VILNIUS UNIVERSITY
NATURE RESEARCH CENTER

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Petrography and petrophysical properties of the Lower Silurian shales of West Lithuania – geological assessment of gas shale prospects

SUMMARY OF DOCTORAL DISSERTATION

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VILNIAUS UNIVERSITETAS
GAMTOS TYRIMŲ CENTRAS

Saulius
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Vakarų Lietuvos apatinio silūro molingų uolienų petrografinė sudėtis ir petrofizinės savybės – dujų skalūnų perspektyvų geologinis vertinimas

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CONTENT

SUMMARY

The Baltic sedimentary basin is a pericratonic depression filled by the Phanerozoic sediments in the western part of the East European craton. The thickness of the sedimentary succession varies from several dozen meters in the periphery (Swedish Baltic coast, North Estonia) to more than 4 km in central Poland. All Phanerozoic geological systems are present in the Baltic sedimentary basin. The long-term sinking of the Earth's crust was associated with the different mechanisms, which changed over time, i.e., the passive continental periphery stage (Cambrian - Ordovician), the stage of the flexural bending, active continental periphery stage (latest Ordovician - earliest Devonian), the stage of the intracratonic basin (Devonian - early Carboniferous), the stage of thermal sinking - from the Late Permian (Poprawa et al., 1999).

Several dozen small oil and gas fields were discovered in the Baltic sedimentary basin. In total, more than 16 oil-filled structures were discovered in Lithuania. Also, several organic matter enriched shale packages are identified. Confusion often arises when describing the terminology of clay rocks – argillite, clay, shale. From a lithological point of view, the most appropriate is the term "argillite" adopted in Lithuanian literature (*i.e.*, a highly compacted clay that does not swell in water). Shale is the name of a rock of a silty-clayey sedimentary rock and also is used for organic-rich commercial bodies (*e.g.* Estonian combustible shales with a wide variety of compositions from clays to limestones). Until now, the Lower Palaeozoic clayey formations of West Lithuania have been studied only as potential oil source rocks, *i.e.*, organic matter composition, oil generation potential, etc. (Kadūnienė, 1978; 2001; Zdanavičiūtė, Lazauskienė, 2009). The specific research approach is required for studies of shales as the gas production formations. The present study responds to these new assessment requirements, which are important not only for assessing prospects of the Baltic region prospects, but also for the regional

characterisation of shales as the potential new hydrocarbon exploitation formations.

Rationale of the study

The previous studies of the Silurian succession were scoped mainly on tracing lithological variations, lithostratigraphic, paleontological subdivision and characterisation of geochemistry of organic matter (quantity, composition, *etc.*). The studies of organic matter are essentially important in assessing the Silurian shales as the hydrocarbon source rock package in the Baltic basin. The content and thermal maturity of organic matter have been studied in detail since seventies and essentially in recent decades (*e.g.*, Zdanavičiūtė and Lazauskienė, 2007; 2009; Šliaupa et al., 2016, Chichon-Pupienis *etc*., 2020). Since the initiation of the successful practical exploitation of unconventional gas (and oil) in organic rich shales (in USA), thermally mature shales are considered not only as the source rocks, but also as the unconventional reservoirs. It requires considerable reconcilement of the study strategy. The Lower Silurian shales are considered as the major geological formation in unconventional gas exploration in the Baltic basin, including West Lithuanian territory. There is little known of the exploitation properties of these shales that requires better knowledge of the mineral composition of shales which directly control exploitation properties (petrophysical, mechanical) of shales.

Relevance of the study

Shales, which meet criteria for the operational suitability of unconventional gas exploitation, are reported from most European countries. It is easier to list countries where they have not been classified as promising geological formations in this respect, i.e., the Czech Republic, countries of the Balkan Peninsula, Switzerland, Finland, Norway, Belarus, Latvia, Estonia. It is accounted to specific geological conditions in different regions. The age of shales varies from the Cambrian (*e.g.*, Sweden, central Poland) to Paleogene (Eocene Ebro basin in Spain, Oligocene Hungarian basin) and Neogene (Transylvanian basin in Romania, small rifts in Hungary, Austria, Slovenia, Croatia).

The largest resources of the Silurian shales are hosted by the Baltic and the Podlasie-Lublin sedimentary basins. Also, potential gas shales of the Silurian age are reported from Spain (Cantabrian massif), Romania (Mossen platform). Furthermore, the Silurian shales rich in organic matter are widespread in North Africa, Arabian Peninsula, China.

Intense shale gas research was carried out in Poland between 2011 and 2014. However, initial expectations were likely too high before key assessment wells were drilled in the region that led to decline in exploration activities during recent years. It has been realised that more sophisticated methodology is needed to facilitate the cost-effective exploitation of shale gas.

The previous approach was focused on assessment of the hydrocarbon generation of the Silurian shales. The present thesis discusses the petrophysical properties of the Silurian clayey rocks of West and central Lithuania to gain a better understanding of the Silurian shales as the unconventional gas reservoir.

Brief description of the study subject

Silurian sedimentary rocks are distributed in most of territory of Lithuania, except for the south-eastern part of the country, where they have been denudated during Carboniferous - Early Permian. The thickness of Silurian succession in Lithuania varies from 100 m in the east to 850 m in the westernmost part (Paškevičius, 1994). Accordingly, the sedimentation environment also was deepening to the west as well as the burial depth in the basin.

The study area encompasses the west half of Lithuania showing predominance of shales in the section and maximum burial depth as well as the highest thermal maturity of organic matter. Black and dark grey graptolitic shales with rare marlstones and limestones accumulated in the deep shelf environment. The depth of the Silurian base varies from about 1500 m to 2000 m, present temperature is in the range of 40°C to 85°C. The Lower Silurian shales are most enriched in organic matter and are characterised by the highest thermal maturity. Petrophysical and mechanical properties were studied using both rock samples and well log data. A wide range of properties were investigated.

Practical relevance

The research of shales enriched by organic matter, considered as a source rock for hydrocarbon generation and sealing rocks of hydrocarbon deposits, has a long history in Lithuania. However, considering the role of shale as the unconventional reservoir, there is no common global approach that challenges future effective studies of this kind of geological formations. The results of the present research can be applied not only for deciding on the prospects of Lithuanian gas shales, but also may contribute to assessment of these rocks in adjacent countries (Poland, Ukraine, Denmark). The Lower Silurian shales are important exploration object not only in Europe as discussed above. The results of detailed analysis of the mechanical and petrophysical properties of clayey rocks can also be of significance in assessing the potential of shales for underground storing radioactive wastes, underground energy storage, etc.

The methodologies used in the dissertation can be used by geoscience researchers and students of Vilnius University, by specialists of the Lithuanian Geological Survey, scientists of the Nature Research Centre to carry out further targeted research of the Lower Silurian or other stratigraphic units. The results can be used effectively in the activities of operators in the development of the exploration and production industry for non-conventional hydrocarbons (oil and/or gas) and the above-mentioned issues of assessment of the potential of underground storage facilities.

Scientific novelty

A complex study of the mineral composition and related petrophysical (and mechanical) properties of the Lower Silurian shales was carried out. There is little known about these parameters in Lithuania (and the whole Baltic basin, in general). Also, studies combining numerous methods are still an exception, rather than systematic, as most of the previous studies were focused on discussing particular parameters in the literature, missing correlation to other characteristics.

Purpose

The purpose of present study is an assessment of the previously scarcely studied or not studied characteristics of the clayey rocks of the Lower Silurian of West Lithuania. It is essentially important to recognise lateral and vertical variations of the key parameters controlling exploitation parameters, aiming at identification of sweet spots that may be recommended for more detail studies.

Tasks

The following tasks were set for the research work:

1) To systematize available data presented in publications in Lithuania and adjacent regions;

2) To perform new laboratory tests of the petrographic composition and petrophysical (and mechanical) properties of the Lower Silurian shale rock samples;

4) To perform rock sample petrographic studies relevant to petrophysical properties of shales;

5) To interpret well log data to expand knowledge on the operational characteristics to a whole Lower Silurian section (by contrast to selective distribution of the available rock samples;

6) To link results of studies of the organic geochemistry indicators to petrophysical properties.

Defended positions

The following key aspects were set for the research work:

1. The main part of the Lower Silurian shales of West Lithuania consists of clay minerals with a high proportion of detrital minerals and low content of carbonates. In the clay fraction, illite predominates with a high proportion of chlorite and low content of mixed layer illite-smectite and kaolinite.

2. The mineralogical composition of shales is the main factor determining their petrophysical and mechanical characteristics. Detrital minerals increase the values of brittleness, and the amount of clay minerals decreases them. High brittleness index, high porosity, high CEC value, low CST are treated as favourable from the operational point of view.

3. Interpretation of geophysical well log data allows to significantly expand the description of shales, especially in those parts of the section where there is no core. Using appropriate logs, we can obtain areal TOC values distribution, distinguish OM-enriched layers, and obtain the distribution of brittleness index.

Data sources

The optimal complex of methods was established for the study based on published recommendations and peculiarities of the Lower Silurian shales. Published and unpublished (industrial reports stored in archives of the Geological Survey of Lithuania) data were collected. Archive materials concern mainly data on rock porosity, density, carbonate and total organic carbon (TOC) content. As the Silurian shales are considered as the major source rocks, there is abundant published and unpublished data on organic matter (TOC content, different organic chemistry parameters, thermal maturity, *etc.*). Also, industrial well log data of deep oil exploration wells were collected. These were interpreted for assessment of variations in organic matter content and rock brittleness index in wells. Drill core samples, stored in Vievis core house, were collected. New measurements of the petrophysical (and mechanical) properties of rock samples, mineralogical composition and TOC content were performed at different certified laboratories.

Material and individual contribution

Data of previous studies, both published and unpublished, were collected and systemized. Drill cores of selected wells were inspected, samples were collected. Laboratory tests (SEM, XRD, mechanical and petrophysical properties) were carried out at different laboratories. Data base of selected well logs was compiled. Collected data were interpreted and cross corelated.

Scientific approbation of the study results

The study results were presented at 3 international and 3 national scientific conferences.

3 papers were published in peer-reviewed periodical scientific journals, 2 of which are listed in Web of Science data base.

Ph.D. thesis structure

Thesis consists of Introduction, 5 chapters, Conclusions, References, a list of author's publications.

Statistics: 105 pages of text, 51 paintings and 8 tables. The Reference list contains 132 literature sources.

1. SHORT OVERVIEW OF THE LOWER SILURIAN SUCCESION OF LITHUANIA

The Silurian sedimentary succession comprises the largest part of the sedimentary infill of the Baltic sedimentary basin (Paškevičius 1997). The thickness of the Silurian succession ranges from about 100 m in East Lithuania to about 850 m in West Lithuania.

The thickness of the prospective Lower Silurian shale package is about 120-200 m in West Lithuania. There are little changes in the thickness across Lithuania that is typical of uncompensated sedimentation. The Lower Silurian succession of West Lithuania is dominated by black shales. There are no sedimentation breaks documented in the Lower Silurian section. The Juuru, Raikküla, and Adavere regional stages (RSt) compose the Llandovery Series and the Jaani, Jagurahu, and Gėluva RSts are defined in the Wenlock Series (Paškevičius 1997) (Figure 1).

In West Lithuania, the basal several metres thick micritic limestones, attributed to the Juuru RSt, are distinct by the gamma-ray log minimum (Figure 1). There is a thin transition zone of marlstones and limestones passing upwards to the most gamma-ray active (200-400 API) black graptolitic shales of the Raikküla RSt, the thickness of which ranges from about 4 to 11 m. This remarkable geological layer is overlain by about 40 m thick Adavere RSt black shales, the gamma-ray activity of which varies between 180–200 API. This package shows a slight regression pattern explained by carbonate content increasing upwards. The Wenlock succession starts with the calcareous black shales attributed to the Jaani RSt. It is about 15 m thick. The overlaying Jagurahu RSt indicates reestablishment of more clayey sedimentation conditions. It is about 60 m thick. The Gėluva RSt shales of about 20 m thick crown the Wenlock succession. The gamma-ray activity of the Wenlock rocks is about 190–210 API.

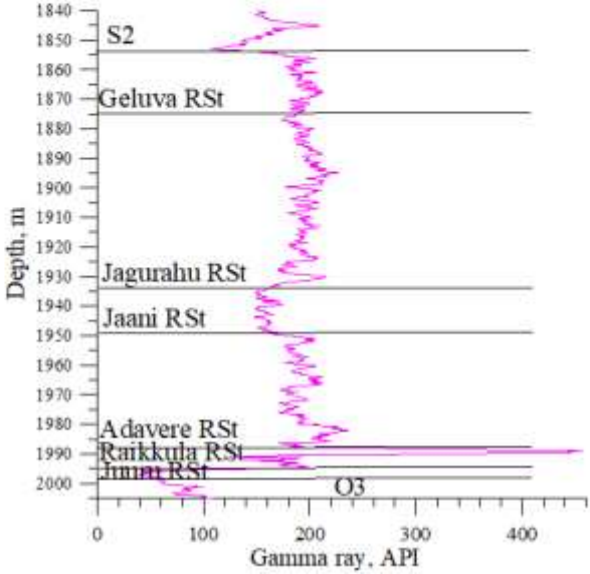

Figure 1 Gamma-ray log of the Lower Silurian section, well Barzdėnai-1, west Lithuania (see Figure 3 for location). Regional stages are defined. O3 shows the top of the Upper Ordovician, S2 marks the basal part of the Upper Silurian

The thermal maturity of organic matter (OM) is variable in West Lithuania. The oil window (Tmax values >435ºC) is defined in most of the territory of West Lithuania. In West Lithuania, two prospective and priority gas shale areas are defined (Fig. 2). The largest priority area is situated in the southern part of West Lithuania, where Tmax values vary between 442°C-455°C. Vitrinite-calibrated reflectance was reported as high as 1.01, 1.15 and 1.94 in three shale samples in the southern part of the priority area (wells Rukai-1, Ramuciai-3 and Vainutas-1, respectively) (Zdanavičiūtė and Lazauskienė 2009). It implies generation of "wet" gas. The second priority area is centred by Klaipeda City. Tmax was measured as high as 450–451°C there.

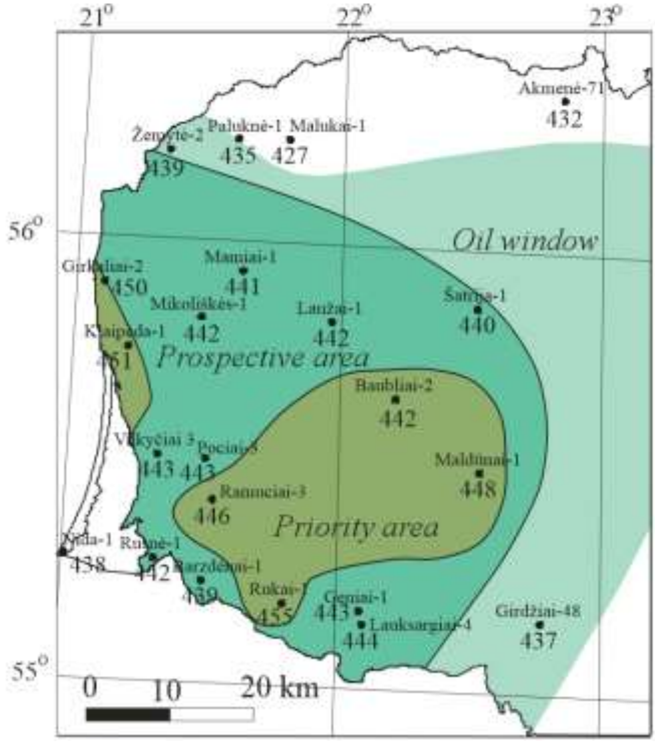

Figure 2 Classification of Lower Silurian gas shale of West Lithuania based on thermal maturity of OM (Šliaupa *et al.*, 2016). Measured Tmax of OM of the Lower Silurian shales are indicated

2. METHODS

Rock samples of the Lower Silurian shales and subordinate carbonates were collected from 26 oil exploration wells from the West Lithuania (Figure 3). Data base of well logs of studied wells was compiled for interpretation of the basic shale parameters (brittleness index, OM content) and stratigraphic control. Drill core samples were analysed in different laboratories (Core Laboratories, Weatherford Laboratories, Laboratory of Lithuanian Geological Survey and Physical Sciences and Technology Centre Laboratory in Vilnius). Petrographic studies were performed at Nature Research Centre.

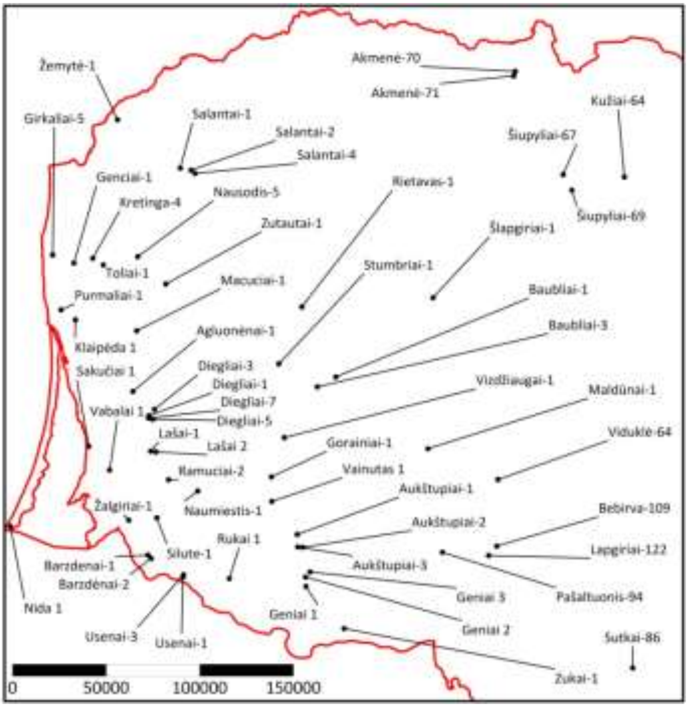

Figure 3 Wells where clay rocks have been sampled or other tests have been carried out.

X-ray diffraction (XRD) data were obtained in cooperation with Core Laboratories (https://www.corelab.com) and laboratory of the Centre for Physical Sciences and Technology in Vilnius. 120 samples were analysed. The mineralogical composition of the Lower Silurian shale samples was also analysed using scanning electron microscope QUANTA 250 with EDX (SEM) extension at Natural Research Centre in Vilnius.

Brittleness index (BI) was estimated for rock samples using correlation equations with the mineral composition derived from representative wells. BI was also calculated by interpreting well log data of 12 wells (gamma-ray and electrical resistivity).

TOC (total organic carbon) content was estimated using XRD data (along with mineralogical characterisation, 120 samples) and 13 well gamma-ray logs.

Cation Exchange Capability (CEC) (36 samples), Cation Suction Time (CST) (9 samples) and Stability to Erosion (7 samples) were measured at Core Laboratories (https://www.corelab.com) and Weatherford Laboratories (https://www.weatherford.com).

Uniaxial compression and extensional testing (3 samples) was carried out at the Laboratory of Lithuanian Geological Survey.

3. RESULTS

3.1. Mineralogical composition of the Lower Silurian shales

15 wells were sampled to determine mineralogical composition of shales. 120 samples collected mainly from Llandovery shales and only a few rock samples of the Wenlock age were analysed. The X-ray diffraction (XRD) method was used, the results of which were supplemented by the SEM (with EDS) petrographic studies.

The Lower Silurian shales contain variable amount of OM that correlates with TOC content (correction factor 0.833) reaching up to 21% of rock volume (Šliaupa *et al*., 2016). Distinct anomalous layer, attributed to the Raikküla RSt, is defined in the lower part of the Landovery succession that is marked by GR spike in the well logs (4-12 m thick) and is also well defined in the acoustic logs. The overlying Llandovery shales have average TOC content 1.8% (Lazauskienė *et al*., 2014; Šliaupa *et al*., 2016). Similar levels of TOC have been reported from the Wenlock shales. SEM petrographic analysis shows that OM concentrates on the bedding planes and form micrometre- to millimetre-scale lenses and also in a dispersed form (Figure 4).

XRD data show that the amount of clay minerals varies from 37% to 57% in the lower Silurian shales. Clay minerals are dominated by illite. The part of illite in the clay fraction varies from 36% to 69%, while mixed layer illite-smectite (I-S) composes 5-35%. I-S contain 5-10% of the smectite layers. Chlorite amounts 12-15% of clay minerals. Kaolinite content is only 2-7% (two samples contain 14% and 21% kaolinite).

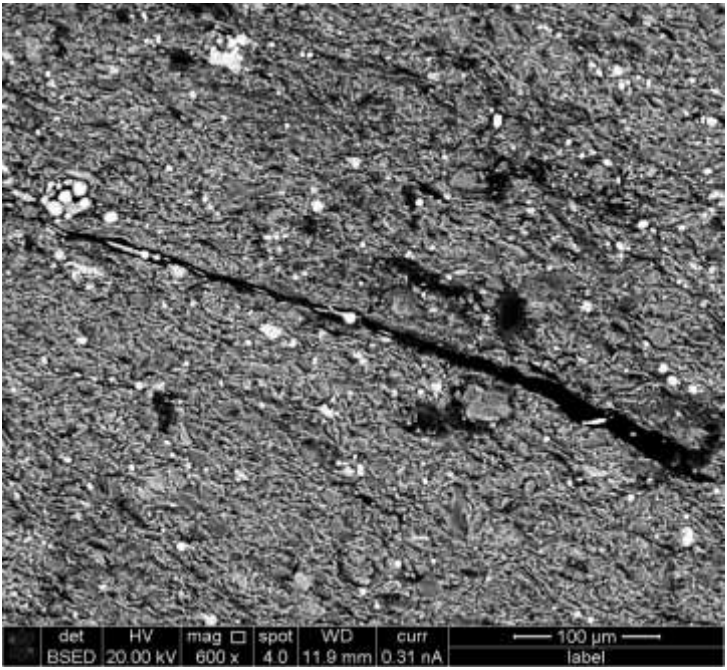

Figure 4 SEM micrographic sowing distribution of OM, well Klaipėda-1

Shales are rich in detrital quartz and feldspars. The quartz content is 11-40% and amount of feldspars is 2-10% (potassium feldspars 1.5-5.5% and plagioclase 0.9-6.4%). The ratio of quartz to feldspars varies between 3.8 and 9.7, but is mainly between 5 and 6. The total amount of detrital quartz and feldspars varies in the range of 13-48% (mainly 35-45%).

The abundance of feldspars depends only little on the content of clays and carbonates, while the amount of clay minerals strongly correlates negatively with concentration of carbonates. On the shale classification ternary diagram by Gamero-Diaz et al. (2013), most of data dots cluster in the silica-rich calcareous mudstones field and some are scattered in the mixed argillaceous mudstones field (Figure 5). Also, there is a wide scatter of dots in the argillaceous siliceous mudstones and mixed mudstones fields. Two dots are classified as a mixed carbonate mudstone. Notably, all dots compose a coherent trend implying a common population (quartz and clay minerals contents decrease with increasing carbonates content), suggesting the common lithotype.

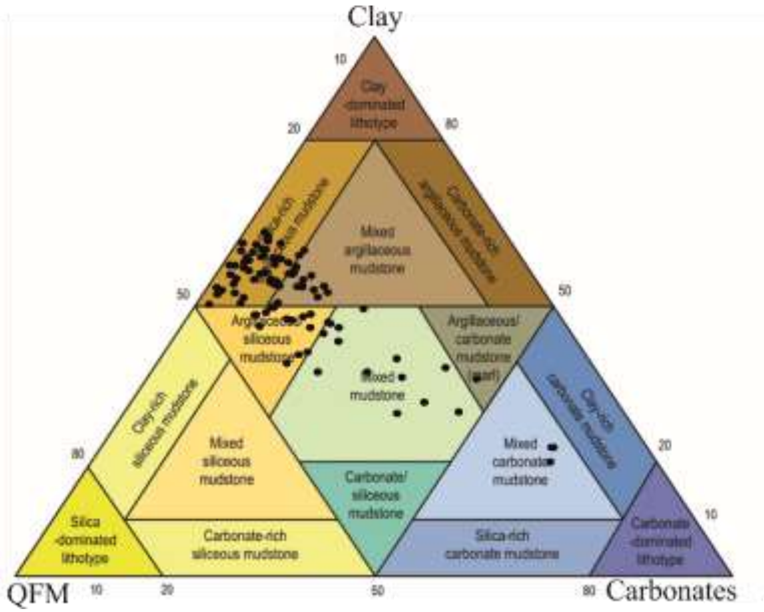

Figure 5 Litological classification of core samples enriched in OM (Gamero-Diaz et al., 2013), the Lower Silurian shales of West Lithuania (black dots)

Calcite is a common mineral in shales. Two generations are distinct, the early and late diagenetic calcite. Most often, early diagenetic calcites form micrometre-scale patches in shales.

Idiomorphic late diagenetic dolomite crystals are common in shales. Most likely, these crystals are associated with the transformation of clay minerals which cause release of calcium and magnesium cations into pore water. The maximum dolomite concentration 9.4% was reported from sample from the well Nida-1, but usually ranges from 1% to 2% (Figure 6).

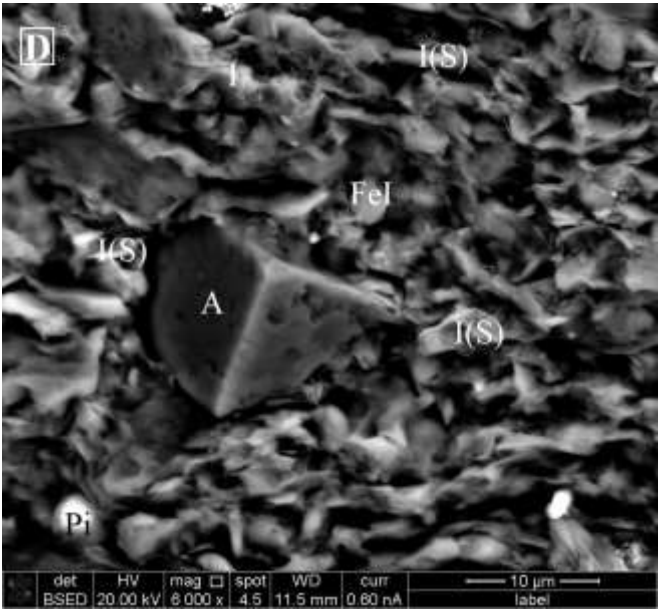

Figure 6 Micrograph of the surface of the SEM sample (vertical layering), Llandovery shale, depth 2041.9 m, well Klaipėda-1. Idiomorphic ankerite (A) in illitic-smectitic mass (I(S). Fel – feldspars, Pi – pyrite

Pyrite is present in most shale samples, its abundance ranges from 0.4% to 8.8. Framboids, which commonly formed during early shale diagenesis (Taylor, Macquaker, 2000), are common (Figure 7). Late diagenetic pyrite is also widespread in the form of small lenses.

Gypsum crystals were detected in some samples. Gypsum crystals are 10-50 µm in diameter and are concentrated on the bedding planes that suggests horizontal circulation of the pore water.

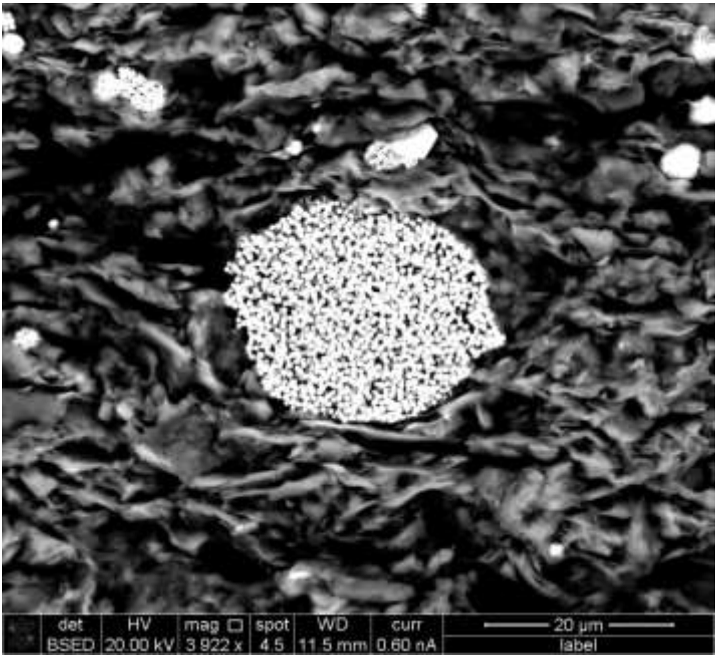

Figure 7 Micrograph of shale, depth 2041.9, well Klaipeda-1. In the central part, secondary (framboids) and late diagenetic (lenses) pyrite is abundant

3.2. Distribution of organic matter

Organic matter (OM) content is a key parameter in assessing the exploitation prospects of shales (Wand and Gale, 2009; Lu et al., 2012). Previous studies of changes in OM content have revealed some certain vertical and horizontal trends in the Silurian shales (Kadūnienė, 1978; 2001). TOC content is mainly less than 1% in the Upper Silurian shales (Creep *etc*., 2016), while it exceeds 1.5-2% in the Lower Silurian shales. A close statistical correlation between gamma-ray intensity and TOC content was established (Šliaupa et al., 2016). This made it possible to better recognise variations in TOC content in the wells in Lower Silurian shales.

Gamma-ray log of 12 wells was interpreted. The well Barzdėnai-1 is provided as a typical example of vertical TOC distribution in the Lower Silurian shales. The maximum TOC values are characteristic to the Raikküla RSt and reach 14% (up to 21% in some wells) (Figure 8). The overlying Adavere RSt and Wenlock shales are less enriched in OM. The average TOC content is about 2% in Adavere RSt and in the Wenlock shales.

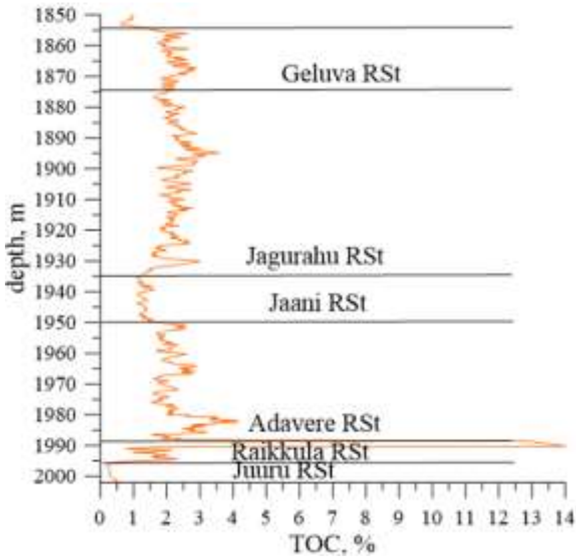

Figure 8 TOC content in the Lower Silurian shales, well Barzdėnai-1. Calculated from gamma-ray well log using correlation equation TOC%=exp(0.12*GRmkr/h*0.15) (after Šliaupa *et al*., 2016)

TOC distribution maps were compiled for the particular regional stages of the Lower Silurian succession in West Lithuania (Figure 9). Despite different stratigraphic levels and slightly different average TOC content, a similar lateral pattern is observed. The maximum TOC content is calculated along the anomalous maximum trending NNE-SSW in the eastern part of West Lithuania. The most distinct concentration of OM is documented in the southern part of this trend (well Žukai-1), on average TOC=6.6-8.9%. The concentration of OM decreases significantly to the east (well Lapgiriai-122). Central Lithuanian wells has a good core yield, shales are enriched in carbonates. In the west, enrichment in OM is systematically decreasing. The lowest TOC concentration is calculated in the north-west, the TOC concentration is less than 2%.

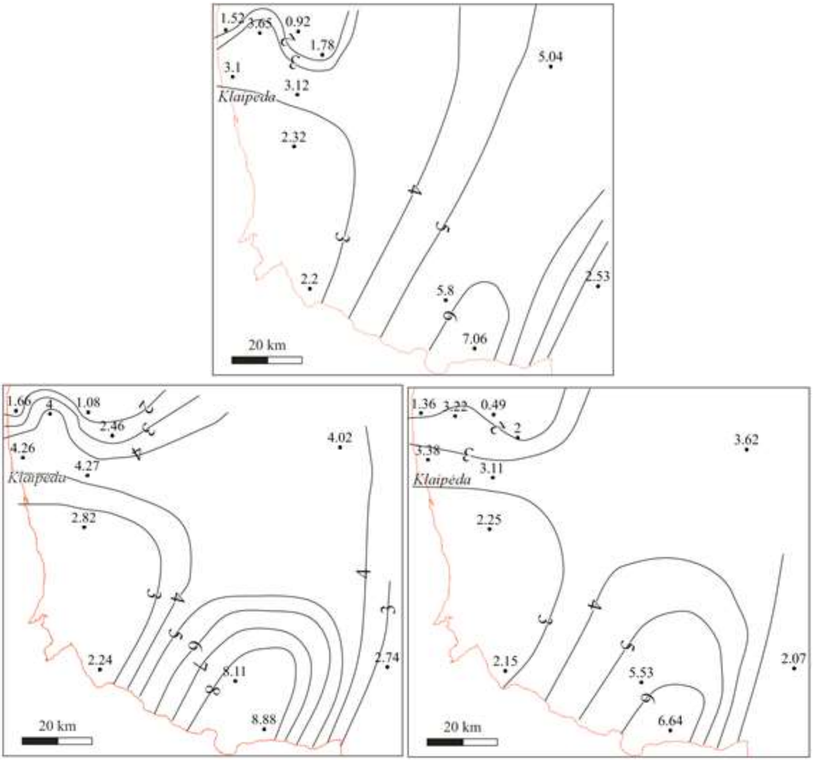

Figure 9 Horizontal TOC (%) the distribution of values calculated from gamma-ray on the Adavere, Jagurahu, and Gėluva RSts

Cross-plots of acoustic wave velocity and electrical resistivity are sometimes used to determine prospective intervals for shale gas exploration (Passey *et al.*, 1990). It is also known as the "D log R" method. The correlation describing the "baseline" Silurian shales of West Lithuania is defined:

$$dT=3522-63.102\times \ln R \quad [1]$$

where dT is the slowness of sonic waves (μs/feet), lnR is the electrical resistivity (ohm-m).

In the representative well Barzdėnai-1, a significant deviation between the measured sonic slowness and calculated slowness DtR is calculated using the formula [1] (Figures 10, 11). The difference is essentially distinct in Jaany RSt and Raikkula RSt that suggest maximum content of OM.

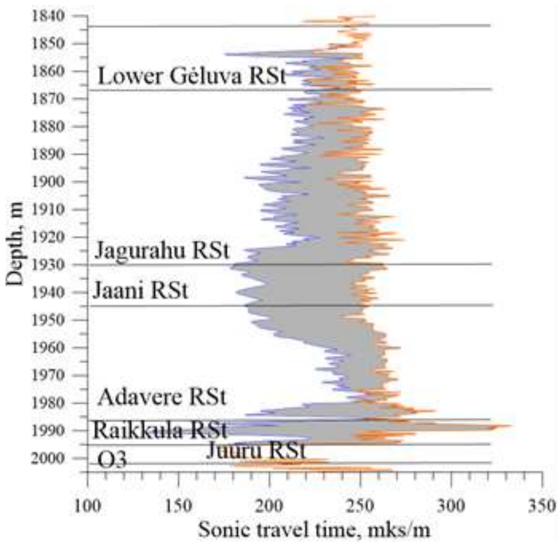

Figure 10 Acoustic wave slowness and calculated acoustic wave slowness according to formula [1], well Barzdėnai-1

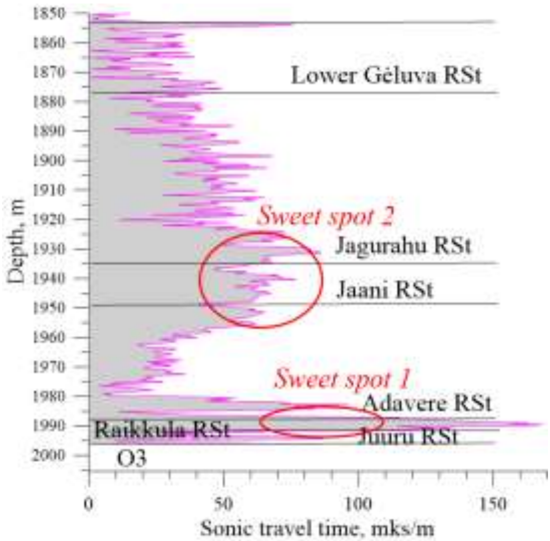

Figure 11 The difference between the measured acoustic wave slowness and the pseudo-acoustic slowness DtR, calculated according to equation [1], reflecting the relative distribution of OM, well Barzdenai-1

3.3. Porosity of shales

Porosity of deeply buried shales is typically low. The depths of the Llandovery shales vary from 900–1200 m in Central Lithuania to 1900–2100 m in West Lithuania. Porosity decreases generally to the west. In Central and North-West Lithuania, porosity of the Lower Silurian shales is 12–16% (Figure 12). In the West Lithuania, it decreases to less than 3%. The width of the transition zone between the west and east zones is 30-40 km.

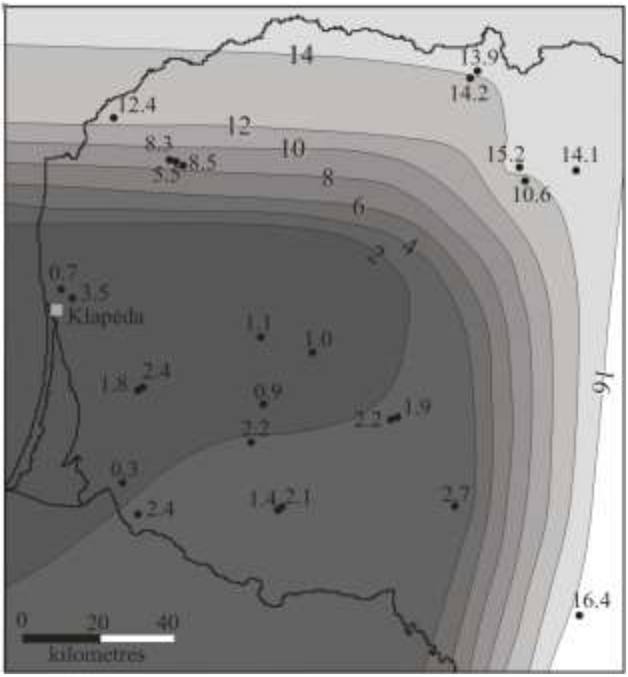

Figure 12 Map of distribution of porosity of Llandovery shales

There is a tendency for the porosity decrease with increasing depth (Figure 13). The porosity of 3-8% of shale rocks is a positive indicator of reservoir performance. These values are close to the widely described Barnett (USA) shales 3-6%, Woodford (USA) shales (~2500 m depth) 3.5-6.5%. Thus, on the basis of this comparison, it can be argued that the shales of the Lower Silurian of West Lithuania are of sufficient operational quality.

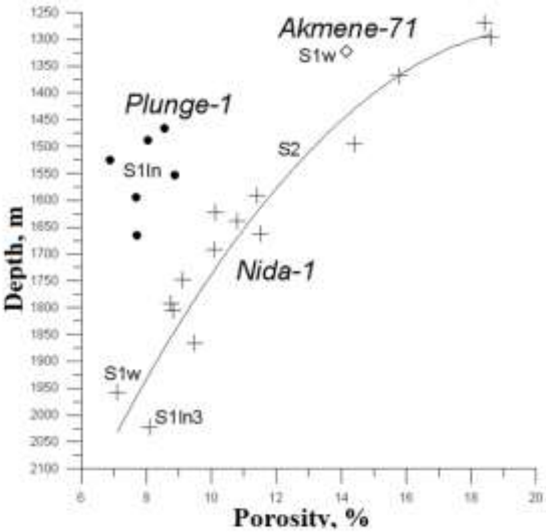

Figure 13 Porosity of the Silurian shales in selected wells. Points – well Plungė-1; crosses – well Nida-1; diamonds – well Akmenė-71. The line indicates the second-degree correlation curve of well Akmenė-71

3.4. Brittleness index

Brittleness is an important mechanical property of gas shales. However, there is no universally accepted brittleness concept or measurement method. There are different techniques to estimate the brittleness index (BI). A comprehensive summary of different BI calculation methods was presented by (Jin *et al*., 2014; Bai, 2016).

The mineral composition of shales strongly influences the ductility/brittleness of shales. Quartz and feldspars increase shale brittleness, while carbonates, clay and OM decrease BI (*e.g.* Yasin *et al*., 2017). Jarvie *et al*. (2007) proposed a simplified BI equation incorporating quartz, carbonate, and clay content data:

$$BI_{Jarvie(2007)} = \frac{Qz}{Qz + Ca + Cly} \quad [2]$$

Wang and Gale (2009) suggested an alternative relationship:

$$BI_{Wang(2009)} = \frac{Qz + Dol}{Qz + Dol + Ca + Cly + TOC} \quad [3]$$

here Qz is the fractional quartz content, Dol is the dolomite content, Ca is the calcite content, TOC is the total organic carbon content, and Cly is the clay content by weight in the shale.

Calculated values of BI_{Jarvie} vary from 0.17 to 0.43 for the Lower Silurian shales of West Lithuania. Most samples analysed show, however, a narrow BI_{Jarvie} range of 0.36-0.39. The BI_{Wang} values change in a similar range from 0.22 to 0.48 (mainly from 0.35 to 0.40). The studied shales are classified as less ductile and less brittle.

Well logs are often used as an alternative technique to derive BI. Interpretation of sonic wave logs of the Lower Silurian shales were performed in eight wells in West Lithuania. These data were used to calculate the logging brittleness index (LBI) of shales. Equations derived for the Carboniferous-age Barnett shales (Jin *et al*., 2014), the global correlation established for the Barnett, Woodford and Eagle Ford shales (Jin et. al., 2014), and the Devonian Woodford shales (Jin and et al., 2014) were applied:

$$\text{Woodford} \quad LBI = -0.012 \times DTC + 1.4921 \quad [4],$$

$$\text{Barnett} \quad LBI = -0.01104 \times DTC + 1.4941 \quad [5],$$

$$\text{Globali} \quad LBI = -0.0142 \times DTC + 1.7439 \quad [6],$$

here LBI (logging brittleness index) is the brittleness index (non-dimensional), DTC is the acoustic wave slowness (feet/μs).

Despite the specific equation used, there is a common trend towards an overall upward increase in LBI (Figure 14).

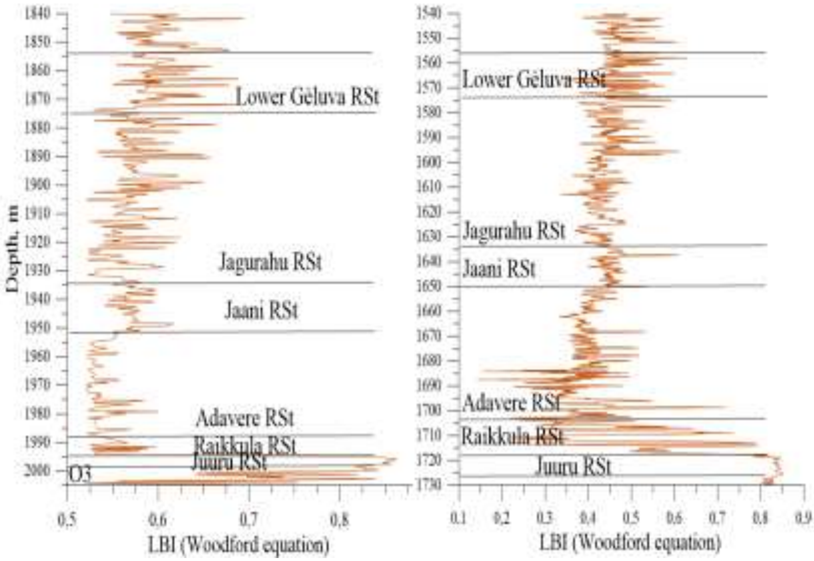

Figure 14 Brittleness index calculated from sonic wave slowness using the Woodford shale correlation. On the left the well Barzdėnai-1 is shown, on the right is the well Šlapgiriai-1. Spikes are caused by technical (electrical) malfunctions of the probing equipment

It is commonly accepted that the BI is closely related to the unconfined compressive strength and tensile strength (Hucka and Das, 1974):

$$BI = \frac{C_0 - \sigma_T}{C_0 + \sigma_T} \quad [7]$$

here C_o is the unconfined compressive strength (UCS) and σ_T is the tensile strength.

The BI 0.33 was calculated for one Wenlock shale sample of the well Rietavas-1. This value is in the range to values estimated from the mineral composition, though lower than the average 0.40 value. The shale sample is classified as marginally less brittle.

When comparing the equations used, the relatively highest LBI values were calculated using the Barnett shale equation, although they are quite close to the LBI values calculated from the global shale correlation equation, with LBI varying from 0,48 to 0,72. The Woodford shale equation calculates relatively lower LBI values.

Three maps of the distribution of LBI values of the Jaani, Adavere and Jagurahu RSTs were compiled, illustrating the lateral changes in the brittleness of shales. The same trends are observed for the particular stratigraphic levels studied, regardless of the specific correlation equation used. LBI is systematically increasing westwards from 0.39 in the east to more than 0.55 in the west (Figure 15). In addition, the effect of the regional W-E direction Telšiai fault zone is evident (the fault is not indicated on the maps).

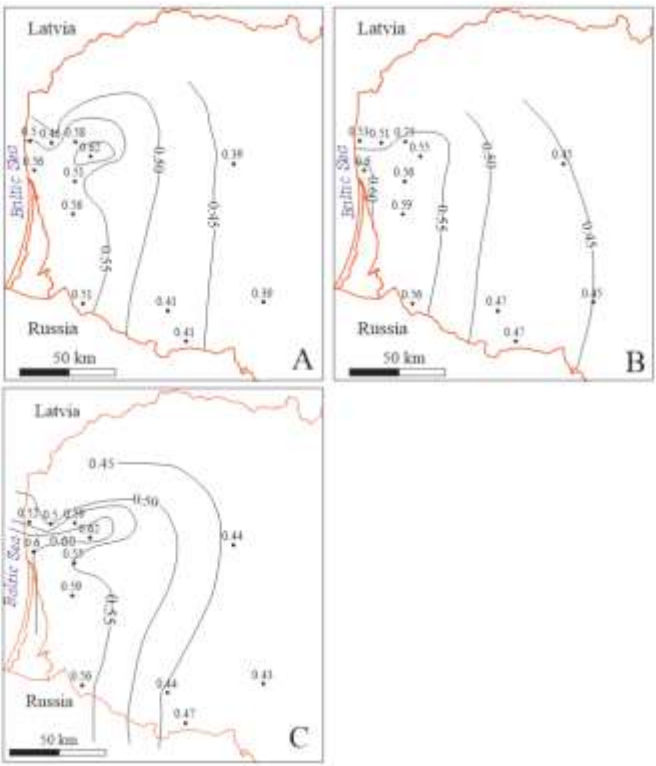

Figure 15 Lateral distribution of LBI (Woodford correlation) calculated for (A) Adavere RSt, (b) Jaani RSt and (C) Jagurahu RSt

3.5. Cation exchange capacity

The cation exchange capacity reflects the gas absorption capacity and depends primarily on the mineralogical composition of shales. The measured values of the cation exchange capacity (CEC) of shales vary from 0,2 to 8,8 milli-equivalents/100 g (meq is the number of ions which total a specific quantity of electrical charges). For comparison, CEC ranges from 0.3 to 9.1 meq/100g with an average of 5.9 meq/100g in the Marcellus Shale and 3.5 meq/100g in the Utica Shale (USA). CEC decreases with depth. No clear difference was observed between the different stratigraphic units. The maximum proportion of samples have values of 2-3 meq/100g. The CEC histogram shows the bimodal distribution with maximum values of 2-3 meq/100 g and 4-6 meq/100 g respectively (Figure 16).

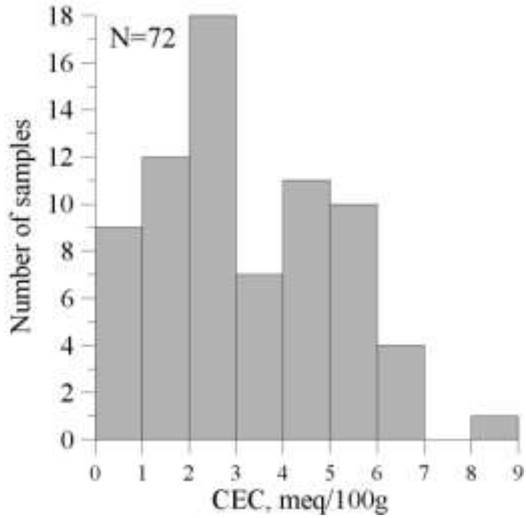

Figure 16 Histogram chart of CEC values

3.6. Erosion resistance of shales

Shale resistance to fluid circulation is an important operational parameter. The studied samples show rather different resistance to erosion. Samples collected from the well Geniai-1 show the highest resistance, while samples of the well Usėnai-3 have the lowest resistance to erosion (Figure 17). Diesel has the lowest erosion effect (0.35-2% by weight lost), and distilled water erodes shales most intensively (1.95-6% of the mass lost). All samples analysed demonstrate moderate erodibility with all kinds of combinations of water additives. A 7% KCl solution shows the best effect in maintaining low shale erosion. These values are consistent with published results showing that K+ cations added to the water-based mortar effectively inhibit destruction of clay (Hallman, 2003).

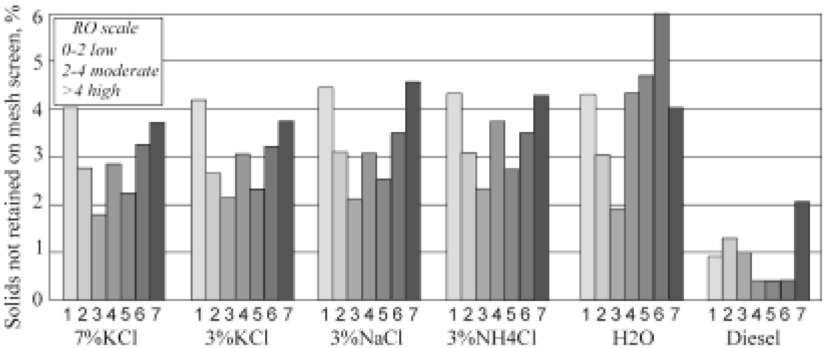

Figure 17 ROS values of the Lower Silurian shales

3.7. Capillary Suction Time

The measured values of the Capillary Suction Time (CST) are relatively low (Table 1) that is accounted to the specific mineralogical composition of the Lower Silurian shales, *i.e.*, predominating illite has a low degree of swelling, while the kaolinite and chlorite swelling do not increase to such a level that they can be distinguished (Hart, 1989).

A 7% KCl solution in water was found to be the most suitable for controlling the low swelling of clays. A 7% solution of KCl reduces the time of absorption of capillaries by 1.6-2.3 times compared to distilled water.

Table 1

Capillary suction time (CST) ratio of the Lower Silurian shales

Well	**Age**	**Depth, m**	**7% KCl**	**3% KCl**	**3%NaCl**	**5%NH_4Cl**	**H_2O**
Barzdėnai-1	S1ln	1985.0	1.68	2.13	2.55	2.49	2.62
Genčiai-1	S1w	1858	1.80	1.83	2.20	2.01	3.70
Genčiai-1	S1ln	1941.9	1.05	1.10	1.41	1.20	2.45
Geniai-1	S1ln	1757.7	0.97	1.01	1.05	0.82	1.75
Naumiestis-1	S1ln	1963.4	0.96	0.97	1.43	1.48	1.61
Ramučiai-2	S1ln	2010.0	1.46	1.75	2.01	1.82	3.42
Šilutė-1	S1w	1956.4	1.41	1.77	1.95	1.54	2.98
Šilutė-1	S1ln	1990.6	1.68	1.60	1.70	1.46	2.12
Usėnai-3	S1ln	1937.1	0.83	0.79	1.02	1.03	1.72

CONCLUSIONS

The Lower Silurian shales consist of 37% to 57% clay minerals and averages 40-50%. Diagenetic illite is the predominant mineral and chlorite is the second most abundant clay mineral. Illite-smectite (mixed layer) and kaolinite minerals are subordinate minerals. The total amount of quartz and feldspars is less than that of clay minerals and on average reaches 35-45%. In studied rocks, the amount of calcite is higher than that of dolomite, although an inverse relationship was observed in some shale intervals. This determines the favourable operational (petrophysical) properties of Lower Silurian shale in Western Lithuania.

Using gamma, electrical resistance, and acoustic wave well log data the distribution of organic matter model was greatly improved. The layer most enriched with organic matter is associated with the 4–10 m thick Raikküla regional stage. A second viable area has been identified in the Jaani regional stage.

Studies of samples from the Lower Silurian shale show that the total organic matter (TOC) content is about 1-2% (except for the anomalous Raikküla layers (up to 21%)). Interpretation of the well log data revealed significantly higher values of TOC concentration in the whole section of the lower Silurian clay column, the TOC content in places exceeds 4-5% and reaches up to 8.9%, which is well above the operational limit concentration of 2%. The saturation of organic matter in clayey rocks is closely related to the spatial variation of Lower Silurian thicknesses - it is concluded that the intensity of its accumulation was determined by the roughness of the basin bottom and increased shale gas productivity is predicted in paleo-pits.

The porosity of clayey rocks of the Lower Silurian decreases with increasing depth of deposition from ~ 15% at a depth of 1.5 km to 2-3% at a depth of ~ 2 km. In most of the territory of Western Lithuania, the porosity is comparable to sedimentation basins, where intensive shale gas exploitation is carried out.

The mineralogical brittleness index (MBI) of the studied rocks (important in predicting the efficiency of hydraulic fracturing) is slightly below the critical limit of 0.40, considering clay shale as a potential formation for gas extraction. The average brittleness of the rocks is related to the plastic components, i.e., abundance of clay minerals and organic matter. Meanwhile, the well log brittleness index (LBI) shows that the brittleness of the rocks exceeds the minimum threshold and the shales of the Lower Silurian can be considered as a formation viable for operation.

The cation exchange capacity of clayey rocks in the Lower Silurian was assessed as very low. The low values are explained by the high content of detrital grains and the predominant illite and chlorite minerals in the clay fraction. The resistance of clayey rocks to hydraulic erosion is positively correlated with the cation exchange capacity. It is considered to be average, which is important in ensuring the sustainability of the hydraulic fracturing zone. The 7% KCl solution reduces the sensitivity values approximately twice compared to distilled water (only expensive organic products have better properties) and is recommended for the practical hydraulic fracturing of Lower Silurian shale.

In general, the investigated petrophysical (and mechanical) properties of Lower Silurian shales can be stated that western Lithuania has good performance properties of these rocks, which were primarily determined by the specific mineralogical composition of the rocks.

LIST OF PUBLICATIONS

Scientific articles indexed in the Scientific Information Institute database "CA Web of Science":

Šliaupa S., Lozovskis S., Lazauskienė L., Šliaupienė R. 2020. Petrophysical and mechanical properties of the Lower Silurian perspective oil/gas shales of Lithuania. Journal of Natural Gas Science and Engineering, 79: Art. no. 103336;

Šliaupa S., Lazauskienė L., Lozovskis S., Šliaupienė R. 2020. Distribution of organic matter and evaluation of brittleness index of the Lower Silurian shales of west Lithuania based on interpretation of well logs. Baltica, 33 (2): 146-165.

Other publications:

Lozovskis S., Šliaupa S, Lazauskienė L., Šliaupienė R. 2020. Lithuanian Lower Silurian gas shale prospects - geological assessment. Geologijos akiračiai, 3-4: 25-36.

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Research projects

2015-2020 “Petrography and petrophysical properties of the Lower Silurian shales of West Lithuania – geological assessment of gas shale prospects”, Vilnius university, Lithuania.

2015-2019 “Influence of mudrocks properties on the accumulation and possible use of unconventional hydrocarbons”, Lithuanian Geological Service, Lithuania.

2015-2017 “European Unconventional Oil and Gas Assessment” coordinated by the Joint Research Centre of the European Commission - EUOGA (European Unconventional Oil and Gas Assessment), Lithuanian Geological Survey, Lithuania.

2013-2016 “Classification and management of geophysical data (geophysical research in wells and electrical exploration)”, Lithuanian Geological Survey, Lithuania.

NOTES

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