

Gypsum facies transitions in basin-marginal evaporites: middle Miocene (Badenian) of west Ukraine

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ABSTRACT

In the middle Miocene Badenian gypsum basin of the Carpathian Foredeep, west Ukraine, three main zones of gypsum development occur in the peripheral parts of the basin. Zone I consists entirely of stromatolitic gypsum formed in a nearshore zone. Zone II is located more basinward and is characterized by stromatolitic gypsum in the lower part of the section, overlain by a sabre gypsum unit. Zone III occurs in still more basinward areas and is characterized by giant gypsum intergrowths (or secondary nodular gypsum pseudomorphs of these) in the lowermost part, overlain by stromatolitic gypsum, sabre gypsum and then by clastic gypsum units. Correlation between these facies and zones has been achieved using lithological marker beds and surfaces. Of particular importance for correlation is a characteristic marker bed (usually 20–40 cm thick) of cryptocrystalline massive gypsum occurring in zones II and III. The marker was not distinguished in zone I, possibly because this bed is older than the entire gypsum section of that zone. These new results strongly suggest that the deposition of giant gypsum intergrowth facies and stromatolitic gypsum facies was coeval. In some sections of zones I and II, limestone intercalations have been recorded within the upper part of the gypsum sections. Considerable scatter of the $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ values of these limestones indicates variable diagenetic overprints of marine carbonates, but a marine provenance of the limestones is confirmed by microfacies analysis. Some of the limestones are coeval with an intercalation of gypsarenitic, mostly laminated gypsum occurring in the sabre gypsum unit of zones II and III. Badenian gypsum formed in extremely shallow-water to subaerial environments on broad, very low relief areas of negligible brine depth, which could be affected by rapid transgressions. Stable isotope ($\delta^{34}\text{S}$, $\delta^{18}\text{O}$) studies of the gypsum demonstrate that the sulphate was of sea-water origin or was derived from dissolution of Miocene marine evaporites. Investigations of individual inclusions in the gypsum indicate decreased water salinity when compared with modern marine-derived, calcium sulphate-saturated water. Groundwater influences are indicated by high calcium sulphate contents of the brines in the evaporite basin. The chemical composition of Badenian waters was thus a mixture of relic sea water (depleted in NaCl), groundwater (enriched in calcium sulphate) and surface run-off.

Keywords Badenian, depositional environments, facies, geochemistry, gypsum.

INTRODUCTION

The interpretation of ancient evaporite sequences has changed tremendously in the last three decades (Kendall & Harwood, 1996). Many evap-

orites that were once regarded as being products of deep hypersaline basins were reinterpreted as sabkha sediments, using supratidal, nodular anhydrites in the Persian Gulf as analogues. Subsequently, nodular anhydrites were recorded that

pseudomorphed upright-growth gypsum crystals, and a subaqueous environmental interpretation for such nodular anhydrites became fashionable. More recently, Kendall (1992) and Kendall & Harwood (1996), following Logan (1987), suggested that some shallow-water gypsum may have formed in desiccated environments subject to infrequent floods, and evaporites possessing subaqueous features may, in fact, be deposited on evaporitic flats that are only flooded infrequently during storms or abnormally high tides.

It has been recognized (Peryt, 1996) that the middle Miocene Badenian (Langhian–Serravallian; Fig. 1) gypsum of west Ukraine was mostly deposited, at least in the lower part of the stratigraphic section, in a vast brine pan. Although individual depositional features and facies types in the Badenian may be explained by comparison with modern salinas (Arakel, 1980; Warren, 1982; Orti Cabo *et al.*, 1984; Logan, 1987), the lateral persistence of thin beds over large areas with only minor changes in thickness and facies indicates that they formed on broad, very low relief areas that could be affected by rapid transgressions.

Such laterally persistent units, which are of great use for correlation purposes, have been recorded in many peripheral parts of the Badenian basin, including west Ukraine, southern Poland and the NE Czech Republic (Peryt *et al.*, 1994, 1997, 1998; Peryt, 1996). However, in the most peripheral parts of the basin in west Ukraine, a different facies occurs: entire sections,

or major parts of them, are built of stromatolitic gypsum. The correlation of sections characterized by mainly crystalline gypsum overlain by clastic gypsum and gypsum breccia with those consisting mostly (or entirely) of stromatolitic deposits remained enigmatic. Therefore, in this study, 29 new sections have been measured located in the transition zone between these two facies (Figs 2 and 3). The aim was to refine correlations and to determine whether (and how) major changes in brine chemistry, as recorded in the first facies, are expressed in the second. A stable isotopic study of three selected gypsum sections was undertaken to provide geochemical constraints on environmental interpretations. A more general aim was to combine the results of this detailed sedimentological study with earlier research on fluid inclusions in the Badenian gypsum of west Ukraine.

GEOLOGICAL SETTING

In the west Ukraine, Badenian gypsum occurs in the Carpathian Foredeep basin and the adjacent part of the East European Platform. The Carpathian Foredeep is a typical peripheral foredeep basin filled with synorogenic molasse sediments. The foredeep was initiated in early Miocene Eggenburgian times (for regional stratigraphy, see Fig. 1) and lasted at least until the end of the middle Miocene (Andreyeva-Grigorovich *et al.*, 1997). As the Carpathian Foredeep basin

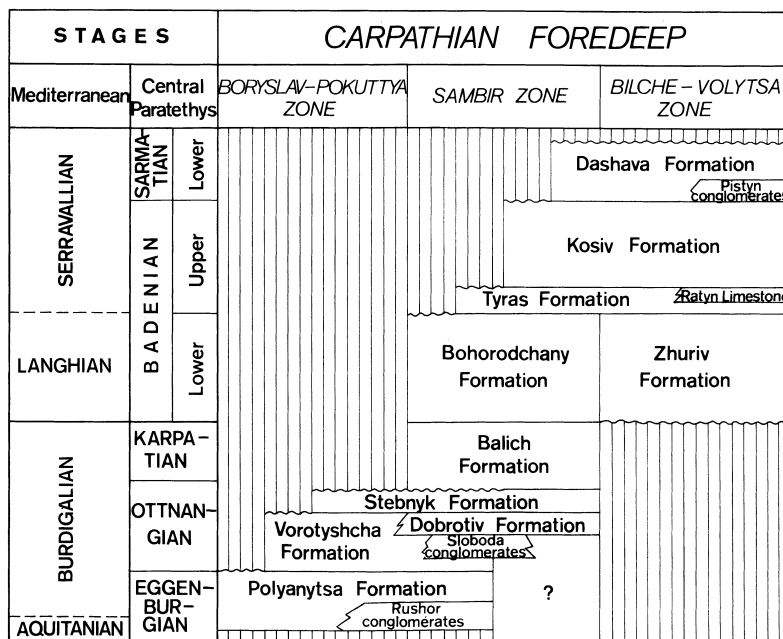


Fig. 1. Regional stratigraphy of Neogene deposits of the Ukrainian part of the Carpathian Foredeep (after Andreyeva-Grigorovich *et al.*, 1997).

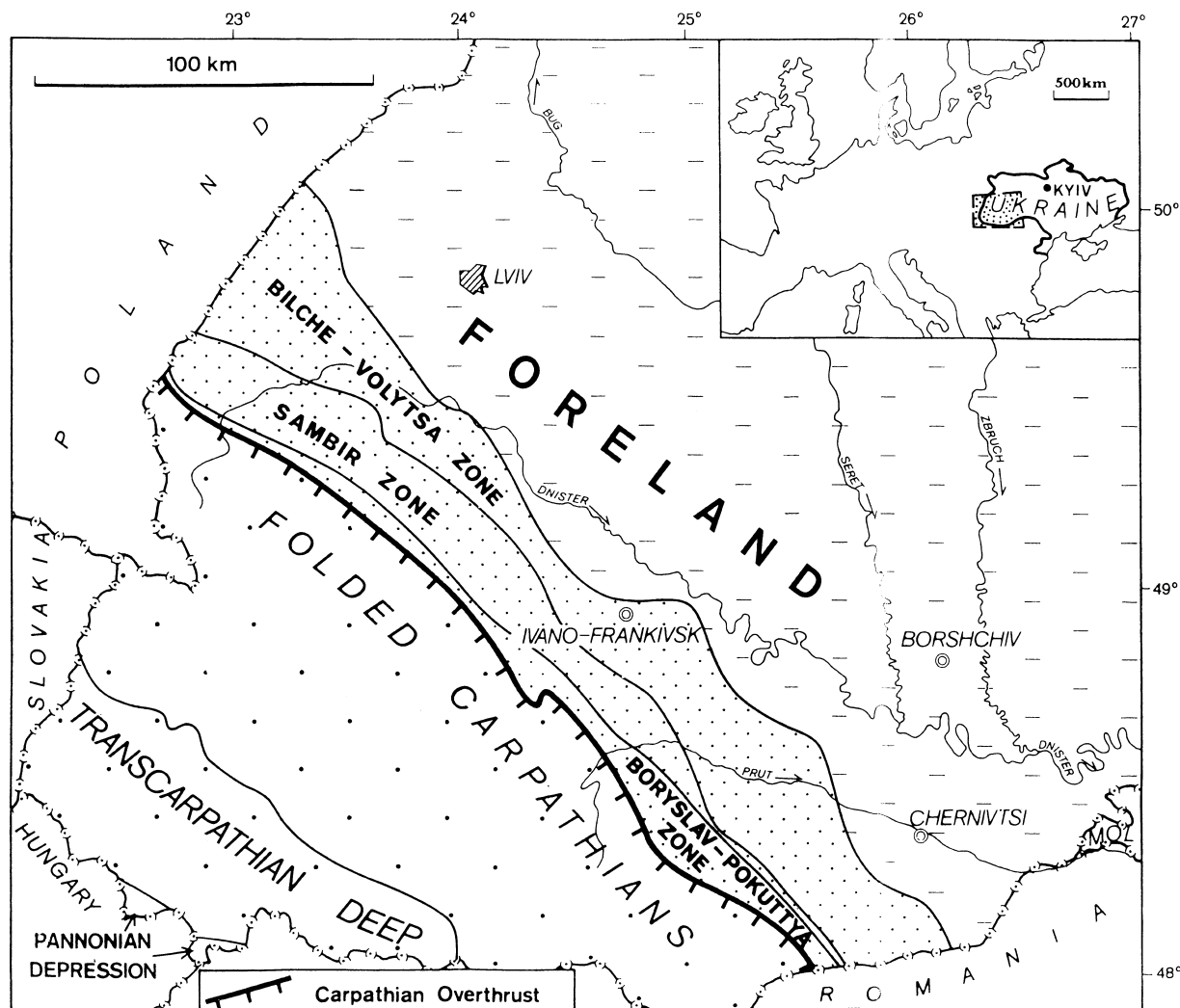


Fig. 2. The main tectonic units of west Ukraine (after Vul *et al.*, 1998). The Carpathian Foredeep area is indicated by the fine stipple; adjacent Folded Carpathians and Transcarpathian Deep areas (SW of the Carpathian Foredeep) are indicated by the coarse stipple, and the Foreland area (NE of the Carpathian Foredeep) is indicated by dashed lines; MOL, Moldova.

transgressed over the East European Platform, facies belts migrated towards the NE, and subsequently towards the SW (Panow & Plotnikow, 1996).

The Carpathian Foredeep in the Ukraine is subdivided into three parts. In the outer part (up to 50 km wide), called the Bilche–Volytsa zone (Fig. 2), Badenian to Sarmatian (Table 1) marine deposits are a few hundreds of metres to 3.5 km thick. In the other two zones of the Carpathian Foredeep (the middle, Sambir zone and the inner, Boryslav–Pokuttya zone), the Miocene sections begin in the Eggenburgian (Aquitanian–Burdigalian). In the Boryslav–Pokuttya zone, sections terminate with Ottnangian deposits, whereas in the Sambir zone, later deposits (Karpatian, Bade-

nian and Sarmatian) occur (Table 1). The allochthonous Sambir and Boryslav–Pokuttya zones are thrust over the autochthonous Bilche–Volytsa zone.

In the Bilche–Volytsa zone, the Badenian sulphates are included in the Tyras Formation (Fig. 1). They are typically laminated, 10–28 m thick and commonly contain clay intercalations (Smirnov *et al.*, 1995). In the part of the Badenian basin located on the East European Platform, the gypsum overlies Devonian, Cretaceous or Badenian deposits (Kudrin, 1955) and is overlain in turn by the Ratyn Limestone (a few tens of centimetres to a few metres thick), which is related to the next marine transgression (Peryt & Peryt, 1994). A nannoplankton study has shown

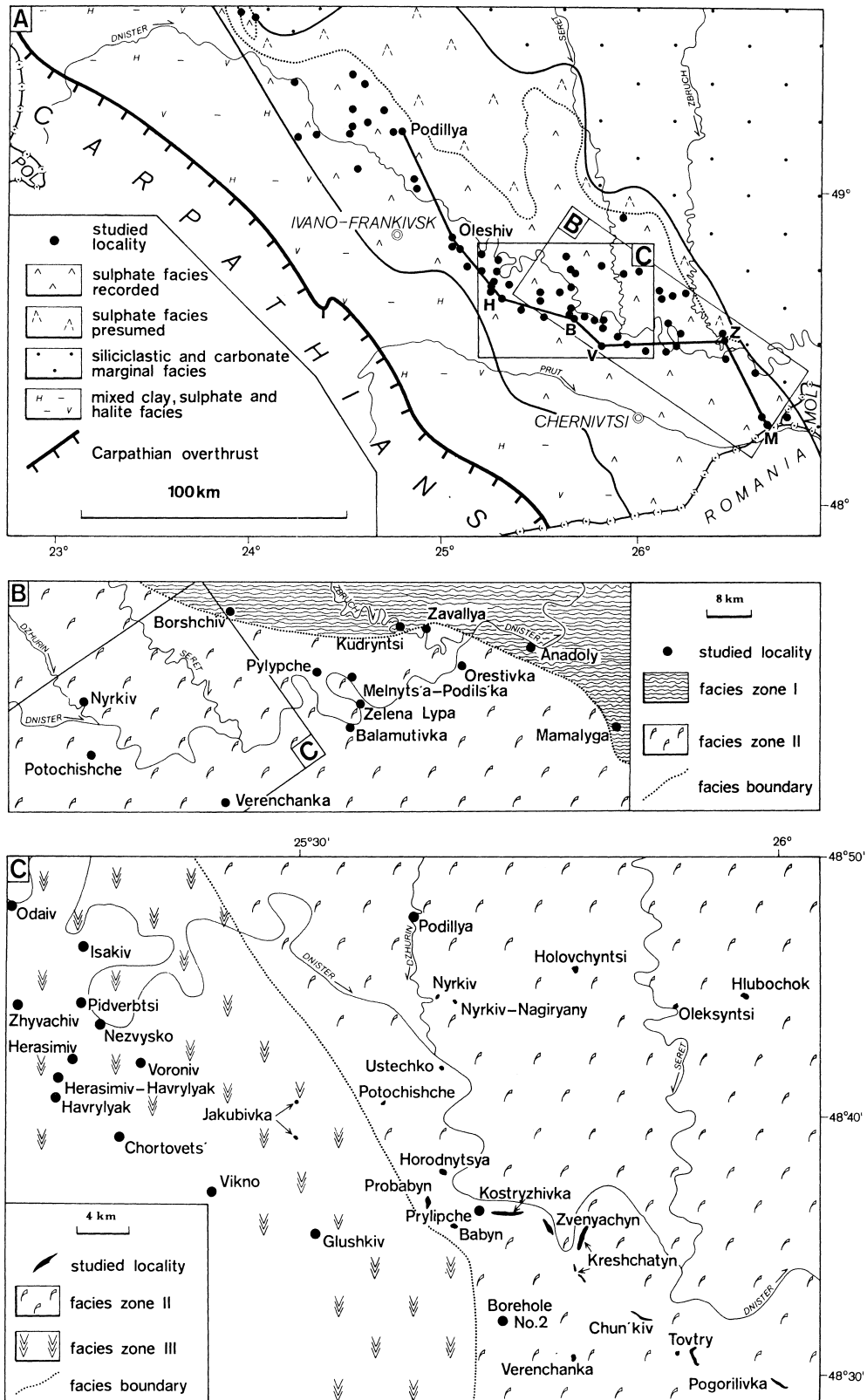


Fig. 3. (A) Badenian evaporite basin in west Ukraine, location of studied outcrops (partly after Peryt, 1996) and the distribution of correlated gypsum sections along the Podillya–Oleshiv–Havrylyak (H)–Babyn (B)–Verenchanka (V)–Zavallya (Z)–Mamalyga (M) line (see Fig. 11); Mol., Moldova. (B and C) Facies zones (described in text) and the studied localities [in (B), only part of outcrops shown in (C) is given].

that the Badenian gypsum corresponds to the lower part of the NN6 zone (cf. Peryt, 1997, 1999). The Ratyn Limestone is overlain by the Kosiv Formation, which is a few tens of metres thick in the marginal part of the Carpathian Foredeep and increases in thickness rapidly (reaching up to 1200 m) towards the central part of the Carpathian Foredeep.

The present pattern of gypsum thickness is complex (Panow & Płotnikow, 1996), mostly because of erosion before the deposition of the Ratyn Limestone (Peryt & Peryt, 1994). In the area shown in Fig. 3B and C, the gypsum sections are usually 20 m thick or less (maximum 25 m) but are much thicker (reaching >40 m) in local grabens where the gypsum sections are more complete. Such a local graben was recorded in borehole no. 2, located 6 km WNW of the Verenchanka quarry section. In this graben, above the stromatolitic gypsum and overlying sabre gypsum units (and their anhydrite equivalents) that form the entire gypsum section at Verenchanka, ≈20 m of mostly laminated sulphate and sulphate breccia (Fig. 4) occur.

The sedimentology of the Badenian gypsum in west Ukraine has been studied by Peryt (1996; which includes a review of earlier studies) and Petrichenko *et al.* (1997). Previous work has shown that autochthonous gypsum facies (crystalline gypsum, stromatolitic and massive alabastine gypsum) were deposited in two main environments. One variety of crystalline gypsum (giant gypsum intergrowths) precipitated from highly concentrated brines in moderate water depths at the initial stage of gypsum precipitation (stage A). Other varieties of autochthonous gypsum facies represent very shallow subaqueous gypsum deposited in a vast brine pan developed during stage B of gypsum deposition. The brine pan was characterized by a facies mosaic that reflected an interplay of concentrated brines from the central part of the evaporite basin, diluted brines resulting from the influx of continental meteoric waters and local evaporation of a restricted shallow-water body. Consequently, salinities varied considerably. A facies continuum, microbial gypsum – grass-like selenite – skeletal selenite – sabre gypsum, indicates increasing salinity of the brine with time. During the formation of the allochthonous gypsum (encompassing clastic gypsum and gypsum breccias), an influx of sea water is indicated by a fauna of molluscs, foraminifera (mostly pelagic globigerinids), ostracods and pteropods, recorded on clays intercalated within the gypsum (Venglinskiy &



Fig. 4. Gypsum breccia in the upper, allochthonous part of the Badenian gypsum section in borehole no. 2 (Fig. 3C); depth 83.5 m.

Goretskiy, 1966). The allochthonous gypsum unit was deposited during stage C.

METHODS

Stratigraphic studies, detailed mapping of selected zones and sampling were carried out in all existing outcrops in the study area. Stratigraphic sections were measured and plotted at a 1:100 scale. The main lithology observed was gypsum. The classification used in this paper follows that of Peryt (1996) and is based on the most common types of gypsum. These types may be grouped into two broad categories: autochthonous and allochthonous. Autochthonous gypsum forms

most of the lower part of the Badenian section, whereas allochthonous gypsum occurs mostly in its upper part (Peryt, 1996). In the area shown in Fig. 3B and C, allochthonous gypsum is rare on account of non-deposition or erosion before the deposition of the Ratyn Limestone; the allochthonous facies is preserved only in local grabens.

A total of 122 gypsum samples from three sections (Verenchanka, Potochische and Mamylyga) was selected for stable isotope analysis. Each sample was first dissolved in distilled water and, after filtration, the solution was acidified with HCl to pH 1, and then a 10% solution of BaCl₂ was added to precipitate BaSO₄. The precipitate was washed with water to eliminate Cl⁻ ions, centrifuged and dried at 110 °C. Barium sulphate was then treated with different analytical procedures to determine sulphur and oxygen isotopes. SO₂ was produced by the reduction of NaPO₃ by barium sulphate at a temperature of

850 °C (Halas & Wolacewicz, 1981). CO₂ was produced in a separate vacuum line, in which BaSO₄ was quantitatively reduced by spectrally pure graphite in a thin platinum boat at 1000 °C to BaS and CO, which was converted to CO₂ by glow discharge between platinum electrodes (Mizutani, 1971). The SO₂ and CO₂ gases were collected into glass ampoules and analysed on a dual inlet and triple collector mass spectrometer (rebuilt Russian MI1305). The isotopic composition of oxygen and sulphur was expressed per mille (‰) relative to the VSMOW and VCDT international standards respectively. Analytical precision was 0.05–0.08‰ for δ¹⁸O and δ³⁴S respectively. Results are shown in Figs 5–7.

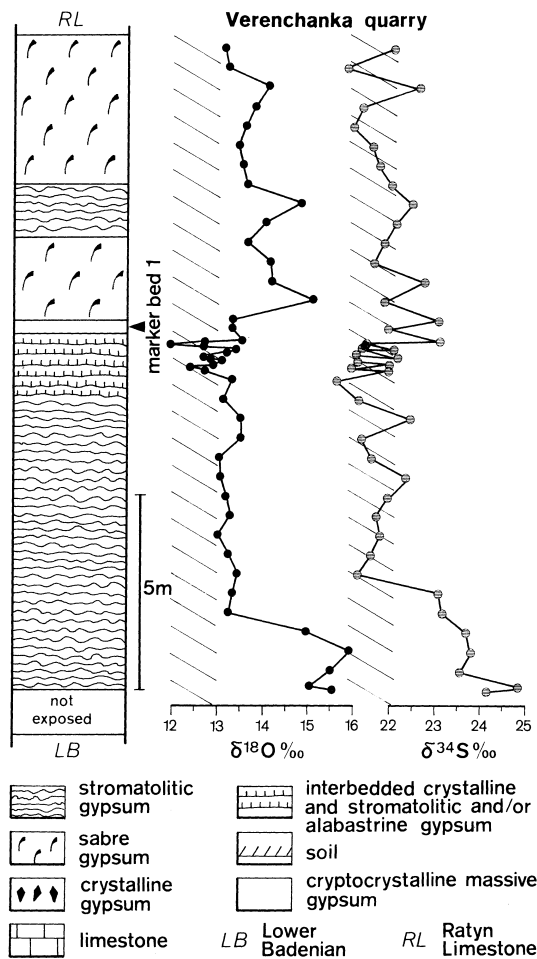


Fig. 5. Badenian gypsum section at Verenchanka and δ³⁴S and δ¹⁸O profiles. Obliquely hatched strips show average Middle Miocene values for δ³⁴S and δ¹⁸O.

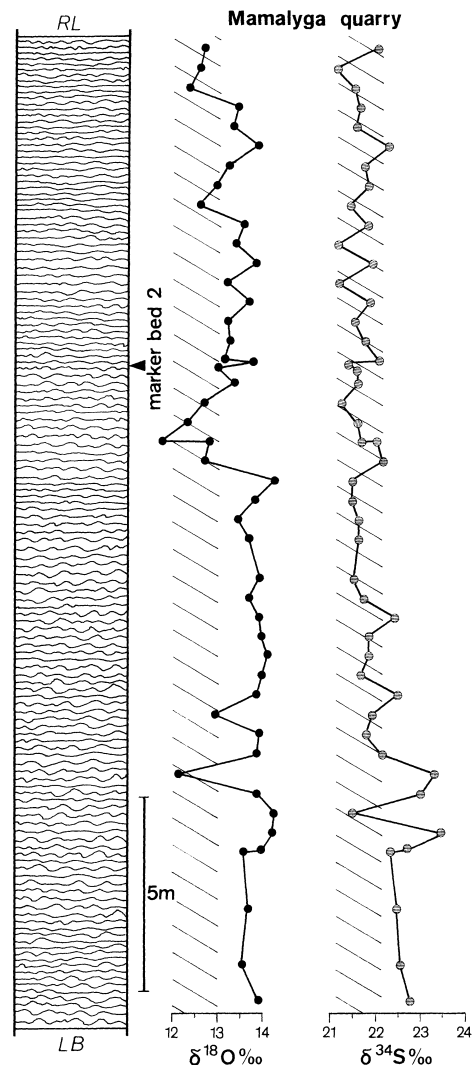


Fig. 6. Badenian gypsum section at Mamalyga and δ³⁴S and δ¹⁸O profiles. Arrow points to marker bed 2 (Fig. 11), which is illustrated in Fig. 16. Symbols and abbreviations as in Fig. 5.

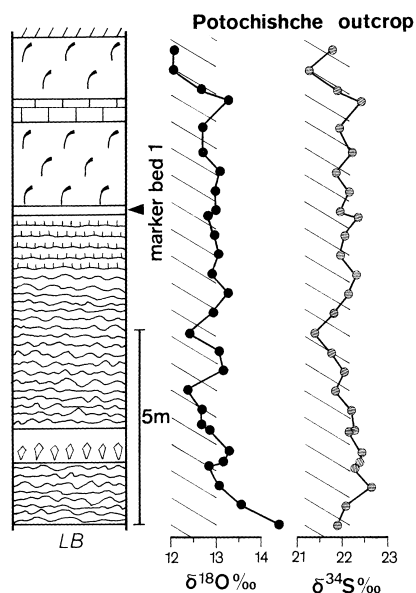


Fig. 7. Badenian gypsum section at Potochishche and $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ profiles. Symbols and abbreviations as in Fig. 5.

In the limestone intercalated within the gypsum, the main objective was to determine the fossil content and to carry out petrological and geochemical investigation. CO_2 gas was extracted from the samples by reaction of calcite with H_3PO_4 (McCrea, 1950) at 25 °C in a vacuum line, according to the NBS-19 reference standard. The gas was H_2O purified on a P_2O_5 trap and collected on a cold finger. Isotopic compositions were determined using a modified Russian MI1305 triple collector mass spectrometer (Durakiewicz, 1996) equipped with a gas ion source. Isobaric corrections were applied to measurements of isotopic ratios. After subsequent normalization to measured certified reference materials, the isotopic composition was expressed per mille (‰) relative to the VPDB international standard. Analytical precision of both $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ in a sample was $\pm 0.08\text{‰}$. The results of the analyses are shown in Fig. 8, which also shows the results of analyses of other Badenian limestones in the study area from the Ervilia Bed and Ratyn Limestone (data partly from Peryt & Peryt, 1994).

CORRELATION OF GYPSUM SECTIONS

There are three main types of gypsum section. The first consists entirely of stromatolitic gypsum (e.g. Mamalyga quarry; Fig. 6) and is characteristic of the area bordering the nearshore sulphate facies limits (Fig. 3B). The stromatolitic gypsum

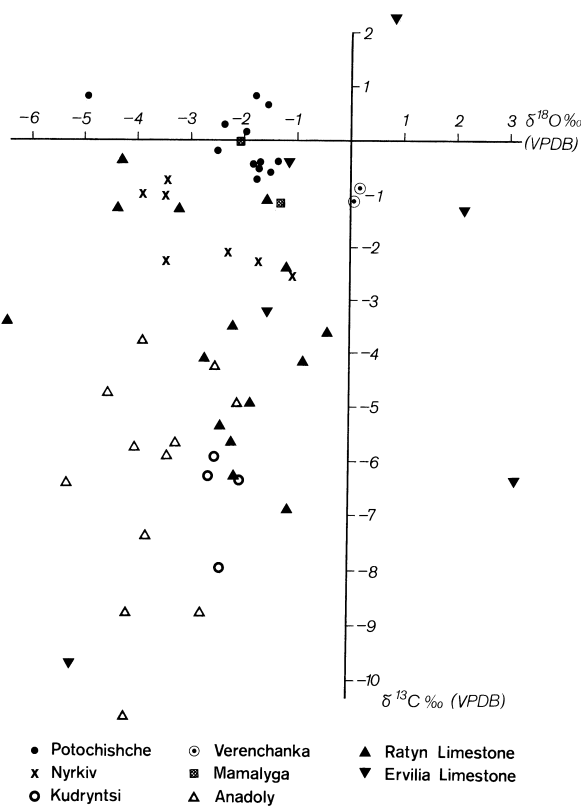


Fig. 8. Plot of $\delta^{18}\text{O}$ vs. $\delta^{13}\text{C}$ values from Badenian limestones occurring within the gypsum succession. Data for limestones occurring below (Ervilia Bed) and above (Ratyn Limestone) the gypsum are also plotted.

exhibits a considerable variety of forms, although planar or slightly crenulated stromatolites prevail. In the lower part of the unit, stromatolites show common intercalations and laminae of clastic gypsum and, as a result, the original planar structure is obliterated (Fig. 13 of Peryt, 1996). The second type of gypsum section is located further towards the basin centre. This type is characterized by the occurrence of stromatolitic gypsum (Fig. 9) in the lower part of the section, and sabre gypsum (occasionally with a clastic gypsum unit above the sabre gypsum) in the upper part (e.g. Verenchanka quarry and Potochishche outcrop; Figs 5, 7 and 10). The third type occurs in still further basinward locations and is composed of giant gypsum intergrowths (or secondary nodular gypsum pseudomorphs of that facies) in the lower part, overlain by stromatolitic gypsum, sabre gypsum (Fig. 11) and clastic gypsum units (Fig. 10). The distribution of these facies is shown in Fig. 3B and C.

The correlation between facies zones was based on the occurrence of characteristic marker

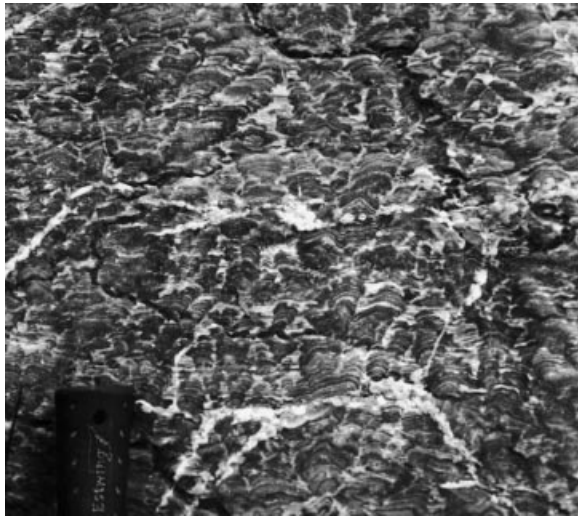


Fig. 9. Stromatolitic gypsum; Zvenyachyn. The hammer shaft is 3.5 cm across.

beds, as well as the observations made in areas transitional between the facies zones (Fig. 3B and C). Of particular importance for correlation is a characteristic marker bed (usually 20–40 cm thick) occurring a few tens of centimetres to a few metres above the giant gypsum intergrowths unit in zone III. This marker (bed 1 in Fig. 10) is developed across much of the Carpathian Fore-deep basin (Peryt *et al.*, 1994, 1998). The marker bed (Fig. 12) is underlain by a unit consisting of interbedded crystalline and stromatolitic and/or alabastrine gypsum, which is in turn underlain by stromatolitic gypsum. Marker bed 1 consists of cryptocrystalline massive gypsum with a distinctive finely crenulated lamination and a dome-like to cauliflower-like top. It was possible to distinguish the same marker bed in zone II, where it overlies a unit consisting of interbedded crystalline and stromatolitic and/or alabastrine (Fig. 13; see also Turchinov, 1999). The

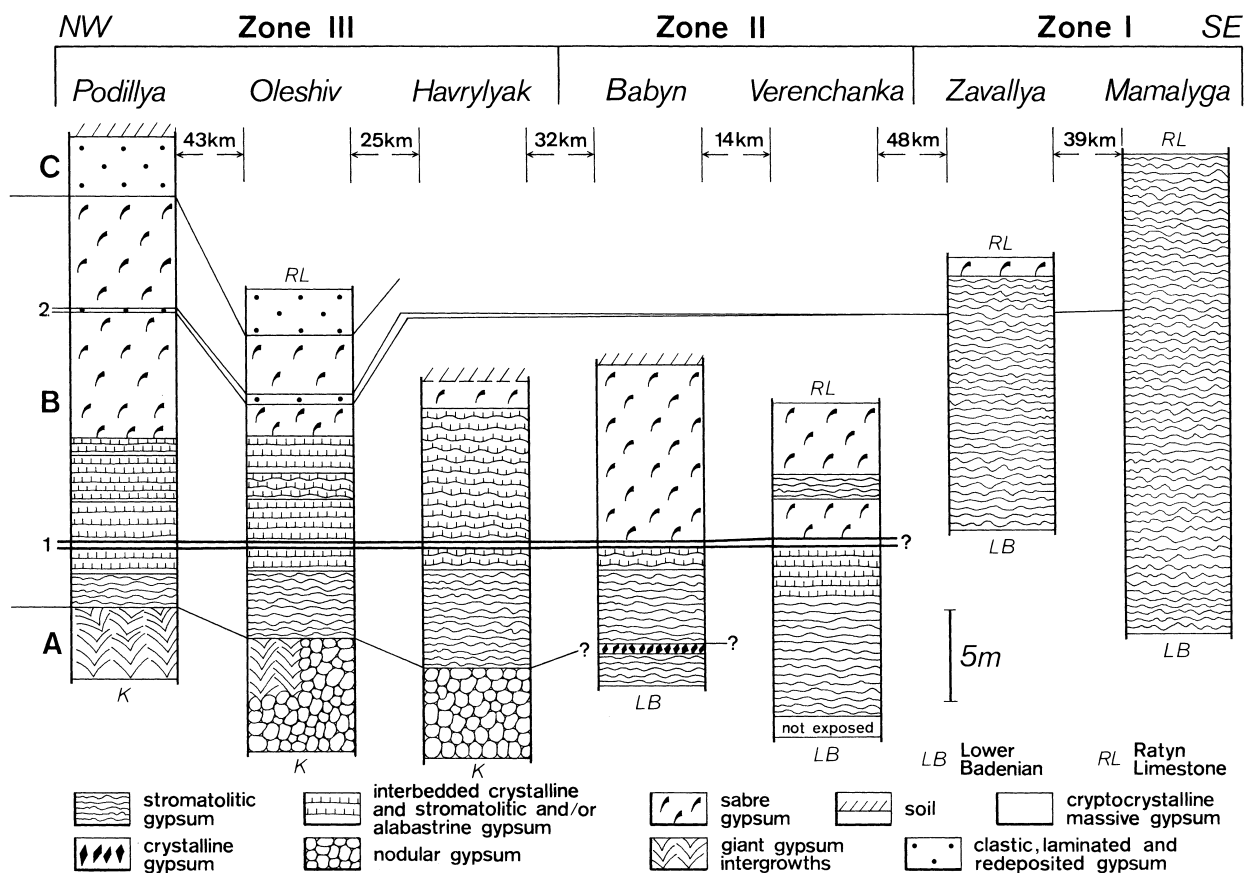


Fig. 10. Correlation of selected Badenian gypsum sections in west Ukraine (see Fig. 3A for location of the correlation line). (A–C) Stages in the Badenian gypsum basin history [see Peryt, 1996; (A) onset of hypersalinity; (B) brining upward and transgression; (C) cannibalism]. 1 and 2, regional marker beds (1, bed of cryptocrystalline massive ‘alabastrine’ gypsum occurring throughout the major part of the marginal area of the Badenian evaporite basin; 2, bed of clastic, laminated and redeposited gypsum forming an intercalation within sabre gypsum unit passing shoreward into thin discontinuous lenticular limestone).



Fig. 11. Bedded sabre gypsum unit with crystals bent predominantly towards the left side of the photo (WSW); Podillya. The hammer shaft is 30 cm long.



Fig. 12. Stromatolitic gypsum unit overlain by intercalated stromatolitic and selenitic gypsum; arrows indicate the boundary of both gypsum units. The right hand of M. Jasionowski lies on the upper surface of the marker bed 1 (Fig. 10) composed of alabastrine gypsum. Oleshiv.



Fig. 13. Stromatolitic gypsum (S) overlain by bedded selenite with thin intercalations of stromatolite and/or alabastrine gypsum (C); this unit is overlain by the marker bed 1 (A) composed of alabastrine gypsum upon which M. Jasionowski is standing. Horodnytsya.

marker was not distinguished in zone I, perhaps because the entire gypsum section of this zone is younger than the marker bed in zones II and III.

Above marker bed 1, selenitic and stromatolitic facies occur (Fig. 10); their upper part is devel-

oped as sabre gypsum (Peryt, 1996). There are continual lateral facies changes between stromatolitic and selenitic gypsum, which can be observed within particular outcrops and, thus, the distance between the marker bed and the sabre gypsum unit fluctuates. Within zone III, it is possible to distinguish a thin (usually 20 cm) unit

of gypsarenitic (marker bed 2 in Fig. 10), mostly laminated gypsum within the sabre gypsum unit. This unit is also rarely recorded in zone II, whereas in zone I, at approximately the same position in the gypsum section, there occurs a distinct surface (e.g. at Mamalyga, Fig. 14A; and at Borshchiv) or a limestone intercalation (at Kudryntsi, Fig. 15A and B). Finally, another important correlation point is the autochthonous/clastic gypsum contact (B/C boundary in Fig. 10). This facies boundary seems to reflect a major change in the basin history (see Peryt, 1996) and, as such, is considered to be essentially isochronous.

The marker beds seem to reflect events that may be related to sudden and widespread changes in water chemistry, which in turn imply major changes in basin hydrology. The marker beds make it possible to interpret the

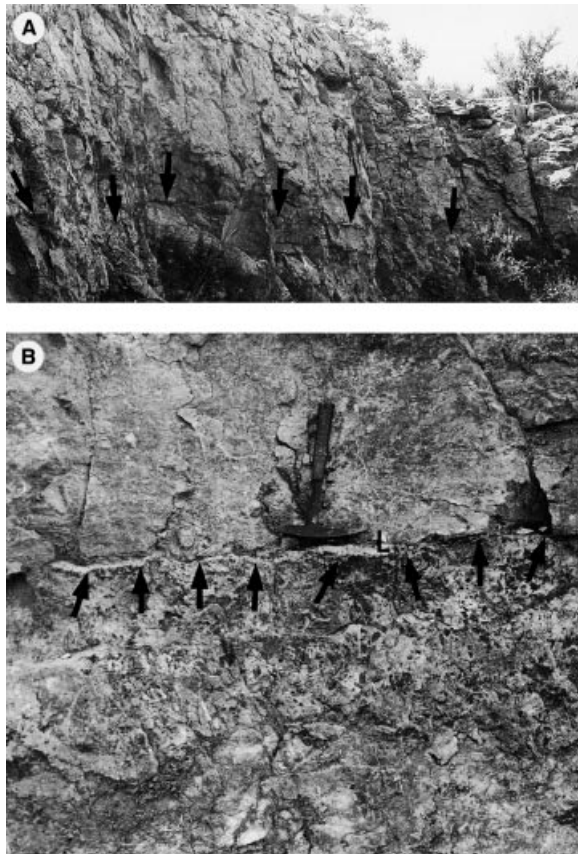


Fig. 14. The distinct boundary (arrowed) within the upper portion of the gypsum section at Mamalyga. (A) General view. (B) Detail of the boundary showing the lenticular nature of the limestone (light coloured area marked L, below the hammer). The brecciated appearance of the gypsum below the boundary is a weathering feature.

facies changes in the portions of the sections occurring between them, and thus to establish a framework for the study of the facies transitions. However, this is possible only if the major part of the section is composed of primary gypsum. When only secondary (nodular) gypsum occurs, as with some outcrops located along the Dnister River (e.g. Balamutivka, Zelona Lypa), it is not possible to subdivide the gypsum succession.

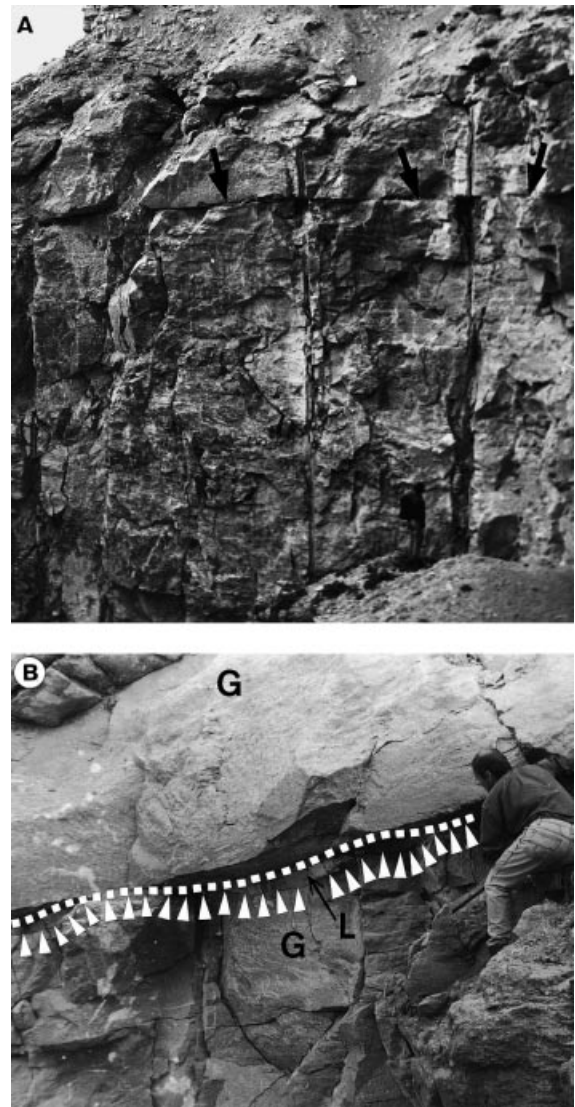


Fig. 15. The distinct boundary (arrowed) within the upper portion of the gypsum section at Kudryntsi. (A) General view of the gypsum outcrop (field assistant standing to the right is 1.75 m tall). (B) Detail of the boundary showing the irregular, lenticular nature of the limestone (G, gypsum; L, limestone) that occurs between the lower arrowed surface and the upper dotted one.

GIANT GYPSUM INTERGROWTH-STROMATOLITIC GYPSUM FACIES TRANSITION

When the gypsum section below marker bed 1 is compared in zones II and III, it can be seen (Fig. 10) that the thickness of the giant gypsum intergrowths (and/or nodular gypsum) decreases, and the thickness of the overlying stromatolitic gypsum increases, towards zone II. This thickness variation might be interpreted as a result of the existence of facies transitions between the giant gypsum intergrowth unit and the stromatolitic gypsum unit, and such a conclusion is supported by the occurrence of an intercalation of crystalline gypsum in the stromatolitic gypsum unit (Fig. 16). This intercalation (0.5–1.0 m thick) occurs ≈ 1.5 m above the base of the gypsum succession and was recorded in a number of sections (Potochishche, Gorodnitsa, Babyn and Zvenyachyn).

The giant gypsum intergrowths are interpreted as having formed by continual precipitation from a brine body, although fragmented giant gypsum intergrowths that occur in the upper part of the giant gypsum intergrowth unit suggest periodic high-energy events (Peryt, 1996). Stromatolitic gypsum seems to have originated from diluted brines in a shallower part of the brine pool (Peryt, 1996). Thus, the temporary establishment of crystalline gypsum facies in the shallower part of the basin, where stromatolitic gypsum normally formed, can be explained by storm-induced flooding of the brine pan and the evaporite flat.

LIMESTONE INTERCALATIONS WITHIN THE GYPSUM

Description

Limestone intercalations within the upper part of the gypsum sections have been recorded at Anadoly, Mamalyga, Kudryntsi, Potochishche, Nyrkiv and Verenchanka. The Anadoly section is located close to the limits of the basin, where transitions between gypsum and siliciclastic (and minor carbonate) facies are commonplace. Limestone forms lenticular bodies, a few tens of centimetres long and up to a few centimetres thick, within the upper (2–8 m below the top) part of gypsum sequence, which is developed mostly as recrystallized gypsum. The limestone is peloidal and bioclastic and exhibits grainstone and packstone textures. Stable isotope values range from -2.04‰ to -5.22‰ $\delta^{18}\text{O}$ and from -3.77‰ to -10.58‰ $\delta^{13}\text{C}$.

At Mamalyga, there are some clear surfaces that can be traced throughout the quarry (e.g. Fig. 14A). One of these surfaces, located 8.3 m below the Ratyn Limestone, shows small (centimetric) relief, and some broad depressions are filled by limestone (a few centimetres thick; Fig. 14B). The limestone shows packstone and grainstone textures and is peloidal and intraclastic with marine bioclasts (foraminifera – mostly benthonic, molluscs and echinoids) in the lower part. In the upper part, the limestone is peloidal and barren of fauna and shows wackestone



Fig. 16. Blocky crystalline gypsum forming an intercalation within the lower part of a stromatolitic gypsum unit. The hammer is 33 cm long. Babyn.

and packstone textures. Stable isotope values obtained were -1.30‰ and -2.03‰ $\delta^{18}\text{O}$, and -1.16‰ and -0.02‰ $\delta^{13}\text{C}$ for the samples coming from the lower and upper parts respectively.

At Kudryntsi, a thin (up to 20 cm) limestone intercalation occurs (Fig. 15A and B) in the upper part of the gypsum sequence (≈ 2.5 m below the Ratyn Limestone). The gypsum surface below the intercalation is clearly channelized (Fig. 15B). This surface may be correlated, based on its position in the section, with the prominent erosion surface (3 m below the base of the Ratyn Limestone) in the Zavallya outcrop, where an almost continuous series of outcrops occurs along more than 1 km of the Zbruch river valley. This surface was formerly regarded as the equivalent of the autochthonous–allochthonous gypsum boundary (Fig. 5 of Peryt, 1996), but locally 1- to 2-m-thick sabre gypsum occurs at the top of the gypsum section at Zavallya (Fig. 10). The limestone at Kudryntsi exhibits mudstone and wackestone textures and contains a marine biota including planktonic foraminifera, pteropods, ostracods and a calcareous nannoplankton assemblage. Stable isotope values obtained were -2.03‰ to -2.56‰ $\delta^{18}\text{O}$ and -5.91‰ to -7.93‰ $\delta^{13}\text{C}$.

In the Potochishche and Nyrkiv sections, a limestone intercalation (70 cm thick) occurs in the sabre gypsum (Fig. 7). The limited outcrops of those parts where the limestone occurs makes it impossible to exclude the possibility that the limestone is a calcareous filling of karst cavities within the gypsum, and was formed during Ratyn Limestone deposition. Such a supposition is supported by its composition, which is mostly peloidal and intraclastic packstone and grainstone with gastropods and molluscs and is identical to that occurring in the Ratyn Limestone (Peryt & Peryt, 1994). On the other hand, the limestone occupies the same position in the gypsum section as the stromatolitic gypsum intercalation recorded at Verenchanka within the sabre gypsum unit. This is interpreted as indicating a decrease in brine salinity (cf. Peryt, 1996), so the carbonate deposition recorded in Potochishche and Nyrkiv could be coeval with gypsum deposition elsewhere. Stable isotope values obtained were -1.03‰ to -3.92‰ $\delta^{18}\text{O}$ at Nyrkiv and -1.38‰ to -4.94‰ $\delta^{18}\text{O}$ at Potochishche, -0.69‰ to -2.55‰ $\delta^{13}\text{C}$ at Nyrkiv and -0.70‰ to 0.85‰ $\delta^{13}\text{C}$ at Potochishche.

At Verenchanka, thin limestone laminae have been recorded below marker bed 1. They form the base of rhythmic sets (13–20 cm thick) composed

of millimetre-thick micritic limestone lamina, followed by a few centimetre-thick stromatolitic gypsum, and then by 10 to almost 20 cm of selenitic gypsum (Fig. 17). The $\delta^{18}\text{O}$ values obtained for two samples of this limestone were $+0.05\text{‰}$ and $+0.19\text{‰}$, and $\delta^{13}\text{C}$ values were -1.10‰ and -0.91‰ respectively.

Interpretation

The microfacies indicate a marine provenance for these limestones. They originated through an influx of marine water into an otherwise evaporitic environment. The driving force to bring that water could be storms, especially in the case of thin limestones filling microrelief of an earlier exposed gypsum surface. In the case of the thicker limestone bodies such as those observed at Potochishche and Nyrkiv, it seems that the best explanation is that there was a longer term supply of open marine water to the sedimentary environment.

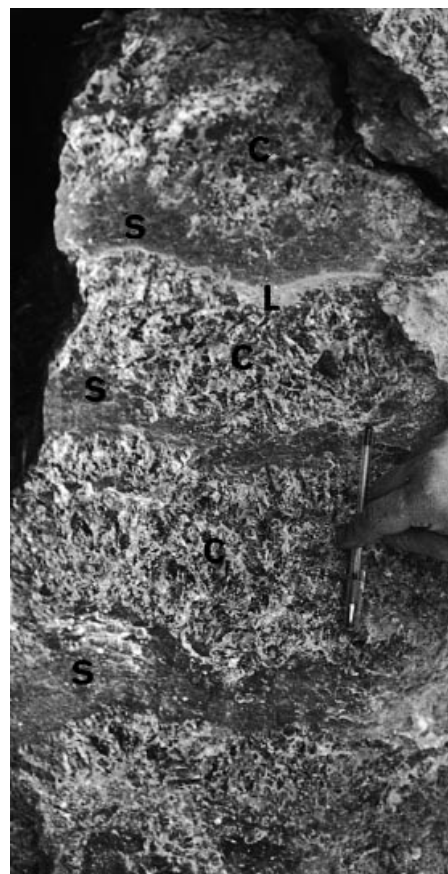


Fig. 17. Detail of the gypsum section below alabastrine marker bed 1, with locally occurring limestone (L). S, stromatolitic gypsum; C, selenitic gypsum. The pen is 15.5 cm long. Verenchanka.

Both $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the limestones occurring within the gypsum show a large range and scatter of values that are comparable with overlying Badenian limestone of marine origin (Fig. 8). The isotope ratios of the studied samples reflect their complex diagenetic histories and are probably not representative of the depositional palaeoenvironment. Only samples from Verenchanka can be regarded as maintaining the isotopic memory of precipitation from evaporating sea water. All the other samples show more negative oxygen and carbon isotope ratios as a result of post-depositional isotope exchange between carbonate rock and circulating fluids. Very similar isotopic variation to that shown in Fig. 8 has been recorded from the Messinian Calcare di Base in Sicily (Censi *et al.*, 1980; Montana *et al.*, 1993), which has been explained as resulting from inputs of meteoric water into the depositional environment and (sub)recent isotope exchange phenomena in a diagenetic and/or vadose/phreatic environment (Montana *et al.*, 1993).

GYP SUM DEPOSITION

The $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values shown in Figs 5–7 show relatively wide ranges (+20.8 to +24.8‰ for $\delta^{34}\text{S}$ and +11.8 to +15.8‰ for $\delta^{18}\text{O}$). The average values for the Mamalyga, Verenchanka and Potochishche sections were, for $\delta^{34}\text{S}$, +21.9, +22.1 and +22.1‰ and, for $\delta^{18}\text{O}$, +13.6, +13.5 and +12.9‰ respectively. These averages are close to the range of δ -values reported for Middle Miocene marine sulphate (cf. Claypool *et al.*, 1980; Attia *et al.*, 1995; Paytan *et al.*, 1998). This suggests that the sulphate was of sea-water origin or was derived from the dissolution of Miocene marine evaporites.

At Mamalyga and Potochishche, both $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ show a general slight decrease upwards (Figs 6 and 7). At Verenchanka, both $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ show a decrease upwards within the unit of stromatolitic gypsum, and then a sharp increase in the $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ values above marker bed 1 is followed by a general gradual decrease in both $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ (Fig. 5). In general, the isotopic composition of both oxygen and sulphur show similar evolution trends throughout the sections (Figs 5–7). The anomalous isotopic values of some gypsum samples probably reflect variation between isotope species, controlled by redox conditions in the basin (Kasprzyk, 1997). In recent marine salt pans from the Mediterranean coast of France and SE Spain, such isotopic variations are attributed to partial bacterial sul-

phate reduction in organic-rich sediments. Sulphate reduction is followed by reoxidation of reduced sulphur compounds in well-mixed water, for which $\delta^{18}\text{O}$ values vary as a function of the evaporation–dilution mass balance (Pierre & Fontes, 1982; Pierre, 1989). Badenian gypsum typically abounds in organic matter, bitumens and reduced iron species, suggesting reducing conditions during gypsum precipitation (Petrichenko *et al.*, 1997). Kasprzyk (1997), who studied the Badenian gypsum in southern Poland, proposed that such conditions favoured anaerobic microbial activity and continual production of H_2S during the sulphate reduction reaction, and simultaneous ^{18}O and ^{34}S enrichments in residual sulphate. This interpretation would also seem appropriate for the sections studied here.

The Badenian gypsum section in the west Ukraine can be divided into two parts, although in most cases, the upper part is absent, having been deposited only locally, in tectonically controlled troughs, such as those near Ustechko and NNW of Verenchanka. The gypsum of this upper, clastic part of the section consists of detritus derived from erosion of the lower part of the gypsum section, and possibly of primary precipitates, and was deposited in a basin with a bottom topography of at least a few tens of metres. In contrast, during the deposition of the lower part of the gypsum section, the basin relief was minor, and the differences in water depth between facies zones I and III probably did not exceed 10 m.

In zone III, the gypsum facies indicate a mostly subaqueous environment, with the exception of the stromatolitic gypsum and alabastrine gypsum facies. Zone II is transitional in character between zones III and I. In zone I, only stromatolitic gypsum facies occur that were deposited from evaporating brine sheets on a subaerially exposed evaporative flat (cf. Peryt, 1996). In zone II, stromatolitic gypsum dominates the lower portion of the gypsum sections, whereas sabre gypsum occurs in the upper portion of the autochthonous part of the gypsum sections (Fig. 10). Sabre gypsum was formed in a very shallow subaqueous environment with brine depths of a few metres at most (Peryt, 1996). Babel *et al.* (1999) suggested that the orientation of sabre gypsum crystals with apices turned in one horizontal direction indicated the direction of inflowing brine. Measurements indicate that the brine generally circulated along the shores of the basin in a counterclockwise direction, although in places a more complex flow pattern developed, indicating a variable bottom

morphology (caused by the presence of shoals and islands). During the course of Badenian gypsum deposition, the sabre gypsum facies migrated shoreward and, therefore, the gypsum sections exhibit a general transgressive sequence.

The sabre gypsum unit contains an intercalation of gypsarenitic, mostly laminated gypsum (marker bed 2), which is correlated with the limestone intercalations recorded in some outcrops (Fig. 10). The inflow waters were normal marine, as indicated by the presence of marine fossils in the limestones so, in such cases, gypsum precipitation resumed once the entire brine again became supersaturated with gypsum. Saturation could have resulted from either evaporative concentration or mixing.

DISCUSSION

Badenian gypsum formed in a standing body of water, as well as in desiccated environments subject to infrequent floods. Solid inclusions in gypsum, such as trapped microorganisms (calcareous nannoplankton and charophytes, among others) in sabre gypsum crystals (Petrichenko *et al.*, 1997) and detrital particles, as well as intercalations of limestone, all indicate that inflowing waters periodically formed a less dense surface layer that also carried detrital material into a density-stratified brine body. Occasionally, the total water body was undersaturated with gypsum and so dissolved gypsum apices. Some inflow episodes resulted in the deposition of marine limestones in some places (e.g. at Kudryntsi) and also led to the formation of the clastic gypsum within the middle part of the sabre gypsum unit. When the entire brine became supersaturated, gypsum precipitation resumed.

The determination of freeze/thaw temperatures of individual primary fluid inclusions in bottom-growth crystalline gypsum from many sections (900 analyses in total) of the Carpathian Foredeep (Kulchetska, 1987, 1988) has yielded information about the total concentration of salt in the Badenian basin brines. The data show that the total concentration of solutions from which bottom-growth gypsum precipitated ranged from 16 to 65 g L⁻¹ (average 45 g L⁻¹). The study of water and alcohol leachates from separate crystals of bottom-growth gypsum containing similar inclusions, as well as results of analyses of brine solutions in individual inclusions, have yielded similar conclusions (Petrichenko *et al.*, 1997). Geochemical studies (Kulchetska, 1987, 1988;

Petrichenko *et al.*, 1997) have shown that the total salinity of the Badenian water during gypsum precipitation was several times lower than is characteristic for modern, calcium sulphate-saturated sea water. For example, a study of water leachates taken from the bottom-growth gypsum at Verentchanka yielded a total dissolved solids content for the water of 53–54 g L⁻¹, consisting of (in g L⁻¹): 36.4 NaCl, 8.1 K₂SO₄, 5.9 MgSO₄ and 3.1 CaSO₄. Lower NaCl/(K+Mg) ratios in brine inclusions when compared with Miocene oceanic sea water may be explained by active halite precipitation at the first stage of evaporite basin development when halite deposits accumulated (Petrichenko *et al.*, 1997). Data on the chemical compositions of brine inclusions indicate that the Badenian water during gypsum precipitation shows some similarity to continental and continental-marine basins such as the Aral Sea (Petrichenko *et al.*, 1997, Table 3). The water of the Aral Sea, which is genetically linked to the marine basins of Eastern Paratethys, is enriched in calcium sulphate (up to 1.6 g L⁻¹), and its density at the beginning of gypsum precipitation is 1.015 (Lepeshkov & Bodaleva, 1952), which is lower than the density of normal sea water (1.0257).

The Badenian water salinity during gypsum precipitation was 3–10 times lower than is characteristic for modern, calcium sulphate-saturated sea water, and salinity varied strongly throughout the entire basin (Petrichenko *et al.*, 1997; Table 1), suggesting variable amounts of freshwater input balancing evaporation. The waters were enriched in calcium sulphate (up to 6 g L⁻¹; Petrichenko *et al.*, 1997), indicating significant groundwater influences. The chemical composition of Badenian waters was thus a mixture of relic sea water (depleted in NaCl), groundwater (enriched in calcium sulphate as a result of the dissolution of older Miocene evaporites, including Badenian ones; cf. Cendón *et al.*, 1999) and surface run-off (low salinity).

IMPLICATIONS

Attia *et al.* (1995) concluded that data from primary fluid inclusions in Middle Miocene gypsum on the eastern coast of the Gulf of Suez, west-central Sinai, indicate that gypsum precipitated from 'recycled' sea water or mixed sea water–non-marine waters, and that there were large fluctuations in water salinity during crystal growth. During periods of low sea level or active

faulting, the basin may have been completely non-marine and, during high sea-level stands, it may have been flooded by sea water to become a marine-fed lagoon (Attia *et al.*, 1995). They considered that this hydrology was related to a rift setting but, undoubtedly, a similar scenario can occur in different geotectonic situations.

The Badenian in the Carpathian Foredeep can be compared with the Messinian of the Mediterranean (e.g. Krijgsman *et al.*, 1999; Playà *et al.*, 2000), as both regions were at times isolated from an adjacent ocean. There are, however, some differences. The Messinian evaporites are considered to be third-order, sea-level lowstand deposits, as thick mappable units are always restricted to the central parts of the basins (Michalzik, 1996). The controlling mechanism for the formation of these deposits was extrinsic (eustatic), and cyclic facies successions of higher order were primarily controlled by intrinsic basin dynamics, although marine incursions (evidenced by faunal content) were also related to extrabasinal control (Michalzik, 1996). Consequently, changes in water depth and corresponding facies successions in the Messinian evaporites may be related to an interplay of eustasy, local tectonics and the rate of sediment supply and evaporation (Michalzik, 1996, p. 218).

The Badenian evaporites are characterized by the occurrence of thick mappable units forming a narrow strip in the axial part of the basin (halite deposits \approx 120 m thick; Oszczyk, 1996) and are developed as a sulphate wedge at the basin peripheries. Halite deposits show the same facies successions, and marker beds can be traced across and between individual basins, as in the Upper Silesia and Wieliczka–Bochnia basins (Garlicki, 1994). This lateral continuity of facies and events is best explained by an extrabasinal control such as eustatic changes. Based on the observed facies changes, it seems that the Badenian evaporite basin was located within a depression in which brine top level was located below contemporaneous sea level. Accordingly, during sea-level rises, a new slush of marine water could enter this depression and bring with it a marine fauna.

Warren (1991) suggested that wide bodies of brines precipitate subaqueous gypsum under near-equilibrium conditions, with brine depths restricted to only a few tens of centimetres, although Kendall & Harwood (1996, p. 310) commented that it is difficult to see how uniform salinities could be maintained over large areas. However, broad, flat areas of negligible brine depth could be affected by rapid transgressions,

as in the MacLeod Basin (Logan, 1987), a modern partial analogue for the marginal part of the Badenian evaporite basin. Accordingly, the Warren (1991) model seems to be valid for the peripheral part of the Badenian evaporite basin. However, at the same time as the marginal Badenian gypsum was being deposited in extremely shallow-water to subaerial environments, deep-water sulphate deposits were accumulating in the central part of the Badenian evaporite basin (Peryt, 2000). It is likely therefore that the Warren (1991) model is also valid for peripheral parts of many other deep evaporite basins.

CONCLUSIONS

The Badenian gypsum section in the marginal part of the Carpathian Foredeep Basin of west Ukraine can be divided into two parts. During the deposition of the lower part, relief was minor, and water depths in the various marginal parts of the basin probably differed by no more than 10 m. The gypsum of the upper part of the infill was deposited in a basin of increased relief, of the order of a few tens of metres. Although extremely shallow-water to subaerial environments prevailed in the peripheral parts of the basin, deep-water sulphate deposits accumulated in the basin centre. In peripheral areas, three facies zones can be distinguished. In zone III, the gypsum facies indicate a mostly subaqueous environment. Zone I, where only stromatolitic gypsum facies occur, was mostly a subaerially exposed evaporative flat. Zone II is transitional in character between zones III and I.

Correlation between the facies zones based on characteristic marker beds strongly suggests that the deposition of giant gypsum intergrowth facies and stromatolitic gypsum facies was coeval. Limestone intercalations in the form of lenticular bodies (usually centimetres thick) within the upper part of the gypsum sections have been recorded in some sections in zones I and II. The microfacies (mainly the presence of marine fauna) of these limestones indicate a marine provenance.

Basin-marginal Badenian evaporites formed in a standing body of water, as well as in desiccated environments subject to floods. The lateral persistence of thin beds over large areas with only minor changes in thickness and facies indicates that they formed on broad, very low relief areas that were affected by rapid transgressions that led to major changes in brine chemistry. The

Badenian basin of the Carpathian Foredeep is comparable to the Messinian basins of the Mediterranean, being cut off from the adjacent ocean at times. Marine incursions were related to extra-basinal factors and, based on rapid facies changes, it seems that the Badenian evaporite basin was located in a depression in which the brine top level was located below contemporaneous sea level. Accordingly, during sea-level rises, new marine water could enter this depression and bring with it a temporary pulse of marine fauna.

Stable isotope ($\delta^{34}\text{S}$, $\delta^{18}\text{O}$) compositions of the Badenian fall within the published ranges for Middle Miocene marine sulphate. The average values for the three gypsum sections studied ranged from +21.9 to +22.1‰ $\delta^{34}\text{S}$ and +12.9 to +13.6‰ $\delta^{18}\text{O}$. The sulphate was of sea-water origin (or was derived from the dissolution of Miocene marine evaporites), but previous fluid inclusion studies indicate that the chemical composition of Badenian waters was a mixture of relic sea water (depleted in NaCl), groundwater (enriched in calcium sulphate) and surface run-off.

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