	9	512
3.0820	3.086	20	$\bar{3}11$	1.1717	1.7646	25	$\bar{2}22$
	3.015	35	311	1.7549	1.7522	10	$\bar{7}21$
2.9724	2.970	17	510	1.7392	1.7406	10	$\bar{2}31$
2.8938	2.885	50	020		1.7330	8	10.0.0
2.8045	2.799	3	$\bar{4}11$	1.7259	1.7242	15	721
2.7404	2.735	45	220	1.7031	1.7004	5	$\bar{3}31$
	2.582	20	610	1.6870	1.6898	7	331
2.5205	2.526	19	$\bar{5}11$	1.6837	1.6811	17	530
2.4862	2.479	30	$\bar{6}01$	1.6470	1.6444	9	422
2.4756	2.463	45	511	1.6379	1.6380	15	$\bar{1}0.0.1$
2.4290	2.413	100	121	1.6328	1.6339	12	$\bar{8}21$
2.4048	2.401	40	420	1.6147	1.6079	8	821
	2.359	35	$\bar{2}21$	1.6032	1.6003	8	630
2.3352	2.339	45	221	1.5969	1.5923	6	712
2.2995	2.292	15	002	1.5835	1.5858	6	$\bar{5}31$
2.2919	2.275	29	$\bar{6}11$	1.5736	1.5775	15	522
2.2651	2.262	5	$\bar{3}21$		1.5705	10	531
	2.219	12	520		1.5449	7	802
	2.199	2	202	1.5208	1.5189	18	730
2.1694	2.166	9	800				

TABLE 3  
Chemical composition of strashimirite (A) ( $n = 12$ ) and cornwallite (B) ( $n = 5$ ) from Redziny (wt.%)

	A	B
CaO	0.21	0.13
MgO	0.38	0.10
Fe <sub>2</sub> O <sub>3</sub>	0.36	0.47
ZnO	<0.01	0.13
CuO	55.36	58.31
Al <sub>2</sub> O <sub>3</sub>	<0.01	<0.01
As <sub>2</sub> O <sub>5</sub>	40.47	35.90
V <sub>2</sub> O <sub>5</sub>	<0.01	<0.01
H <sub>2</sub> O	3.21	4.93
Ca	0.02	0.03
Mg	0.05	0.02
Fe	0.03	0.04
Zn	<0.01	<0.01
Cu	3.90	4.92
Al	<0.01	<0.01
(AsO <sub>4</sub> ) <sup>3-</sup>	1.99	2.09
(VO <sub>4</sub> ) <sup>3-</sup>	<0.01	<0.01
(OH) <sup>-</sup>	2.00	3.67

$n$  — number of point analysis.

analysis does not include crystallization water, and two (OH)<sup>-</sup> groups have been added to compensate for positive charge of cations. A small deficit of Cu<sup>2+</sup> is probably supplemented with other divalent cations (Fe, Mg, Ca). Several point analyses indicated also the presence of trace amounts of vanadate and chloride ions. Point microprobe analyses of strashimirite give the Cu/As ratio in a range of *ca.* 1.9—2.1 (the theoretical Cu/As ratio for this mineral is 2.0). Other copper arsenates with the identical Cu/As ratio are: olivenite Cu<sub>2</sub>(AsO<sub>4</sub>)(OH), euchroite Cu<sub>2</sub>(AsO<sub>4</sub>)(OH) · 3H<sub>2</sub>O and arhbarite Cu<sub>2</sub>(AsO<sub>4</sub>)(OH) · 6H<sub>2</sub>O, but they have not been identified in X-ray analyses.

Cornwallite, Cu<sub>5</sub>(AsO<sub>4</sub>)<sub>2</sub>(OH)<sub>4</sub>, in contrast to strashimirite does not contain crystallization water, thus its identification is simpler and the results obtained are easier to be compared with the literature data (Berry 1951; Guillemin 1956; Claringbull et al. 1959). The amount of (OH)<sup>-</sup> groups has been calculated from the rule of electronegativity of the formula unit. As it was in the case of strashimirite, it has been found that a deficit of copper is supplemented with small admixtures of Fe, Zn, Mg and Ca ions, and an excess of arsenate ions lowers the amount of (OH)<sup>-</sup> groups. The Cu/As ratio in the chemical analyses of cornwallite oscillates between 2.4 and 2.6 (the theoretical Cu/As ratio is 2.5). Another mineral with the same Cu/As ratio — cornubite — is dimorphous with cornwallite. As an admixtures of this phase in the samples



studied could be minimal at the highest, reliable confirmation of cornubite has not been possible.

## CONCLUSIONS

Precipitation of both minerals presented here is a consequence of the presence of  $\text{Cu}^{2+}$  and  $[\text{H}_2\text{AsO}_4]^-$  ions in mineralizing solutions that originate in diversified transformations of primary minerals in zone of weathering. The primary arsenides and sulphides are oxidized with formation, among others, of arsenic acid  $\text{H}_2[\text{AsO}_4]^-$ , also  $\text{Cu}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Fe}^{2+}$  and other ions, migrating within the weathered zone. An inventory of various mineral forms precipitating under such conditions is controlled by thermochemical parameters, first of all by activities of ions, pH of the environment and solubility of the compounds formed. Precipitation and dissolution of various arsenate minerals may be generally described as:  $M_p(\text{AsO}_4)_q(\text{OH})_{r-n}\text{H}_2\text{O}(\text{s}) + (2q + r)\text{H}^+(\text{aq}) = p\text{M}^{2+}(\text{aq}) + q\text{H}_2\text{AsO}_4^-(\text{aq}) + (n + r)\text{H}_2\text{O}$  (Magalhaes et al. 1988). It can be seen that the formation of arsenate compounds is accompanied by a general increase of acidity of the environment and a decrease of activity of  $\text{Me}^{2+}$  ions. Therefore, in the  $\text{Cu}^{2+}$ - $\text{H}_2[\text{AsO}_4]^-$  system (Fig. 1) a full sequence of precipitating compounds should start with clinoclase,  $\text{Cu}_3(\text{AsO}_4)(\text{OH})_3$ , whose presence in Rędziny has

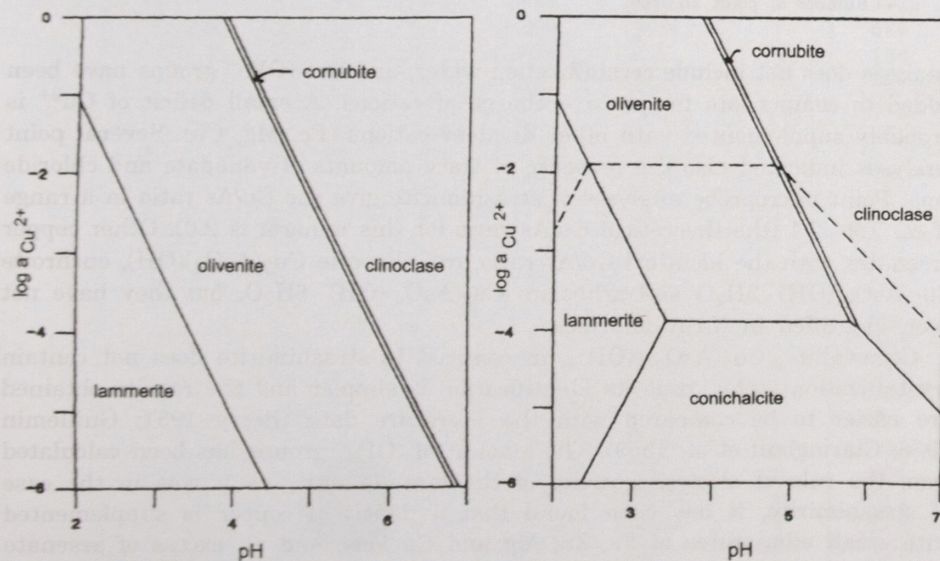


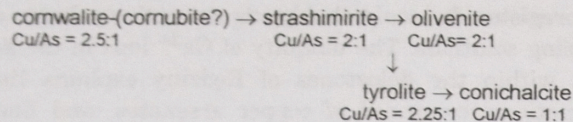
Fig. 1. Stability field diagram for the copper (II) arsenate minerals. Limits of pH have been set to those corresponding to the predominance in aqueous solution of the  $\text{H}_2[\text{AsO}_4]^-$  (aq) ion (after Magalhaes et al. 1988)

not been so far registered due, probably, to respectively lower activity of  $\text{Cu}^{2+}$  ions in mineralizing solutions. The ubiquity of  $\text{Ca}^{2+}$  ions in the zone of oxidation of ore minerals within the dolostones of Rędziny explains the lack of more diversified associations, composed of copper arsenates, and limits the mineral inventory to systems with olivenite, tyrolite and, most common of them, conichalcite. The presence of  $\text{Ca}^{2+}$  in the surrounding environment considerably widens the stability field of the latter mineral at the expense of clinoclase, cornubite-cornwallite and olivenite, while the presence of copper in mineralizing solutions practically excludes the possibility of precipitation of any calcium arsenates.

In the case discussed, cornwallite and strashimirite precipitated earlier than tyrolite that surrounds them in the form of feathery overgrowths on cornwallite-strashimirite spherulites. The two minerals, particularly cornwallite, must have crystallized when the activity of copper ions or pH of the environment were locally higher. Under such conditions either one or both  $\text{Cu}_5(\text{AsO}_4)_2(\text{OH})_4$  dimorphs, i.e. cornwallite and/or cornubite, can form; both are very rare, usually co-occur and may be accompanied by clinoclase and olivenite. Their scarceness is explained by the diagram of thermodynamic stability of copper arsenates (Fig. 1) that indicates that both compounds form in a very narrow stability zone between clinoclase and olivenite. In addition, chemistry of mineralizing solutions may favour precipitation of either of them in a specific situation, and in the case of Rędziny it was cornwallite. Crystallization of cornwallite, associated with a gradual decrease of activity of  $\text{Cu}^{2+}$  ions and lowering of pH of mineralizing solutions, should result in formation of olivenite as the next mineral. In Rędziny, however, cornwallite is accompanied by strashimirite, occurring in a similar situation in Bulgaria, where it always was described within accumulations of tyrolite and cornwallite (Minčeva-Stefanova 1968). The studies on the stability of arsenate  $\text{Cu}^{2+}$  minerals (Magalhaes et al. 1988) indicate that strashimirite is most probably metastable in relation to olivenite. After all, it is usually found alongside olivenite, and in the Bulgarian occurrence it is present in a complex paragenetic sequence: cornwallite-olivenite-tyrolite-strashimirite-conichalcite. This sequence is similar to the one observed in Rędziny, with the exception that in Bulgaria strashimirite seems to be later than tyrolite (tyrolite is overgrown by strashimirite).

Observations made in Rędziny indicate that strashimirite is most probably later than cornwallite and metastable in relation to potential olivenite (strashimirite has been found with cornwallite only in spherulites occurring within tyrolite accumulations). A sudden change of geochemical conditions, associated probably with appearance of  $\text{Ca}^{2+}$  ions in the environment, closed the stage of formation of simple copper arsenates and commenced crystallization of tyrolite, considered either metastable in relation to conichalcite and olivenite or formed within the narrow stability field between the two minerals. As a result, the tentative path of evolution of copper arsenates in the Rędziny dolostones seems to be as follows:





The appearance of  $(\text{CO}_3)^{2-}$  ions in the final stages of secondary reactions resulted in bounding of the remaining amounts of copper in the form of common malachite.

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## STRASZIMIRYT I CORNWALLIT (ARSENIANY MIEDZI) Z RĘDZIN (DOLNY ŚLĄSK, POLSKA)

Straszczanie

W kopalni dolomitu w Rędzinach napotkano dwa rzadkie arseniany miedzi — straszimiryty i cornwallity. Minerale te występują w strefach mineralizacji kruszcowej, przede wszystkim w żyłkach tyrolitowych tworząc niewielkie, do około 1 mm wielkości, białe-zielone i jasnozielone sferolity, zbudowane z bardzo drobnych, włóknistych, poprzerastanych z sobą osobników o długości dochodzącej do 20  $\mu\text{m}$ . Identyfikacji obu minerałów dokonano na podstawie badań rentgenowskich oraz chemicznych. Skład chemiczny straszimiryty określony mikrosondą ekelektronową odpowiada jednostce formalnej  $(\text{Cu}_{3,90}\text{Mg}_{0,05}\text{Fe}_{0,03}\text{Ca}_{0,02}\text{Zn}_{0,01})[(\text{AsO}_4)_{1,99}(\text{OH})_{2,00}] \cdot 2,5\text{H}_2\text{O}$ ; minerał ten charakteryzuje się też typowymi wartościami stałych sieciowych:  $a = 9,719(2) \text{ \AA}$ ,  $b = 18,806(5) \text{ \AA}$ ,  $c = 8,937(3) \text{ \AA}$ ,  $\beta = 97,31(3)^\circ$ . Podobnie cornwallit nie odbiega chemicznie od składu teoretycznego, jego jednostka formalna jest następująca  $(\text{Cu}_{4,92}\text{Mg}_{0,02}\text{Fe}_{0,04}\text{Ca}_{0,03}\text{Zn}_{0,01})[(\text{AsO}_4)_{2,09}(\text{OH})_{3,67}]$ , a parametry komórki elementarnej są równe:  $a = 4,603(3) \text{ \AA}$ ,  $b = 5,785(3) \text{ \AA}$ ,  $c = 17,344(1) \text{ \AA}$ ,  $\beta = 91,66(5)^\circ$ . Obydwa minerały są produktami reakcji zachodzącymi pomiędzy jonami  $\text{Cu}^{2+}$  i  $[\text{H}_2\text{AsO}_4]^-$  obecnymi w strefie utlenienia rozproszonej mineralizacji siarczkowej i siarczkowo-arsenkowej znajdującej się w dolomitach rędzińskich.