

Geochemistry of Neogene evaporites of the Transcarpathian Trough in Ukraine

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Abstract. The initial brine during halite sedimentation in the Zaluzh salt-forming basin in the Chop-Mukacheve depression was of $\text{Na}^+ \text{-Cl}^- \text{-Mg}^{2+} \text{-K}^+ \text{-SO}_4^{2-}$ composition, and by ratio of K^+ and Mg^{2+} was close to the modern seawater saturated to the stage of halite precipitation. The progressive evaporation of the brine has led to a significant increase of the total concentration of solutions in the basin, but the halogenesis has not reached the high stages of its evolution and finished on the halite stage. During diagenesis the sediments were recrystallized under the impact of solutions, with the decreased concentration of the major compounds.

Key words: Miocene, evaporites, fluid inclusions, geochemistry, Transcarpathian basin.

Introduction

The Transcarpathian basin together with the Transylvanian and East Slovakian basins was a part of the united system of intracarpathian evaporite basins in the Middle Miocene (Kityk *et al.*, 1983). Two basins of the second order can be distinguished in the Transcarpathian basin: Zaluzh and Solotvyne, separated by Khust bar.

The distribution of salt-bearing deposits in the north-western part of the Transcarpathian trough in the Chop-Mukacheve depression is restricted to Mukacheve-Svalava region (Kityk *et al.*, 1983). Salt-bearing rocks are developed in the eastern part of the depression (Zaluzh, Irshava, Vynogradove) and other areas and sulfate-carbonate rocks occur in its southern part, in the Vynogradove area (Fig. 1). The Badenian deposits occur at a depth of 1000–1800 m. Their thicknesses range from 170 to 310 m. The deposits consist of interbedded dark grey, black, and rarely white rock salt, siliciclastic rocks and sometimes tuff-mudstones. In lower part of section the layers of gypsum and anhydrite are sometimes found. In some places (Zaluzh area), layers of tuff-sandstone and grey vitroclastic tuff were found.

The geochemical peculiarities of rock salts and the conditions of their formation have been studied mainly in the southeastern part of the trough (Solotvyne deposit). The literature data on the geochemistry of evaporites from the north-western part of trough are scarce (Kityk *et al.*, 1983; Khrushchov & Petrichenko, 1980; Petri-

chenko, 1989), and therefore the purpose of this investigation was to establish the geochemical peculiarities of formation of rock salt in this part of the trough. I have studied the core samples of rock salt from borehole 6-T in the Mukacheve region (Svalava, Health Centre "Carpathians"). During research work on thermal water at a depth of 1047–1318 m under the cover of Recent volcanic rocks of Vgorlat-Gutinsk ridge where the occurrence of salt has not previously been supposed, an evaporite sequence of about 300 m thick was recorded.

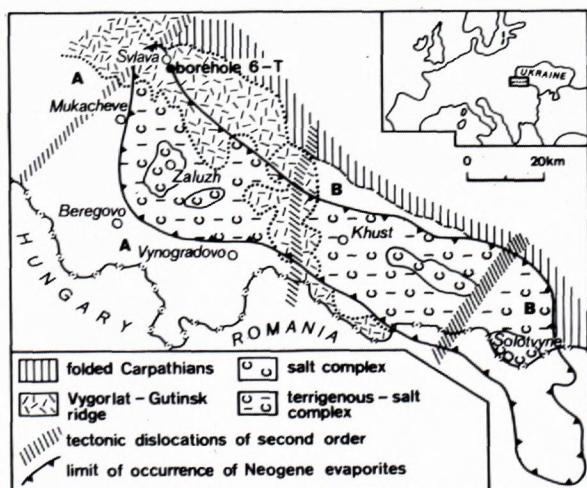


Fig. 1. Schematic map of distribution of Neogene evaporites in the Transcarpathian Trough (after Kityk *et al.*, 1983).
A - Mukacheve depression, B - Solotvyne depression.

The composition of inclusion brines in halite has been determined by means of methods suggested by Petrichenko (1973) and Yermakov & Dolgov (1979). The data on Br^- and $\text{Br}1000/\text{Cl}$ in halite and the composition of rock salt have been also used. The obtained information was correlated with data from the southeastern part of the trough (Solotvyne deposit).

Lithology

The studied rock salt is recrystallized. Its sedimentary origin is indicated by occasional occurrences of zonal constructions (chevrons) that have been preserved in grains and crystals and are macroscopically visible. Their diameters reach 2 cm. Under the microscope it can be seen that the zonation is caused by numerous microinclusions of brine and gas, that look like negative cubic crystals. These primary inclusions in halite indicate its primary crystallization from the brine.

The rock salt is fine- to coarse-grained and grain sizes vary from 1 mm to 2.0 cm. The structure is massive, cellular or laminated. The colour of the rock is light or dark grey and is caused by clay admixture. The clay in the salt is present in dispersed form (and the grains are grey and slightly transparent) or as thin seasonal layers, sometimes balls or chains inside the halite grains or around them. The clayey fraction in the insoluble residue of rock salt from the depth of 1075-1205 m consists of illite and mixed-layered growths of chlorite and trioctahedral montmorillonite. The amount of mixed-layered minerals decreases with depth. It should be noticed that chemical and X-ray analyses indicate that hydromica prevails and the chlorite admixture is present in the clayey fraction of the rock salt of Solotvyne deposit. My data confirm results obtained by Bilonizhka (1979) who recorded the hydromica with a significant admixture of magnesium-ferriferous chlorite in the clay fraction of salt rocks. In the same fraction from overlying terrigenous rocks the hydromica, chlorite, mixed-layered minerals (hydromica-montmorillonite), kaolinite and possibly montmorillonite are present. These facts permit the conclusion that during evaporation in the evaporite basin or during diagenesis of salt rocks, the montmorillonite and kaolinite were not stable and were transformed into illite and chlorite. It should be noted that Bilonizhka (1992) pointed out the presence of clastic magnesium-ferriferous chlorite and authigenic magnesium chlorite in the clay fraction from the rock salt of Solotvyne deposit.

The content of bromine and the value of the bromine-chlorine coefficient in halite is an important index for conditions of rock salt formation (Valiashko, 1962; Bilonizhka, 1975). Actual bromine contents in rock salt of different formations often differ from experimental and theoretically calculated contents of bromine in halite

crystallized at different stages of seawater saturation. The changes of the bromine-chlorine coefficient in halite from Chop-Mukacheve depression are shown in Fig. 2. The cleanest halite grains and crystals with the lowest amount of terrigenous admixture have been used for analyses. The sedimentary as well as recrystallized halite have been sampled. The chlorine and bromine content in halite are shown in Table 1. I compared these data with experimental and theoretically calculated contents of chlorine and bromine in the halite crystallized at different stages of seawater saturation (Table 2). The comparison shows that the studied halite is similar to that typical for a halite stage of crystallization of chlorides in marine-derived salt basin. The variation of the bromine-chlorine coefficient may be explained by the processes of postsedimentary alteration of halite that led to the bromine depletion. For Solotvyne deposit (mine 8), the average value of bromine in water-soluble part of halite (10 determinations) is 0.011% (maximum 0.020%, minimum 0.004%). The values of the $\text{Br}1000/\text{Cl}$ coefficient are 0.075-0.374, and the average is 0.187. These data also correspond to experimentally determined ones for the halite stage of chloride precipitation. Much lower is the content of bromine in halite, sampled in mine 9 of this deposit. The average value of bromine (22 analyses) is 0.003% (maximum 0.014%, minimum 0.001%). The $\text{Br}1000/\text{Cl}$ coefficient ranges from 0.016 to 0.205, and the average is 0.048. For the majority of samples from mine 9, these values are lower than in the samples from initial zones of halite crystallization from seawater. The decreased content of bromine and values of the $\text{Br}1000/\text{Cl}$ ratio are caused by recrystallization of halite.

Table 1. The content of chlorine and bromine in halite from the rock salt of the Chop-Mukacheve depression

Depth (m)	Content, % Cl^-	Br^-	$\text{Br}1000/\text{Cl}$ coefficient
1047-1050	59.33	0.0055	0.093
1068-1079	59.41	0.002	0.034
1075-1078	57.13	0.007	0.122
1087-1090	58.84	0.009	0.153
1110-1114	56.35	0.004	0.071
1113-1114	55.86	0.0055	0.098
1127-1131	59.59	0.004	0.067
1135-1140	56.42	0.0025	0.044
1145-1149	55.67	0.003	0.054
1150-1154	55.67	0.003	0.054
1164-1167	55.38	0.005	0.090
1171-1174	55.22	0.008	0.145
1189-1194	57.07	0.003	0.053
1200-1205	57.08	0.006	0.105
1274-1278	56.75	0.002	0.035
1302-1306	58.30	0.0025	0.043
1314-1318	58.35	0.0035	0.060
Average	56.71	0.005	0.081

Table 2. The content of bromine (%) and the values of bromine-chlorine coefficient in chlorides of marine genesis.

Stage of chloride precipitation	After Valiashko (1962)		After Bilonizhka (1975)	
	Br ⁻	Br 1000/Cl coeff.	Br ⁻	Br 1000/Cl coeff.
Halite	0.068-0.026	0.11-0.43	0.006-0.016	0.099-0.26
Sylvite	0.027-0.037	0.44-0.61	0.016-0.025	0.26-0.41

sions in sedimentary halite have a cubic shape. They contain solutions with individual gas bubbles. The presence of this gas may be explained by overheating of inclusions after sedimentation. Sometimes a solid phase - needles of gypsum or anhydrite - is noticed. There are more variable inclusions in diagenetic halite. Except of relics of sedimentary halite, several types of inclusions are developed.

Both primary and secondary inclusions are distinguished. Secondary ones are located in healed fissures, by aggregate state they are fluid and gas-fluid with a not-

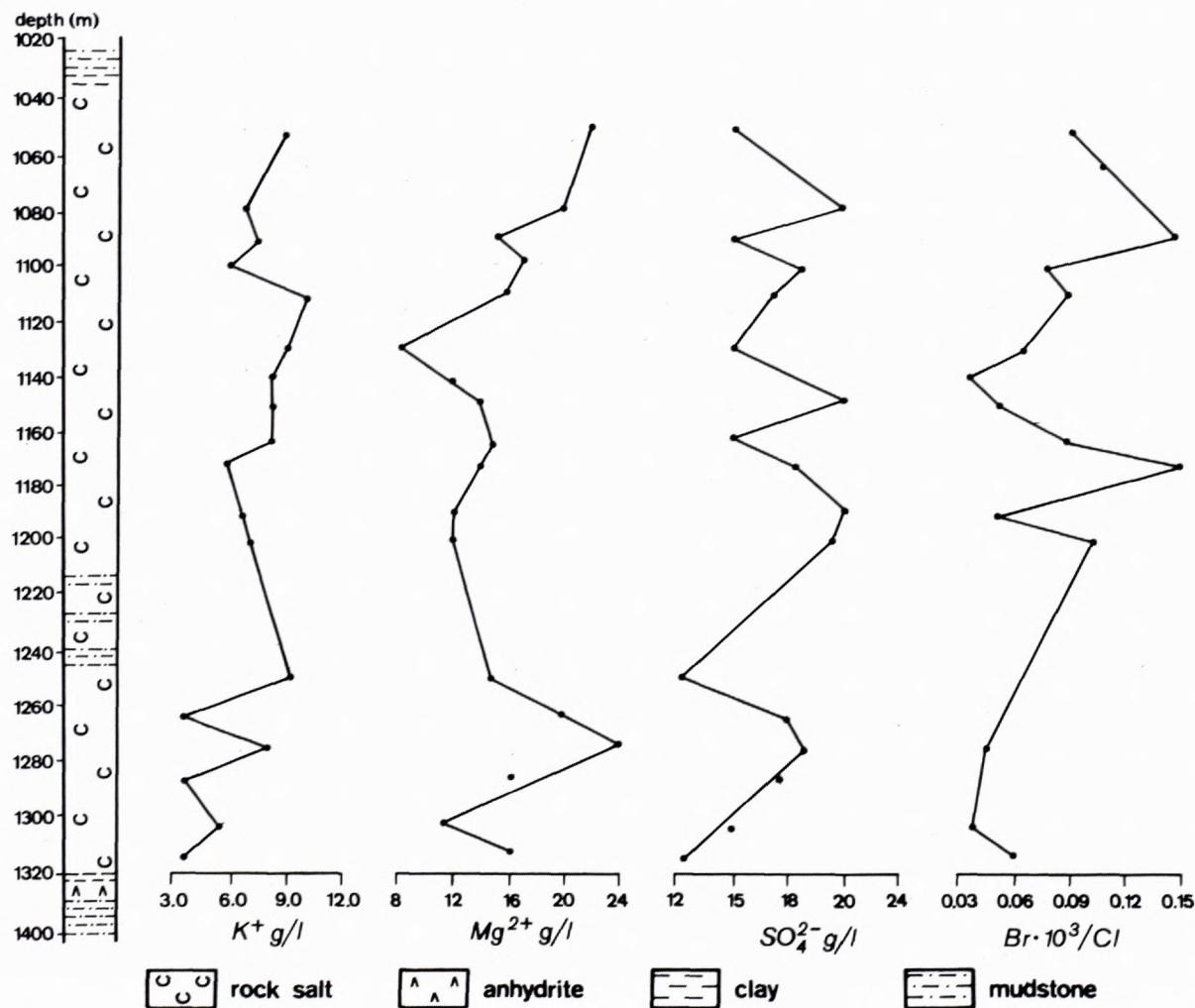


Fig. 2. The content of potassium, magnesium and sulfate ions in brine inclusions and the values of bromine-chlorine coefficient in recrystallized halite in the section of borehole 6-T (Svalava, Mukacheve region).

The results of inclusion study

Under the microscope it is seen that the rock salt consists mainly of recrystallized (diagenetic) halite with chevron relics. Often the parts of sedimentary crystals with rhythmic location of inclusions are preserved. Inclu-

constant ratio of phases. Primary inclusions by aggregate state are one-phase (solid), two-phase (gas-liquid), three-phase (gas-liquid with solid phase), multiphase (gas-liquid with several solid phases) and inclusions with hydrocarbons. Two-, three- and multiphase inclusions in recrystallized halite are distributed without any order. By

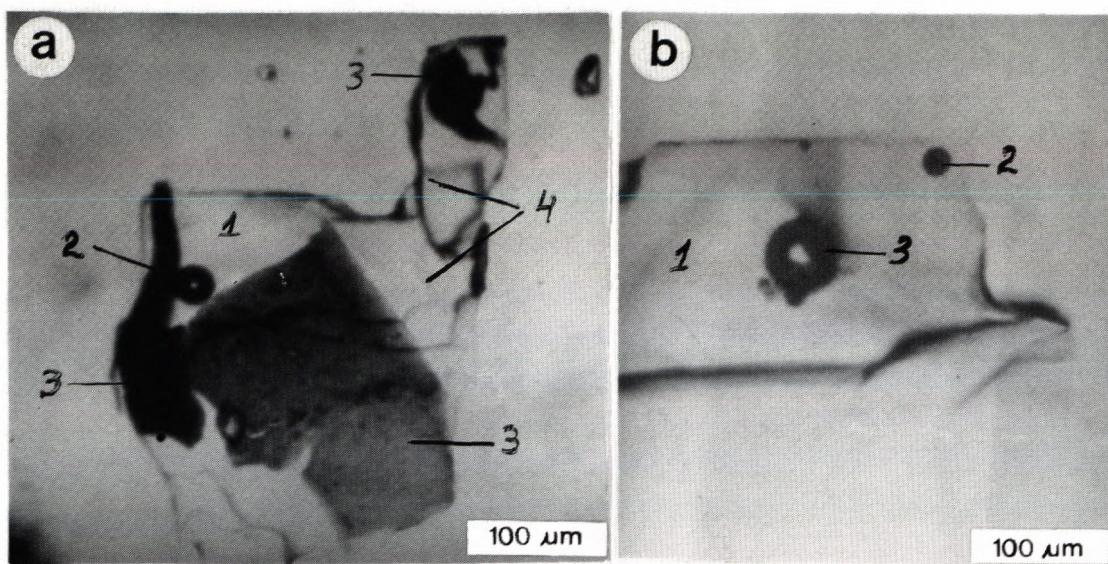


Fig. 3. Inclusions in diagenetic halite of Chop-Mukacheve depression (borehole 6-T); a - depth 1287-1291 m: 1 - water solution; 2 - gas phase, 3 - hydrocarbon crust; 4 - anhydrite; b - depth 1200-1205 m): 1 - water solution; 2 - oil; 3 - gas phase.

composition the solid phase in these inclusions is similar to the composition of solid inclusions in halite. The liquid phase is represented by water solution or oil, and gas phase - by gas mixture.

The most interesting are inclusions with hydrocarbons. In the Badenian rock salt they were discovered for the first time. They have been found in samples from depths of 1200-1205 m, 1274-1278 m, and 1287-1291 m (Fig. 3). Inclusions with hydrocarbons are mainly irregular in shape and rarely cubic. Hydrocarbons form globules or thin red-brown crusts. The aggregate state of inclusion hydrocarbons may be liquid and solid. In the first case, hydrocarbon globules are formed by light-yellow, yellow and dark-yellow fluid (in translucent light), that is immiscible with the inclusion solution. Except for fluid hydrocarbon globules and solution, the solid phase composed of black matter is often present in inclusions. Hydrocarbons make up less than 2% of inclusions. The number of globules is different. Globules have mainly homogeneous construction; however, there are globules with zonal construction, and their centre, as a rule, is darker in colour (dark brown or black) than peripheral parts which are light-brown or red-brown. Under luminiscence microscope in ultraviolet light, fluid hydrocarbon globules have yellow, brown-yellow, red-yellow luminescence, that testifies to their organic nature and is typical for oils with high content of pitch-asphaltene compounds and show the process of oxidation of hydrocarbons (see Botneva, 1979). Solid globules of hydrocarbons are brown, dark-brown and black. In reflected light they are translucent in brown colours. When the globules are extracted from the inclusion, they often divide into two parts and the droplet of yellow fluid is separated from them.

Table 3. The chemical composition of brine inclusions in sedimentary halite of the Badenian rock salt from the Chop-Mukacheve depression.

Depth m	K ⁺	Content of elements, g/l	
		Mg ²⁺	SO ₄ ²⁻
1047-1050	9.0	24.0	16.0
1068-1079	8.0	24.0	24.0
1075-1078	9.0	21.0	25.0
1087-1090	9.0	15.0	16.0
1110-1114	8.5	20.0	22.0
1113-1114	14.0	22.5	24.0
1127-1131	11.0	20.5	18.5
1135-1140	8.0	16.5	19.5
1145-1149	8.0	11.0	20.5
1150-1154	8.5	13.0	21.0
1164-1167	9.0	14.0	23.0
1171-1174	9.0	14.0	17.0
1189-1194	9.0	14.0	20.5
1200-1205	9.0	16.5	20.0
1274-1278	13.0	26.0	23.5
1302-1306	6.5	14.0	16.0
1314-1318	4.0	17.0	14.0
Average	9.5	18.0	20.0

Primary solid inclusions in diagenetic halite are disordered separations or are located on the boundary between terrigenous layers and recrystallized halite. Individual solid inclusions in the halite and the solid phase in two-, three- and multiphase inclusions are represented by authigenic minerals such as anhydrite, dolomite, quartz and pyrite, or terrigenous material. Anhydrite forms grains or crystals, fine-aggregate sepa-

rations, balls or nodules. More frequently the tabular, prismatic crystals with jagged edges and also needle crystals 0.1 to 1.5 mm long, that form bundle-like accumulations, are present. In halite plates, one can see the growth of needle anhydrite from clayey terrigenous layers towards the halite. Dolomite is located on the contacts of halite grains, along the cleavage planes, on the boundary of terrigenous layers and halite and forms aggregates, individual inclusions, randomly located in the recrystallized halite. Its sizes range from 0.1 to 0.5 mm. Crystal shapes are rhombohedral. Quartz forms transparent short-prismatic crystals with well developed rhombohedral heads, aggregates of several crystals, and rose-like growths; their sizes range from 0.5 to 1.0 mm. Pyrite looks like small black balls that often form segregations. They are located within clayey layers or are present as individual inclusions in diagenetic halite.

Composition of solutions

The data on the liquid phase in inclusions allow the reconstruction of the composition of mineral-forming solutions (Petricenko, 1973; Yermakov & Dolgov, 1979). Specifically, the analyses of inclusions in sedimentary halite permit determination of the composition of brine during the stage of halite precipitation, and, in recrystallized ones, the composition of solutions can be determined in which the alteration of halite took place during diagenesis. It has been determined that Na^+ , Mg^{2+} , K^+ , Cl^- , SO_4^{2-} are the major compounds in inclusion solutions of sedimentary halite (Table 3). The content of K^+ , Mg^{2+} and SO_4^{2-} in relict mineral-forming solutions significantly changes (Fig. 4). The rhythmic increase of the content of major compounds in brine inclusions from the lower to the upper part of the section was recorded. For example, in the interval of 1302–1318 m the content of potassium is 4.0–6.5 g/l; of magnesium is 14.0–17.0 g/l, and of sulfate ion is 14.0–16.0 g/l; and in interval of 1047–1079 m potassium concentration is 8.0–9.0 g/l; of magnesium is 24.0 g/l, and of sulfate ion is 16.0–24.0 g/l. The total concentration of major elements (K^+ , Mg^{2+} , SO_4^{2-}) increases from 35.0 g/l in the lower part to 49.0 g/l in the upper one.

The same chemical composition is determined in inclusions from sedimentary halite of the southeastern part of the trough (Solotvyne deposit, mines 8 and 9), but the average contents of all major compounds are significantly higher there. The average content of potassium (42 analyses) is 15.5 g/l, with the minimal concentration of 8.0 g/l and maximal one of 25 g/l. The magnesium content ranges from 19.5 to 36.5 g/l; its average concentration is 28.5 g/l (35 determinations). The content of sulfate-ion changes from 18.0 to 63.0 g/l, and averages 36.5 g/l (33 analyses).

Brine inclusions in diagenetic halite from the northwestern part of the trough as well as in sedimentary ones, have Na^+ - Mg^{2+} - K^+ - Cl^- - SO_4^{2-} -composition (Table 4). When compared to sedimentary halite, the brine inclusions in diagenetic halite are characterized by decreased concentration of major elements. In the interval 1302–1318 m the potassium content is 4.0–5.0 g/l; of magnesium 11.0–16.5 g/l; of sulfate ion 12.5–15.0 g/l; and in the interval 1074–1079 m the potassium content is 5.0–9.0 g/l; of magnesium 20.5–22.0 g/l; of sulfate ion 15.0–20.0 g/l (Fig. 2). The same composition is typical for brine inclusions in recrystallized halite from the Solotvyne deposit. The average content of potassium (22 determinations) is 10.0 g/l, maximum 19.5 g/l, minimum 4.0 g/l. The magnesium content ranges from 9.5 g/l to 26.5 g/l, averaging 18.0 g/l. Sulfate ion is present in amounts from 15.0 to 56.0 g/l; the average of 20 analyses is 27.5 g/l.

Table 4. The chemical composition of brine inclusions in recrystallized halite of the Badenian rock salt from the Chop-Mukacheve depression

Depth m	Content of elements, g/l		
	K^+	Mg^{2+}	SO_4^{2-}
1047-1050	9.0	22.0	15.0
1068-1079	5.0	20.5	21.0
1075-1078	8.0	17.0	20.0
1087-1090	7.0	16.0	15.0
1110-1114	5.5	17.0	19.0
1113-1114	10.0	20.0	22.0
1127-1131	9.0	19.0	18.0
1135-1140	8.0	16.0	17.5
1145-1149	8.0	8.0	15.0
1150-1154	8.5	12.0	19.0
1164-1167	8.0	14.0	21.0
1171-1174	6.0	15.5	15.0
1184-1189	7.0	14.0	19.0
1189-1194	7.0	12.0	21.0
1200-1205	7.0	12.0	20.0
1250-1254	9.5	15.0	12.5
1265-1269	3.5	20.5	18.0
1274-1278	8.0	24.0	19.0
1287-1291	4.0	16.0	17.0
1302-1306	5.0	11.0	15.0
1314-1318	4.0	16.5	12.5
Average	7.0	16.0	17.5

The composition and changes of the major element contents in brine inclusions from sedimentary halite show that during the stage of halite precipitation the brine of the Transcarpathian basin in its northwestern part (Zaluzh salt-bearing subbasin) had contents of potassium (4.0 g/l) and magnesium (17.0 g/l) and by ratio of these elements was close to the modern seawater saturated to

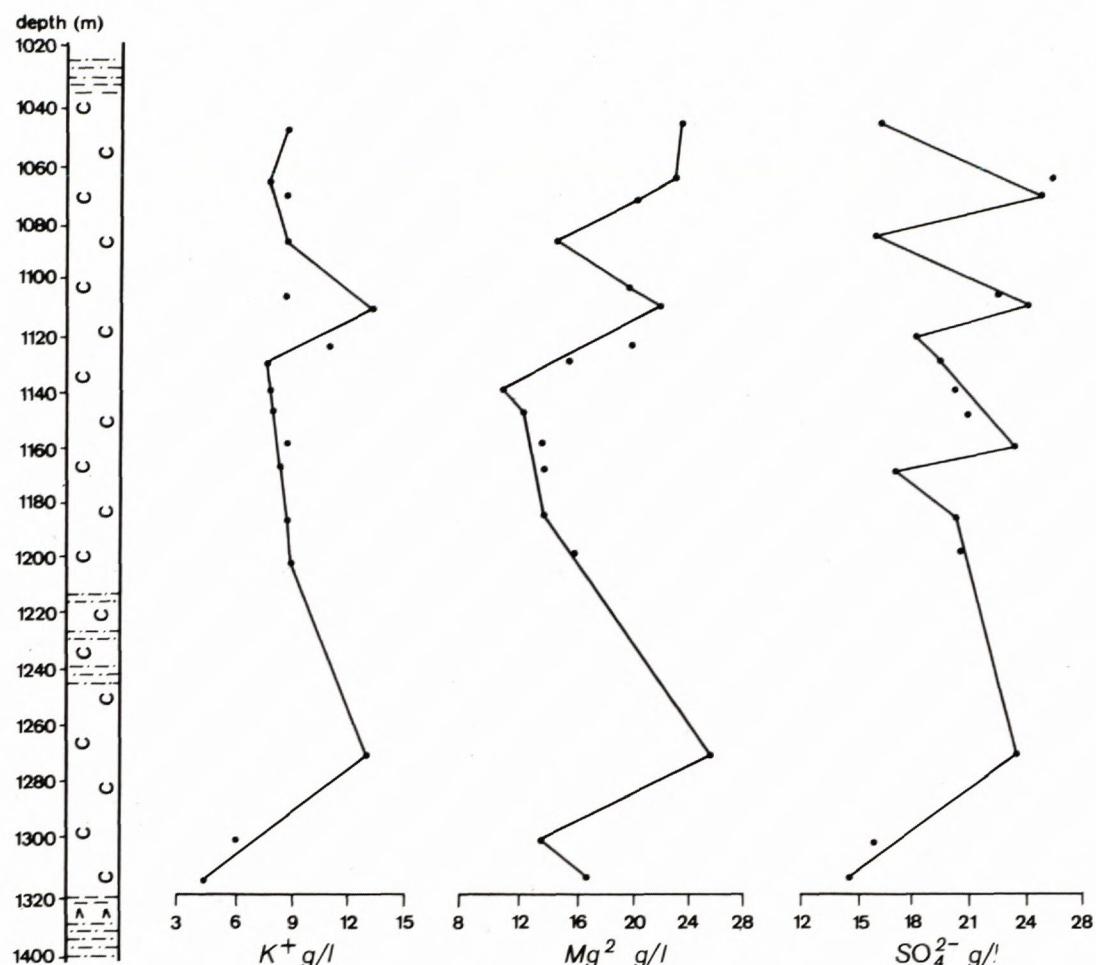


Fig. 4. The content of potassium, magnesium and sulfate ions in brine inclusions of sedimentary halite in the section of borehole 6-T. For explanation see Fig. 2.

halite stage. Concerning the sulfate ion, its content was somewhat lower (14.0 g/l) than in recent seawater. During halogenesis, the concentration of major elements gradually increased and during some stage of basin evaporation the potassium content in the brine reached 14.0 g/l, of magnesium 28.0 g/l, and of sulfate ion 24.0 g/l. The process of progressive evaporation was periodically broken by dilution of the brine by water of low concentration.

The location of the composition points in the diagram (Fig. 5) shows that the ratio of major elements in the brine did not actually differ from their ratio in modern seawater. The majority of the points is situated near the line of composition of directly metamorphised seawater (Valiashko, 1962). Halite precipitation terminated from the brine of higher concentration compared to the initial one, but the evaporation process did not reach the high stages and finished at halite stage. This is indicated by

results of the determination of bromine and the bromine-chlorine coefficient, which correspond to the halite stage of basin evolution and are close to experimentally determined for the normal halite zone. The temperature of the brine of the Zaluzh salt-bearing basin at the halite stage was below 40°C, and its depth was from 7 to 18 m (Petricenko, 1988).

The Solotvyne subbasin was also characterized by the sulfate type of brine during stage of halite precipitation. However, as sometimes the content of all major elements at halite stage reached much more higher values than in Zaluzh subbasin (potassium - 25.0 g/l; magnesium - 36.5 g/l; sulfate ion - 63.0 g/l), it may be concluded that the evolution of this salt-forming basin reached the later stages. The depth of the basin was 10-15 m, temperature of the brine did not exceed 40°C. Halite sedimentation took place in oxidizing conditions.

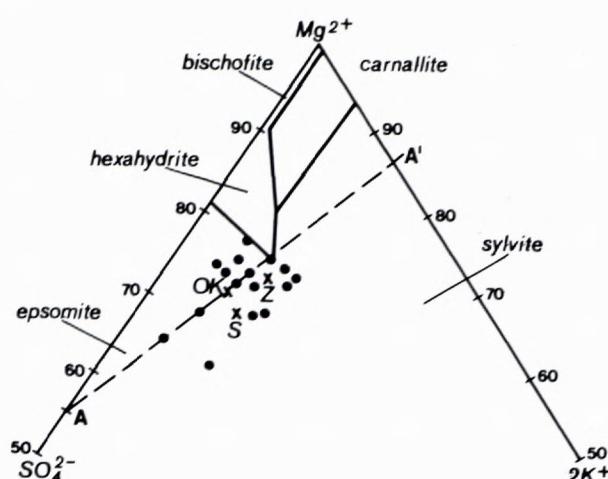


Fig. 5. Location of data points characterizing the chemical composition in salt-forming basin on stage of halite precipitation on the Valiashko (1962) diagram. OK - modern seawater composition; A-A' - the line of metamorphism of seawater (Valiashko, 1962); Z and S - average composition of solutions on halite stage of evolution of the Zaluzh and Solotvyno subbasins, accordingly.

Halite changes in diagenesis

The rock salts of the Chop-Mukacheve depression are significantly recrystallized. Rock structures and the composition of solid and individual fluid inclusions in diagenetic halite are evidence that the alteration of physico-chemical conditions took place during rock formation. Due to tectonic subsidence the salt sediments were subject to the pressure and temperature impacts. Taking into account the maximum present depth of studied rock salt (1314–1318 m), it may be supposed that the pressure during recrystallization reached 33,000–35,000 kPa. The temperature rose to 75–83°C (Petricenko, 1989).

The mineral-forming solutions during diagenesis as well as sedimentation were of $\text{Na}^+ \text{-} \text{K}^+ \text{-} \text{Mg}^{2+} \text{-} \text{Cl}^- \text{-} \text{SO}_4^{2-}$ -composition. The concentration of major elements became lower than during the sedimentation stage explainable by processes of cation exchange and metamorphism of the brine. The composition of solid inclusions in diagenetic halite represented by anhydrite, dolomite, quartz, and pyrite indicate that the mineral-forming environment during the postsedimentary stage changed from oxidizing to reducing conditions, and pH from acid to alkaline conditions. Specifically, pyrite crystallization needs strongly reducing conditions and an alkaline environment. Inclusions with oil hydrocarbons in recrystallized halite indicate strongly reducing mineral-forming conditions in salt deposits. The migration of oil hydrocarbons that were the source of feeding for sulfate-reduction bacteria producing H_2S , caused a significant decrease in the oxidation-reduction potential of the

environment and increase in the alkalinity of solutions. During the local recrystallization of salts it led to the preservation of hydrocarbons separately or together with solutions in inclusions.

The rock salt of the Solotvyno deposit has also been recrystallized under the increased pressure during diairism. The solutions had a sulfate composition but their total concentration decreased and the content of some elements changed in comparison to the precipitation stage. The recrystallization of halite occurred during oxidizing conditions and acid environment: pH of solutions measured after the opening of inclusions in halite ranges from 5.0 to 5.4 ± 0.2 . Eh changes from 0 to +100mv (Kityk *et al.*, 1983).

Conclusions

Studies of relict mineral-forming solutions in sedimentary halite show that the brine in Neogene Transcarpathian salt-forming basin (Zaluzh as well as Solotvyno subbasins), on the basis of composition belonged to a sulfate type, and were close to the brines from which modern marine evaporites precipitate. It has been found that the initial brine of the Zaluzh basin was depleted in SO_4^{2-} in comparison to modern seawater. Salt precipitation in basins terminated at the halite stage, but in the Solotvyno basin it reached a higher stage that was close to potash precipitation. The brine temperature did not exceed 40°C. Primary sedimentary deposits were significantly changed during postsedimentary time. Recrystallization occurred under increased pressure and temperature. Mineral-forming conditions changed from oxidizing to strongly reducing, and the pH of the environment from alkaline to acid. The recrystallization of primary sediments took place under the impact of solutions that have inherited the type, chemical composition and particularities of the behaviour of the main elements from brine of the salt-forming basin. They differed from solutions of sedimentary basins by a decreased concentration of major elements. In local parts of a salt deposit during recrystallization, hydrocarbons penetrated into burial solutions and, together with mineral-forming solutions or independently, were preserved in halite as inclusions.

Acknowledgments.

I thank the International Science Foundation (Grant No. UCM000) for the support and T. Dobroszycka for the drawings.

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