

NEW DATA ON THE MINERALOGY OF THE SALT DEPOSIT FROM SOVATA (MUREȘ COUNTY-ROMANIA)

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Abstract. The Badenian salt massif from Sovata is among the main salt diapirs from the Transylvanian Basin. It belongs to the eastern diapiric alignments (Șieu–Odorheiu Secuiesc–Sovata–Praid). The salt tectonics was mainly controlled by the uplift of Eastern Carpathians accompanied by high heat flow due to Neogene volcanism, generating the gravity spreading of the overburden salt. In Sovata area younger deposits are cropping out too: Pannonian sediments and Neogene volcanic breccias and agglomerates. The salt deposition was controlled by climate, tectonics, eustatism and palaeogeography. As indicated by the salt deposit mineralogy, the sedimentary conditions covered the range from the last stage of carbonates precipitation to halite. Simultaneously, an input of clastic material into the sedimentary basin from the surrounding area, took place. Such environment explains the presence of two genetically-different sets of minerals in the salt deposit from Sovata: of authigenic (halite, anhydrite, gypsum, calcite, and dolomite), as well as allogenic (quartz, clay minerals, feldspars etc.) origins. Under the microscope, two different types of fluid inclusions in halite were identified: primary inclusions in undissolved “core” of halite crystals, and secondary fluids distributed either parallel to the new cleavage planes of the recrystallized halite, or as clouds next to the planes/cracks generated by deformational events.

Key words: salt deposit, halite, sulfates, carbonates, allogenic minerals, Sovata, Romania

1. INTRODUCTION

Sovata is located on the eastern side of the Transylvanian Basin, close to the Călimani–Gurghiu–Harghita volcanic chain of Eastern Carpathians.

Like in the whole basin, in this area the Middle Miocene history begins in the Early Badenian with the deposition of acid pyroclastic rocks of the Dej Formation (Popescu, 1970), covered by the Middle Badenian evaporites – salt and gypsum – of the Ocna Dejului Formation (Mészáros, 1991). The age of the salt deposition was estimated at around 13.6-13.4 My (Balintoni & Petrescu, 2002). This process had been controlled by climate, tectonics, eustatism and palaeogeography. The classic concept is that the salt concentrated in a shallow lagoon environment, under arid climate. However, several geologists believe that the Transylvanian salt accumulated in a rather deep-marine basin, with stable tectonics and stable rate of sedimentation (Dragoș, 1969; Krézsek & Filipescu, 2005). Reconstructions based on pollen data indicate

warm-temperate climate (Petrescu & Bican-Brișan, 2005).

Primary, the salt deposit thickness was around a few hundred meters. The present uneven thickness recorded both in seismic surveys as well as in boreholes, is due to salt tectonics. In Transylvanian Basin the salt extends on 16,500 km², with 250 m average-thickness but reaching 1,000-2,000 m in diapire bodies. The salt volume is estimated at 4,120 km³. Probably, the originary extension area of the salt was even larger than today (Drăgănescu, 1997). The salt is cropping out along the western, southern and eastern borders of the Transylvanian Basin (e.g. Dej, Unguraș, Cojocna, Turda, Ocna Mureș, Ocnișoara, Ocna Sibiului, Mărtiniș, Praid, Sovata, Brâncovenești, and Sărățeni) due to diapirism. Diapir structures are much better developed on the eastern side, where diapirism was strongly enhanced by regional tectonics (compressive stress field) and Neogene volcanic activity (Szakács & Krézsek, 2007). The uplift of Eastern Carpathians accompanied by high heat flow due to arc volcanism

generated the gravity spreading of the salt overburden. Three structural domains were outlined: extensional weld (upslope), contractional folds (central), and contractional to thrust structures (downslope; Krézsek & Bally, 2006).

2. PALEOCLIMATIC CONSIDERATIONS

Several tentative on the environment reconstructions for the salt bearing Badenian deposits had been made. As a hypothesis, for the salt deposition time span Balintoni & Petrescu (2002) mentioned “temperate-mediterranean and altitudinal zoned, with two seasons” climate, underlining that the main control factor for the halite deposition was the basin isolation. This climate followed a warmer subtropical episode occurred in the Early Badenian (Moravian) corresponding to the Middle Miocene Optimum, reported both in marine and continental records (Chira et al., 2000). The tendency of climate deterioration continued in Late Badenian (Kosovian) too. A reversal of water circulation in Central Paratethys, from antiestuarine (Early and Middle Badenian, including the salt deposition in Romania) to estuarine (Late Badenian) had been reported (Báldi, 2005).

3. GEOLOGICAL FRAMEWORK

The salt massif from Sovata belongs to the eastern diapiric alignments (Șieu–Odorheiu Secuiesc–Sovata–Praid), with northwest/southeast trends

(Ciupagea et al., 1970). The Muntele de Sare („Salt Mountain”) has an ovate outline; most of its surface is covered by woods, infields and buildings belonging to Sovata Spa wellness resort. Salt rock is cropping out on the left side of Săcădat Valley. The salt built-up a prominent landscape, probably due to recent flow of salt mass. This salt is purer than that from Praid (it contains 96.3% NaCl). The geological reserves concern around 24 billion of tones. Each year, 50,000-100,000 tones of salt are washed away (Gherasie & Stoica, 1981). In the area, younger deposits are cropping out too: Pannonian sediments and Neogene volcanic breccias and agglomerates (Fig. 1).

The Pannonian beds are bearing mollusk shells (*Congeria* sp., *Paradacna* cf. *syrmiensis*, *Valenciennesia* or *Provalenciennesia* sp.). The microfauna (mainly ostracods) are indicative solely or the Early Pannonian (Zotta, 1964). Along Sovata Valley, Nagy (1956) reported three horizons: (1) lower horizon, 2-3 m thick, represented by grey marls; (2) middle horizon, with reddish and grey sands and lens-like fine sandstones; (3) upper horizon, wherein thin layers of sand and sandstones are interleaving with thin beds (1-10 cm thickness) of marls. The Pannonian subsides eastward beneath the mass of volcanic breccias and agglomerates.

The volcanic breccias extend on most part of the region, exceeding 100 m in thickness. They have diverse lithologies, but as forests are covering large areas at the surface, it is impossible to make sharp distinctions between the different types.

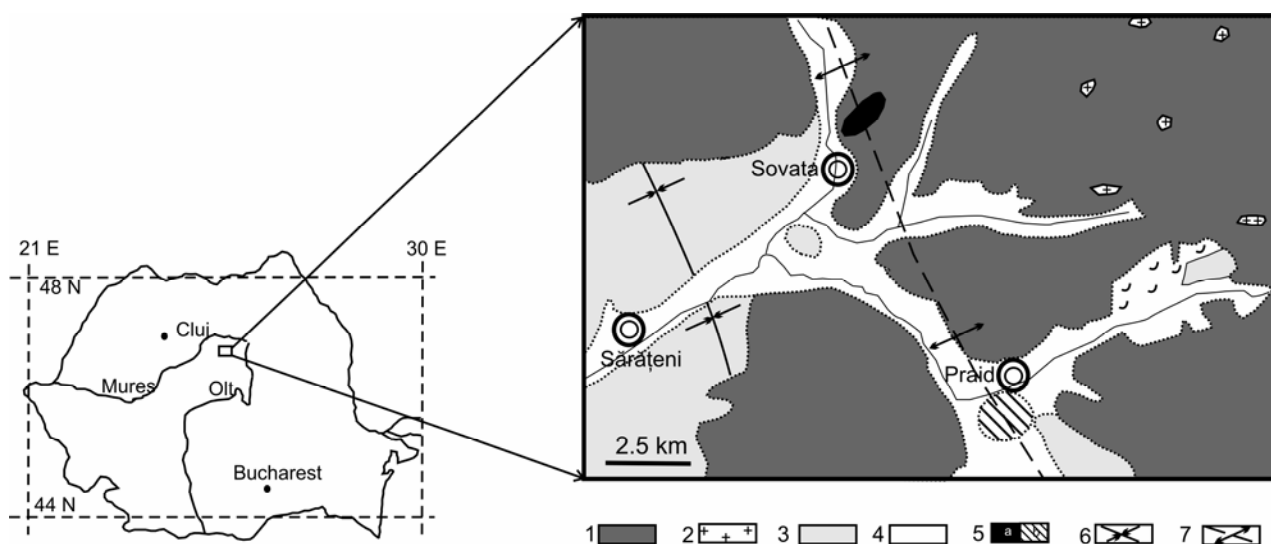


Figure 1. Location of the studied area on the map of Romania and the geology of Sovata–Praid zone (according to the geological map 1:200 000 Odorhei). 1-Neogene volcanic breccias and agglomerates; 2 – Pannonian amphibole andesite; 3 - Pannonian sands, clays and conglomerates; 4 – Quaternary deposits; 5 – Salt open (a) and covered (b) diapirs; 6- Syncline; 7 – Anticline.

Götz (1956) reported the following breccia-types: (1) andesitic volcano-breccia with hornblende; (2) andesitic volcano-breccia with hornblende and pyroxene; (3) andesitic volcano-breccia with pyroxene; and (4) andesitic volcano-tuffs and breccia with hornblende. The compact magmatic rocks (dykes, andesite lava flows) show large variability and can be found in many outcrops.

Sovata is notorious for its salt lakes too. The Ursu Lake was formed in 1875, after the collapse of the surface and obstruction of two river streams. This lake has heliothermal features. Because of erosion at the surface of the salt massif, the salinity of the lake increased significantly, and the freshwater input formed a distinct shallow layer on the top of the highly-saline water. The surface freshwater impedes the emission of the salt-water heat (due to the sun) and hence at the lake surface, the temperatures are 20-24°C, while at 2-3 m deep the water can reach 40-60°C. Nowadays, these temperatures are relatively smaller, due to the degradation of water stratification (Horváth, 2004).

According to the Law 5/2000, an area of 79 ha at Sovata has status of national natural reserve of fourth category (Bleahu, 2004). It includes both the salt lakes (5.7 ha) and the surrounding forests that cover the salt massif.

4. ANALYTICAL TECHNIQUES

The analyzed samples were collected from the Muntele Sării salt outcrop in Sovata. In order to identify the mineralogical assemblages in the salt deposit, different methods of samples preparation and subsequent analytical techniques were used. Thin sections made on natural samples were investigated under polarizing microscope in order to study especially the anisotropic minerals (sulphates, carbonates, etc.) and their spatial relationship with the main mineral constituent represented by halite. Fluid inclusions in halite crystals were microscopically investigated under parallel polars. X-ray powder diffractions (XRD) using both Philips and Dron 3 diffractometers ($2\theta = 4 - 90^\circ$) were also performed on natural samples as well as on detrital material extracted from salt samples. The detrital material consists especially of allogenic minerals (e.g. clay minerals, quartz, feldspars and micas) and thus, the obtained samples were investigated in air-dried and ethylene glycol-solvated state. Scanning Electron Microscopy (SEM) by using Jeol JSM 5510 LV with additional EDX detector – Oxford Instruments INCA 300 were used to investigate natural samples for (semi)quantitative chemical composition of the main mineral (halite), as well as of the clay minerals.

5. MINERALOGY OF SALT DEPOSIT

The salt from Sovata consists of crystal aggregates forming a gritty mass with variable color from white to grey or blackish, due to impurities content (detrital material). The color changes reflect turnovers that took place during salt precipitation as consequences of climatic variation as well as of changes in the detrital input into the sedimentary basin. Different genetic types of minerals are present in the salt deposit from Sovata:

- *authigenic minerals*, precipitated from saturated brines and genetically related with evaporitic conditions in the sedimentary basin. Under such conditions, halite precipitated as the main component of the salt deposit. Anhydrite, gypsum and carbonates are also presents in subordinate quantities.

- *allogenic minerals* represented by clay minerals, quartz, feldspars, micas etc., as the result of detrital input into the sedimentary basin.

Halite (NaCl) is the main authigenic mineral and it was identified under the microscope, in the XRD patterns as well as by using electron microscopy investigations. Its crystals are variable in size, up to a few centimeters (2-3 cm), and optically isotropic, with cubic cleavage (Plate I, Fig. A). Halite crystals include associated sulphates (e.g. anhydrite) and different allogenic minerals (e.g. clay minerals, quartz, feldspars, micas etc.). The XRD patterns of natural samples (Fig. 2) reveal the main peaks of halite, defined by the following $d(\text{Å})$ values: 3.25; 2.82; 1.99 and 1.70.

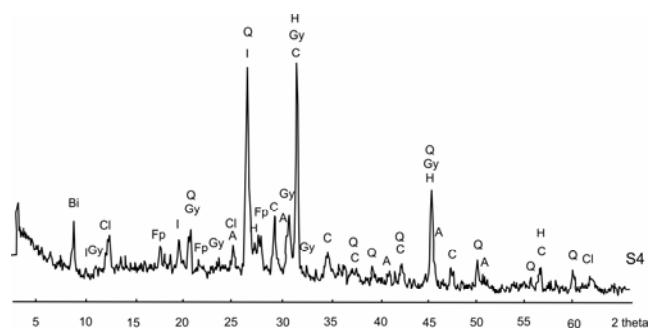
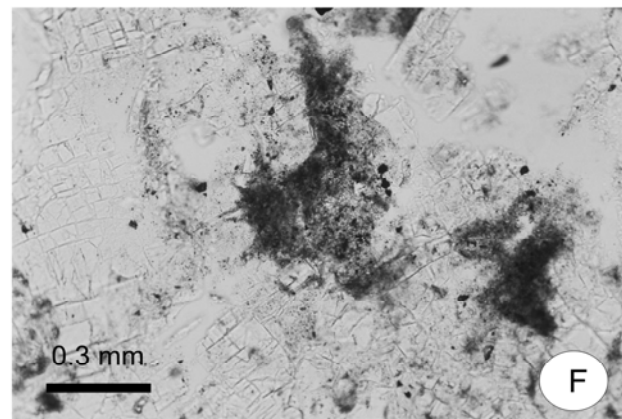
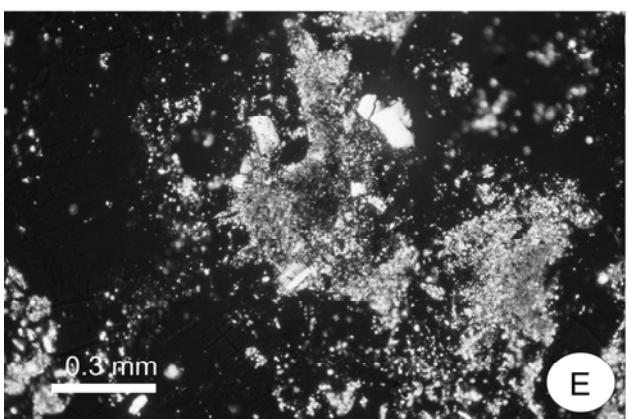
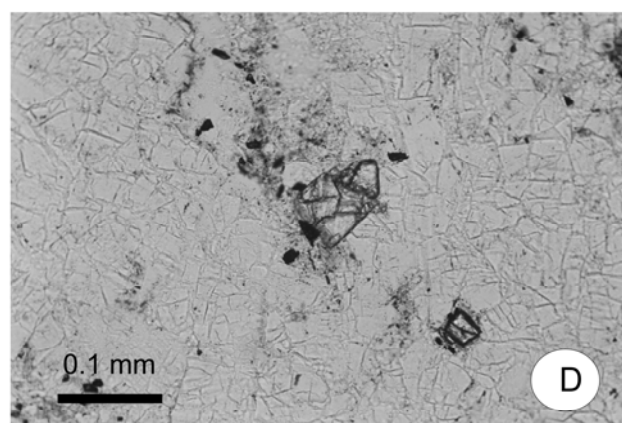
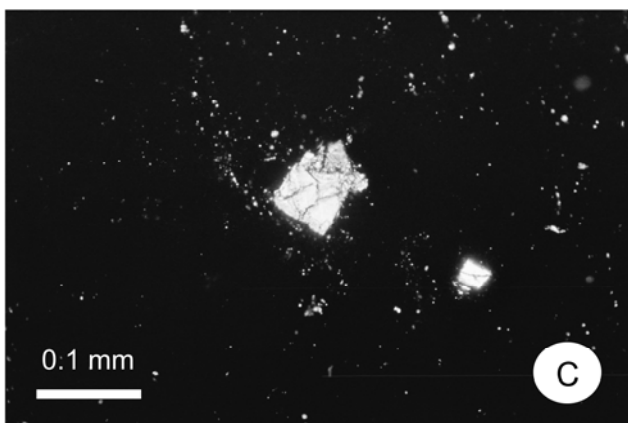
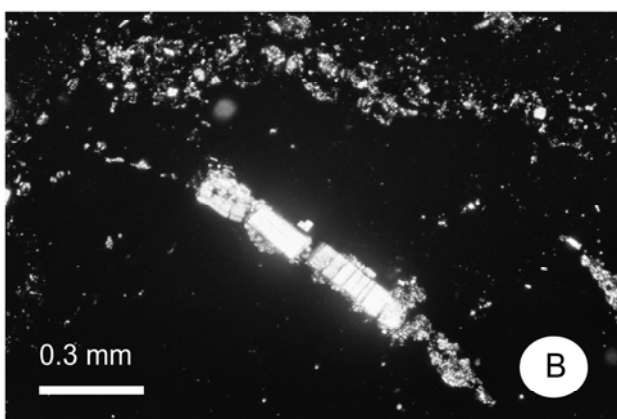
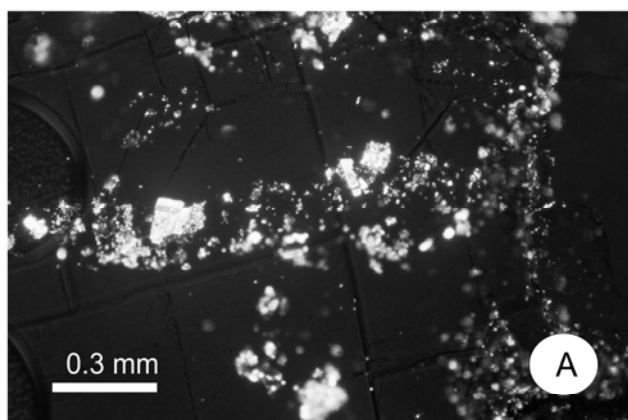
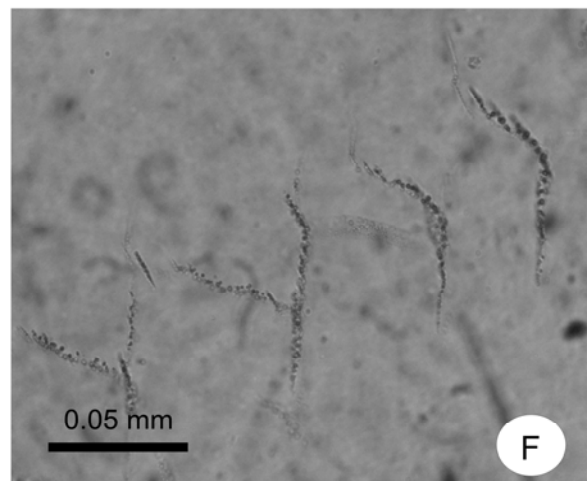
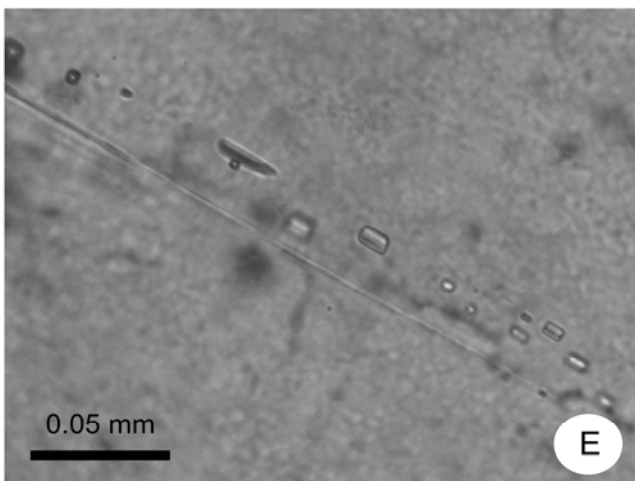
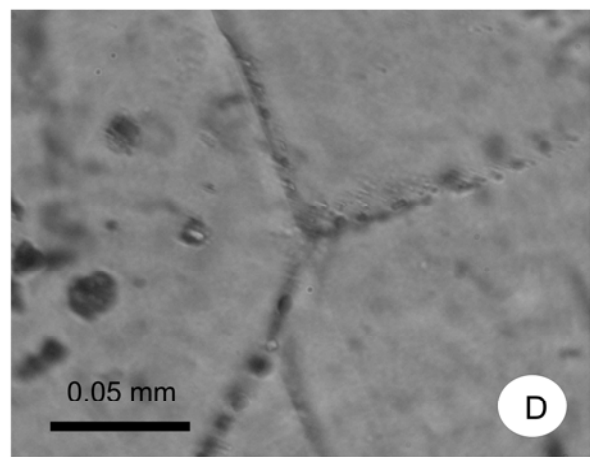
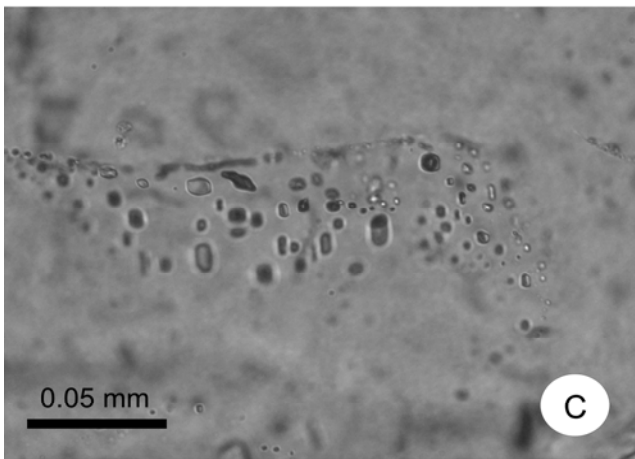
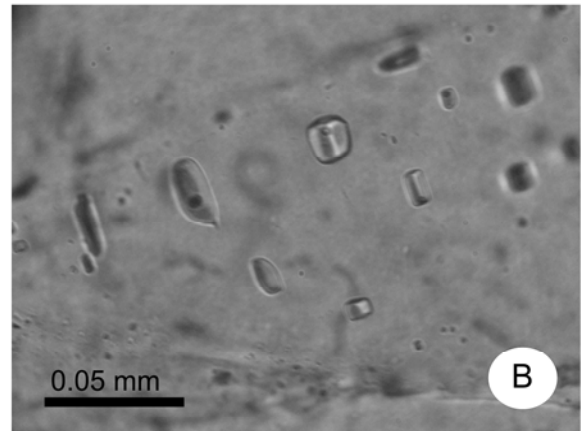
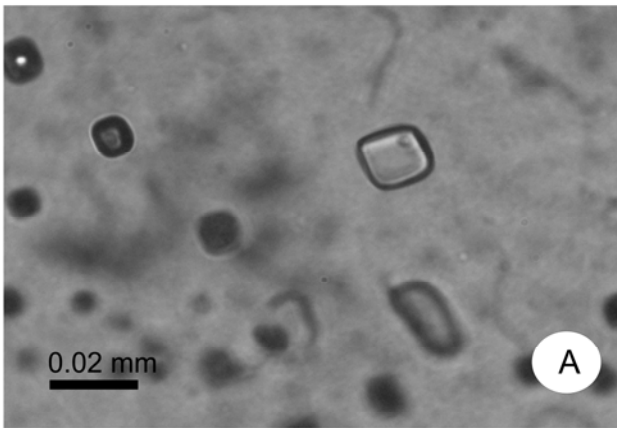


Figure 2. XRD pattern of salt rock from Sovata. H - halite, Gy – gypsum, A – anhydrite, C – calcite, Bi – biotite, Q – quartz, Fp – feldspars, I – illite, Cl – chlorite

Halite contains fluid inclusions. They are cubic or irregular in shape and most of them (especially the primary ones) contain bubbles of gas (Plate II, Fig. A, B). Under the microscope, two different types of inclusions were identified:



A. Cross-polarized light microphotograph of isotropic halite (with cubic cleavage);
 B. Cross-polarized light microphotograph of tabular crystal of anhydrite;
 C – D. Isolated crystals of carbonates in the salt mass (C- cross polars; D – parallel polars);
 E – F. Sulphates and clay minerals in halite (E - cross polars; F – parallel polars);



A - B. Microphotograph of fluid inclusions with bubbles of gas in a colorless crystal of halite (parallel polars);
 B. Core of primary halite with cloud of fluid inclusions, delimited by an undulatory surface and unaffected by dissolution processes;
 D. Grain boundaries at 120° angles in diagenetic recrystallized halite;
 E. Secondary fluid inclusions parallel with the cleavage planes of recrystallized halite;
 F. Undulatory planes/cracks generated by plastic deformational events, with clouds of fluid inclusions.

- primary inclusions, still preserved in the primary halite. The primary halite is considered the part of the salt rock that was not affected by dissolution processes.

Such primary halite is usually truncated by an undulatory surface (Plate II, Fig. C); corresponding fluid inclusions have different spatial orientation as compared to the inclusions from recrystallized halite (see below) around the “core” of primary halite.

- secondary fluid inclusions, located in recrystallized halite. Recrystallization is a low temperature diagenetic process generating clear crystals with smooth grain boundaries and frequent 120° (Plate II, Fig. D) triple junctions (Roedder, 1984). Usually, the spatial distribution of secondary fluid inclusions is controlled by recrystallization processes - when inclusions locate parallel to the cleavage planes of the newly formed crystals (Plate II, Fig. E), or by plactical deformational events, which typically generate structures of undulatory planes/cracks accompanied by parallel clouds of fluid inclusions (Plate II, Fig. F). The EDX spectrum of a cubic crystal of halite from a natural sample points out to the following composition: Na = 41.1 wt% and Cl = 58.9 wt% (Plate III, Fig. A).

Anhydrite ($CaSO_4$) is one of the main sulphates in the rock besides gypsum, and carbonates. Isolated tabular crystals with cleavages at right angle and parallel extinction were identified under the microscope (Plate I, Fig. B). Anhydrite crystals are variable in size, up to 0.5–0.6/1.0 mm. The XRD patterns obtained on rock salt or on the detrital material extracted from salt evidence some typical reflexions for anhydrite (d/Å): 3.48; 2.19; 1.99 and 1.75 (Figs. 2 and 3).

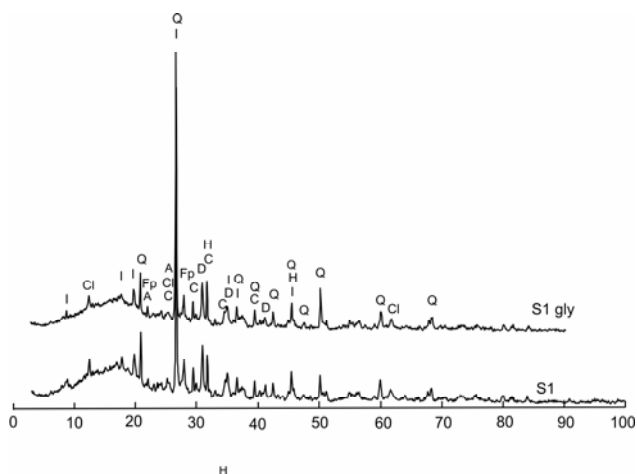


Fig.3. XRD patterns of detrital material in the salt from Sovata, in air-dried (S1) and ethylene glycol-solvated (S1 gly) state. H - halite, A - anhydrite, C - calcite, D - dolomite, Q - quartz, Fp - feldspars, I - illite, Cl - chlorite

Gypsum ($CaSO_4 \cdot 2 H_2O$) is tabular in shape or forms microscopic granular aggregates. Some peaks typical for gypsum were also identified in the XRD patterns of natural samples (d/Å): 7.61; 4.28; 2.88 and 1.90 (Fig. 2).

Carbonates – Calcite ($CaCO_3$) and *dolomite* ($CaMg[CO_3]_2$) are also present as authigenic minerals. They occur as isolated crystals in the halite mass, rarely as rhombohedral crystals clearly visible under the microscope (Plate I, Figs. C and D). In the XRD patterns on the detrital material extracted from salt samples (Figs. 2 and 3), the most representative peaks of the above mentioned carbonates are present: calcite(d/Å: 3.03; 2.82; 2.28) and dolomite (d/Å: 2.88; 2.56; 2.19; 2.05; 2.01; 1.81; 1.78; 1.56).

Quartz (SiO_2) is the most important allogenic mineral, identified by using XRD on natural samples as well as on the detrital material extracted from salt (Figs. 2 and 3). The most representative peaks of quartz are clearly present in the XRD pattern (d/Å: 4.25; 3.34; 2.45, 2.28, 1.97, 1.81; 1.67; 1.66; 1.54; 1.45; 1.38; 1.37; 1.19; 1.84).

Plagioclase feldspars ($NaSi_3AlO_8-CaSi_2Al_2O_8$) are present as allogenic phases and they were identified based on a few peaks (Figs. 2 and 3) present in the XRD patterns (d/Å: 4.03; 3.77; 3.23 and 3.19).

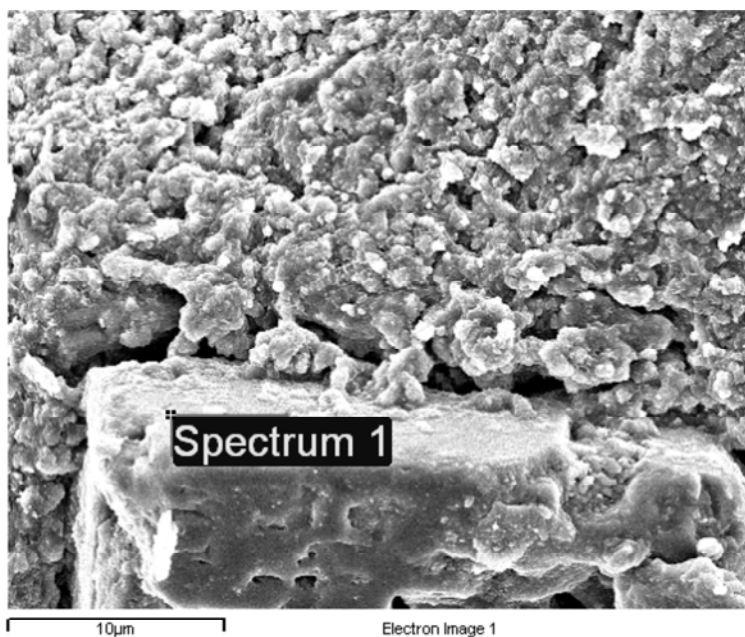
Clay minerals were microscopically identified in the salt rock, they occur as lamellar aggregates up to 200 μm in size (Plate I, Figs. E and F). Their presence was also confirmed in the XRD patterns obtained on extracted detrital material in both air-dried and ethylene glycol-solvated samples as well as by using SEM+EDX investigations. The comparative XRD patterns do not show any significant differences. Using the typical (00l) reflexions in the range of $2\theta = 5.36-28.59^\circ$, the following minerals were identified (Fig. 4):

- illite, defined by d(Å)/(00l): 9.98/(002); 4.98/(004) and 3.34/(006). The additional peaks, 4.48/(110) and 3.67/(023) are also present.

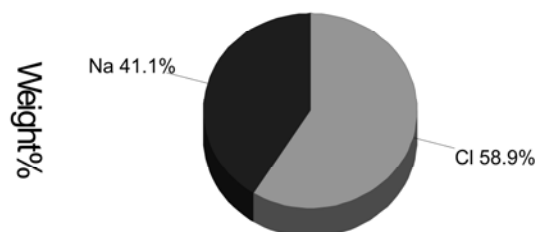
- chlorite, defined by d(Å)/(00l): 7.08/ (002); 4.72/(003) and 3.53/(004). The peak at 1.538/(060) is also very typical for chlorite.

Five chemical spectra obtained using EDX investigations on fine lamellar crystals of clay minerals (Plate III, Fig. B) indicate the presence of illite or mixing aggregates of clay minerals as well as possibly some transitional compositions (Table 1). The presence of K^+ is typical for illite as fixed cation, while Fe^{2+} (most probably) and Mg^{2+} are also mentioned by Moore & Reynolds (1997) as potential cations in octahedral coordination in the illite structure. Al^{3+} (and Fe^{3+}) could be in both tetrahedral (substituting Si^{4+}), and octahedral coordinations.

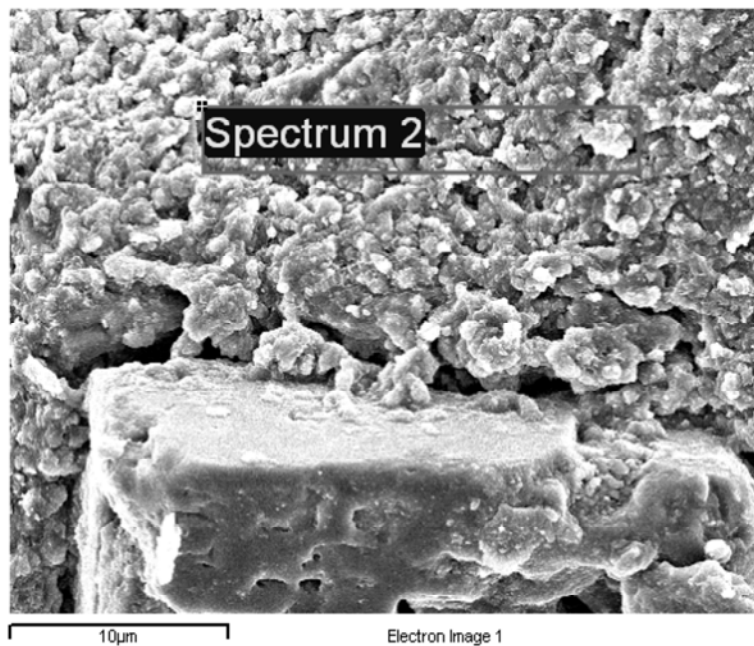
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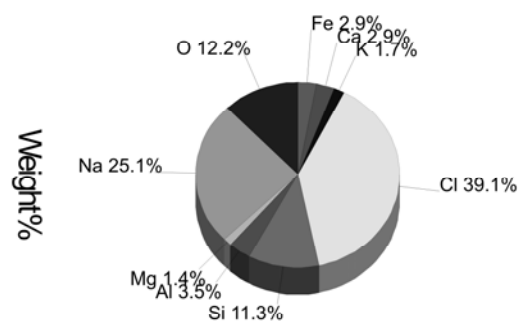
Quantitative results



B



Quantitative results



- A. SEM image of halite cubic crystal and EDX chemical composition
- B. SEM image of detrital material from salt and EDX chemical composition

Table 1. EDX chemical composition of the halite and clay minerals from Sovata (wt%)

	Na	Cl	Si	Al	Mg	Fe	K	Ca	O	Si/Al	Al/Fe + Mg	Mineral
1	41.10	58.90	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-	-	Halite
2	25.10	39.10	11.30	3.50	1.40	2.90	1.70	2.90	12.20	3.23	0.81	Fe- and Mg-rich Illite/Celadonite
3	0.00	0.00	30.30	10.30	4.60	6.60	4.50	7.20	36.50	2.94	0.92	Fe- and Mg-rich Illite/Celadonite
4	3.90	24.10	23.30	14.30	0.00	3.00	8.10	0.90	22.30	1.63	4.77	Illite
5*	0.00	0.00	36.90	19.50	0.00	4.40	10.60	1.30	27.40	1.89	4.43	Illite
6	27.80	40.70	12.10	6.50	0.00	1.40	2.90	1.00	7.60	1.86	4.64	Illite

• represents the results for sample 4, recalculated without Na and Cl

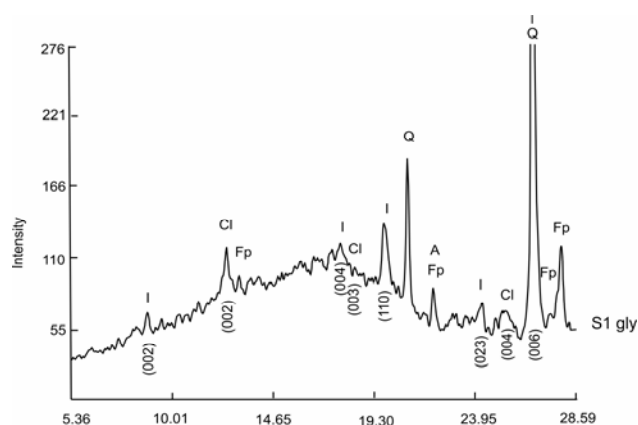


Figure 4. XRD patterns of detrital material from Sovata salt in ethylene glycol-solvated state (S1 gly) in the range of $2\theta = 5.36\text{--}28.59^\circ$. I-illite, Cl-chlorite, Fp-feldspars, Q-quartz, A-anhydrite.

An increase of Al^{3+} content (in both octahedral and tetrahedral positions) will decrease the Si/Al ratio of illite, while the presence of Fe^{3+} , Fe^{2+} and Mg^{2+} will lead to an increasing Si/Al ratio. The EDX spectra performed of the lamellar crystals (Table 1) of natural samples from Sovata (Plate III, Fig. B) indicate the presence of illite with variable contents of $\text{Fe}^{3+}+\text{Fe}^{2+}$ (1.4–6.6 %) and Mg^{2+} (up to 4.6 %). The Si/Al ratio (Table 1) shows variables values from 1.63 to 3.23, in all cases higher than 1.26 calculated according to Rieder et al. (1998). Thus, the current compositions point to a typical illite, with low contents of iron and magnesium (also indicated by the relatively high values of the Al/Fe+Mg ratio; Table 1). The other analyzed crystals, with low values of Al/Fe+Mg ratio and higher Si/Al ratio (2.94–3.23) respectively point to the presence of a Fe- and Mg-rich illite. Such compositions could be also characteristic for celadonite, which has a typical Si/Al ratio of 3, as

calculated according to Antony et al. (1995). Higher values than 3 for celadonite could be the result of Al^{3+} substitution by Fe^{3+} in the tetrahedral position, as well as the exclusive presence of Fe^{2+} and Mg^{2+} in octahedral coordination, leading to low Al/Fe+Mg ratios. Celadonite is a clay mineral very often mentioned in the composition of Badenian volcanic tuffs from the Transylvanian Basin (Mârza et al., 1991; Ghergari et al., 1991). The presence of Ca^{2+} and/or Mg^{2+} in small amounts could also indicate the presence of carbonates, previously identified as authigenic minerals in the salt from Sovata.

6. CONCLUSION

The Badenian diapiric salt massif from Sovata (Mureş County) belongs to the Şieu-Odorheiu Secuiesc-Sovata-Praid diapiric alignments from the Transylvanian Basin. Based on polarizing microscopy, X-Ray diffraction, SEM and EDX investigations, two different types of mineral phases were identified in the salt deposit from Sovata: of authigenic (halite, anhydrite, gypsum, calcite and dolomite) and of allogenic origin (quartz, feldspars, clay minerals). Halite is the main component and it was diagenetically recrystallized due to salt diapirism. Fluid inclusions found in halite indicate the presence of primary core crystals, delimited by undulatory surfaces. The recrystallized halite, with 120° triple junctions limits, contains fluid inclusions parallel with the new cleavage plans or as clouds next to the planes/cracks of deformational origin. The presence of carbonates (calcite and dolomite), anhydrite and gypsum as subordinate authigenic minerals, indicate that the sedimentary conditions covered the depositional range from the last stage of carbonates precipitation to halite formation. Chemistry of the clay minerals indicates the presence of illite with variable Fe^{3+} and Mg^{2+} contents.

The high content of Mg²⁺ of the phase with high Si/Al rates could also indicate the presence of celadonite.

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Received at: 25. 03. 2010

Revised at: 28. 05. 2010

Accepted for publication at: 16. 06.2010

Published online at: 19. 06. 2010