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Tidal Flat Facies and Barite Mineralization in Gorski Kotar

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Barite mineralization in Gorski Kotar is a stratabound ore deposit conformably situated at the contact between the Permian and Lower Triassic sediments. On the basis of new field observations and detailed studies of former investigators, the writers propose tidal flat facies, accompanied by evaporative dolomitization as a millieu where early diagenetic barite mineralization took place.

Baritna rudna mineralizacija u Gorskom kotaru nosi oznake tipičnih uslojenih leži-šta. Konkordantno je uložena na kontaktu permskih i donjo trijaskih sedimenata. Na bazi novih terenskih zapažanja i detaljnih studija ranijih istraživača, predlaže se facies plimskih zaravni, sa pridruženom evaporitnom dolomitizacijom, kao sredina u kojoj je došlo do formiranja ranodijagenetske baritske mineralizacije.

INTRODUCTION

The purpose of this paper is to connect results of previous studies related to geology, mineralogy and genesis of the Gorski Kotar barite ore deposits and associated rocks with new field observations, which will be followed by appropriate laboratory elaboration.

The new evidence, gathered during the summer field campaign 1985, when authors reexamined recent and old mining sites and outcrops in the area of interest, motivated them to propose the tidal flat facies with related sedimentary processes as a framework for barite mineralization model. The model embraces all prerequisites to define genesis of the barite ore deposit, like: source of metal, way of transport and ore deposition, and formation time relative to the host rocks.

GEOLOGICAL SETTING AND MINERALIZATION

The Gorski Kotar barite deposits are situated along the skirts of the Upper Paleozoic terrain, adjacent to the contact of the Upper Paleozoic and the Lower Triassic sediments. The biggest deposits are in vicinity of Homer and Mrzle Vodice villages, while minor occurrences crop out between Zelin and Crni Lug locally.

The uppermost part of the Permian sediments consists of clastic deposits: shales, siltstones, and sandstones. They are followed by dolomites conformably, whose thickness varies between 5 and 10 m, and at places obtains values over 15 m. The dolomites are succeeded by micaceous dolomitic sandstones and then surmounted by typical Lower Triassic sandstones in continuation.

The barite deposits are interstratified in the lower part of the dolomite horizon, most often lying directly over the Paleozoic sediments. The deposits are equilateral, ranging in size from several m to 300 m. Thickness of the ore bodies varies between 1 and 3 m and sporadically reaches even 5 m. Marginal parts of the barite layers wedge out gradually or progress into sequence of smaller lenses or nests inside the dolomite. Contact barite-footwall rocks is sharp mostly, especially if barite rests on the Paleozoic clastics. In upward direction, across the hanging-wall dolomites, transition is gradual. The coarsegrained sandstones, beneath the barite layer at Mrzle Vodice locality, is pervaded by barite and dolomite matrix locally, whereas tiny quartz pebbles may be observed in the barite layer as well.

The richest ore body yield 70 to 90 % of barite. Its quantity in ore bodies, however, is usually much smaller and varies greatly (20 to 70 %). Poorer ores are lenses and irregular accumulations of barite, distributed over the dolomite without pattern. The dolomites in vicinity of ore bodies are finegrained with equidimensional grain size. Pyrite is abundant in deposits surrounding Mrzle Vodice.

CASE HISTORY

There have been many opinions about the age and genesis of Gorski Kotar barite deposits. Salopek (1960) attributed barites and associated dolomites to the Upper Triassic. Jurković (1959, 1962) considered the major ore deposit group to be of the Late Paleozoic age and of submarine-exhalative origin, while minor occurrences in upper parts of the basal dolomites as a metasomatic product of secondary-hydrothermal activity. Reliable age determination of the Lower Triassic dolomites with barite and overlain sandstones, as well as expressed suggestions regarding continuous transition between Paleozoic and the Lower Triassic strata (Ščavničar, B., Šušnjara, 1966; Ščavničar, B. 1973) assisted writers in envisaging of the proposed ore forming model and its age.

Šušnjara and Šinkovec (1973) stated that the basal dolomite belongs to a transitional zone between the Permian and the Lower Triassic and is an exclusive bearer of mineralization. Further on, the dolomites with barite were deposited in very shallow, partly or completely land-locked basins during regressive conditions.

Narrow range of $\delta^{34}S$ values indicates a homogeneous marine source of the barite sulfur probably of the Triassic age, (Šiftar, 1981).

Savić, et al. (1982) consider the basal dolomites with barites, overlying Paleozoic clastics, to be of the Upper Triassic age, while minor occurrences in limonites and beneath it belong to the Middle Permian time.

GEOLOGICAL, PALEOGEOGRAPHICAL AND PALEOCLIMATIC CONDITIONS

Middle and Late Permian sedimentation in Gorski Kotar is characterized by great quantity of clastic material, originating from newly exposed country areas, uplifted during Hercynian orogenic phases (Late Carboniferous, Middle Permian). It consists mostly of shales sandstones and conglomerates, deposited in a relatively shallow, epicontinental Dinaric sea, which was divided, according to Grubić, et al. (1973), into two parts, »inner« and littoral« by an elongated insular country. The progressive denudation reshaped cordilleras into less expressive relief with fully developed weathering processes. The sedimentary basins, filled up by persistently maturing clastics, grew shallower. The final member of the Upper Paleozoic clastic rocks are green-gray and gray-violet finegrained siltstones and pelites (Ščavničar, B., 1973).

Obliteration of the relief stopped clastic deposition, which along with concurrent change in climate, humid-arid, moderate-warm, triggered carbonate sedimentation all over Central and Southern Europe. Certain shift in time of that event from the Middle Permian till the Early Triassic, even at short distances, f. e. along the traverse Žirovski Vrh, Gorski Kotar, Velebit, may be well explained by slight differences in character of erosional base and depositional basin, type of weathering process and epeirogenic movements.

Since the Middle Permian time, inherited Hercynian lineament, with submeridional orientation had been evolving gradually, through geotectonic development into geological spaces characteristic for tectonic evolution over the Mesozoic and the Cainozoic eras, (Grubić, 1980). Gorski Kotar at that time was the westernmost part of the »Croatian through« representing a littoral zone of a vast country stretching across the northern Yugoslavia, as a part of the European continent.

Extensive low-lying coastal areas and warm-arid climate enabled immense evaporite deposition in lagoons, coastal sabkhas and inland sabkhas along the Zechstein sea from Ireland to Poland. The Dinaric Basin in its »inner« and »littoral« parts was under similar conditions, with ubiquitous evaporites, outcroping at many places in recent time (Samobor, Srb, Knin, Sinj, etc.), (Polšak, Pezdić, 1978; Herak, 1973, 1986).

Evaporite formation in a peritidal facies is an intricate mechanism, easily disturbed, obstructed, or even reversed by slight change in environmental conditions. On the other hand, dolomitization, as penecontemporaneous alteration of aragonite mud sediments by hypersaline ground waters, the best developed in embayments, lagoons and tidal flats, is preponderant process for recognition of the peritidal facies, due to dolomite chemical resistance and its durability in weathering cycles. According to Friedman and Sanders (1967), the most dolomite deposits in geological records owe their origin to this process and should be regarded as evaporite deposits.

Among several recent analogues, like: Coorong Lagoon-Australia, Bonaire-Netherlands Antilles, Bahamas, Southern Florida and the Persian Gulf, the last two would be the most appropriate in the considered model, since they belong to shelf-edge carbonate platform type, resting on continental, shelf bedrocks, preferentially clastics. The Persian Gulf sedimentary environment is compatible with the model even regarding climatic conditions.

All the Lower Triassic carbonate lithotypes with stratabound barite mineralization in Gorski Kotar adjacent to conformable contact with Permian clastics, originated in described conditions near mean sea level. It is a sedimentary millieu that comprises pertinent barite ore forming process as well.

A TENTATIVE SEDIMENTARY AND RELATED ORE FORMING MODEL

To stay in the scope of the article our attention will be paid to a particular geological situation in surrounding of Lokve and Mrzle Vodice villages with still current barite production.

In order to describe the model, a step-wise presentation has been chosen.

T h e f i r s t s t a g e responsible for barite mineralization formation began with clastic deposition during Late Permian time (Fig. 1a). The Upper Permian sediments (Š č a v n i č a r, B., 1973) underlying productive barite horizon are rhythmical alternations of sandstones, siltstones and shales with intercalations of fine to coarse-pebbled conglomerates. Their dark gray and at places almost black colour, in addition to abundant plant debris and ubiquitous pyritization point out reducing diagenesis but not necessarily euxinic bottom water conditions. On the contrary, some textual elements like grain size and flora remnants are indirect sign of shore-line proximity and its high energy water. Red, gray and green clastics, underlying the pyrite-bearing reduzate horizon at Glavica deposit might have witnessed influence of underground oxidizing terrestrial water, or could have even been exposed subaerially as country rocks during an earlier regression cycle. As a matter of fact, other characteristic of pyrite and barite mineralization and associated host rocks at Opaljenac and Glavica deposits bear some elements of Sabkha environment.

An abrupt change in sedimentary style, introducing carbonate deposition as a predominant process is t h e s e c o n d s t a g e of the model (Fig. 1b). Structural features of basal dolomites and other lithotypes like, cross-bedding, ripple marks and oolitization processes, indicate shallow-marine environment with agitated water (Š č a v n i č a r, B., 1973), (Fig. 1, Table I.). At Homer locality as yet undetermined framebuilding organisms founded first colonies, developing into a bioherm-like reef structure (Fig. 1, Table IV.), which would represent a preferable site for later baritization. At that time, the first carbonate sediments were calcareous, preferentially aragonitic in composition.

T h e t h i r d o r »m i n e r a l i z a t i o n« s t a g e is closely related to dolomitization process (Fig. 1c). The lime sediments deposited in near-shore, shallow water were soon subjected to subaerial evaporative pumping in a tidal flat environment during a regressive phase of a fluctuating sea level cycle. Early diagenetic dolomitization as a replacing process, caused by hypersaline brines with high Mg^{2+}/Ca^{2+} ratio, is followed by concurrent manifestations easily recognizable like:

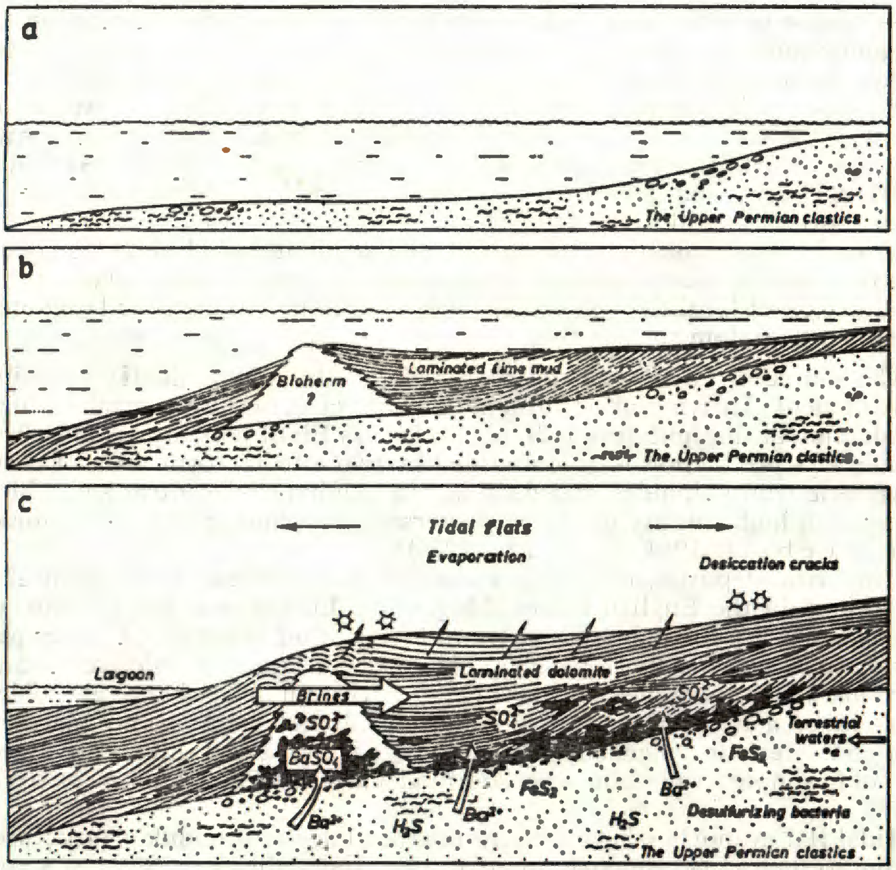

Fig. 1. Three stages of ore deposit development; a— deposition of the Upper Permian clastics, b — regression and sedimentation of laminated lime mud, c — evaporitic dolomitization accompanied by barite ore forming processes.

Sl. 1. Tri stupnja razvoja baritnog rudnog ležišta; a — taloženje gornjopermskih klastita, b — regresija i taloženje laminiranog vapnenačkog mulja, c — evaporitna dolomitizacija sa pridruženim rudonosnim procesima.

— significant increase in porosity of dolomite in comparison to aragonite precursor, due to remarkable difference in aragonite-dolomite specific volume. It facilitates any subsequent infiltration process, f. e. baritization,

— early diagenetic lithification, making dolomitized layers rigid, has characteristic geomechanical effect on underlying soft water-rich sediments. It is noticeable by frequent intraformational brecciation and slumping structures (Fig. 1, Table I.). Subsequent upward dewatering of underlain soft deposits may have certain importance in transport of barium into final resting mineralization place,

— upper surface of tidal flat sediments, exposed to intensive drying usually shows desiccation cracks, as discovered on the Lower Triassic clastic in terbeds (Babić, 1968),

— release of high strontium concentration during dolomitization of aragonite mud may be even sufficient for formation of celestobarite evaporitic deposits, as in the Central Europian Zechstein (Puchelt, Müller, 1964). Celestite may also replace an older generation of evaporitic minerals (West, at. al., 1968) or be an isomorphical substitute in barite minerals. Barite from Gorski Kotar is also rich in strontium (Šiftar, 1981). Šiftar and Šinkovec (1973) reported authigenic celestite as an accessory mineral in the same deposit,

— underlain reduzates are often former stromatolitical algal mat sediments. It still cannot be claimed that foot-wall, pyrite-bearing, dark layers at investigated localities are of that origin, although some fine laminated clastics at Kozolom suggest that.

The source of barium should be sought in this emphatically reducing environment. Thriving of desulfurizing bacteria upon burial enables high production of dissimilatory H_2S, which lowers Eh of connate waters. Profitable H_2S production is accompanied by two effects, formation of early diagenetic iron sulphides and decrease in sulphate concentration, which along with high salinity dissolves dispersed, tiny, authigenic barite minerals (Puchelt, 1964; Scherp, 1974).

Evaporitic deposits, related to dolomitization process, have been also discovered in the English Lower Magnesian Limestones (equivalent of Zechstein-Kalk). Stratabound barite mineralization outcrops at many places from Newcastle to Nottingham along the contact of calcareous and carbonaceous mudstones and overlying Zechstein 1 carbonates, which stretche as a narrow zone for a hundred kilometers. At some places it has characteristic epigenetic appearance, which is very illustrative and useful in supporting of our proposed model as well (Fig. 2), (Harwood, 1978).

Tidal flat evaporites have usually microcellular and nodular structures owing to their concretionary origin in tidal flat sediments. Similar structures have been discovered in massive barite ore at Homer locality (Fig. 2, Table I) with tiny pyrite impregnations and stylolites. It is still undetermined whether barite replaces former evaporites or its formation prevented evaporite growth.

Worth mentioning is also the outlook of the mineralized profile at Opaljenac locality (Fig. 1, Table II.). Underlain reduzate sandstone bears several percentages of framboidal pyrite, whose concentration increases toward the interface sandstone-dolomite. Close to the contact it is almost massive pyrite (Fig. 2, Table II.), gradually incorporating barite microcells and thin layers of barites and passing into barite-dolomite ore. The interface is a boundary between low Eh—pH and high Eh—pH environments. Porous sandstones served as a good conduit for mineralizing solutions carrying Ba^{2+} ions. This appearance, together with some other features like, stylolite cuttings of dolomite-barite mottled ores as a late diagenetic feature and dolomite-barite lamellar ore structures at Homer (Fig. 1, 2, Table III.), proves early diagenetic origin of mineralization, as well as continuous sedimentation through Late Permian-Early Triassic time.

Higher concentration of manganese, now in form of encrustations in gossans at Opaljenac, consisting of barite-bearing hypergene limonites and severaly weathered dolomite (due to acid weathering processes) originated

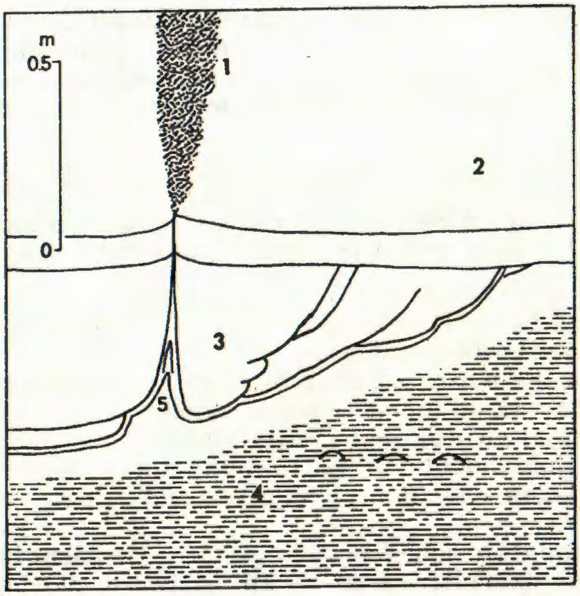

Fig. 2. Stylised drawing of a vertical section exhibiting barite ore appearance in the Lower Magnesian Limestones (English equivalent of Zechstein-Kalk); 1 — barite breccia, 2 — thickly bedded dolomite, 3 — dolomite channel cut into, 4 — calcareous mudstones, 5 — penetration flame structure of the ore body, (after Harwood, 1978).

Sl. 2. Shematski crtež pojavljivanja baritne mineralizacije u Donjem magnezijskom vapnencu (ekvivalent Zechsteinskog vapnenca u Engleskoj); 1 — baritna breča, 2 — debeli uslojeni dolomit, 3 — kanal u dolomitu usječen u vapnenačkom muljavcu, (4) — 5 — penetraciona »plamena« struktura rudnog tijela, (prema Harwood, 1978).

from the same barium and H_2S rich reducing solutions, since Mn^{2+} and Ba^{2+}, in contrast to chalcophile element ions, are mobile in such circumstances.

The same holds true for mineralized biolithite at Homer locality (Fig. 1, Table IV.), which served as conduit and infilling space, nourished by Ba-rich solutions. The brines were expelled from a soft, sulphate-poor, underlain mud by heavy load of rigid carbonate layer and evaporative pumping during subaerial exposure. Big euhedral barite minerals (Fig. 2, Table IV) are characteristic for growth in voids and vugs of frame-work structures (W a l t e r, 1976).

The last but not the least important support for the model is sulfur isotopic data obtained by Š i f t a r (1978, 1981) and Š i f t a r and S r z i ć (1981). A rather narrow range of $\delta^{34}S$ values indicate homogeneous source of the sulphate sulfur, corresponding to the composition of the Early Triassic sea water sulphates. The westernmost barite occurences in the surrounding of Mrzle Vodice show slightly increased $\delta^{34}S$ for sulphides and sulphates and negative correlation with Sr content. According to Šiftar, this phenomenon might be ascribed to conditions encountered in a rather closed system. Appearance of heavier sulphate-sulphide sulfur isotopes might be also explained by Sabkha sulfur isotopic fractionation (Wedepohl, 1971). Heavier sulfur in sulphate-sulphide pairs, red clastics underlying dark pyrite-bearing sandstones are characteristics of landward Sabkha flats which are under influence of terrestrial water.

CONCLUSION

In order to summarize, presented ideas on the sedimentary and ore forming model a short review of important events in successive order is given:

— deposition of Upper Permian clastics
— regression and carbonate deposition
— lowering of the mean sea level by evaporative drawdown and exposure of lime mud to intensive evaporation
— dolomitization, early diagenetic lithification
— loading of rigid dolomitized horizon on a soft reduzate sediment, rich in water, where reduction of sulphate and release of barium takes place
— expulsion and diffusion of Ba-rich, sulphate deficient brines into overlying porous dolomite, soaked with landward moving, hypersaline, lagoonal brines rich in sulphates, caused by overburden pressure and evaporative pumping
— barite precipitation in porous dolomite, possible replacement of former evaporites, and concretionary growth.

Stylolites as a late diagenetic feature crossing through dolomite-barite patches in mottled ores, nodular and microcell barite structures and laminated dolomite-barite ores point out early diagenetic origin. Barium is remobilized in underlain, reduced sediments by bacterial activity and lateral secretion mechanism and then precipitated in dolomites again. Clear stratabound relationship of the mineralization with the sharp boundary »clastics-dolomites«, which represents geochemical barrier low Eh—pH and high Eh—pH, as well as its wide lateral extention support this statement. High energy water, reflected in sedimentary structures in basal barite-bearing dolomites, dismisses possibility of direct barite precipitation in the sea water and preservation of barite laminae in intact position.

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Facies plimskih zaravni i baritska mineralizacija u Gorskom Kotaru

L. A. Palinkaš, B. Šinkovec

U kratkom prikazu iznesene su osnovne ideje na kojima se bazira sedimentni model sa pridruženim procesima stvaranja rudne mineralizacije. U tu svrhu su navedena glavna zbivanja u tekućem vremenskom redoslijedu:

— taloženje gornje permskih sedimenata, sa dispergiranim barijem,
— regresija i taloženje karbonata,
— spuštanje srednjeg nivoa mora u toku evaporitne aktivnosti i izlaganje karbonatnog mulja intenzivnoj evaporitnoj transpiraciji,
— dolomitizacija i rano dijagenetska litifikacija,
— opterećenje mekanog, vodom bogatog mulja, u kojem se reduciraju sulfati a oslobađa barij, sa poluočvrslim dolomitiziranim horizontom,
— istiskivanje barijem bogatih slanih otopina u nadležeće dolomite, kroz koje cirkuliraju slane, lagunarne vode bogate sulfatom, zbog gravitacijskog opterećenja i evaporitnog pumpanja,
— precipitacija barita u poroznom dolomitu i konkrecijski rast.

Na ranodijagenetski postanak mineralizacije ukazuju stiloliti (kasno-dijagenetska karakteristika) koji presjecaju dolomitno-baritne nakupine u uprskanoj rudi, zatim nodularne i mikročelijske baritne teksture te laminirana dolomitno-baritna ruda. Barij je remobiliziran u podinskim klastitima pomoću bakterijske aktivnosti i mehanizma lateralne sekrecije te precipitiran u nadležećim dolomitima, što se očituje u jasnoj slojnoj povezanosti mineralizacije i njezinoj pridruženosti kontaktu »klastiti-dolomiti«, koji predstavlja geokemijsku barijeru tipa »niski Eh—pH, visoki Eh—pH«.

Visoka energija vode, koja se ogleda u karakterističnim sedimentnim teksturama u dolomitima s baritom, kao na primjer kosa laminacija, valni tragovi (ripple marks) te izražena oolitizacija, isključuju mogućnost direktnog precipitiranja barita iz morske vode i sačuvanja baritnih lamina u intaktnom stanju.

TABLE — TABLA I

Fig. 1. Early diagenetic dolomitization, caused by evaporative transpiration in tidal flats, induced partial stiffening of newly formed dolomites in a vadose zone. Heavy load of overlying carbonate rocks on soft water-rich mud deposits deformed primary structures and brought about intraformational slumping. At the left and right of the picture are typical crossbedding structures.

Sl. 1. Rano dijagenetska dolomitizacija, uzrokovana evaporitnom transpiracijom u plimskim zaravnima, dovela je do djelomičnog ukrućivanja novo-formiranih dolomita u vadoznoj zoni. Pod opterećenjem iznad ležećih karbonata na meko, vodom bogato blato, došlo je do deformacije primarnih struktura i intraformacijskog slamanja. Na lijevoj i desnoj strani slike vide se tipične teksture kose laminacije.

Fig. 2. Microcellular and nodular structures typical for evaporites, in this case barite, growing in tidal flat sediments. Homer locality.

Sl. 2. Mikrocelularna i nodularna tekstura evaporita, u ovom slučaju barita, karakteristična za konkrecioni rast u sedimentima plimskih zaravni. Homer lokalitet.

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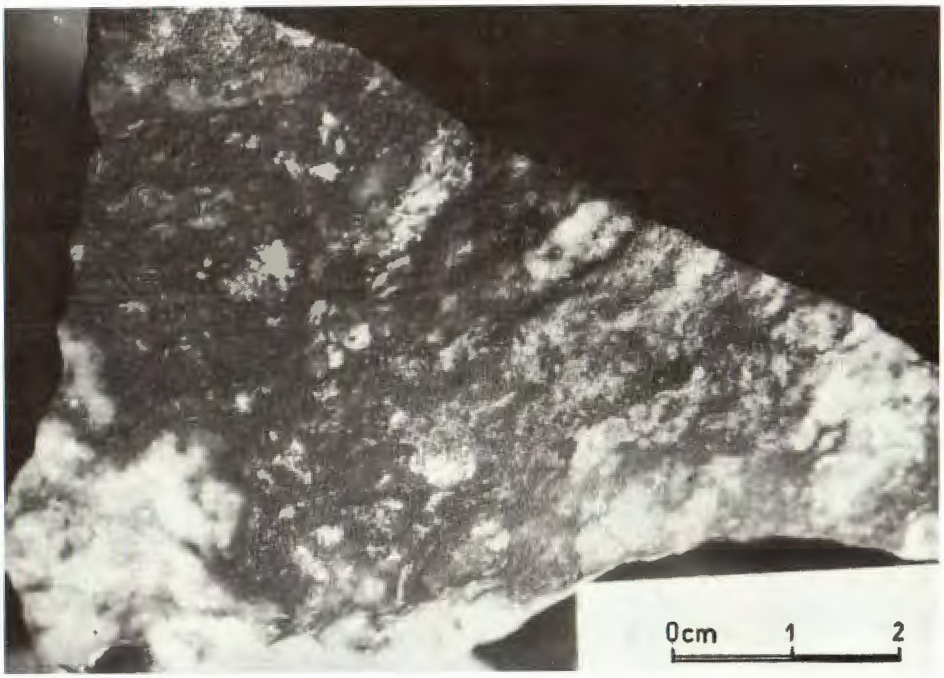

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TABLE — TABLA II

Fig. 1. Gossan of limonite, corroded dolomite, barite and manganese encrustation. It was a former massive pyrite-barite ore as may be seen at less weathered parts of the outcrop. Underlying sediments are pyrite-bearing reduzate sandstones, occasionally pervaded by barite cement too. Opaljenac locality.

Sl. 1. Željezni šešir od limonita, korodiranih dolomita, barita i manganskih prevlaka. To je produkt oksidacije masivne piritsko-baritsko-dolomitske rude što se može uočiti na manje trošnim dijelovima šešira. Podinski piritizirani pješčenjaci su na nekim mjestima prožeti baritskim cementom. Lokalitet Opaljenac.

Fig. 2. Massive pyrite and barite interlayers and microcells discovered in less weathered parts of the gossan.

Sl. 2. Masivni pirit sa baritnim proslojcima i mikročelijama, otkriven u manje trošenim partijama željeznog šešira.

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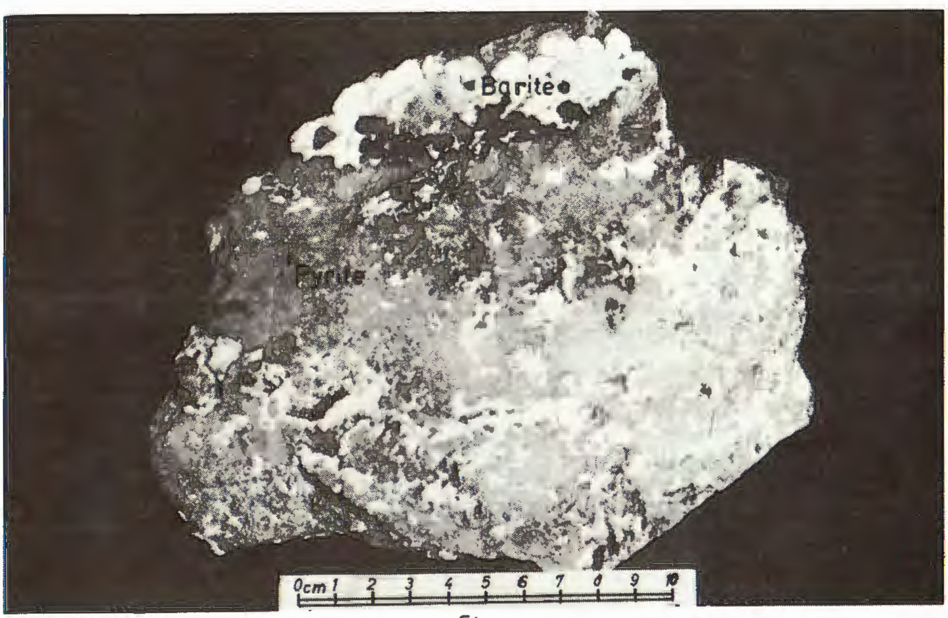

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TABLE — TABLA III

Fig. 1. Stylolites, as a late diagenetic feature, passing through patches of dolomite and barite in mottled ores (upper part of the basal dolomites) is a clear sign of early diagenetic origin of mineralization.

Sl. 1. Stiloliti, (kao kasno dijagenetska karakteristika), presjecajući nakupine barita i dolomita u gornjem dijelu bazalnih konglomerata, ukazuju na rano dijagenetsko porijeklo mineralizacije.

Fig. 2. Laminated dolomite-barite ore. Lamination is often present in lower parts of the basal dolomites, where other high energy water sedimentary structures like cross-bedding and ripple marks are well developed.

Sl. 2. Laminirana dolomitno-baritna ruda. Laminacija je često prisutna u donjem dijelu bazalnih dolomita gdje se susreću različite sedimentne teksture karakteristične za visoku energiju vode, kao kosa laminacija i valni tragovi.

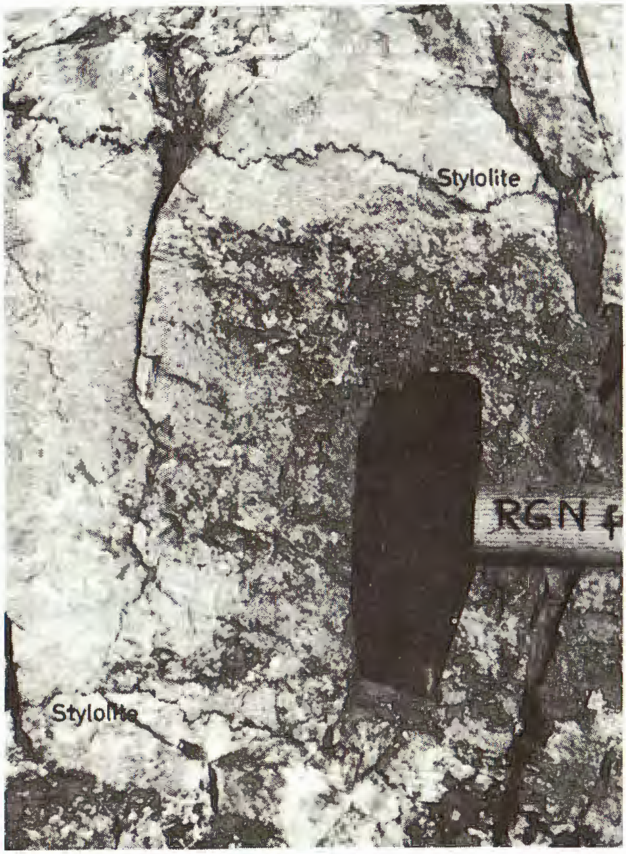

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TABLE — TABLA IV

Fig. 1. Biolithite of as yet undetermined origin, served as a favorable space for barite ore deposition. Homer locality.

Sl. 1. Biolitit nepoznatog porijekla, na lokalitetu Homer, poslužio je kao naročito pogodno mjesto za odlaganje baritne rude.

Fig. 2. Big euhedral, finger — thick barite crystals typical for growth in open vugs and voids of framework structures. This is a rich ore in bioherm-like rocks.

Sl. 2. Veliki, idiomorfno razvijeni kristali barita, karakteristični za rast u slobodnom prostoru rešetkastih biostruktura.

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