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PROSPECTS OF LITHUANIAN SILURIAN SHALE GAS, BALTIC SEDIMENTARY BASIN

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Abstract. *The Baltic sedimentary basin contains numerous hydrocarbon fields in Lithuania. The Lower Silurian shales are considered as potential for shale gas exploration. The thickness of shale units within the oil window ranges from 115 to 180 m in west Lithuania. The average total organic carbon (TOC) content of the Lower Silurian shales is about 1.0–1.2%, with an elevated content averaging 6.7% in the Aeronian black shales. The organic matter (OM) is Type II kerogen. Pyrolysis analysis shows moderate thermal maturity parameters;* T_{max} *is from 428 °C in central Lithuania to 455 °C in the west of the country, implying the oil generation stage. It is concluded that the burial conditions were favorable for oil generation, while the gas potential of Silurian shales is rather limited.*

Keywords: *gas shales, Baltic sedimentary basin, Silurian, pyrolysis, TOC.*

1. Introduction

The Baltic sedimentary basin comprises a number of oil fields in Lithuania, Kaliningrad District of Russia, Gotland of Sweden, and Poland. The source rocks are composed of Cambrian, Ordovician and Silurian organic rich shales [1–4]. The Silurian shales represent the thickest part of the sedimentary cover of the basin. The shape of the basin and the sedimentation pattern were controlled by the flexural loading of the western margin of the Baltica continent induced by the overriding of Eastern Avalonia and formation of North German-Polish Caledonides during the Silurian time [5]

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that is reflected in the thickening of the sediment package and deepening of the deposition environment to the west. The thickness of the Silurian succession reaches 3 km in the westernmost part of the basin. The thermal maturity of Silurian shales changes westwards with increasing burial depth from immature, through the oil window, wet gas window to dry gas window, and even overmature [6]. The TOC content in the Silurian shales in the basin ranges from 0.5 to 20% [6].

The lower part of the Silurian succession of west Lithuania reached the oil window stage in the Devonian time as implied from hydrocarbons (HC) generation modelling [7]. Furthermore, high vitrinite reflectance values were reported from a few wells, suggesting the gas window stage in the southern part of west Lithuania [8]. According to [9] the technically recoverable shale gas resources were estimated in 2011 at 4 Tcf in Lithuania. Yet, no resources of shale gas were suggested in 2013 due to unrealiable geological data [10]. The present study is focused on a detailed identification of the prospective intervals of the Silurian section and evaluation of their parameters as regards the shales gas prospects.

2. Silurian succession of Lithuania

Silurian sedimentary rocks are distributed in most of the territory of Lithuania, except the southernmost part of the country that resulted from the Late Palaeozoic erosion (Fig. 1). The shallow periphery of the basin is dominated by carbonaceous lithologies that grade to shales in the basin's deeper part (Fig. 2). The subsidence rate gradually increased from the Early Silurian to the Late Silurian that was caused by the progressing orogeny driven flexural loading of the western margin of the East European Craton [5, 11].

The Llandovery section is only up to 55 m thick. The basal part is composed of the Rhuddanian limestones of about 5 m in thickness [12] and these are not discussed in the present study. They grade to the Aeronian graptolitic black shales with rare clayey limestones. The thickness ranges from 1 to 11 m. This layer is the most distinct feature on the gamma ray logs by API values attaining 300–450. The Telychian dark grey graptolitic shales are characterised by a lower TOC content that is also reflected in the lower gamma ray values (200–250 API). The thickness of the shales is in the range of 30–40 m in west Lithuania, reaching 70–80 m in the deepest part of the basin in Poland.

The Wenlock section is composed of dark grey graptolitic shales showing gamma ray values similar to the Telychian's (200–250 API). Rare thin limestone layers are present. The thickness of the Wenlock succession is 80–130 m. For the sake of comparison it should be noted that the Wenlock succession is as thick as 700–800 m in the westernmost part of the basin in northwest Poland.

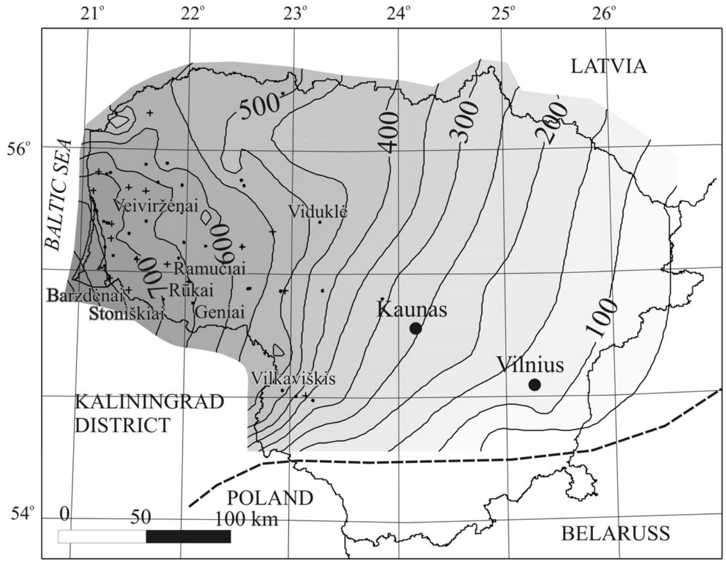

Fig. 1. Isopachs of the Silurian succession of Lithuania. The hatched line shows the southern boundary of distribution of Silurian deposits. The studied wells are shown as follows: TOC data collected from old publicly available industrial reports (dots), newly studied wells (crosses). Wells mentioned in the text are marked.

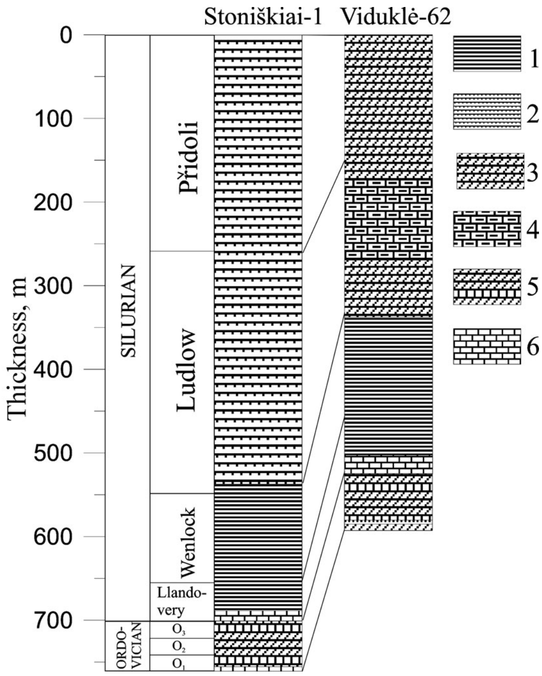

Fig. 2. Lithological sections of wells Stoniškiai-1 (west Lithuania) and Viduklė-62 (central Lithuania) (see Fig. 1 for locations). Symbols: 1 – shales, 2 – carbonaceous shales, 3 – marlstones, 4 – clayey limestones, 5 – intercalation of limestones and marlstones, 6 – limestones.

The thickness of the Upper Silurian succession reaches 720 m in west Lithuania. The succession represents a homogeneous package of carbonaceous shales of dark grey color containing rare limestone layers. This part of the Silurian is characterized by a higher content of carbonaceous material that is reflected in low gamma ray values, 150–170 API, except the lowermost part of the section showing about 190 API.

3. Data and methods

Numerous TOC measurements of Silurian shales have been carried out owing to extensive oil exploration activities in west and central Lithuania (Fig. 1). 384 TOC measurements combined with estimation of the insoluble residue content were collected from the industrial reports. A 3–5% HCl solution was used to estimate the insoluble residue content by removing the carbonate component.

The publicly available industrial data were supplemented with the new 113 organic matter (OM) content measurements of Silurian samples collected from 14 wells in west Lithuania (Fig. 1). TOC content was determined using the approach by Tjurin [13]. Furthermore, 17 shale samples (weighed to 100 mg) were subjected to pyrolysis analysis to determine the kerogen type, TOC content, and thermal maturity of organic matter. Pyrolysis was carried out using an MX7304AM mass spectrometer (Ukraine); temperature regime was controlled by a precise converter TERKON (OOO "Termex", Tomsk, Russia). Pyrolysis was based on the Rock-Eval 6 technology [14]. TOC (wt%), S1 (mg HC/g rock), S2 (mg HC/g rock), and T_{max} (°C) were measured. Hydrogen index (HI; S2/TOC × 100) and production index (PI; S1/(S1 + S2)) were calculated from the measured parameters.

The industrial sonic velocity, electric resistivity and gamma ray well log data were available for the present study to identify the vertical distribution of the clayey material and organic matter in the Silurian section.

4. Results

4.1. Distribution of organic matter

The distribution of organic matter shows some distinct lateral and vertical trends [15]. TOC increases from the east to the west which is related to the deepening of the sedimentation environment in the Baltic sedimentary basin. The maximum content of OM was reported in the Aeronian shales (Fig. 3). The overlying Telychian and Wenlock shales are less enriched in organic matter. The Upper Silurian shales show generally lower TOC values. These variations in TOC content directly correlate with the insoluble residue content (Fig. 3). The Upper Silurian shales are characterised by lower insoluble residue content values, while higher values are typical of the Lower Silurian shales, which reversely correlate with the carbonate content.

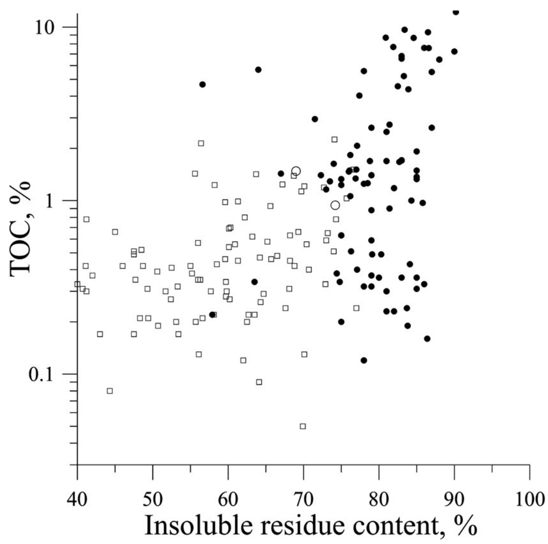

Fig. 3. Insoluble residue content vs TOC of Silurian shales, west and central Lithuania. Black dots indicate Llandovery samples, open circles symbolize Wenlock samples, squares denote Upper Silurian samples.

Figure 3 shows that there is a distinct difference between the Upper and Lower Silurian (Aeronian-Telychian) shales in TOC and insoluble residue content. The average TOC content of the Upper Silurian shales is 0.3 wt%, ranging from 0.05 to 1.5 wt%; only 6% of the samples have TOC values exceeding 1 wt% and only a few samples have TOC higher than 2 wt%. There are only a few estimates of TOC content of the Wenlock shales reported in industrial reports. TOC ranges from 0.9 to 1.1 wt%. Despite the low thickness there are abundant data provided from the Llandovery shales, as the underlying Rhuddanian limestones were considered as a potential reservoir for oil exploration and therefore drilled with core sampling. TOC ranges from 0.15 to 20 wt% in the Aeronian-Telychian shales.

The insoluble residue content marks a sharp boundary between the Upper and Lower Silurian shales. Most of the samples show values lower than 70% in the Upper Silurian, while this content is higher in the Lower Silurian, being primarily related to the different carbonate component contents in the shales.

The average values of TOC content were calculated to be as high as 6.7, 1.0, 1.2 and 0.6 wt% for, respectively, Aeronian, Telychian, Wenlock and Upper Silurian shales in west Lithuania. These values correlate with the average insoluble residue content values of 82, 79, 71 and 62 wt%, respectively. The TOC content of shales is significantly lower in central Lithuania. The average contents of TOC and insoluble residue are 0.8 and 63.0 wt% in the Lower Silurian shales and 0.5 and 61.8 wt% in the Upper Silurian shales, respectively.

Despite the large amount of TOC data, individual wells are characterised by a few to a dozen measurements that do not provide detailed information on the distribution of OM in the section. It especially applies to the Wenlock section that is represented by only a few samples from west Lithuania.

Well log data can be employed to evaluate source rock potential [16]. Sonic velocity vs resistivity crossplots are often used to identify the organic rich intervals prospective for shale gas exploration [17]. The baseline correlation of non-source Silurian shales was calculated for the well Barzdėnai-2 to illustrate the distribution of OM in the section in southwest Lithuania (depth interval 1070–2000 m) (Fig. 4). The lower part of the Silurian section shows a distinct enrichment in OM. The highest concentration of OM is found in the Aeronian and Upper Telychian shales. The OM content gradually decreases upwards in the Wenlock succession. Increased enrichment in OM is established in the succession's lower part and only low enrichment is calculated in its upper part and the lowermost Upper Silurian, whereas the rest of the Upper Silurian has no anomalous enrichment in organic matter.

Still, there are rather scarce sonic logging data available for the Silurian section in west Lithuanian wells, as this technique was mainly targeted at characterisation of the Middle Cambrian oil prospective sandy reservoir. Therefore the application of this method, as regards estimation of the distribution of organic matter in Silurian shales, is rather limited.

On the other hand, all oil exploration wells were subject to gamma ray logging. The gamma ray logs are suggested as an efficient tool in calculating distribution of organic matter. The insoluble residue (mostly clay minerals) and organic matter mainly contribute to gamma ray intensity. Gamma ray log data and TOC measurements were collected from 11 representative wells of west Lithuania that unravelled close correlation, which can be described by the following regression equation:

$$\text{TOC (wt\%)} = \exp(0.15 \times \text{GR}) \times 0.12 \text{ (correlation coefficient + 0.81)}, \quad (1)$$

where GR stands for gamma ray (API).

Figures 5a–b provide an example of correlation between TOC and gamma ray logs in two wells located in west Lithuania.

The defined correlation was applied to calculate TOC content (Figs. 5c–d). It provides a better insight into the distribution of organic matter in the section. Moreover, most of the wells were drilled with no drill core sampling in west Lithuania, while the proposed approach enables identification of the organic matter content in shales in wells with no drill coring. Calculations indicate maximum TOC content in the Aeronian shales exceeding 10 wt%, while the measured and calculated TOC values are mainly 1.5–2.0 wt% in the Telychian. The basal 15–20 m thick part of the Wenlock section is depleted of TOC that is mainly below 1%. The rest of the Wenlock section indicates TOC values between 1.5 and 2.2 wt%, showing maximum organic matter content in the lower part, decreasing upwards that is in good agreement with the sonic velocity-resistivity data discussed above.

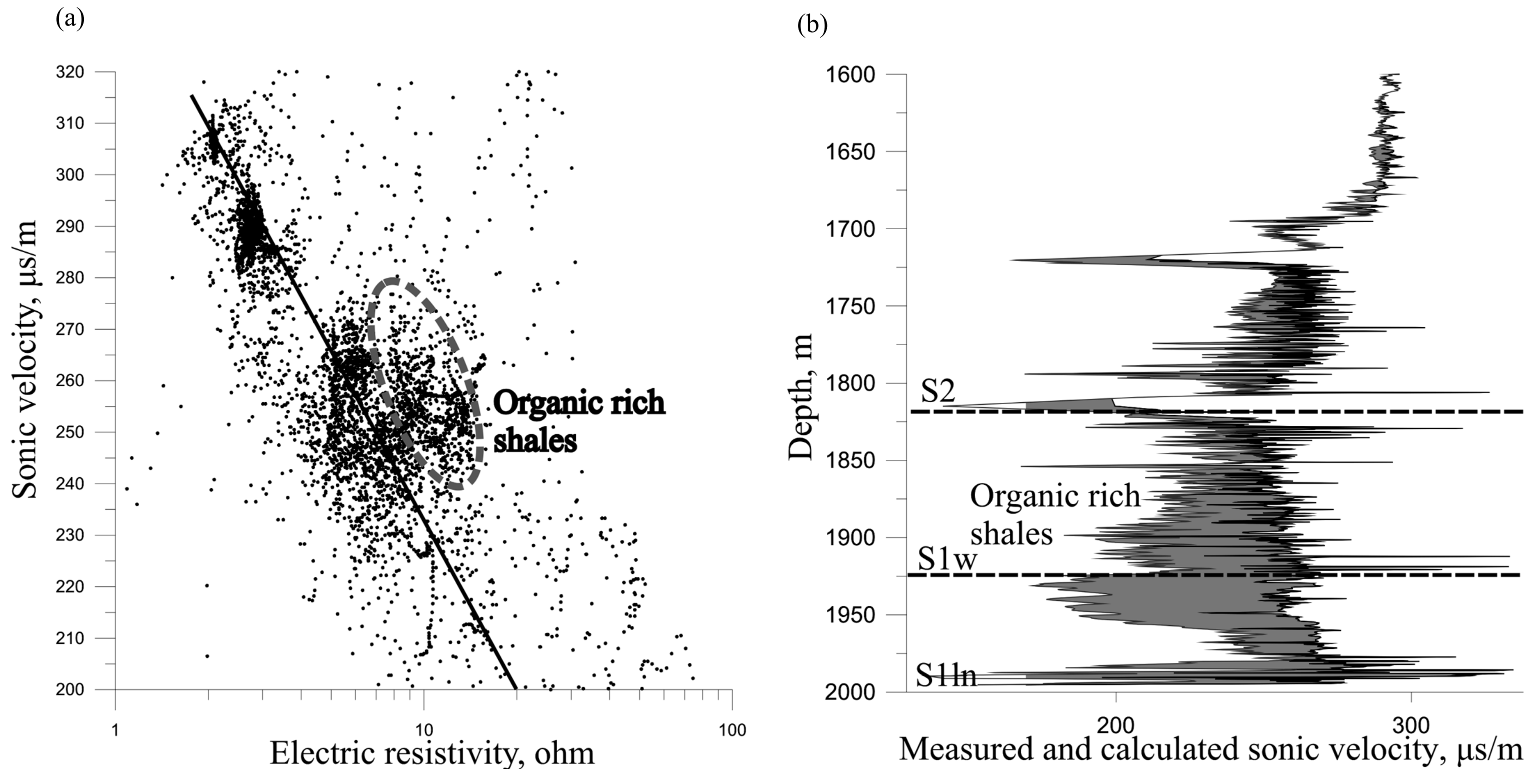

Fig. 4. Identification of organic rich shales in the Silurian section, well Barzdėnai-2, southwest Lithuania: (a) crossplot of electric resistivity and sonic velocity (the basic non-source shale line ($dT = 352 - 63.102 \times \ln R$) is indicated; the hatched line symbolizes the Lower Silurian organic rich shales); (b) calculated (a grey line) and measured (a black line) sonic velocities. Organic rich shales are marked in grey, stratigraphic boundaries by hatched lines (Llandovery, Wenlock and Upper Silurian).

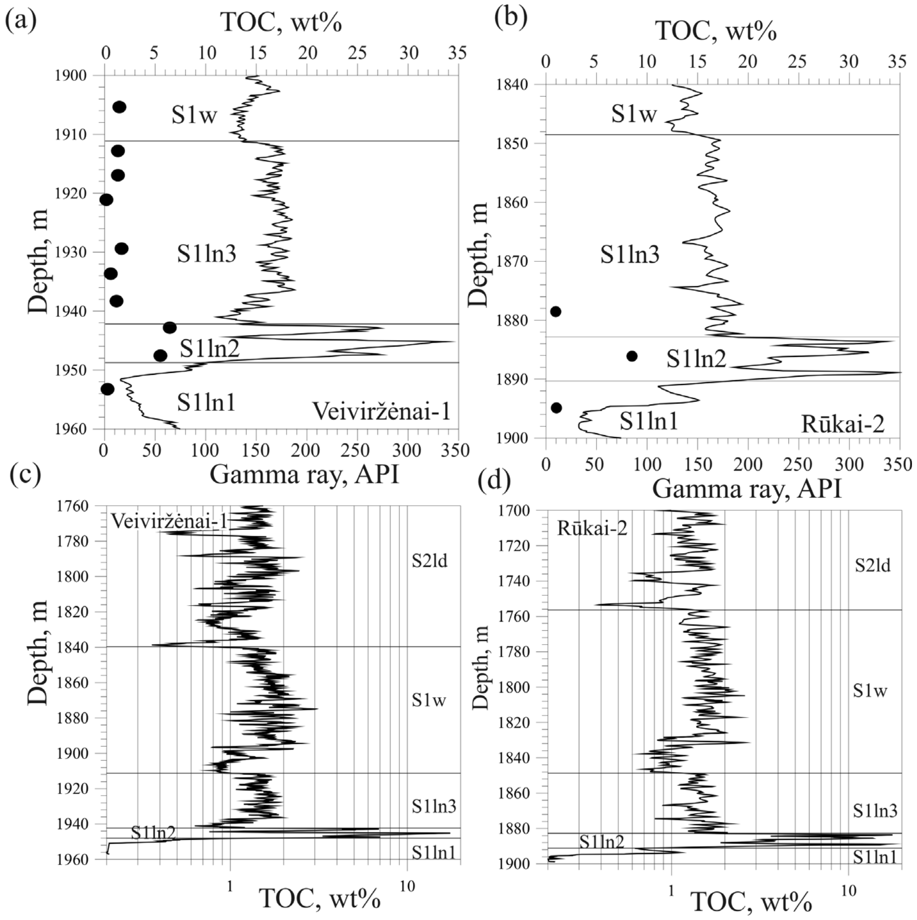

Fig. 5. (a), (b): Gamma ray logs and TOC contents (wells Vieviržėnai-1 and Rūkai-2, west Lithuania); (c), (d): TOC content in Silurian shales converted from gamma ray logs (wells Vieviržėnai-1 and Rūkai-2, west Lithuania). The stratigraphy of layers is indexed.

4.2. Characteristics of organic matter

The content and type of organic matter are the primary parameters controlling the gas potential of shales. Previous studies of OM indicate the predominance of Type II oil- and gas-prone kerogen in the Palaeozoic rocks in the Baltic region [1, 2], including Lithuanian territory [15]. The kerogen is composed of phytoplankton, zooplankton, and bacterial remnants [18]. The organic matter is mainly concentrated along the bedding surfaces (Fig. 6).

According to these studies the oil in west Lithuania contains 70–85% (it is 50–60% of that in central Lithuania) of saturated compounds with predominance of light n-alkanes and are almost devoid of steranes and terpenes [8]. Pyrolysis analysis indicates rather low peak S1 values, mainly in the range of 0.28–2.88 (mg HC/g rock) that points to a low content of trapped

HC in the Silurian shales (Table 1). Two studied samples have S1 peaks 4.58 and 9.24 mg HC/g rock, respectively. It may refer to an extensive expulsion of HC from shales during the prolonged deep burial.

S2 peak values show variations from 0.02 to 55.53 mg HC/g rock (Table 1), suggesting a different HC generation potential of Silurian shales. S2 closely correlates with TOC content (correlation coefficient +0.88). The lowest values, 2.15–5.56 mg HC/g rock, were determined in the Upper Silurian shales. The Wenlock shales show a similar scatter of S2 values, 2.67–10.57 mg HC/g rock (Table 2). The highest S2 peak, 6.38–55.53 mg HC/g rock, was found in the Aeronian shales.

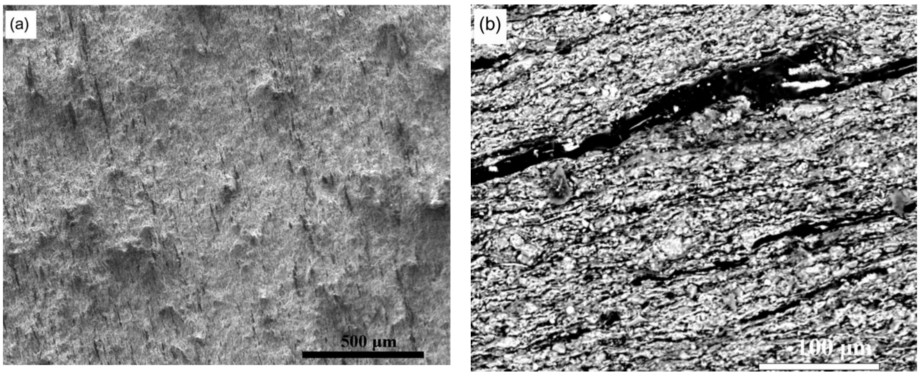

Fig. 6. SEM micrographs showing the distribution of organic matter (black lenses) in Aeronian shales; (a) well Vilkaviškis-131, central Lithuania; (b) well Geniai-2, west Lithuania.

Table 1. Results of pyrolysis analysis of Lower and Upper Silurian shales

Well	Depth, m	Age	TOC, wt%	S1, mg HC/g rock	S2, mg HC/g rock	HI, mg HC/g TOC	PI, mg/g TOC	T_{max}, °C
Geniai-1	1564	S2ld	0.98	0.28	2.15	219	0.12	439
Geniai-1	1583	S2ld	1.21	0.35	2.73	226	0.11	439
Geniai-1	1601	S2ld	1.14	0.3	2.76	242	0.1	440
Geniai-1	1754	S1ln2	6.72	2.88	19.77	294	0.13	443
Malukai-1	1612	S1ln2	9.72	2.25	55.53	571	0.04	425
Mikoliškės-1	2031	S1ln2	11.19	4.58	37.57	336	0.11	442
Paluknė-1	1660	S1ln2	7.36	1.73	35.82	486	0.05	432
Paluknė-1	1662	S1ln2	2.03	0.33	6.38	315	0.05	437
Vilkaviškis-131	789	S2pr	1.27	9.24	5.56	438	0.62	418
Vilkaviškis-131	1192	S1ln2	4.07	0.7	13.75	338	0.05	429
Vilkaviškis-131	1197	S1ln2	7.57	1.61	31.49	416	0.05	428

Table 2. Results of pyrolysis analysis of Lower Silurian shales

Well	Depth, m	Age	S2, mg HC/g rock	S2A, mg HC/g rock	S2M, mg HC/g rock	S2N, mg HC/g rock	T_{max}, °C
Barzdėnai-1	1974.8	S1w	2.67	0.29	0.66	1.72	439
Barzdėnai-1	1976.0	S1w	10.57	1.42	2.79	6.36	422
Genčiai-1	1810.0	S1w	7.72	1.35	1.87	4.50	432
Genčiai-1	1978.0	S1ln3	0.02	0.15	0.11	0.38	434
Vilkyčiai-3	1894.5	S1ln2	43.19	3.41	13.42	26.36	443

Remark: S2 peak is subdivided into the following compounds: A – aromatic, M – methane, N – naphthene.

Naphthene is the main compound contributing to S2 peak, while aromatic compounds have a minor part (Table 2). The ratio S2M/(S2A + S2N) ranges from 0.21 to 0.45, pointing to the high potential of methane production.

Silurian shales yield HI values of 219 to 571 mg HC/g TOC. The T_{max} vs HI plot suggests Type II kerogen (Fig. 7a). HI values decrease with increasing T_{max}. The production index (PI = S1/(S1 + S2)) is rather low, ranging from 0.05 to 0.13 that corresponds to the early mature stage [19]. This classification is supported by the TOC vs S2 peak plot (Fig. 7b).

S1 + S2 parameter value is indicative of source rock quality. The Upper Silurian shales are classified as poor quality (< 2.5 mg HC/g); one sample obtained from central Lithuania shows excellent quality (> 6 mg HC/g). All Wenlock and Llandovery samples collected from west and central Lithuania are mainly classified as an excellent source rock (2.67–55.53 mg HC/g rock).

S1 + S2 parameter characterises the source rock that underwent maturation, HC generation, and expulsion. For evaluation of the original shale potential the mass-balance calculations can be used to determine the original organic matter content [20, 21]. The following two equations are used to calculate the original organic matter content:

$$f = 1 - HI_x\{1200 - [HI_o/(1 - PI_o)]\}/HI_o \times \{1200 - [HI_x/(1 - PI_x)]\}, \quad (2)$$

$$TOC_o = (83.33(HI_x)TOC_x/[HI_o(1 - f) \times (83.33 - TOC_x) + HI_x(TOC_x)], \quad (3)$$

where f, HI_x, PI_x and TOC_x are the fractional conversion, measured hydrogen index, measured production index and measured TOC, respectively, and HI_o, PI_o and TOC_o are the original hydrogen index, original production index and original TOC, respectively. 1200 is the maximal amount of hydrocarbons that can be formed assuming organic matter comprises 83.3% of HC composition. HI_o and PI_o for most immature source rocks are equal to 500 and 0.02, respectively [21].

The calculated original TOC_o values do not differ much from the measured TOC content that relates to low production index (Table 3). Only a small part of OM was converted to hydrocarbons.

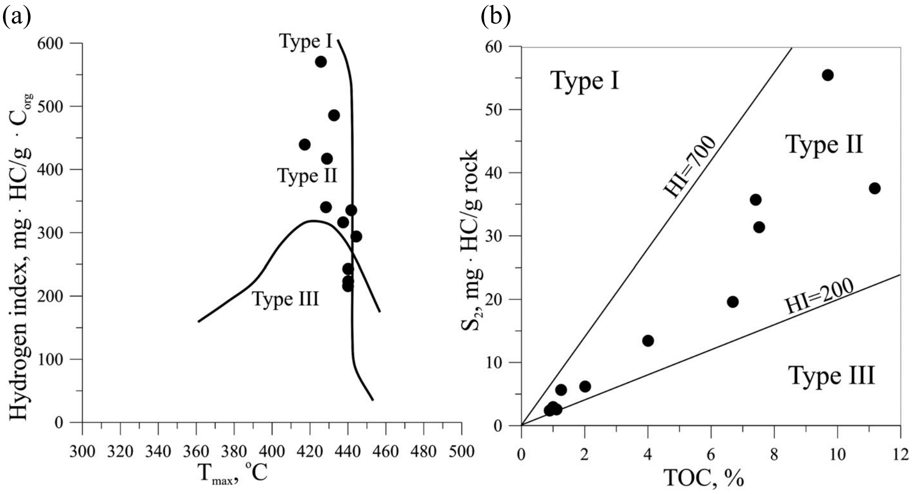

Fig. 7. (a) Diagram for HI vs T_{max} for Silurian shales. The organic matter type curve for HI/T_{max} is from [22]; (b) crossplot of S2 pyrolysate vs TOC defining kerogen type. Graph criteria are from [21] for west Lithuanian Silurian shales.

Table 3. Measured TOC_m and calculated original TOC_o values of Silurian shales

Well	Depth, m	Age	TOC_m, wt%	TOC_o, wt%
Geniai-1	1564	S2ld	0.98	1.39
Geniai-1	1583	S2ld	1.21	1.70
Geniai-1	1601	S2ld	1.14	1.58
Geniai-1	1754	S1ln2	6.72	8.61
Malukai-1	1612	S1ln2	9.72	8.96
Mikoliškės-1	2031	S1ln2	11.19	13.56
Paluknė-1	1660	S1ln2	7.36	7.60
Paluknė-1	1662	S1ln2	2.03	2.59
Vilkaviškis-131	789	S2pr	1.27	1.40
Vilkaviškis-131	1192	S1ln2	4.07	5.03
Vilkaviškis-131	1197	S1ln2	7.57	8.50

4.3. Thermal maturity of organic matter

The thermal maturity of the organic matter of the Lower Palaeozoic shales indicates that the western part of Lithuania was placed within the oil window (Fig. 8). There are two thermal maturity anomalies defined in west Lithuania. The Klaipėda anomaly reaching T_{max} 451 °C is located in the coastal area. The Rukai anomaly as large as 25 × 35 km is located in the southern part of west Lithuania. T_{max} values reach 455 °C within this anomaly. The thermal maturity for oil is classified as the early stage in most territory of west Lithuania. It reaches peak to late maturity level in only the central part of the Rūkai anomaly. These data are in concert with the low production index as discussed above.

T_{max} data agree with vitrinite-like reflectance measurements reported in [8, 22]; the reflectance is in the range of 0.6–0.9%, reaching 0.93–1.15% in the central part of the Rūkai anomaly.

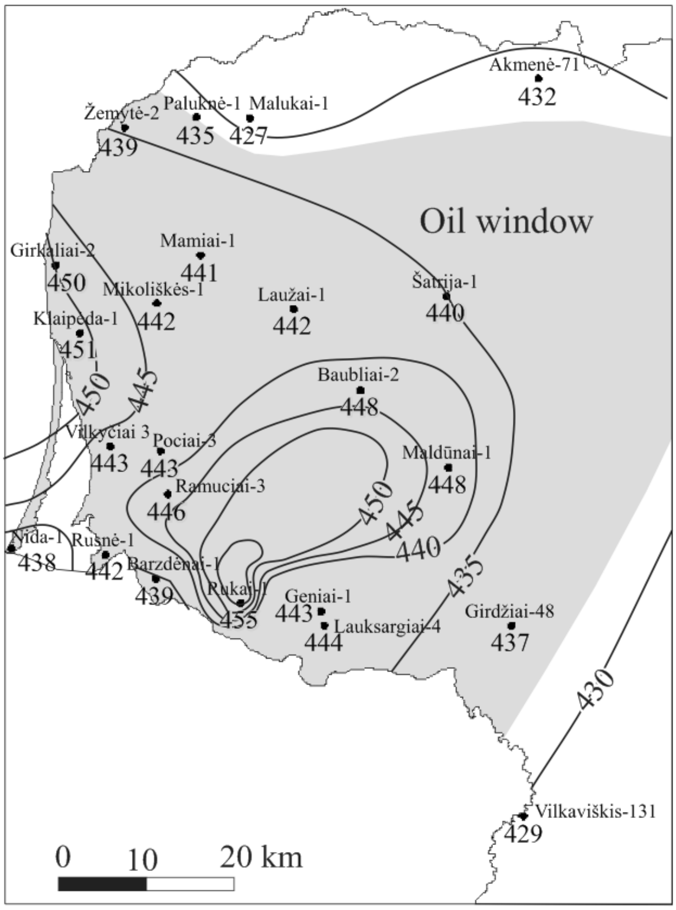

Fig. 8. Thermal maturity (T_{max}) of Lower Palaeozoic shales of west and central Lithuania based on [8] and new data. A grey polygon marks the oil window area.

5. Discussion and conclusions

Available geological data indicate that the Lower Silurian graptolite shales represent the most prospective source rock body for shale gas exploration in west Lithuania.

GASH criteria [23] can be assumed in evaluating shale gas prospects as follows:

- TOC > 2 wt%;
- type of kerogen II (II–III);
- thermal maturity of organic matter Ro% – 1.0%(1.5%) < Ro < 3.3%;
- depth < 3–4 km;
- thickness > 20 m;
- shale brittleness controlled by admixture of quartz and carbonates ca 30–40%.

TOC content exceeding 2 wt% is identified in only the Aeronian succession that is 1–8 m thick in the western part of Lithuania. The average content of organic matter is about 1.5–2.2 wt% in the Telychian and Wenlock shales, which is somewhat lower than the recommended lower limit for gas shale exploration. On the other hand, the kerogen is classified as type II that is considered as a positive parameter. The Upper Silurian shales are regarded as a non-prospective succession due to their low TOC content, < 1 wt%.

The thickness of Lower Silurian shales is in the range of 115–180 m that well exceeds the recommended lower limit. The depth of shales is within the prospective exploration range of 1–2 km (Fig. 9).

One of the most important parameters controlling the shale gas exploration prospects is the thermal maturity of organic matter. The reflectance of the vitrinite-like matter of the Lower Silurian shales is in the range of 0.6–0.9% in west Lithuania. Previously some anomalous values as high as 1.15–1.94% were reported, which, however, are not supported by T_{max} measurements, whose maximum values reach 455 °C that suggests the burial of the Lower Silurian shales in the oil window only. Some minor gas volumes are associated with oil, while no dry gas conditions were reached during the burial history of the basin in west Lithuania. The low thermal maturity is supported by a low gas factor in the Cambrian oil fields of west Lithuania [24].

Comparison shows the Lithuanian part of the Baltic sedimentary basin and the basin's western part in Poland to have rather similar shale gas prospects (Fig. 9). The parameters are compatible in terms of tectonic setting and TOC content, while the Lithuanian part is characterised by more favourable depths and thickness. The Polish part of the basin shows, however, higher thermal maturity. Furthermore, the gas fields are present in this part of the basin, while only oil fields have been discovered in the Lithuanian part of the Baltic sedimentary basin.

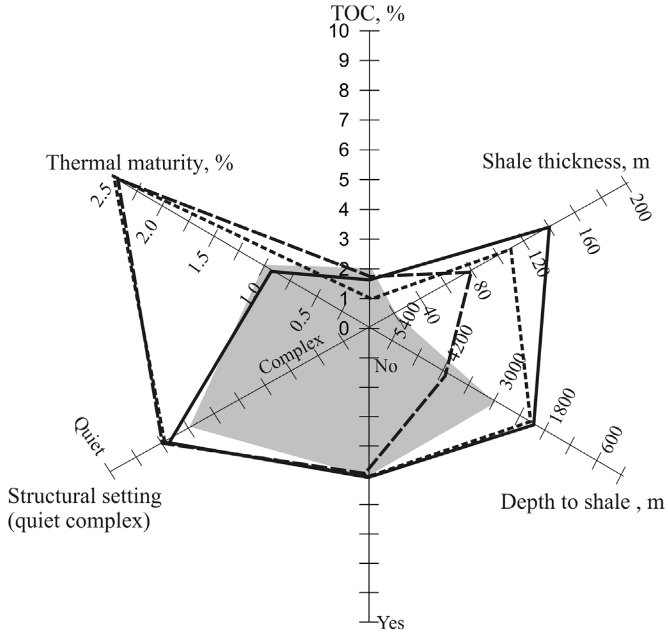

Fig. 9. Geological and geochemical risk parameters of shale gas exploration of Ordovician-Silurian shales for west Lithuania (a bold line) and Polish offshore and Pomerania representing, respectively, the eastern and western parts of the Baltic sedimentary basin. Polish data are from [25]. A grey area shows values in the high-risk zone.

It can be concluded that the obtained results indicate some positive parameters regarding shale gas prospects in Lithuania. However, the basic parameters, primarily the thermal maturity of organic matter, show only the oil generation burial stage of the Silurian shales. The low maturity is associated with an estimated low production index PI. Furthermore, the oil-gas ratio is low in Lithuanian oil fields, implying limited methane gas generation; no gas fields were discovered in Lithuania. The exploration activities should be therefore focused on evaluation of the Silurian shale oil potential instead of shale gas prospects.

Acknowledgments

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