Silurian dolostones of eastern Lithuania

Donatas Kaminskas, Giedrius Bičkauskas and Antanas Brazauskas

Department of Geology and Mineralogy, Faculty of Natural Sciences, Vilnius University, M. K. Čiurlionio 21/27, LT-03101 Vilnius, Lithuania; donatas.kaminskas@gf.vu.lt, giedrius.bickauskas@gf.vu.lt, antanas.brazauskas@gf.vu.lt

Received 14 October 2009, accepted 16 March 2010

Abstract. Silurian dolostones representing the Jaani (Verknė and Jočionys formations (Fms)) and Minija (Pabradė Formation (Fm.)) regional stages were studied in the Jočionys 299 borehole located in eastern Lithuania. In addition to petrological studies, dolostones were subjected to XRD, XRF and ICP-MS analyses. X-ray diffraction analysis revealed that dolomite crystals in dolostones were very close to stoichiometric and well ordered and could be interpreted as early diagenetic. Strontium in dolostone of the Pabradė Fm. is comparable to that of other ancient dolomites, and is much lower than Sr concentrations in typical modern marine dolomites. Slight enrichment in Sr and S in the Verknė and Jočionys Fms is due to the presence of celestine ($SrSO_4$) and gypsum. Evaporative (sabkha), seepage-reflux, mixing-zone, burial and seawater dolomitization models of modern and ancient examples from literature were considered. For the Jočionys Fm. we suggest seepage-reflux and burial(?) models. Evaporative (sabkha) and mixing-zone dolomitization models may be applied to the Verknė Fm. and the Pabradė Fm., respectively.

Key words: Baltic Silurian basin, dolostones, geochemistry, XRD, stable isotopes.

INTRODUCTION

Dolomitization of Silurian rocks of the southern part of the Baltic Silurian Basin is poorly understood. Lapinskas (2000) and Paškevičius (1997) report epigenetic dolostones in the lower and upper Silurian, which originated from various types of limestone and dolomitic marl. Much research has been done on dolomitization and a number of models have been proposed to explain the processes involved (e.g. Tucker & Wright 1999; Machel 2004). Different dolomitization models rely on three basic factors: the source of Mg (generally seawater), water/fluid movement through the sediment package and concentration of kinetic inhibitions to dolomite precipitation. Thermodynamically, dolomite should be a stable and widespread precipitate from seawater. However, several kinetic factors (hydration of Mg^{2+} ions in seawater, high ionic strength of seawater, relative efficiency of aragonite and high-Mg calcite precipitation, inhibition effects of SO_4^{2-} ions) mitigate its formation (Scholle & Ulmer-Scholle 2003). The role of sulphate as an inhibitor according to Machel (2004) is rather overrated.

In this paper we study the dolomitization of the Silurian carbonate sequence in eastern Lithuania by means of XRD, geochemical major and trace element analysis and stable isotope ($\delta^{18}O$ and $\delta^{13}C$) composition of dolomitic rocks in order to reveal the mechanisms of dolomitization, including distinction between meteoric, marine or evaporitic environment and possible dolostone precursor sediment. We hope that the results obtained by us will trigger a discussion on dolomitization of Silurian dolostones. It is worth to notice that west of the study area there are oil emplacements, which possibly could be related to dolomitization of Silurian rocks. Stentoft et al. (2001) reported that dolomite of the Kudirka Atoll in Lithuania was probably precipitated at the time when reef sediments were buried to an adequate minimum depth for the start of chemical compaction of calcite.

GEOLOGICAL SETTING

The East Baltic Silurian basin, our study area in general, was a gulf-like pericratonic sea on the western margins of the Baltica landmass. The facies history and palaeogeography of the basin was most recently summarized by Paškevičius (1997) and Lapinskas (2000).

The Jočionys 299 borehole is located in the eastern part of Lithuania (Fig. 1). The stratigraphy and generalized lithology is presented in Fig. 2. The Pabradė Formation (Fm.) is attributed to the Minija Regional Stage, the Verknė and Jočionys Fms are assigned to the Jaani Regional Stage. The Verknė Fm. lies conformably on the Jočionys Fm. The boundaries of the formations are taken from Lapinskas (2000). The Pabradė Fm. and the lower part of the Jočionys Fm. consist of microcrystalline dolostone intercalated with mudstone. The Verknė Fm. is represented by microcrystalline dolostone with intercalated mudstone and gypsum. To illustrate sedimentary

Fig. 1. Location of the Jočionys 299 borehole. Coordinates (latitude, longitude): 54°40′45″; 25°09′28″.

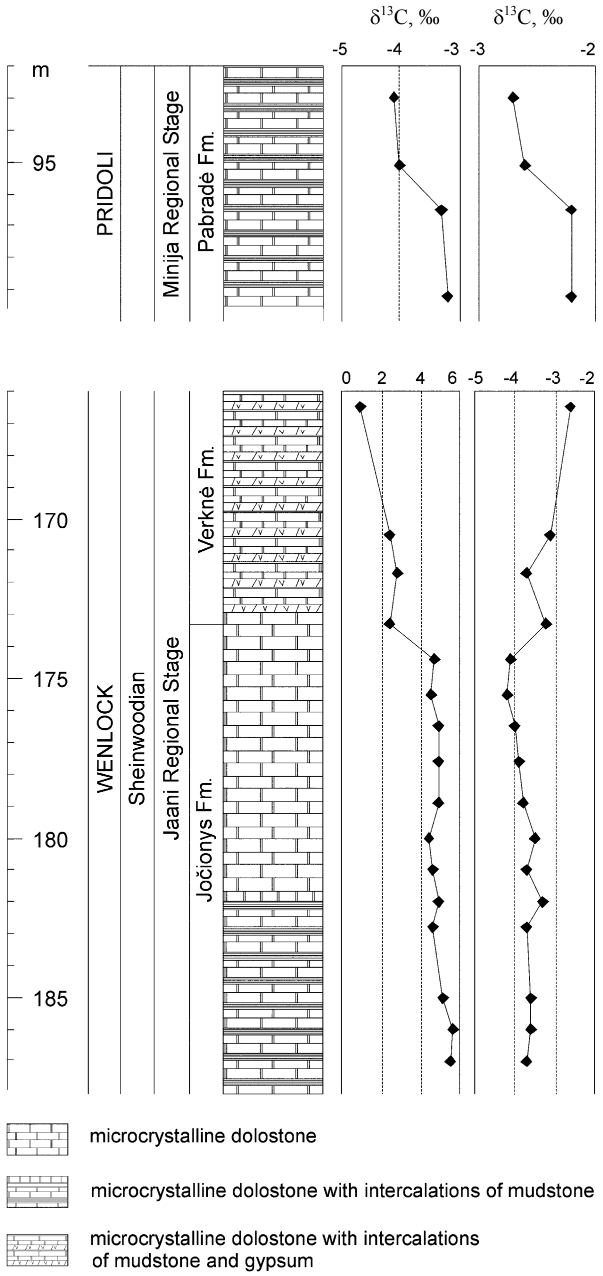
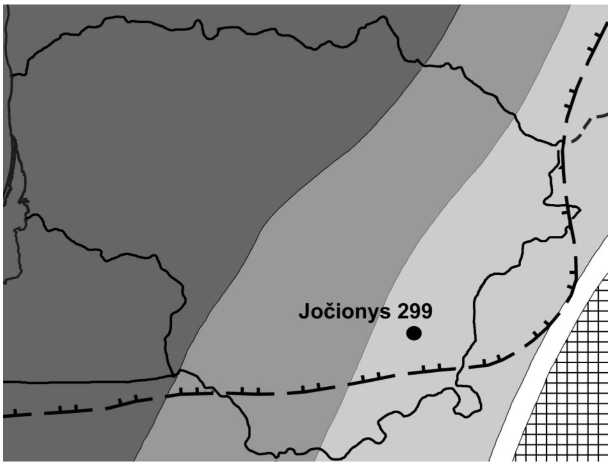


Fig. 2. Stratigraphy, general lithology, carbon and oxygen isotopes of the Jočionys 299 core.

environments of Jaani and Minija times, we compiled three palaeomaps (Figs 3–5). The deposition of Jaani time sediments was coincident with the generally regressive sea-level trend (Paškevičius 1997; Lapinskas 2000). At the beginning of Jaani time carbonaceous sediments accumulated in shallow shelf environment during maximum widening of the basin (Fig. 3) (Lapinskas 2000; Lazauskienė 2003). Later deposition was in the lagoonal and tidal-range environments (Fig. 4) (Lapinskas 2000). According to literature, numerous laminated gypsum intercalations suggest evaporitic conditions (Flügel 2004; Warren 2006). As stated by Lapinskas (2000), in Minija time sedimentation in the borehole location area took place in flooded plains and sabkhas as well as in lagoons (Fig. 5).

ANALYTICAL METHODS

Altogether 20 samples were collected from the Jočionys 299 drill core. Petrographic description of dolostones was given by examining thin sections under a polarized-light microscope. X-ray diffraction analysis was used in order to get information on the crystallographic ordering of dolomite crystals by using the height ratio of the d_{015} and d_{110} diffraction peaks (Tucker 1995). The analysis was performed using an X-ray diffractometer D8 (Bruker AXS) at the Institute of Chemistry (Vilnius, Lithuania). The dolomite stoichiometry was determined by measuring the exact position of the d_{104} peak and the equation of Lumsden (1979). The whole-rock major and trace elements were determined by XRF at the Institute of Geology at Oslo University (Norway) and using ICP-MS at Acme Analytical Laboratories Ltd. (Canada). Using ICP-MS, a 0.25 g split was heated in HNO_3–$HClO_4$–HF to fuming and taken to dryness. The residue was dissolved in HCl and analysed. Carbon and oxygen isotope analyses of dolomite were performed by Tõnu Martma at the Laboratory of Mass Spectrometry, Institute of Geology at Tallinn University of Technology (Estonia).



Key to Figs 3–5

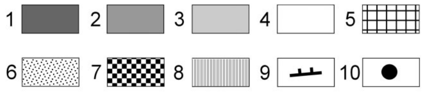


Fig. 3. Palaeogeographic map of Jočionys time (Jaani age). 1, Deep-shelf depression; 2, deeper, outer shelf; 3, shallow shelf; 4, intermediate zone between seashore and land; 5, land; 6, high salinity lagoons and tidal flats; 7, bioherms and biostromes; 8, zone of low hydrodynamic regime; 9, recent boundaries of distribution; 10, Jočionys 299 borehole.

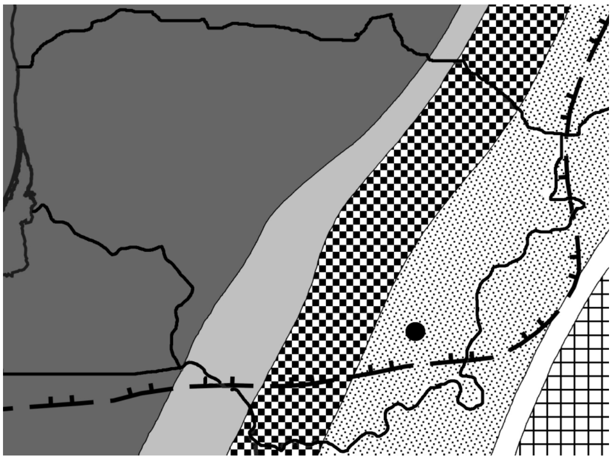


Fig. 4. Palaeogeographic map of Verknė time (Jaani age). For key refer to Fig. 3.

The samples were analysed by a Thermo Scientific mass spectrometer Delta V Advantage with the GasBench II preparation line. An amount of 600 µg of carbonate was reacted with 100% H_3PO_4 at a temperature of 70 °C. The results are given in the usual δ-notation, as per mil deviation from the PDB standard. The reproducibility of replicate analyses was generally better than ±0.1‰. The method is explained in more detail in Kaljo et al. (1997) and Martma et al. (2005).

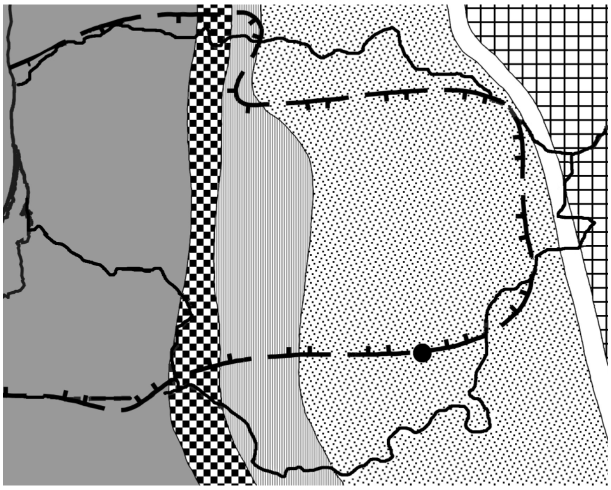


Fig. 5. Palaeogeographic map of Pabradė time (Minija age). For key refer to Fig. 3.

RESULTS

Dolostone types

Three types of 20 sampled dolostones were differentiated on the basis of distinctive stratigraphic and petrological characteristics (Figs 6–8). In addition, they also show geochemical differences, which are compared below.

Jočionys Fm.

Interval 173.0–181.6 m. Dolostone is microcrystalline with small vugs from 1 to 3 mm and from 1 to 1.5 cm in diameter. It contains rare broken fragments of crinoids and brachiopods.

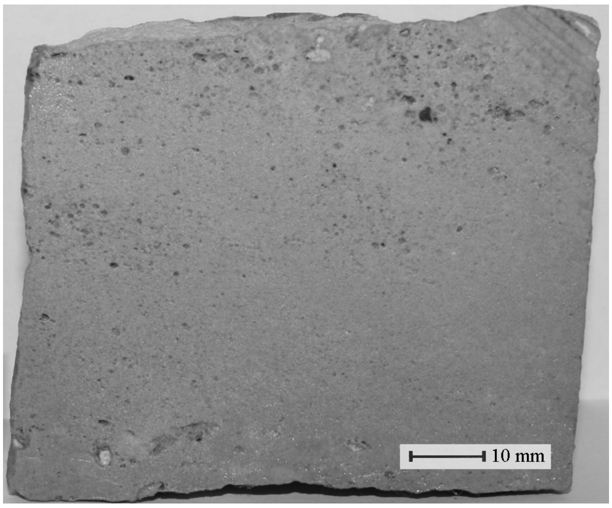


Fig. 6. Jočionys Fm. dolostone (depth 173.8 m).

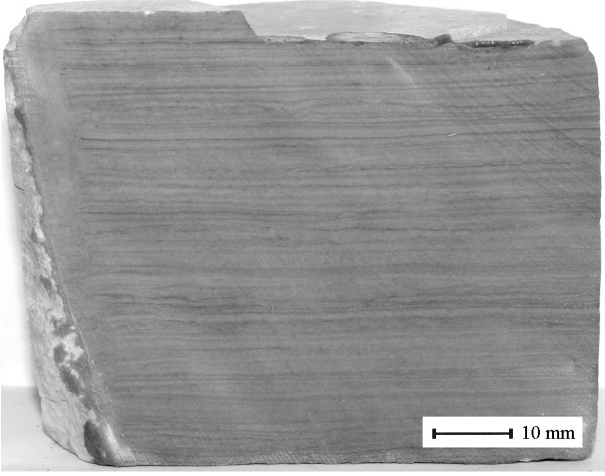


Fig. 7. Verknė Fm. dolostone with mudstone (depth 171.2 m).

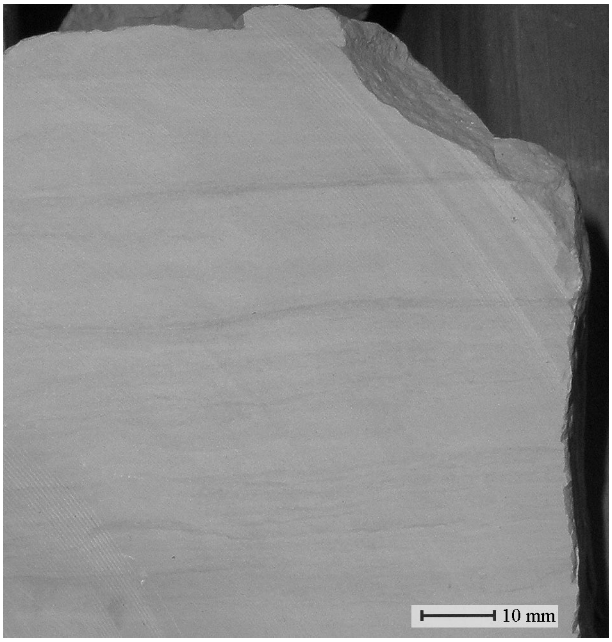


Fig. 8. Pabradė Fm. dolostone (depth 93.0 m).

Interval 181.6–187.0 m. Dolostone is microcrystalline with intercalations of laminated carbonate mudstone. Small vugs (1–2 mm), formed as a result of dissolution of carbonate fauna, are found.

Verknė Fm.

Interval 158.3–173.0 m. Dolostone is microcrystalline with intercalations of thin laminae of siliclastic mudstone and gypsum. Intercalations of mudstone contain elongated (0.05–0.4 mm) celestine ($SrSO_4$) crystals.

Pabradė Fm.

Interval 90.3–101.6 m. Dolostone is microcrystalline with thin carbonate mudstone intercalations. Small vugs are filled with sparry calcite and dolomite. Broken fragments of brachiopods, gastropods and ostracods occur as grains in mudstone.

The stoichiometry and ordering of dolomite crystals

The mineral dolomite, $CaMg\,(CO_3)_2$, is commonly not stoichiometric (ideal), but has an excess of Ca, with Ca to Mg ratio reaching 58 : 42, or less commonly, an excess of Mg with the Ca : Mg ratio up to 48 : 52 (Tucker 1995). The Ca excess was calculated by the equation of Lumsden (1979):

$$CaCO_3 \text{ mol\%} = 333.33 * d_{104} - 911.99,$$

where d_{104} is the position of the peak in angstrom units.

The ordering of dolomite crystals was calculated from XRD data. The height ratio of the ordering peaks d_{015} and d_{110} gave a measure of the degree of the ordering of the dolomite crystal. The greater the ratio, the higher the degree of order (Tucker 1995). No linear relationship between the degree of order and mol% $CaCO_3$ was found (Fig. 9) The dolomite mineral in all studied

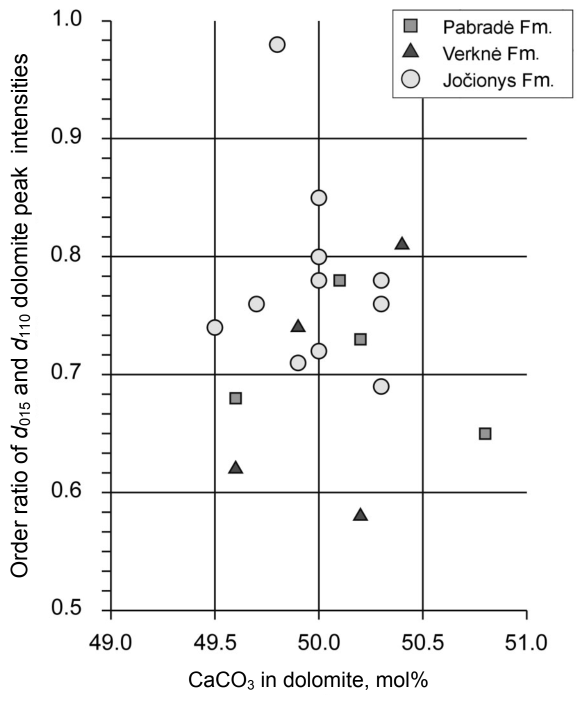


Fig. 9. XRD data from the studied dolostones showing stoichiometry (as mol% $CaCO_3$) plotted against ordering.

dolostones is very close to stoichiometric. The median value of the ordering of dolomite is 0.77 for the Jočionys Fm., and 0.68 and 0.71, respectively, for the Verknė and Pabradė Fms.

Major and trace elements

The whole-rock median values of major and trace elements as well as loss on ignition are presented in Table 1. Dolostones are depleted in most of the major and trace elements when compared to the Silurian limestones of Lithuania (Kaminskas 2002). The quantities of major elements in dolostones of the Pabradė and Jočionys Fms do not differ much, while dolostones of the Verknė Fm. have elevated values of all elements but magnesium and calcium oxides. The quantities of trace elements of the formations mentioned above show slightly different patterns. The median values of V, Cr, Zn, Rb, Ba, Sr, Y, Zr and S are the highest in the Verknė Fm. dolostones.

Table 1. The median values of major and trace elements of the studied dolostones

Element*	Pabradė Fm. (*n* = 4)	Verknė Fm. (*n* = 4)	Jočionys Fm. (*n* = 12)
SiO_2	4.84	12.44	3.64
Al_2O_3	1.29	2.90	0.93
$Fe_2O_3^T$	0.79	1.26	0.73
MnO_2	0.04	0.05	0.05
MgO	19.56	17.62	20.27
CaO	28.16	24.87	28.79
Na_2O	0.04	0.13	0.02
K_2O	0.46	0.78	0.14
TiO_2	0.07	0.17	0.05
P_2O_5	0.03	0.03	0.01
LOI	44.60	39.78	45.37
V	27	26	13
Cr	20	34	19
Co	2	1	2
Ni	20	14	11
Cu	4	3	3
Zn	10	20	13
Rb	14	34	24
Pb	2	2	13
Ba	49	113	36
Sr	88	136	121
Y	4	8	1
Zr	13	41	9
Nb	1	3	6
Th	1	2	2
U	1	2	1
S	250	1921	1152

* Major elements and loss on ignition (LOI) are in % (bold font), the rest are in ppm. 'T' stands for total iron.

Isotopic composition

The $\delta^{18}O_{PDB}$ of values of the studied Silurian dolostones range from –6‰ to –2.2‰, of $\delta^{13}C_{PDB}$ from –4.1‰ to +5.7‰ (Fig. 10). The $\delta^{13}C_{PDB}$ values of the Pabradė Fm. (–4.1‰ to –3.2‰) are distinctly more negative than those of the Verknė Fm. (+1.0‰ to +2.9‰) and the Jočionys Fm. (+4.1‰ to +5.7‰). However, the $\delta^{18}O_{PDB}$ values of all formations overlap. The values of the Jočionys Fm. are the most negative (–4.2‰ to –3.3‰). The Verknė and Pabradė Fms have $\delta^{18}O_{PDB}$ values ranging from –3.7‰ to –2.6‰ and –2.7‰ to –2.2‰, respectively.

The variation of $\delta^{18}O_{PDB}$ and $\delta^{13}C_{PDB}$ in the Jočionys 299 section is presented in Fig. 2. A decrease from 5.7‰ to 1.0‰ of $\delta^{13}C_{PDB}$ values is observed from the Jočionys Fm. to the Pabradė Fm., respectively. In the Jočionys Fm. $\delta^{18}O_{PDB}$ values are relatively constant, but slightly increasing up to –2.6‰ in the Verknė Fm. The upper part of the Pabradė Fm. has more negative values than the lower part.

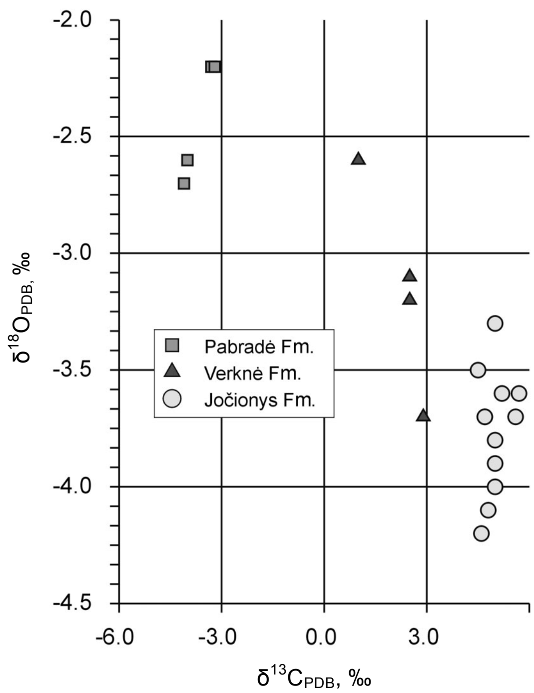


Fig. 10. $\delta^{13}C_{PDB}$ and $\delta^{18}O_{PDB}$ data plot of the studied dolostones.

DISCUSSION

Some of the chemical and hydrological conditions of dolomite formation are poorly understood, and petrographic and geochemical data commonly allow of more than one genetic interpretation (Machel 2004). An effect of poor ordering and non-stoichiometry is that dolomite crystals are metastable and more soluble than 'ideal' dolomite. Stabilization of calcian dolomite to a more 'ideal' type will mostly take place by dissolution reprecipitation (Tucker & Wright 1999). According to Lumsden & Chimahusky (1980), finely crystalline dolomites having 50–51 mol% $CaCO_3$ are usually early diagenetic, near-surface in origin and could be associated with evaporates, while finely crystalline dolomites with 54–56 mol% $CaCO_3$ are not associated with evaporates. Our results show that dolomite from the studied dolostones has the degree of ordering >5 and $CaCO_3$ ranging from 49.6 to 50.8 mol%, and could thus be interpreted as early diagenetic. It is typical of ancient platformal dolomites, which are commonly interpreted to indicate recrystallization of early-formed, poorly-ordered and non-stoichiometric dolomite (Warren 1999).

Median Sr value in dolostone of the Pabradė Fm. (88 ppm) is comparable to that of other ancient dolomites, and is much lower than Sr concentrations in typical modern marine dolomites (~500–800 ppm) (Land 1980). Slight enrichment in Sr and S in the Verknė and Jočionys Fms is due to the presence of celestine ($SrSO_4$) and gypsum. The latter minerals were detected in thin sections and by XRD analysis (Kaminskas 2002). The low median value of sulphur in the Pabradė Fm. (250 ppm) compared to the Verknė and Jočionys Fms (1921 and 1152 ppm, respectively) could be related to dissolution of evaporitic minerals in contact with fresh meteoric waters. V, Cr, Rb, Ba, Y and Zr show strong correlation ($r > 0.7$) with Al, Si and Ti and are mainly related to clayey and clastic fraction (Kaminskas 2002).

We considered evaporative (sabkha), seepage-reflux, mixing-zone, burial and seawater dolomitization models of modern and ancient examples from literature (Tucker & Wright 1999). For the Jočionys Fm. we suggest seepage-reflux and burial(?) dolomitization models. Sparse remains of fauna (brachiopods and crinoids) in thin sections suggest that dolostones of this formation originated from limestone. It is possible that initially limestone was partially dolomitized when mixed sea and meteoric waters filtered downwards. Later dolomitization took place in shallow burial conditions. The replacement of limestone resulted in a series of distinctive structures and textures, i.e. the formation of vugs, cavities, pores which sometimes were emplaced by celestine and gypsum. The latter minerals indicate that the dolomitizing fluids from which the dolostones originated were enriched in sulphates.

Numerous thin gypsum intercalations in the Verknė Fm. refer to lagoonal-tidal settings under arid climate conditions. According to Lapinskas (2000), the basin during the formation of Verknė sediments was semiclosed, of tidal-lagoon type. Several anomalous values of celestine ranging from 2.9% to 23.3% have been detected in these sediments (D. Kaminskas unpublished data). The above-mentioned characteristics indicate that dolostones may result from penecontemporaneous dolomitization of lime mud on supratidal flat (sabkha) environments by high saline seawater. The $\delta^{13}C_{PDB}$ values from +1.0‰ to +2.9‰ indicate that dolomitizing fluid was of marine origin, suggesting thus an evaporative (sabkha) model of dolomitization. On the other hand, the $\delta^{18}O_{PDB}$ values from –2.7‰ to –2.2‰ and $\delta^{13}C_{PDB}$ values from –4.1‰ to –3.2‰ of dolostones of the Pabradė Fm. show that dolomitization could have occurred in the mixing zone when dolomite precipitated from a diluted solution. Dolomite precipitation is favoured from diluted solutions, e.g. if seawater is mixed with fresh water (Folk & Land 1975). Negative values of $\delta^{13}C_{PDB}$ may also be associated with sulphate-reducing bacteria due to incorporation of CO_3^{2-} from organic matter diagenesis (Bustillo et al. 1999; Veizer et al. 1999).

According to Azmy et al. (1998), the $\delta^{18}O_{PDB}$ and $\delta^{13}C_{PDB}$ values for well-preserved Silurian brachiopod samples range from –2‰ to –6.6‰ and from –1‰ to 7.5‰, respectively. The data of the Jočionys and Verknė Fms are within these intervals, supporting the Silurian seawater source for dolomitization. Moreover, the composition of $\delta^{13}C_{PDB}$ in Silurian dolostones of the same age from Podolia (Ukraine) are, according to Kaljo et al. (2007), in the same range (–0.7‰ to 5.9‰) as that of the Verknė and Jočionys Fms. More negative $\delta^{18}O_{PDB}$ values (from –4.4‰ to –7.1‰) of Podolia dolostones, however, may indicate a somewhat higher temperature or a different isotopic composition of dolomitizing fluids. Another possibility is that the $\delta^{18}O$ variation reflects the recrystallization of early dolomite or precipitation of further generations of dolomite during burial (Tucker & Wright 1999).

CONCLUSIONS

1. Dolomite crystals of all three types of dolostones are well ordered and have 49.6 to 50.8 mol% $CaCO_3$.
2. The major element composition of the studied dolostones is very similar, while dolostones of the Verknė Fm. are slightly enriched in Cr, Zn, Rb, Ba, Sr, Zr and S.
3. All three types of dolostones have $\delta^{18}O_{PDB}$ values in the range of –6‰ to –2.2‰ and $\delta^{13}C_{PDB}$ values in the range of –4.1‰ to +5.7‰.

4. For the Jočionys Fm. we suggest seepage-reflux and burial(?) models. Evaporative (sabkha) and mixing-zone dolomitization models may be applied to the Verknė Fm. and the Pabradė Fm., respectively.

Acknowledgements. The study was partly financed by the Lithuanian State and Studies Foundation (grant No. T-26/08). We thank two anonymous reviewers and Kalle Kirsimäe for their constructive comments that considerably improved the manuscript.

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Ida-Leedu Siluri dolokivid

Donatas Kaminskas, Giedrius Bičkauskas ja Antanas Brazauskas

Siluri dolokive uuriti Leedu idaosas asuvas Jočionys 299 puurläbilõikes, milles dolomiitsed kihistud kuuluvad Jaani (Verknė ja Jočionyse kihistu) ning Minija (Pabradė kihistu) lademesse. Lisaks petroloogilisele uurimisele viidi läbi ka röntgenstruktuuri-, röntgenfluorestsents- ja induktiivselt seotud plasma massispektromeetria analüüs. Röntgenstruktuurianalüüsi tulemused näitavad, et dolomiit on koostiselt väga lähedane stöhhiomeetrilisele koostisele, hästi korrastatud ja ilmselt varadiageneetiline. Strontsiumisisaldus Pabradė dolokivides vastab tüüpilistele paleosoilistele-mesosoilistele dolomiitkivimitele ja on palju madalama sisaldusega kui tüüpilistes tänapäevastes merelistes dolomiitides. Verknė ja Jočionyse kihistus täheldati dolokivide vähest strontsiumi- ning väävlisisaldust, mida põhjustab ilmselt tsölestiini ja kipsi esinemine nendes kivimites. Võrdlusest dolomiitide tekkemudelitega võib järeldada, et Jočionyse dolomiidid tekkisid magneesiumirikaste lahuste diageneetilise infiltratsiooni ja/või mattumise(?) mudeli järgi, kuid Verknė ning Pabradė kihistus vastavalt evaporiidistumise ja segunemisvööndi mudeli kohaselt.