MG-MINERALS OF RECENT HYDROTHERMAL FORMATIONS OF THE CU-PORPHYRIC MINERALISATION AT RECSK, HUNGARY

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ABSTRACT

A great variety of minerals precipitated from thermal waters (35—44 °C) were observed in the drifts of the copper-porphyritic skarn ore deposit at Recsk. Beside calcite-aragonite and various salts with different solubility, the Mg-minerals (nesquehonite—dypingite—hydromagnesite—giorgiosite-northupite—astrakhanite—/vanthoffite; langbeinite/—epsomite—palygorskite) have outstanding importance at this locality. The present work focussed on the study and interpretation of the hydrothermal formation of the Mg-minerals otherwise known as hypergenitic evaporites.

AUSZUG

In den Stollen des porphyrischen-skarn Cu-Erzlagers in Recsk ist eine ausserordentlich grosse Variation von hydrothermalen (35—44 °C) Mineralienabsonderungen vorhanden. Unter den Kalzit-Aragonit-, und anderen Mineralsalzen von veränderlicher Lösbarkeit spielen die Magnesiumminerale (Nesquehonit—Dypingit—Hydromagnesit—Giorgiosit—Northupit—Astrakhanit—/Vanthoffit; Langbeinit/—Epsomit—Palygorskit) eine beachtliche Rolle. In dieser Arbeit wird die hydrothermale Entstehung dieser, von anderen Lokalitäten meistens als hypergen-evaporitischen Phasen beschriebenen Mg-Mineralien untersucht und erklärt.

INTRODUCTION AND REVIEW OF CHEMICAL AND PHYSICAL CIRCUMSTANCES

As the Au-enargite and luzonite deposits of Recsk became barren, in the vicinity a new and intense exploration campaigne was initiated by KISVARSÁNYI, G., KISS, J. in 1953—55 years. As a result, several new ore deposits were discovered; polymetallic ore formations (PbS—CuFeS2—FeS2) in the Middle-Mátra Mts. (Parádsasvár), Au-Mo(Re) containing Cu-porphyritic mineralisations, CuFeS2—FeS2 skarn formations and a ZnS—PbS—CuFeS2 polymetallic ore deposit in the deep drillings of the Recsk area going down to over 1200 m. The later mineralisation are associated with Paleogene—Eocene (Sr⁸⁷—Sr⁸⁵=0.705/0.708) contaminated (dioritic-andesitic) magmatic intrusions (endoskarn), metasomatic contact of Permain-Triassic limestone and dolomite (grossularite-hybschite-andradite-diopside-pyroxmangite-epidote-actinolite-tremoli te-antigorite-wollastonite-quartz-calcite exoskarn) as well as their calcareous foliated rocks respectively. A substantial portion of these deposits has not been limited yet towards the depth yielding an estimated exploitable mineral stock of well over 100 million tons (mean values = 1.30 to 1.70 % Cu).

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Thermal springs, also utilized as curative waters, of the near and far vicinity of Recsk (Bükkszék 37 °C, Demjén 69 °C) and a great number of methan-radon containing alkaline hydrocarbonic thermal water occurrences at Mátraderecske, Mátraballa, Parádfürdő and Parádsasvár indicated, that intensive hydrothermal activity should be expected during the mining process of the aforementioned mineral deposits. (Several thermal water wells, some of them extremely (80—90 °C) hot, are known in the Pannonian Basin and also a 180 °C hot saline-carbon-dioxid steam outburst was found near Fábiánsebestyén, (Fig. 1a).

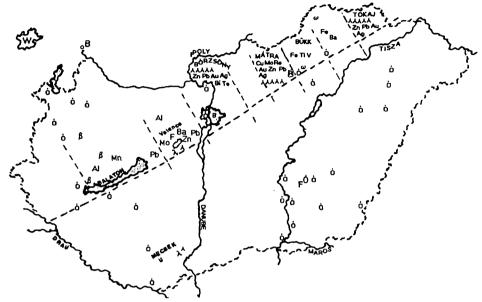


Fig. 1-a. A sketch of metallogeny of Hungary showing several important thermal water wells and springs.

Legend: R: Recsk A: granite (a) gabbro, webslite fit basalt (c) and site (c) thermal waters

Legend: R: Recsk, λ: granite, ω: gabbro, wehrlite, β: basalt, α: andesite, ό: thermal waters, FO: Fábiansebestyén well with 180 °C water

In two of the drifts driven into the ore deposit, 690 and 890 m below the sea-level respectively, gaseous (CO₂-methan) thermal water (35—44 °C) outbursts occur with various water production. A study of their chemical composition and of their spectacular mineral precipitations might yield valuable data for the interpretation of the hydrothermal process.

The chemical composition and the temperature of these thermal waters are obviously affected by the lithological environment (e.g. water from limestone-dolomitic rock is rich in Mg—Ca—Cl, while those coming through volcanic and skarn formation skarn formations have higher alkali (Na⁺>K⁺) and trace element (Pb—Zn—Cu) content). Table 1. summarizes the chemical composition of these waters, the maximum and minimum ionic concentrations (a), their mean values (b) and the gross ion content (x).

An active hydrothermal solution is a pH 6.4—7.8 system with major Na—Ca—Mg chloro-hydrocarbonatic-sulphatic, and minor K⁺—NH₄⁺—Fe²⁺ content. Besides free CO₂ it may also contain methane and radon gases.

Thermal waters of the deeper drifts (-890 m) have higher mean Ca^{2+} — Fe^{2+} — NH_4^+ and SO_4^{2-} ion content, while on the upper levels (-690 m), due to the mineral

TABLE 1

Chemical composition of the epithermal soutions at the -690 m and -890 m levels

| Sample | | Gallery (m) | | Cation | Anion | pH | Ca ²⁺ | Mg ²⁺ | Fe ²⁺ | NH4* | Na* | K⁺ | (HCO ₃) | (SO ₄) ⁻ | CI. |
|---------------|------------|----------------|------------|----------------------|----------------------|---------|------------------|------------------|------------------|-----------|-----------|---------|---------------------|---------------------------------|-----------|
| (No) | _ 1 | | | mg/l | | pri | mg/l | | | | | | | | |
| 1-44 (29) | 35—44 ℃ | -690 | (a) (b) | 3010-6662 3526.6 | 5319-12106 7302.8 | 6.4-7.5 | 76-5900 | 164-717 | 0.0-0.20 | 18.0-110 | 1800-3600 | 10-193 | 48.8-5392 | 138-1085 | 195-11920 |
| | 1 | | (b) | 10829.4 (x) | | 6.8 | 590 | 265.3 | 0.08 | 75.1 | 2465 | 107 | 3545 | 927 | 2792 |
| 47-71 (18) | 35—44 ℃ | -890 | (a) (b) | 3700-6440 4764.04 | 4105-9153 7097.1 | 6.5-7.8 | 40-664 | 14.6-551 | 0.0-0.20 | 14.5-65.8 | 3000-5400 | 118-145 | 1756-5051 | 420-768 | 1580-4600 |
| | (57 °C) | | (b) | 1186 | 1.1 (x) | 7.3 | 290 | 306.2 | 0.03 | 35.9 | 3970 | 127 | 3626 | 564 | 2910 |

a = Deviation; b = Mean; $(x) = \Sigma$ ion

precipitations in the course of the 200 meters long migration, the water is a relatively dilute Na⁺-Mg²⁺-K⁺ chloro-hydrocarbonic and sulphatic solution.

The Br content of the surface occurrences is 11.5 mg/l.

The dripstone formations precipitating from these waters transform in a relatively short time the mining tunnels into "caves of the fairy tales". These dripstone precipitations are usually rich in forms; there are stalagmites, stalactites, ooides, oolites, pisolites, coral and cole-flower-like bizarre clusters and coatings. The precipitations larger in size have rhythmic band-like structure. Some of their inner layers are brownish-black coloured by mineral oil derivatives (hydrocarbons) while the surface is sometimes yellowish-red (goethite-"hydrogoethite") or green, greenish-blue (malachite-brochantite-atacamite-spangolithe).

Similarly to the chemical composition of the thermal waters, the mineral composition and the paragenetic sequence of the dripstones of the deeper (-890 m) and upper (-690 m) drifts are different (Table 2). On the basis of their mineral composition the dripstones could be divided into nine groups and within these

/a/ on the upper levels (-690 m) the calcite-aragonite composition with minor

Mg-mineral constituents is characteristic

Paragenetic sequence of the dripstones minerals at the -690 m and -890 m levels

| GALLERY | Paragenesis | | | | | | |
|---------|---|--|--|--|--|--|--|
| | PRINCIPAL MINERALS: [Calcite > Aragonite] > Gypsum > NaCl > (Nesquehonite>> Thenardite > Northupite > Dypingite, α Sulfur | | | | | | |
| -690 m | ACCESSORY MINERALS: Arcanite, Brochantite, Botallackite, Burkeite, Giorgiosite, Hidromagnesite, Mirabilite, Metathenardite, Glaserite, Palygorskite, (Parasepiolite), Saponite (x), Sepiolite (x), Tschermigite, (?= Antigorite (x), Glauberite, Hydrohalite, Kaolinite (x), Zeolithe (x) | | | | | | |
| | PRINCIPAL MINERALS: Nesquehonite > (Dypingite-NaCl) > Northupite + (Aragonite > Calcite) > Thenardite > Astrakhanite (Blödite)>(Gypsum + α Sulfur) | | | | | | |
| -890 m | ACCESSORY MINERALS: Arcanite, Atacamite, Brochantite, Botallackite, Chalcantite, Cristobalite (x), Epsomite, Gaylussite, Goldichite, Hydrogoethite, Langbeinite, (Parasepiolite) (x), Pirssonite, Saponite (x), Sepiolite (x), Spangholite, Stevensite? (x), Syngenite, Vanthoffite | | | | | | |
| | Botallackite= Cu ₂ (OH) ₃ Cl, Burkeite= Na ₆ [CO ₃ (SO ₄) ₂], Goldichite= KFe(SO ₄) ₂ .4H ₂ O Spangolithe= Cu ₆ Al[(OH) ₁₂ Cl,SO ₄].3H ₂ O ?=Alunite, Dickite, Hexahidrite, Eriochalcite= CuCl ₂ .H ₂ O (x) = Insoluble remain in acid | | | | | | |

/b/ the aragonite-calcite dripstones of the deeper drifts may be characterized by more intense Mg-mineral (nesquehonite-dypingite-northupite-astrakhanite) precipitations. Considerably less frequent is the occurrence of NaCl-thenardite-gypsum and alpha-sulphur precipitations.

THE MINERALS

The present study focuses particularly on the Mg-minerals because of their special mineral paragenetic importance. These minerals are:

Nesquehonite MgCO₃.3H₂O

Dypingite $Mg_5[(OH)_2(CO_3)_4]$. $5H_2O(8H_2O)$

Hydromagnesite Mgs[(OH)(CO₃)₂]₂.4H₂O Giorgiosite Mgs(CO₃)₄(OH)₂.5H₂O

Northupite Na₃Mg[Cl(CO₃)₂]
Astrakhanite (Blödite) Na₂Mg(SO₄)₂.4H₂O

Vanthoffite $Na_6Mg(SO_4)_4$ Langbeinite $K_2Mg_2(SO_4)_3$ Epsomite $MgSO_4.7H_2O$

Palygorskite (Mg,Al)₂[(OH)Si₄O₁₀].4H₂O Sepiolite Mg₄[(OH)₂Si₆O₁₅].6H₂O

(Parasepiolite)

Saponite (Mg>Fe)3(O.5Ca,Na)0.33

 $[(OH)_2(Al,Si)_4O_{10}].x(H_2O)$

Nesquehonite (MgCO₃.3H₂O)

The nesquehonite is the most frequent Mg-mineral of the thermal waters at Recsk. Its several generational appearances can be traced as follows:

/a/ in the ooides it appears in the form of lathy crystals with radial ordering $(400-500 \mu m)$. On its upgrown crystals terminating forms can also be recognized (*Plate I. Figs. I, 2*),

/b/ on the surface of aragonite-calcite dripstones it forms powder-like but nodular coatings.

/c/ the single phase Ne ooides are sometimes terminated by a dypingite band

sharply separated from the nesquehonite bulk (*Plate I. Fig 3*),

/d/ the crystallites of the wheat-white powder-like nesquehonite grains and spores are idiomorphic-hypidiomorphic needle-like accicular formations (Plate

I-II, Figs. 4, 5) here and there holey skeleton crystals (Plate II, Fig.6),

/e/ crystallites showing signs of recrystallization or resolution according to the pattern of decrescence can often be observed (*Plate II*, Fig. 7). Under laboratory conditions at room temperatures in distilled water a sample consisting of Ne>Dy (-890 m) transformed into pure dypingite in seven days. This recrystallization into dypingite goes together with the appearance of pseudohexagonal-monoclinic (orthorhombic?) crystallites. It is possible that the morphology of the dypingite phase formed by the recrystallization of the nesquehonite is significantly different from that of the primary modification:

$$5(MgCO_3.3H_2O)+xH_2O=Mg_5[(CO_3)_4(OH)_2].5H_2O(8H_2O)+(H_2O+CO_2)+xH_2O$$

According to the literature, nesquehonite primarily is a crack filling mineral of serpentinized alkali magmatites; sometimes appears as thin layers in asbestos, magnesite and coal-antracite deposits; precipitations of saline lakes or disseminations and crack fillings in dolomites of tropical territories.

Dypingite $[Mg_5(CO_3)_4(OH)_2.5H_2O(8H_2O)]$

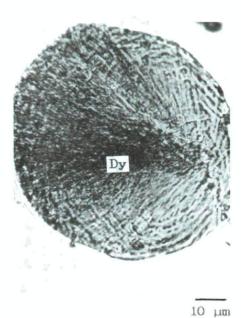


Fig. 1-b. Microphotograph of a nesquehonite (Ne) ooide with dypingite (Dy) crystallites on its surface

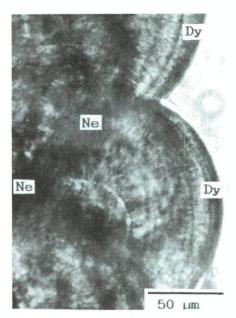


Fig. 3. Microphotograph of a nesquehonite (Ne) — dypingite pisolite. The rhythmic band-like texture is mainly nesquehonite, the outer surface layer is dypingite

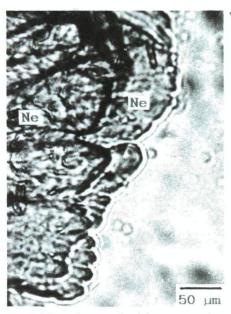


Fig. 2. Microphotograph of the cross section of a nesquehonite (Ne) pisolite. Several hypidiomorphic crystals are overgrown on each other in a layer-like manner.

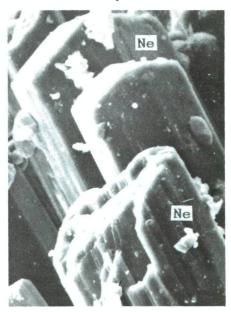


Fig. 4. SEM picture of idiomorphic nesquehonite (Ne) crystallites [(011), (010), (110)]. The grains on their surface with anhedral morphology are supposedly hydromagnesite.



Fig. 5. TEM photograph of a nesquehonite (Ne) ooide.



Fig. 7. TEM photograph showing the resorption-recrystallization formations of the single phase nesquehonite (Ne).

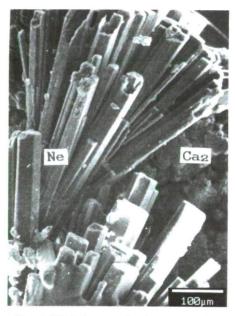


Fig. 6. SEM photograph of upgrown idiomorphic nesquehonite crystallites and skeleton crystals on the surface of an aragonite dripstone.



Fig. 8. TEM photograph of dypingite crystallites terminated by (hk0) (hkl) planes. Some clusters of alpha-sulfur (S) can also be seen.

Several generational appearances of dypingite can be recognized at Recsk (Plate I. Figs. 1 and 3) as follows:

/a/ the dypingite forming the outer layer of some nesquehonite ooides (Plate

I. Fig. 3) is a primary formation,

b/ the pseudomorphic dypingite after nesquehonite is of epigene origin,

/c/ fine needle-like accicular upgrown crystallites on the surface of nesquehonite-aragonite (Plate II, Fig. 8),
/d/ dypingite phase identified within some nesquehonite ooides by X-ray

diffraction, TEM and SEM.

The dypingite was first described from a magnesite deposit (Dyping-dal, Snarum, Norway) together with hydrotalcite [Mg₆Al₂(OH)₂(CO₃).4H₂O], marassite [Mg₆Al₂(OH)₂(CO₃).4H₂O], szájbelyite [Mg₂(B₂O₅)H₂O] and haematite (Fe₂O₃). It was also found in a Japanese locality in the company of chrysotile, nesquehonite, brugnatellite [Mg₆Fe³⁺(OH)₁₃(CO₃).4H₂O] and pyroaurite [Mg₆Fe₂(OH)₁₆(CO₃).4H₂O]. The structure of dypingite, its hydrothermal formation (together with nesqehonite) is the first such observation, its structure has not been completely clarified yet.

Hydromagnesite Mg₅[(CO₃)₂OH]₂.4H₂O.

It appears together with nesquehonite-dypingite in the form of well defined crystallites (Plate III, Fig.9).

Giorgiosite: $Mg_5(CO_3)_4(OH)_2.5H_2O$

Its presence in the powder-like coatings on the surface of the aragonite-calcite dripstones was identified by X-ray diffraction together with nesquehonite-dypingite-hydromagnesite as well as northupite.

Northupite [$Na_3Mg\{Cl, (CO_3)_2\}$]

It is the most characteristic mineral of the Recsk thermal waters. At least two generations of it can be recognized:

/a/ idiomorphic octahedral crystals that can directly be derived from the

thermal water (Plate III, Figs. 10, 11 and 12).

/b/ hypidiomorphic-anhedral crystallites which could form with the reorganization of nesquehonite under the influence of saturated NaCl solutions in an epigene way. Its formation may be explained by the following chemical process:

$$2(MgCO_3.3H_2O)+3NaCl=Na_3Mg[Cl, (CO_3)_2]+MgCl_2+6H_2O$$

A portion of the chlorine might be substituted by (SO₄)²- but the occurrence of the chlorine free tychite Na₆Mg₂[SO₄, (CO₃)₄] at Recsk has not been observed yet. The clear transparent crystals of northupite have low refractive index (n=1.51-1.53), are 60-100 μ m in size and crystal forms of (100) and (111)-(110) can be recognized on them (Plate III, Figs. 11 and 12). After the morphology it can not be distinguished from tschermigite (n=1.46—1.47) [NH₄Al(SO₄)₂] which usually occures as grains 20—40 µm in size. These phases were identified by X-ray diffraction, and further confirmed by the presence of NH₄ and Al.

The northupite was originally found on the surface of karstic dolomites overflooded by hypersaline solutions of Tchad (Africa). It was observed together

with barringtonite and nahcolite; its likely formation process is

$$MgCO_3.2H_2O+2NaHCO_3+NaCl=Na_3Mg[Cl, (CO_3)_2]+H_2O$$



Fig. 9. Hydromagnesite

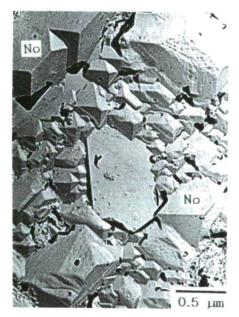


Fig. 11. TEM picture of a cluster of idiomorphic northupite (No) crystals with resorption formations and (111) twinning.

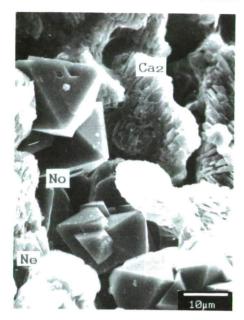


Fig. 10. SEM photograph of northupite clusters in the company of nesquehonite (Ne) and aragonite.

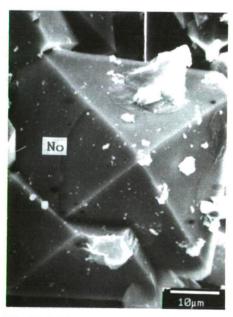


Fig. 12. SEM photograph of a northupite octahedron. The anhedral grains on its surface are hydromagnesite (?) and giorgiosite (?).

This rare mineral was found in clay formations of borax- and salt lakes, in oil shales of California, Wyoming in the company of shortite-trona-bradleyite-pirrsonite and gaylussite.

Astrakhanite (Blödite, Simonyite) = $Na_2Mg(SO_4)_2.4H_2O$

For the time being it is a nearly single phase polycrystalline formation in the deeper (-890 m) drifts. It appears in megascopic nodular formations as well as in film-like coatings. It can be observed in differentiated distribution on the surface of calcite-aragonite dripstones in the company of:

/a/ halite-brochantite-gypsum

/b/ halite-goldichite [KFe(SO₄)₄.2H₂O]-brochantite-arcanite

/c/ halite-nesquehonite-northupite

/d/ halite-gaylussite-pirssonite

The single phase astrakanite is a polycrystalline formation of hypidiomorphicanhedral grains with a "psammitic" texture. Some rare sulphates and carbonates may accompany it. The formation of these rare phases may be explained by a possible recrystallization initiating effect of the epitherms on the astrakhanite:

 $Na_2Mg(SO_4)_2.4H_2O+[Ca^{2+}-K^+-NH_4^+-CO_3]_{aa}$ = Pi, Ga-Th-Mi-La-Sy-Va-Gi+X+Y

Pi (Pirssonite)=Na₂Ca(CO₃)₂.2H₂O

Ga (Gaylussite) = Na₂Ca(CO₃)₂.5H₂O

Th (Thenardite) = Na₂SO₄

Mi (Mirabilite) = Na₂SO₄.10H₂O

La (Langbeinite) = $K_2Mg(SO_4)_2$

Sy (Syngenite) = $K_2Ca(SO_4)_2.H_2O$

 $Va (Vanthoffite) = Na_6Mg(SO_4)_4$

 $Gi (Gypsum) = CaSO_4.2H_2O$

 $X = MgSO_{4(ag)}, MgCl_{2aq}$ $Y = \text{hypothetic phases} = KH(SO_4) \text{ (Mercallite)}$

K₈H₆(SO₄)₇ [Misenite]

 $(NH_4)_3Na(SO_4)_2 - [NH_4$ - Glaserite]

See them in Plate IV—V.

Vanthoffite $[Na_6Mg(SO_4)_4$ and Langbeinite $[K_2Mg(SO_4)_2]$

Nearly isometric water-clear grains, crystallites are on the surface of astrakhanite.

Epsomite (MgSO₄.7H₂O)

Its presence was diagnostized by X-ray diffraction in the dripstones of the -690 m and -890 m drifts and it was accompanied by gypsum-glauberite-mirabilite-syngenite or sometimes by burkeite and goldichite.

Palygorskite (sepiolite-parasepiolite-saponite)

It is a characteristic accompanying mineral of the polymetallic ore veins of the Mátra Mts. (Parádsasvár, Mátraszentimre). At Recsk it was identified in the acid insoluble residue of the outer-edge crystalline limestone (exoskarn) as well as of the dripstones in the company of quartz-opal-kaolinite-zeolite(?). Its crystals are fine feather-like formations (Plate IV, Fig. 14) or fibrous constituents of



Fig. 13. TEM picture of anhedral astrakhanite (blödite) grains with psammitic texture.



Fig. 14. TEM photograph of some feather-like palygorskite (Pa) crystallites. Acid insoluble residue of a calcite-aragonite dripstone.

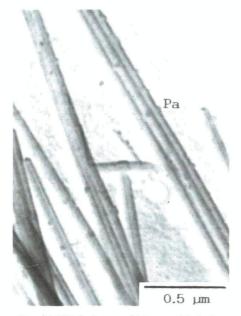


Fig. 15. TEM picture of idiomorphic lathey palygorskite crystals with oriented intergrowths (Parádsasvár).

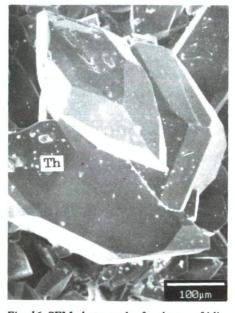


Fig. 16. SEM photograph of a cluster of idiomorphic thenardite (Th) crystals.

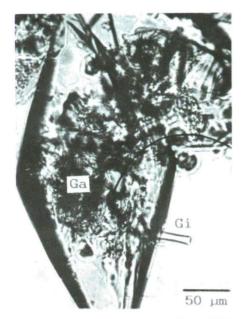


Fig. 17. Microphotograph of gaylussite (Ga) in the company of gypsum (Gi).

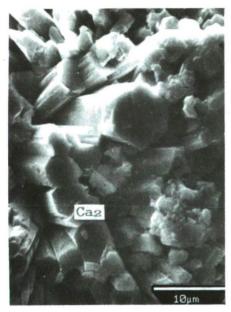


Fig. 19. SEM photograph of upgrown aragonite (Ca₂) crystallites and cyclic twins on the surface of a calcite dripstone.

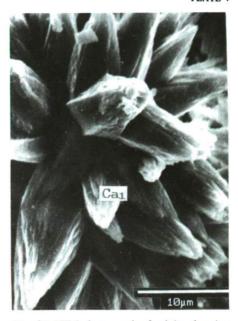


Fig. 18. SEM photograph of calcite showing (4041) and (hkil) forms.



Fig. 20. Microphotograph of powder-like halite (Ha) and gypsum (Gi) precipitation on the surface of a nesquehonite ooide. The outer edge of the halite crystals showing decrescence is slightly brownish coloured and anisotropic (=H hydrohalite (?)).

cotton-like coatings (Parádsasvár, Plate IV, Fig. 15). The chemical composition

of the single phase palygorskite is:

SiO₂ 60.90 % TiO₂ 0.01 % Al₂O₃ 13.33 % Fe₂O₃ 0.13 % 0.39 FeO MnO 0.09 % MgO 10.03 % CaO (CaCO₃) 0.71 % Na₂O 0.04 % K_2O 0.04 % P_2O_5 0.006 % H₂O 14.31 %

Its calculated chemical formula is:

Σ

$$\begin{split} [Mg_{0.961}Al_{0.996}] \ [Ti_{0.003}Fe_{0.025}Fe_{0.020}Mn_{0.005}Na_{0.008}K_{0.005}] \\ \ [(OH)_{0.407}Si_{3.995}O_{10}]^{IV.} \ 2.203H_2O, \end{split}$$

99.986 %

that is, it can be characterized by a slight cation surplus in the [VI] coordinated site. The occurrence of the palygorskite in recent dripstones may be regarded as a dissolution product of the dolomite that arrived to the dripstones in a migration process as an allothigenous constituent. On the other hand, however, the palygors-kite of the limestone and the dolomite (sepiolite, saponite) can recrystallize into diopside, actinolite or tremolite in the high thermal stress affected contactized territories (skarn), accordingly it may be regarded as a forerunner of these minerals.

SUMMARY

The majority of the ionic structure mineral precipitations of the Recsk epihydrothermal waters are similar to the evaporation products of hypersaline (NaClsoda-borax) lakes (nesquehonite-northupite). The astrakhanite (blödite), epsomite, thenardite are typical covering salt products of evaporite deposits.

The nesquehonite-dypingite-hydromagnesite (giorgiosite) (at Recsk with 4— 10 ppm Cr and 7—250 ppm Ni content) are crack-filling minerals of near to surface serpentinized magmatites of oceanic origin, or accompanying minerals magnesite-asbestos deposits. Some of these minerals (e.g. nesquehonite) were also described from antracite and coal deposits.

The Mg-containing mineral precipitations of the ascendent hydrotherms of the Cu-porhyric skarn ore field may be regarded as indications of special genetic-geochemical conditions:

substantial portion of their constituents may be leached out of the Cretaceous serpentinized, chloritized magmatites of oceanic origin from near the Darno fault (gabbro-wehrlite-diabase).

they may have been mobilized, along the Darno fault, from the Miocene evaporite deposits of the Inner-Carpathian Mts., following the Eocene Cu-porphyric skarn ore deposition process.

The Darno is a "rejuvenated rift structure" (?). The hydrothermal appearance of these Mg-minerals may be regarded as a special ("Recsk") local parage-

netic phenomenon.

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