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Petrographic characteristics of the Ordovician and Silurian deposits in the Baltic Basin (N Poland) and their relevance for unconventional hydrocarbon accumulations

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The Ordovician and Silurian deposits in the Baltic Basin are represented by siltstones, mudstones, limestones, marls, sandstones and, most commonly, claystones with a markedly shaley structure. Claystones that are particularly rich in organic matter are considered as potential reservoirs for shale gas accumulations. Samples from 11 boreholes, including core samples of the Piaśnica, Słuchowo, Kopalino, Sasino, Prabuty, Jantar, Pelplin and locally Pasłęk, Kociewie and Puck formations, were analysed by optical microscopy, cathodoluminescence (CL), scanning electron microscopy (SEM) and energy-dispersive spectrometry and X-ray diffraction analyses (XRD). The Piaśnica, Sasino, Jantar, Pasłęk and Pelplin formations are characterized locally by a dominance of black bituminous claystones with a high content of organic matter, and a generally consistent clay mineral composition. Individual samples only show variations in silt fraction and carbonate content. Very important is the negligible proportion of swelling minerals in the clay fraction; the percentage of smectite in the mixed-layered illite/smectite minerals does not exceed 15%. It is important for the rocks to have adequate [SiO_2 >10%; Quartz (Q) + Feldspars (Fs) + Carbonates >40%] brittleness. The content of quartz, feldspars and carbonates (here >40%) suggests that the claystones are susceptible to hydraulic fracturing. The claystones show micropores between detrital grains, between the flakes of clay minerals, within pyrite framboids, and organic matter and secondary micropores within grains.

Key words: petrography, mineralogy, shale-type accumulations, total organic carbon, Ordovician and Silurian rocks, Baltic Basin.

INTRODUCTION

The crystalline basement of the East European Craton in Poland is overlain by a thick Ediacaran–lower Paleozoic sedimentary succession. The lower Paleozoic shales deposited in the Baltic Basin and the Podlasie–Lublin Basin, or alternatively the Baltic-Podlasie-Lublin Basin (Poprawa, 2010) form an undeformed to weakly deformed 700 km long belt situated along the western slope of the East European Craton (EEC) in Poland.

The Ordovician and Silurian deposits of the Baltic Basin (*sensu* Jaanusson, 1976) that extend along the south-western rims of the EEC in Poland, southern Sweden, the Baltic states, Belarus and Ukraine are developed as various facies belts and represent a wide spectrum of environments from the edge of the shelf in the west part, through neritic environments, to littoral ones in the east (Jaanusson, 1976; Nestor and Einasto, 1997; Jaworowski, 2000; Lazauskienė et al., 2003; Modliński, 2010; Radkovets, 2015).

Areas with the highest potential for hydrocarbon exploration and production among unconventional reservoirs in Poland are related to the occurrence of Cambrian, Ordovician and Silurian fine clastic deposits (Poprawa, 2010, 2020; Papiernik et al., 2019; Podhalańska et al., 2020). Potential source rocks and hydrocarbon occurrences in Poland have been the subject of geological and geochemical research from the early 1990s. The most intense research was concentrated between 2010 and 2015 and was related to the availability of new data from new boreholes drilled by oil companies.

Because of their elevated total organic carbon (TOC) content, the Furongian, Ordovician and Silurian mudstones are potential reservoirs for unconventional hydrocarbon accumulations and have become the subject of intense multidisciplinary research, aimed at evaluating their potential for hydrocarbon generation and accumulation (i.a., Poprawa and Kiersnowski,

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2008; Karnkowski et al., 2010; Kosakowski et al., 2010, 2016; Więcław et al., 2010; Matyasik and Słoczyński, 2010; Poprawa, 2010, 2020; Labuda, 2015). Based on strictly determined criteria, prospective zones for the occurrence of unconventional hydrocarbon deposits have been identified in the lower Paleozoic shale succession (Podhalańska et al., 2020).

These potential source rocks were studied by the current authors in 2013–2018 to determine rock successions with the most favourable parameters for hydrocarbon accumulation (Podhalańska et al., 2016a, b, 2018, 2020; Sikorska et al., 2016).

We describe the petrographic characteristics of rocks representing Ordovician and Silurian formations in the Baltic Basin of Poland. Previously published petrographic and mineralogical data are complemented by quantitative XRD, cathodoluminescence and scanning electron microscopy data. The laboratory data is combined with analysis of geological documentation of boreholes drilled by the Polish Geological Institute – NRI. The results of previous petrographic studies conducted by Langier-Kuźniarowa (i.a., 1967, 1971, 1982, 2015) and Sikorska et al. (2016) were also incorporated. The study by Radkovets et al. (2017) contains a petrographic and mineralogical analysis of the organic-rich Silurian black shales of Western Ukraine, which were deposited in the same sedimentary basin as the strata of the Baltic Basin studied. Our research focused on the western and central parts of the Baltic Basin, where clay facies dominated during sedimentation. Siltstones, limestones, marls and sandstones were examined as well as the dominant shales. Particular attention was paid to the shale formations considered to be the most prospective for unconventional hydrocarbon reservoirs, considering whether the petrographic features of the strata analysed affect oil and gas prospectivity.

GEOLOGICAL AND DEPOSITIONAL BACKGROUND

Descriptions of Baltic Basin geology and evolution have been published previously, e.g. by Poprawa et al. (1999), Lazauskienė et al. (2002, 2003), Karnkowski et al. (2010), Modliński and Podhalańska (2010), Mazur et al. (2018) and recently in detail by Poprawa (2020). In this contribution, we briefly describe the most significant geological events and processes that affected the marginal part of the East European Craton, and controlled sedimentation, including of hydrocarbon source rocks.

The Cambrian, Ordovician and Silurian deposits in the Baltic Basin are part of an extensive sedimentary succession that was deposited in a pericratonic marine basin developed in early Paleozoic times on the southwestern slope of the Baltica palaeocontinent, located at that time at the medium and low southern latitudes (Cocks, 2000; Golonka et al., 2019; Fig. 1). This was a vast epicontinental sea where carbonate, marly, clayey and silty sediments were deposited. It covered today's Baltic countries and the Baltic Sea, reaching southern Sweden, northern and eastern Poland, and western Ukraine. The uppermost Ediacaran and lower Paleozoic deposits of the basin are in contact with tectonically deformed Ordovician and Silurian deposits of the Koszalin–Chojnice Zone along the faults of the Teisseyre-Tornquist tectonic zone (TTZ; Znosko, 1997; Dadlez, 2000). The eastern limit of the basin coincides with the natural

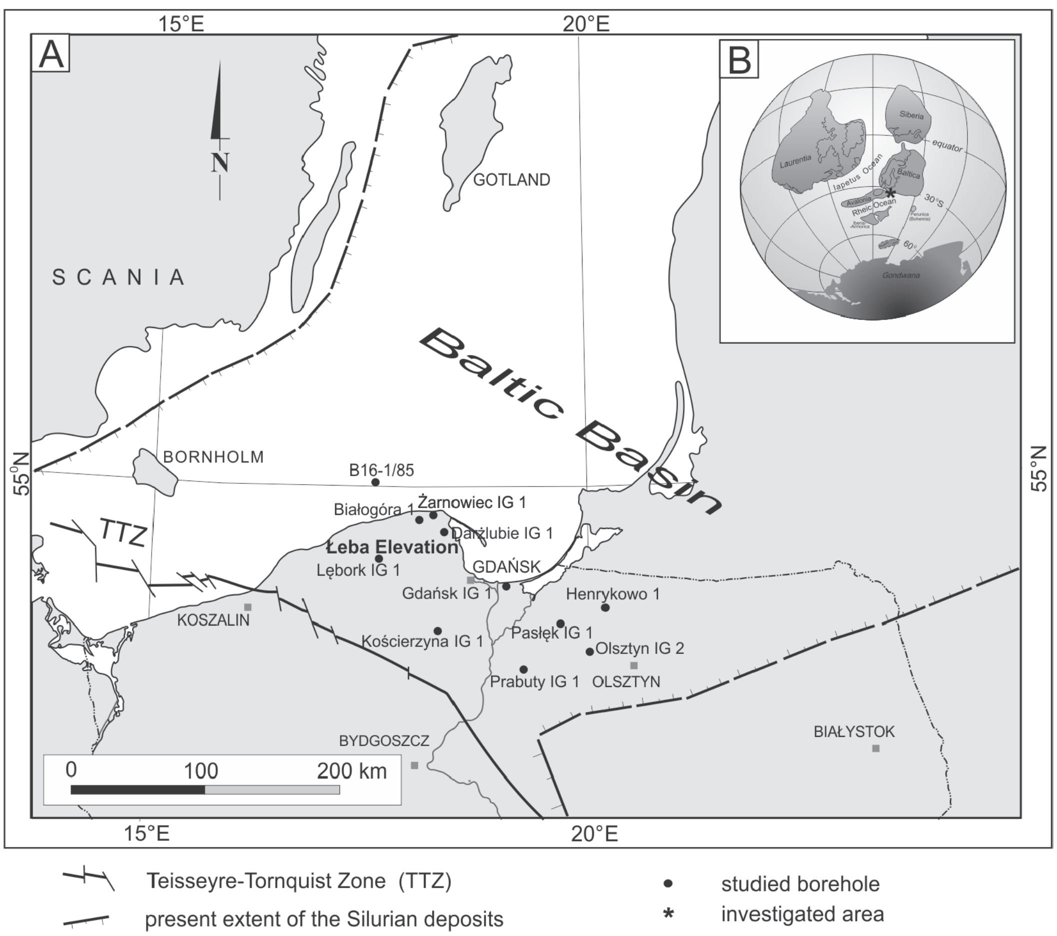


Fig. 1A – locality map of the boreholes investigated; B – palaeogeographical map of the Late Ordovician/Early Silurian after Cocks (2000)

boundaries of Silurian nearshore facies (Paškevičius, 1997; Lazauskiene et al., 2003; Modliński, 2010; Radkovets, 2015).

The formation of the Baltic–Podlasie–Lublin Basin and the early Paleozoic deposition were associated with the late Neoproterozoic break-up of the Rodinia/Pannotia supercontinent (Poprawa et al., 1999; Jaworowski, 2002; Jaworowski and Sikorska, 2003; Poprawa et al., 2020). After a period of terrestrial sedimentation in the late Ediacaran and early Cambrian (Jaworowski and Sikorska, 2003), there was a change to transitional and marine deposition, and the course of sedimentation was dependent on transgressive-regressive cycles in the basin (Jaworowski, 2002; Modliński and Podhalańska, 2010). Sedimentation of marine siliciclastic deposits continued until the middle Cambrian (depositional sequence I, Jaworowski, 2002). Marine deposits of the middle Cambrian, Furongian and Lower Ordovician (Tremadocian) represent depositional sequence II (Jaworowski, 2002). The occurrence of Furongian and lower Tremadocian deposits in the Baltic area is limited to a small area of the Łeba Elevation and the maritime economic zone of the Baltic Sea.

The geotectonic conditions in the Baltic–Podlasie–Lublin Basin changed in the Middle/Late Ordovician. As a result of the oblique collision of Avalonia with Baltica, this area became the foredeep basin of an approaching orogen (Poprawa et al., 1999; Poprawa, 2020). Due to flexural bending of the Baltic crust, subsidence rate increased in the basin, reaching its maximum in the Ludlow and Pridoli (Silurian) (Poprawa et al., 1999; Jaworowski, 2000, 2002; Poprawa, 2020). The development of the basin in the marginal zone of Baltica was characterized by a zonal lithofacies pattern, with a westwards-increasing proportion of clastic deposits and an eastwards-increasing proportion of carbonates (e.g., Paškevičius, 1997; Modliński, 2010; Modliński and Podhalańska, 2010; Porębski et al., 2013; Radkovets, 2015; Poprawa, 2020). This trend persisted, especially in the Ordovician. In the Silurian, the migration of the Caledonian collision front was the main determinant of sediment deposition. Initially, deposition of claystones and mudstones (Jantar and Pasłęk formations) took place on an open shelf, though with a limited supply of clastic material and under conditions of very low oxygenation of both the bottom waters and the sea floor. Oxygenation in the basin was variable throughout the Llandovery and Wenlock (Pasłęk and Pelplin formations); however, the supply of siliciclastic material systematically increased, expanding the zone of claystone-mudstone-siltstone facies towards the east (Modliński, 2010; Porębski et al., 2013; Poprawa, 2020). The supposed source area of clastic material is the Caledonian accretionary prism that formed along the collision zone of Baltica with Avalonia (Poprawa et al., 1999; Jaworowski, 2000; Poprawa, 2006; Porębski et al., 2013), while the carbonate material was derived from the marginal zone and shallows in the basins. The lower Paleozoic section studied is incomplete. Part or all of Pridoli deposits, and locally also the upper part of the Ludlow, are missing as a result of erosion related partly to isostatic post-collisional uplift (Poprawa, 2020).

The presence and migration of depositional environments in the sedimentary basin conducive to the deposition of fine-grained sediments enriched in organic matter was conditioned by both the tectonic evolution (e.g., Lazauskienė et al., 2003; Poprawa, 2020) and global climatic and eustatic changes in sea level along with their effects on biological productivity (Page et al., 2007; Armstrong et al., 2009; Loydell et al., 2009; Podhalańska, 2009; Cichon-Pupienis et al., 2020). Deposition of each shale formation of the lower Paleozoic unconventional hydrocarbon system in Poland – the Piaśnica Formation (Furongian–Lower Tremadocian), the Sasino Formation (Caradoc), the Jantar Formation (Llandovery), the lower part of the Pelplin Formation (Mingajny Formation, according to Poprawa, 2020) – coincides with rising or high global sea level.

The area investigated is located on the western part of the East European Craton, the basement of which gradually descends westwards and is located at depths ranging from several hundred meters in the east to ~5000 m in the west. The sedimentary cover of the basin lies horizontally or sub-horizontally and, as well as the uppermost Ediacaran to lower Paleozoic, comprises also Permian-Mesozoic and Cenozoic sedimentary successions. Knowledge of the lithology of Cambrian, Ordovician and Silurian strata in the Baltic Basin comes from a few tens of boreholes. The key boreholes for the western, distal part of the basin are Słupsk IG 1, Lębork IG 1 and Kościerzyna IG 1, while Darżlubie IG 1 is the key borehole for the Łeba Elevation. For the central and eastern parts of the Baltic Basin, the key boreholes include Pasłęk IG 1, Żelazna Góra 4 and Prabuty IG 1.

ANALYTICAL METHODS AND DATA

The research material (borehole core samples) was derived from 11 boreholes: B16-1/85, Białogóra 1, Darżlubie IG 1, Gdańsk IG 1, Henrykowo 1, Kościerzyna IG 1, Lębork IG 1, Olsztyn IG 2, Pasłęk IG 1, Prabuty IG 1 and Żarnowiec IG 1 (Fig. 1A) drilled in the Baltic Basin, located both in the onshore and offshore parts of the basin. Core samples of the Piaśnica, Słuchowo, Kopalino, Sasino, Prabuty, Jantar, Pasłęk, Pelplin, Kociewie and Puck formations were analysed (Fig. 2). Uncovered polished thin sections were prepared from 140 rock samples (stained blue with resin). Petrographic studies were based on observations in a polarizing microscope (PL) and enabled determination of microlithofacies (types of rocks) and their textural and structural features (e.g., grain size, sorting, porosity). In addition to standard microscopic examinations, 50 thin sections were examined under cathodoluminescence (cold cathode – *CCL 8200 mk3* Cambridge Image Technology). These allowed deciphering of the original features of the rock structure to better understand and interpret the diagenetic processes to which the deposits were subjected. In the case of very fine-grained rocks, the CL images revealed the presence of dispersed carbonates, phosphates, kaolinite and feldspars.

Dye analysis, using Evamy's solution, was also used to identify carbonates. These studies were supplemented with observations using a scanning electron microscope (SEM) LEO 1430 to visualize the pore space and, in particular, microporosity. The studies were carried out on rock chips (35) and thin sections (40). In doubtful cases, minerals were identified by chemical composition analysis (EDS) using an energy microprobe (*Oxford ISIS 300*). To describe the clay minerals, the results of X-ray diffraction analysis (XRD), performed at the Oil and Gas Institute – National Research Institute in Kraków, were used (*X'Pert MPD Philips* diffractometer). These data were also used to determine the rock brittleness as a crucial parameter for shale susceptibility to fracture propagation in hydraulic fracturing (Andrews, 2013, 2014; Dyrka, 2016). A summary of the analyses is given in Table 1.

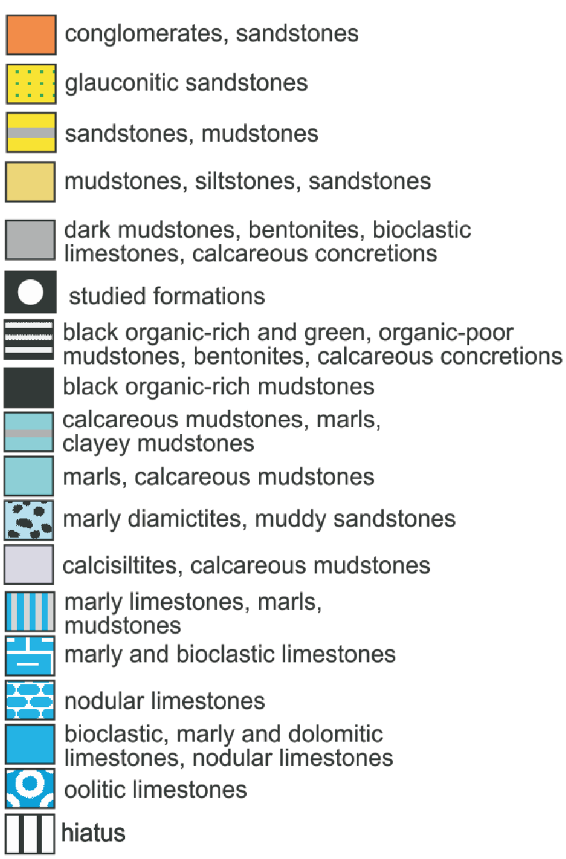
Uneven sampling of the rocks from the different formations resulted from varying availability of core material.

Table 1

List of petrographic and mineralogical analyzes made on selected formations

Age	Formation	Number of analyses			
		PL	CL	SEM	XRD
SILURIAN	Puck	2	2	1	1
	Kociewie	16	4	6	16
	Pelplin	70	36	17	69
	Pasłęk	24	11	6	28
	Jantar	2	1	1	2
ORDOVICIAN	Prabuty	2	1	–	2
	Sasino	18	13	4	22
	Kopalino	2	1	–	–
	Słuchowo	1	1	1	2
	Piaśnica	3	1	–	–
		140	71	36	205

PL – polarizing microscope, CL – cathodoluminescence, SEM – scanning electron microscope, XRD – X-ray diffraction analysis



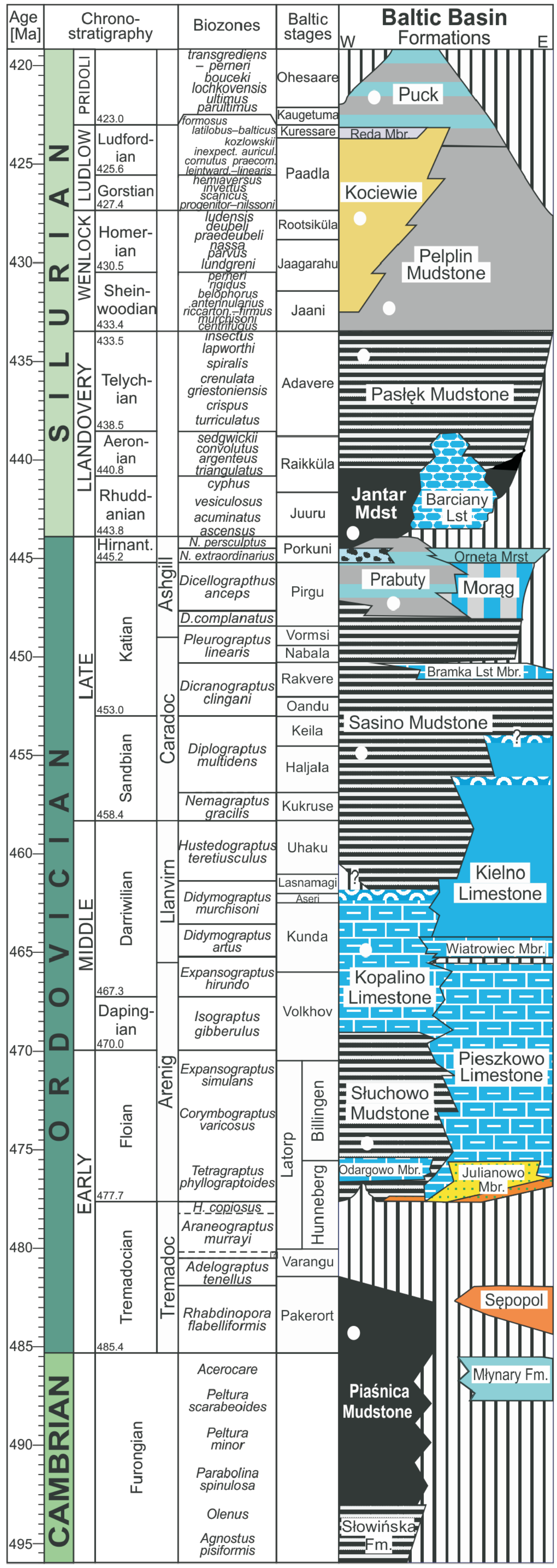
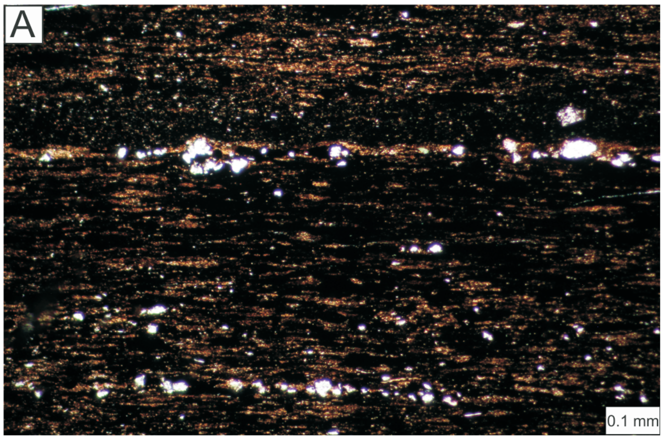


Fig. 2. Lithostratigraphic subdivision of the Ordovician and Silurian succession in the part of the Baltic Basin studied (after Porębski and Podhalańska, 2019), Ordovician–Silurian chronostratigraphy, graptolite zonation and correlation of the Ordovician and Silurian standard stages to the Baltic regional stages after Cooper et al. (2012) and Melchin et al. (2012). The standard graptolite zonation is modified to include the local zones of Urbanek and Teller (1997) and Porębska et al. (2004)

Hinant. – Hirnantian, Lst – Limestone, Mdst – Mudstone, Mbr. – Member, Fm. – formation



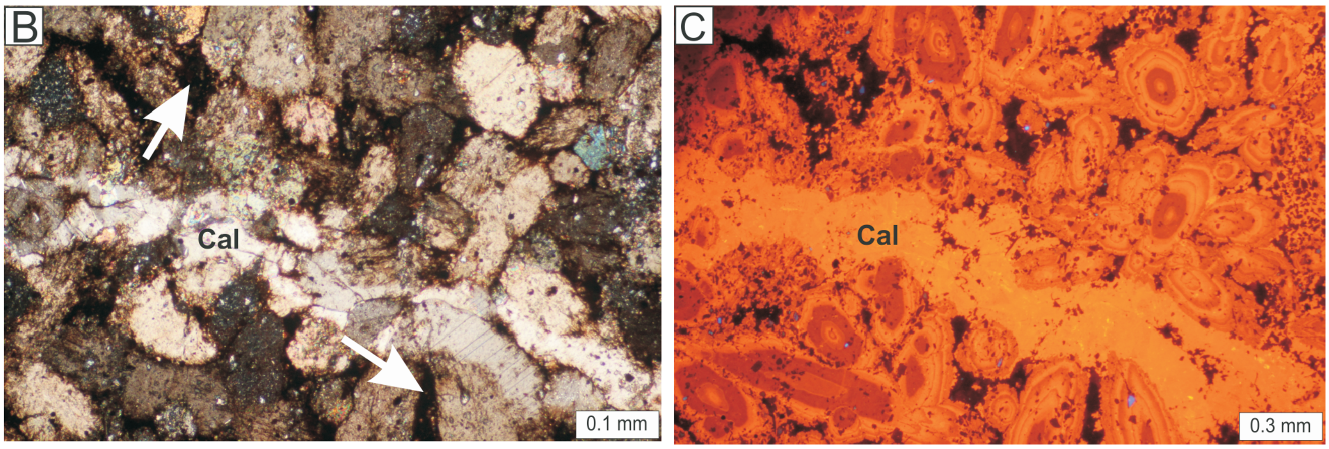


Fig. 3. Photomicrographs of rocks from the Piaśnica Formation

A – horizontally laminated black bituminous claystone, Żarnowiec IG 1 borehole, depth 2725.6 m (without analyzer); **B**, **C** – sparitic limestone with numerous bitumen accumulations (arrowed), sparite grains show zonal structure in CL; Cal – calcite vein, B16-1/85 borehole (offshore), depth 1860.4 m (B – crossed polars; C – CL image)

RESULTS

PIAŚNICA FORMATION

The Piaśnica Formation is composed of black claystones with interbeds and lenses of dark grey limestone and numerous pyrite concretions. Sandstones are a minor lithology found in the bottom part of the formation. According to Sikorska and Jaworowski (2007) the deposits accumulated in a euxinic marine basin and probably on a distal carbonate ramp.

The claystones are characterized by horizontal lamination and are strongly bituminous (Fig. 3A). They consist of a mixture of clay minerals with an admixture of terrigenous material (quartz and feldspars visible in CL), pyrite, carbonates, phosphates and abundant organic matter. Carbonates (calcite, minor dolomite) occur in the form of very small, dispersed concentrations, or fill microfractures. The claystones contain channel (Sikorska et al., 2016). The limestones are represented mainly by sparites and biosparites (calcitic remains of trilobites and brachiopods), less often by microspar. Accumulations of bitumens occur between sparite grains; some of the grains show feather structure (Fig. 3B, C). The limestones occasionally contain psammitic quartz detritus and scattered phosphatic shells. In places, they are fractured and re-cemented with calcite (Fig. 3C).

SŁUCHOWO FORMATION

The Słuchowo Formation is composed of black and dark grey bituminous clays, laminated in places, and in which very fine grains of blue-luminescent potassium feldspar can be seen. XRD analysis (Table 2) of a single selected sample shows that the average clay mineral content is 25.5%, and the sum average of quartz, feldspars and carbonates is 26.1%. A characteristic feature of the claystones is the occurrence of laminae enriched in mud-sized material with greenish glauconite grains and with phosphates. Parts of the glauconite grains are phosphatized (Fig. 4A, B). The claystones also contain barite aggregates with numerous pyrite crystals.

In electron microscope observations, the claystones show microporosity between clay mineral flakes. The pores are isometric (~4 µm) and canal-shaped, with an average length of ~12 µm.

KOPALINO FORMATION

The Kopalino Formation is represented almost entirely by various types of limestone, occasionally with interbeds of clay rocks and scour marks. The dominant rock type is organodetrital limestone (mainly with trilobite and brachiopod debris), grey and brown in colour, spotty in places due to uneven pigmentation with iron compounds. Carbonate clasts are also present in biosparites. There are numerous pelitic and marly limestones in the Kopalino Formation. Fractures in the limestones and the interiors of some bioclasts are filled with secondary calcite or ankerite (Fig. 5). Yellow-orange-luminescent calcite predominates in CL images, while ferruginous orange-brown-luminescent calcite is rarely present. Phosphates (lilac in CL) occur sporadically.

SASINO FORMATION

The Sasino Formation is represented by black and black-grey claystones (Fig. 6), silty clays, calcareous clays and clayey limestones, frequently with faunal debris and an admixture of mud and tuffite. The detrital material includes fine grains

Table 2

Mineralogical composition of the rocks of the Ordovician and Silurian formations, determined by XRD [in %]

Mineral composition	ORDOVICIAN			SILURIAN				
	Formation							
	Słuchowo	Sasino	Prabuty	Jantar	Pasłęk	Pelplin	Kociewie	Puck
	Number of samples							
	2	21	2	4	27	65	16	1
Quartz	16.0–21.0	12.3–49.9	22.3–27.4	16.6–29.7	14.2–39.7	18.7–30.8	10.5–25.7	18.3
Quartz av.	**18.5**	**31.3**	**24.8**	**21.58**	**22.6**	**24.2**	**17.9**	
Plagioclase	1.2–2.6	2.1–5.7	2.2–4.9	2.0–4.0	0.0–6.8	0.0–19.7	3.5–6.7	4.8
Plagioclase av.	**1.9**	**3.4**	**3.6**	**2.83**	**4.2**	**7.0**	**5.2**	
K-feldspar	1.2–9.6	0.0–4.9	1.7–2.7	1.1–2.8	0.0–5.5	0.0–13.4	0.7–1.9	1.2
K-feldspar av.	**5.4**	**2.1**	**2.2**	**1.98**	**2.5**	**2.7**	**1.4**	
Calcite	0.0	0.0–33.0	0.7–2.7	0.0–0.4	0.0–44.0	0.4–28.4	1.4–47.5	9.0
Calcite av.	**0.0**	**5.7**	**1.7**	**0.1**	**6.7**	**5.3**	**12.1**	
Dolomite/ankerite	0.0–0.6	0.0–11.3	1.1–1.5	0.5–5.0	0.0–9.2	0.0–10.3	1.8–5.9	5.0
Dolomite/ankerite av.	**0.3**	**3.1**	**1.3**	**2.28**	**3.3**	**5.5**	**3.3**	
Hematite	0.0	0.0	0.0	0.0	0.0–5.8	0.0	0.0	0.0
Hematite av.	**0.0**	**0.0**	**0.0**	**0.0**	**0.2**	**0.0**	**0.0**	
Sulphides	16.2–31.8	0.0–43.4	2.9–3.6	0.0–22.3	0.0–9.0	0.0–8.1	0.0–4.7	2.3
Sulphides av.	**24.0**	**7.53**	**3.3**	**6.4**	**2.5**	**4.2**	**2.4**	
Sulphates	0.0–0.9	0.0–1.7	0.0	0.0–2.1	0.0–1.3	0.0–7.2	0.0–3.2	0.0
Sulphates av.	**0.45**	**0.55**	**0.0**	**0.53**	**0.1**	**1.0**	**0.6**	
Mica+illite	32.8–48.8	22.9–48.3	47.0–50.5	41.3–51.5	0.0–49.6	0.0–46.2	20.1–47.9	47.2
Mica+illite av.	**20.5**	**33.0**	**48.8**	**44.25**	**37.0**	**35.0**	**39.0**	
Illite/smectite	2.6–3.6	1.6–9.4	1.1–2.5	0.5–10.1	0.0–11.5	0.5–33.0	1.6–5.0	3.3
Illite/smectite av.	**3.1**	**4.3**	**1.8**	**4.6**	**4.7**	**3.6**	**2.3**	
Chlorite	0.8–3.0	3.1–10.0	8.6–9.7	8.0–18.7	0.0–22.5	1.9–20.1	6.3–21.5	8.9
Chlorite av.	**1.9**	**6.8**	**9.2**	**13.35**	**11.1**	**8.8**	**14.7**	
Kaolinite	0.0	0.0–1.8	0.0	0.0–1,9	0.0–2.1	0.0–5.1	0.0–1.4	0.0
Kaolinite av.	**0.0**	**0.6**	**0.0**	**0.48**	**0.7**	**0.8**	**0.3**	
clay minerals	25.5	44.8	59.7	62.7	53.6	48.3	56.4	59.4
quartz, feldspars, carbonates	26.1	45.6	33.6	28.8	39.3	44.6	39.9	38.3
Average smectite content in the illite/smectite								
Number of samples	–	5	1	1	3	12	5	–
Smectite	–	<15	<15	~11	~11	10 – 15	<10	–

av. – average

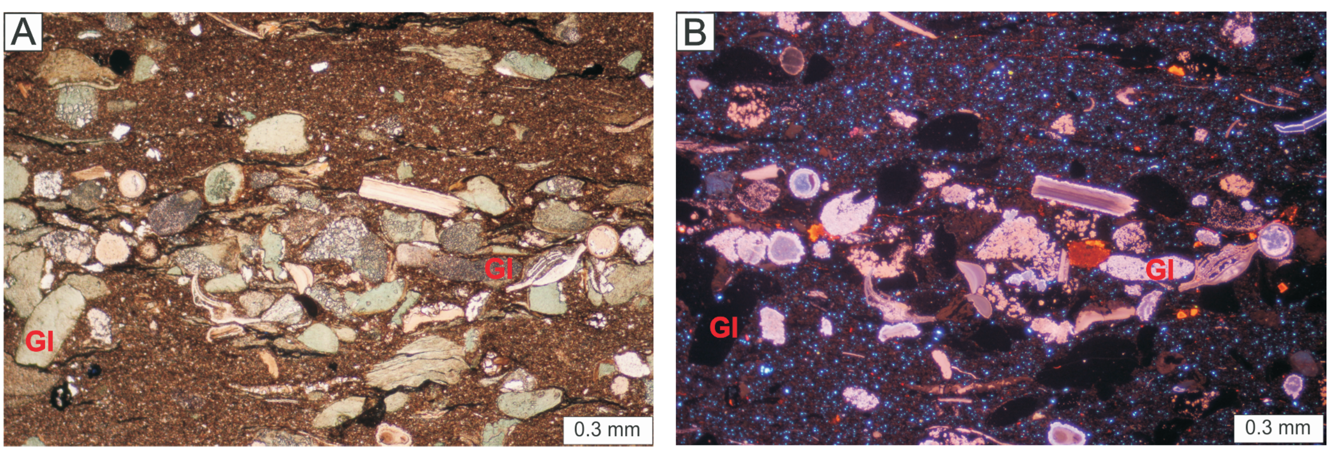


Fig. 4. Photomicrographs of rocks from the Słuchowo Formation

A, **B** – claystone with a glauconite-containing lamina (Gl); **B** – very fine, blue-luminescent feldspar grains, non-luminescent glauconite (Gl) and phosphatized milky pink glauconite grains are visible; Białogóra 1 borehole, depth 2701.5 m (A – without analyzer, B – CL image)

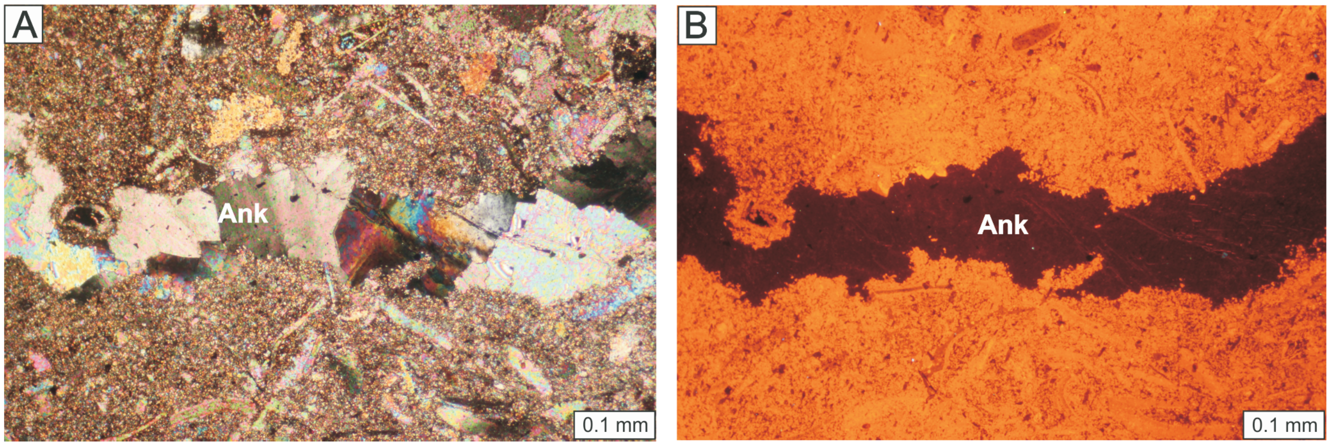


Fig. 5. Photomicrographs of biosparite from the Kopalino Formation

A, **B** – limestone cut by an ankerite vein (Ank), Pasłęk IG 1 borehole, depth 2720.8 m (A – crossed polars, B – CL image)

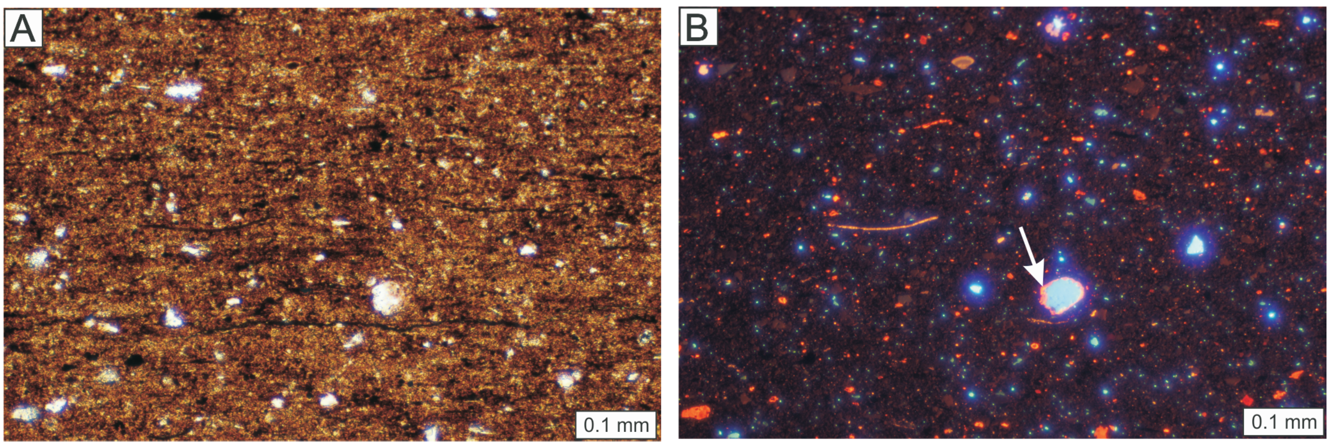


Fig. 6. Photomicrographs of rocks from the Sasino Formation

A, **B** – claystone showing directional structure and containing a small admixture of mud; blue-luminescent feldspar grains, greenish-luminescent plagioclase grains, and orange-luminescent fine carbonate grains, a large feldspar grain is partly replaced by calcite (arrowed), Białogóra 1 borehole, depth 2642.8 m (A – without analyzer, B – CL image)

of quartz and potassium feldspar (blue CL), plagioclase (green CL), mica (muscovite) and apatite.

Many feldspars have been partly calcitized, as seen in CL images (Fig. 6B). The pelite fraction consists of clay minerals with an admixture of carbonate micrite and/or organic matter and iron compounds. The results of the mineral composition analysis of the clay fraction (Table 2) show the presence of illite, illite/smectite mixed-layered minerals (<15% smectite content), chlorite and kaolinite. XRD analyses show that the average clay content is 44.8%. In addition, the rocks contain rhombohedral dolomite crystals, numerous pyrite aggregates and frequent inclusions of organic matter. Apart from micrite and carbonate microspar, interbeds of clayey limestones contain numerous bioclast fragments. Carbonates are represented by calcite, Fe-dolomite, dolomite and ankerite (Appendix 1* and Fig. 7).

XRD analyses show that the average sum of quartz, feldspars and carbonates is 45.6%. In places, there are larger concentrations of phosphates, and accessory anhydrite or celestine. Microporosity was observed between the grains (Fig. 8) and between the clay mineral flakes. The size of isometric pores is ~3.19 μm, and the length of channel micropores is ~8.67 μm.

PRABUTY FORMATION

The Prabuty Formation is lithologically variable and represented by alternating beds of marly limestone, claystone, micrite limestone and marl, generally grey in colour, in places colour-mottled (brown/cherry). Sandstones, limestones with glauconite, clay laminae containing glauconite, sand-silt quartz detritus, partly carbonatized feldspars, pyrite and bioclasts are found at the top (Fig. 9A). The claystones are dark brown in colour and show a directional structure emphasized by parallel arrangement of clay minerals and streaks of organic matter with pyrite pigment. XRD analyses (Table 2) show that the average clay content is 59.7%. Illite, illite/smectite (<15% of smectite) and chlorites have been identified among the clay minerals. The claystone texture is pelitic, in places pelitic-silty, with a

* Supplementary data associated with this article can be found, in the online version, at doi: 10.7306/gq.1613

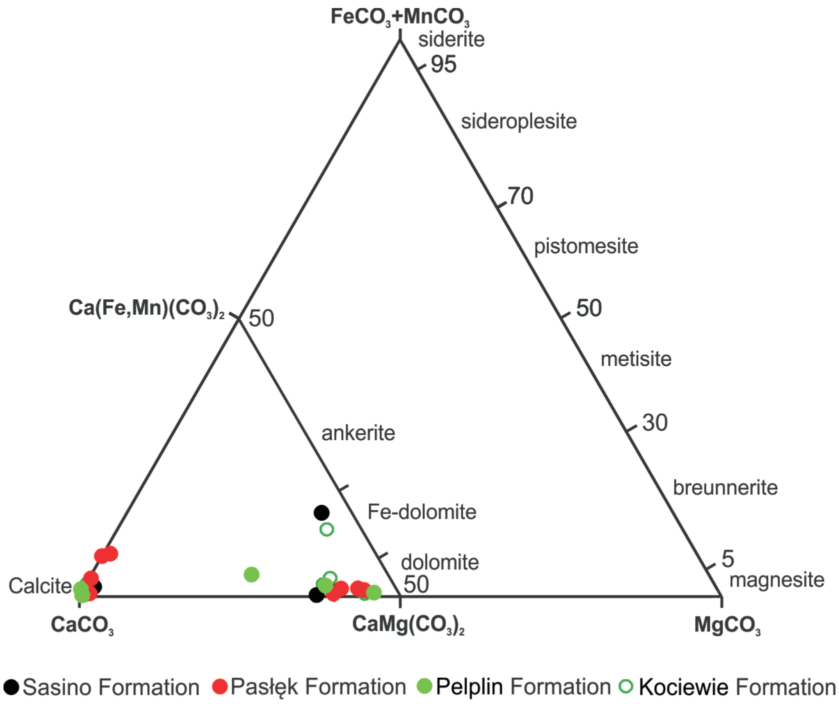


Fig. 7. Triangular diagram of the chemical composition (mol%) of the carbonates

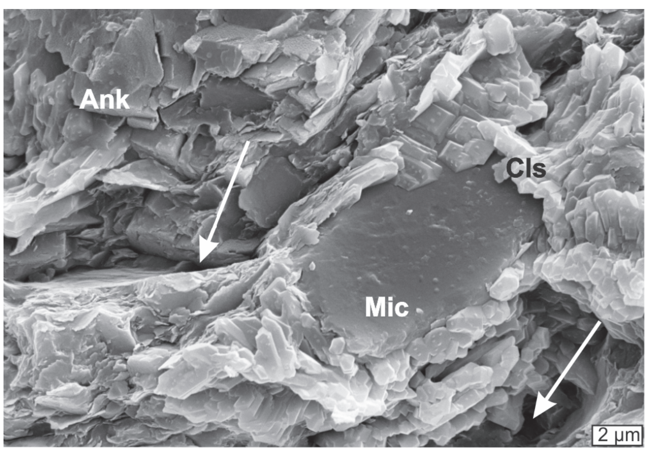


Fig. 8. Claystone from the Sasino Formation

Mic – mica, Ank – ankerite, Cls – celestine grains and intergranular porosity (arrowed) are visible, Białogóra 1 borehole, depth 2657.8 m (SEI image)

small admixture of mainly potassium feldspar grains. There is also an admixture of carbonate micrite in the form of fine anhedral grains. XRD analyses (Table 2) show that the average sum of quartz, feldspars and carbonates is 33.6%.

The micritic limestones and marls contain recrystallized shell fragments composed of calcite ranging in CL colours from yellow to orange and brown. They also contain dispersed detrital material represented by fine quartz and feldspar grains (Fig. 9B). Fractures in these rocks are filled with calcite.

JANTAR FORMATION

The Jantar Formation is represented by black-brown claystones, mainly bituminous with numerous grains of pyrite, and locally by dark brown calcareous siltstones. The claystones show a streaky or laminated directional structure and a pelitic or pelitic-silty texture. The laminae and streaks are composed of organic matter and iron compounds or carbonate micrite (Fig. 10). An admixture of detrital material is represented by quartz grains and potassium feldspars (light blue-luminescent) and by muscovite and biotite flakes. Carbonate grains, mainly calcite, small pyrite aggregates and fine anhydrite crystals were also noted. The calcareous siltstone is composed of carbonate micrite, quartz grains and an admixture of clay minerals. It contains small concentrations of pyrite and admixtures of potassium feldspar. XRD analyses (Table 2) of selected claystone samples show the average content of clay minerals is 62.7%. The main clay mineral is illite. Chlorites occur in considerable amounts. Mixed-layer illite/smectite minerals (~11% of smectite) and trace amounts of kaolinite also occur. The claystones show microporosity between clay mineral flakes. The size of isometric pores is ~3.25 µm, while the length of channel macropores is usually >15 µm.

PASŁĘK FORMATION

Rocks of the Pasłęk Formation are represented by black-brownish claystones, and in places by dark brown calcareous siltstones. Limestone interbeds are locally found. There are also interbeds of bioturbated mudstone and claystone, greenish in colour. The mudstone-claystone rocks show random or directional structure, and are laminated. Laminae in the claystone are enriched in silty material or are of siltstone (Fig. 11). Detrital material of the silt fraction is represented by grains of quartz, potassium feldspar and plagioclase, with accessory apatite and potassium feldspars (Appendix 2) contain, on average, 63.8 wt.% SiO_2, 18.9 wt.% Al_2O_3, 16.1 wt.% K_2O, 0.7 wt.% Na_2O, 0.3 wt.% FeO, 0.1 wt.% TiO_2, 0.1 wt.% MnO. Plagioclases (Appendix 2) contain, on average, 66.9 wt.% SiO_2, 20.6 wt.% Al_2O_3, 0.1 wt.% K_2O, 11.2 wt.% Na_2O, 1.1 wt.% CaO, 0.1 wt.% FeO, 0.1 wt.% TiO_2. Moreover, there are also calcite grains and dolomite rhombohedra (Appendix 1 and Fig. 7). XRD (Table 2) analyses show that the average sum of quartz,

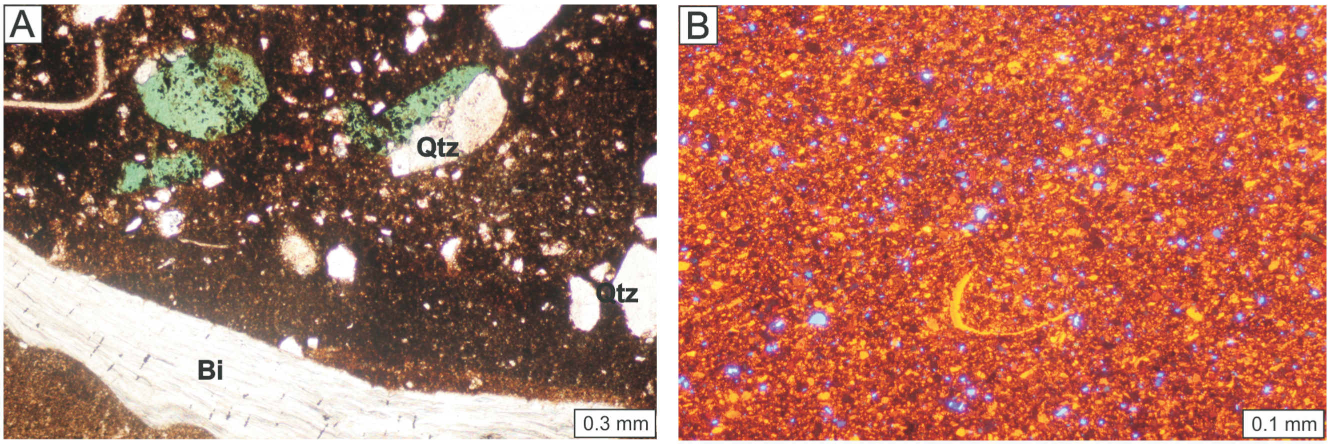


Fig. 9. Photomicrographs of rocks from the Prabuty Formation

A – part of clay lamina in marl, partly carbonatized green glauconite grains, quartz grains (Qtz) and bioclasts (Bi) are visible, Gdańsk IG 1 borehole, depth 3089.0 m (crossed polars); **B** – micrite limestone containing quartz and feldspar silt (blue CL) and a piece of shell visible in the centre (yellow-luminescent calcite), Prabuty IG 1 borehole, depth 3361.5 m (CL image)

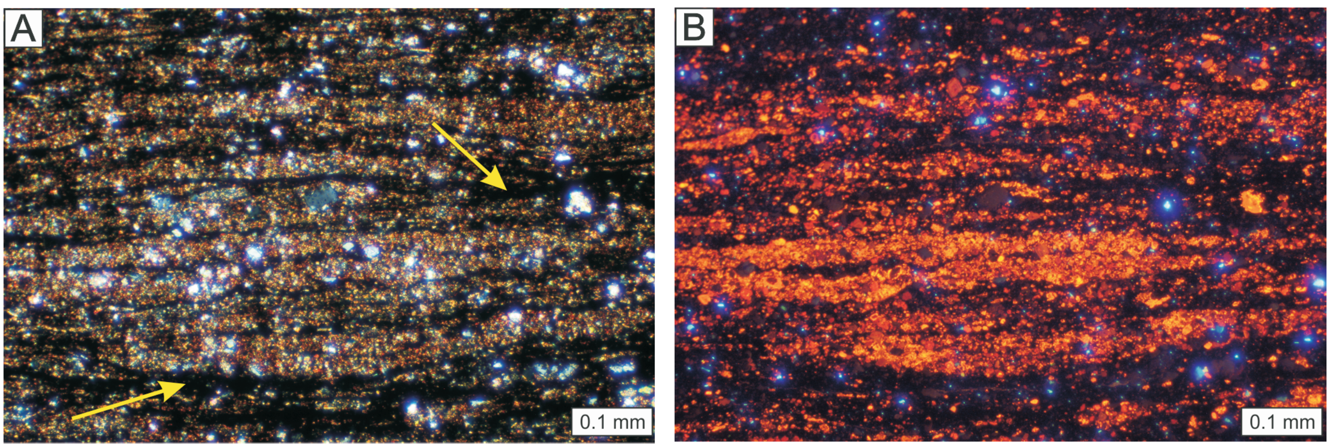


Fig. 10. Photomicrographs of rocks from the Jantar Formation

A – claystone with carbonate laminae and organic matter streaks (arrowed), with a small admixture of mud; **B** – blue-luminescent feldspar grains, orange-yellow-luminescent carbonates are visible in CL, Białogóra 1 borehole, depth 2613.9 m (A – crossed polars, B – CL image)

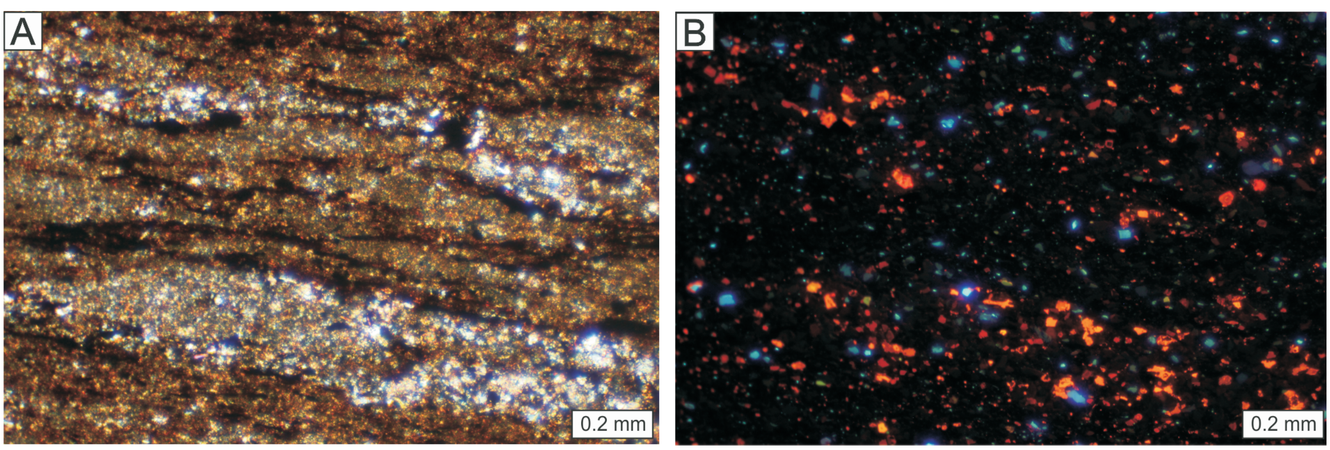


Fig. 11. Photomicrographs of rocks from the Pasłęk Formation

A – laminae and lenticular forms composed of siltstone; **B** – blue-luminescent K-feldspars, greenish-luminescent plagioclase grains, yellow-luminescent calcite, red-orange-luminescent dolomite are visible in CL, Olsztyn IG 2 borehole, depth 2354.9 m (A – crossed polars, B – CL image)

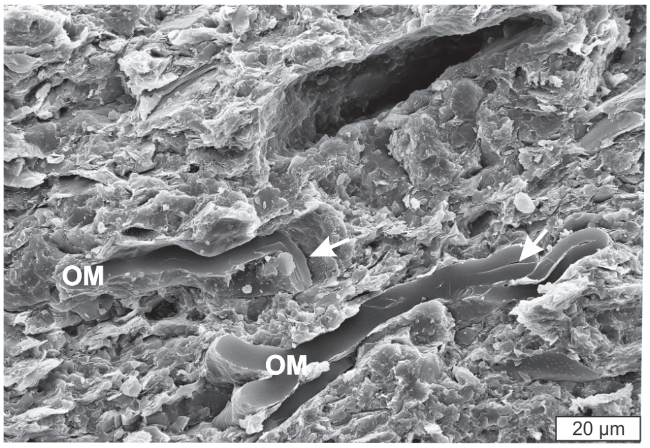


Fig. 12. Silty claystone from the Pasłęk Formation

Organic matter (OM) with visible microporosity (arrowed), Olsztyn IG 2 borehole, depth 2347.6 m (SEI image)

feldspars and carbonates is 42.3%. The pelitic fraction (Table 2) is represented by clay minerals (illite, illite/smectite, chlorite, kaolinite) and iron hydroxides. The content of smectite in the mixed-layer illite/smectite is ~11%. XRD analyses show that the average clay content is 53.6%.

There are also concentrations of organic matter (Fig. 12). Microporosity of organic matter is visible under a scanning electron microscope (Fig. 12), also between clay mineral flakes. The average diameter of isometric pores is ~2.5 µm, and the length of channel micropores is ~25 µm.

PELPLIN FORMATION

The Pelplin Formation is characterized by the presence of dark grey mudstones, commonly laminated, with graptolites and carbonate material, dispersed or as concretions (Fig. 13), as well as grey-brownish claystones.

They show a pelitic, locally pelitic-silty texture and a directional structure accentuated by parallel arrangement of clay minerals and mica flakes. The pelitic fraction (according to XRD analysis; Table 2) is represented by clay minerals (illite, mixed-layered minerals: illite/smectite, chlorites, and kaolinite). The content of smectite in the mixed-layer illite/smectite mineral

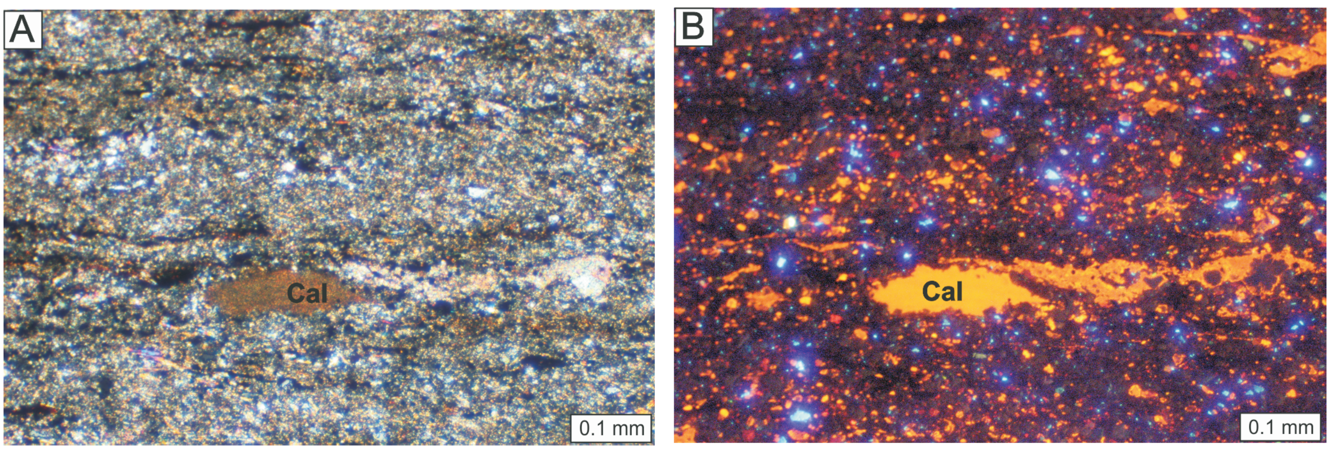


Fig. 13. Photomicrographs of rocks from the Pelplin Formation

A, B – claystone; orange-yellow-luminescent calcite laminae (Cal) are visible in CL, Białogóra 1 borehole, depth 2268.1 m (A – crossed polars, B – CL image)

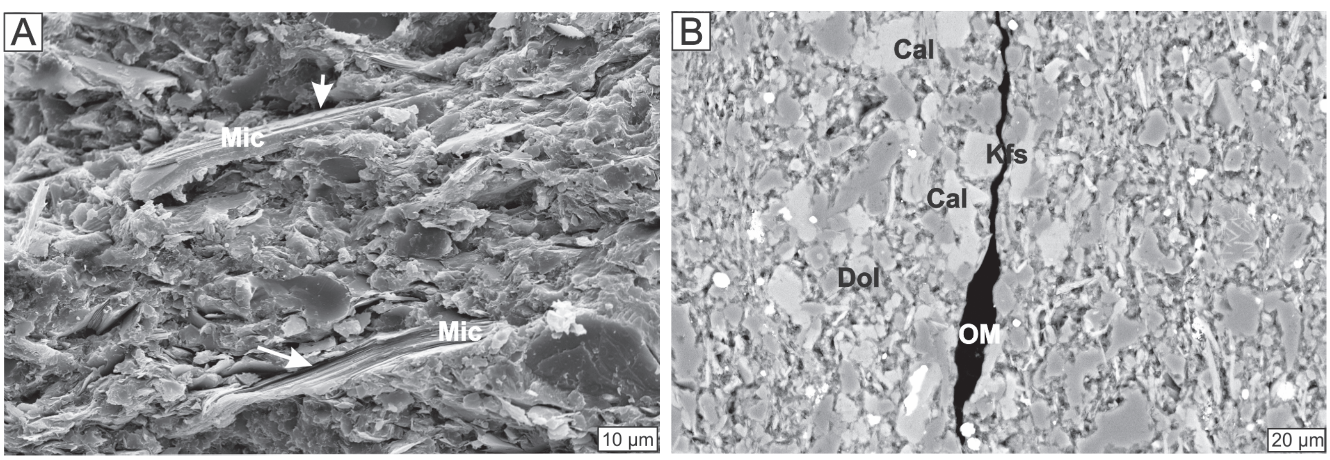


Fig. 14. Rocks from the Pelplin Formation

A – claystone, parallel arrangement of mica flakes (Mic) and microporosity (arrows) are visible, Olsztyn IG 2 borehole, depth 2208.3 m; **B** – fragment of calcareous claystone, grains of K-feldspar (Kfs), calcite (Cal), and dolomite (Dol), and organic matter (OM) are visible, Gdańsk IG 1 borehole, depth 2981.8 m (A – SEI image, B – BEI image)

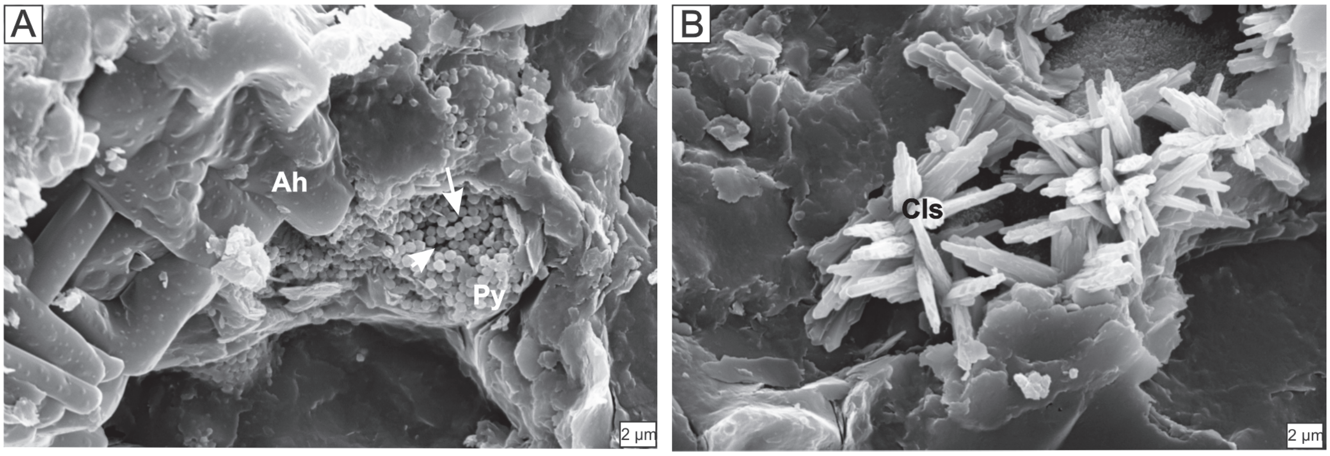


Fig. 15. Claystone from the Kociewie Formation

A – anhydrite crystals (Ah) and framboid pyrite (Py) with microporosity (arrows); **B** – celestine crystals (Cls), Lębork IG 1 borehole, depth 3056.4 m (SEI images)

ranges from 10 to <15%. The average clay mineral content is 48.3%. Carbonates occur as micrite and subhedral and anhedral crystals represented by calcite and dolomite, sporadically by ankerite (Appendix 1 and Figs. 7, 14B). An admixture of silt fraction consists of grains of quartz, K-feldspars and plagioclases (Appendix 2). Potassium feldspars contain, on average, 67.7 wt.% SiO_2, 18.5 wt.% Al_2O_3, 16.2 wt.% K_2O, 0.4 wt.% Na_2O, 0.1 wt.% FeO, 0.1 wt.% TiO_2, 0.1 wt.% MnO. Plagioclases contain, on average, 68.0wt.% SiO_2, 20.2 wt.% Al_2O_3, 0.2 wt.% K_2O, 10.4 wt.% Na_2O, 0.4 wt.% CaO, 0.1 wt.% FeO, 0.03 wt.% TiO_2, 0.1 wt.% MnO. XRD (Table 2) analyses show that the average sum of quartz, feldspars and carbonates content is 45.5%. Framboidal pyrite, as well as anhydrite and accumulations of organic matter, are also present. Between mica flakes, microporosity is observed. The dominant size of micropores is of the order of a few micrometres (>4.35 µm). Elongated pores (canals), >16 µm in length, are rare and occur between the parallel clay mineral flakes (Fig. 14A).

KOCIEWIE FORMATION

The rocks of the Kociewie Formation are represented mainly by claystones, locally silty, calcareous, or by interbeds of silty claystone and claystone. They show pelitic-silty texture, locally pelitic, with a directional texture emphasized by the parallel orientation of clay mineral flakes, micas or laminae enriched in detrital silty or calcareous material. The pelitic fraction is represented by clay minerals (Table 2) and carbonate micrite. XRD analyses show that the average clay content is 56.4%. Apart from illite, there are also chlorites (average 14.7%) among the clay minerals. There are also mixed-layer illite/smectite minerals containing <10% of smectite, and trace amounts of kaolinite. In places, carbonates occur as subhedral and anhedral crystals represented by calcite and dolomite (Appendix 1 and Fig. 7). Rhombohedral dolomite crystals are often characterized by a zonal structure. The marginal parts of such crystals are enriched in Fe and may have a composition corresponding to Fe-dolomite or ankerite.

The silt fraction is represented by quartz and feldspar grains, and by micas. Potassium feldspars contain, on average, (Appendix 2) 65.3 wt.% SiO_2, 18.9 wt.% Al_2O_3, 15.7 wt.% K_2O, 0.2 wt.% Na_2O, 0.1 wt.% FeO, 0.2 wt.% TiO_2, 0.1 wt.% MnO. Plagioclases contain, on average, 68.7 wt.% SiO_2, 18.8 wt.% Al_2O_3, 0.2 wt.% K_2O, 11.4 wt.% Na_2O, 0.3 wt.% FeO, 0.2 wt.% TiO_2.There are also numerous accumulations of pyrite and celestine (Fig. 15) and anhydrite crystals (Fig. 15A).

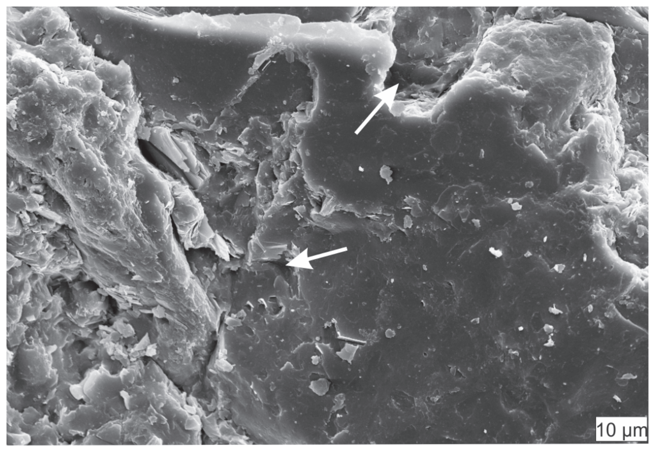


Fig. 16. Rocks from the Kociewie Formation

Calcareous siltstone; microporosity due to selective diagenetic dissolution is visible (arrowed) in calcite, Kościerzyna IG 1 borehole, depth 4174.2 m (SEI images)

The study also revealed porosity that developed diagenetically, especially by selective dissolution of detrital components (quartz, feldspars) or cement (Fig. 16). There is also microporosity between clay mineral flakes and within framboidal pyrite (Fig. 15A). The average size of isometric micropores is ~4.75 µm, while the length of channel micropores is ~7.50 µm.

PUCK FORMATION

The Puck Formation is a monotonous series of grey clay rocks, often with a greenish hue. The material analysed consists of only two samples that probably represent rocks of this formation. These are compact, grey-green, non-laminated claystones classified into calcareous claystones due to numerous dispersed very fine calcite grains, as visible in CL images (Fig. 17). The clay groundmass contains also silt-sized quartz and feldspar grains.

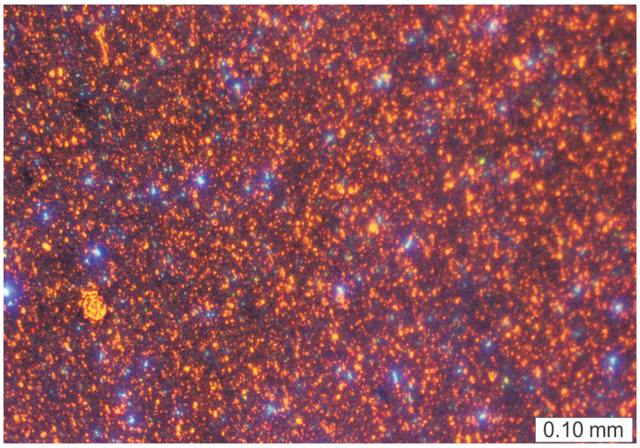


Fig. 17. Photomicrograph of calcareous claystone from the Puck Formation

Fine calcite grains (yellow-orange CL) and K-feldspar and plagioclase grains (blue and green CL) in the clay groundmass, Białogóra 1 borehole, depth 1645.7 m (CL image)

The XRD analysis of one sample (Table 2) show that the clay content is 59.4%. The claystones show microporosity between clay mineral flakes (~3.5 μm).

DISCUSSION

Within the context of earlier papers and scientific reports (Podhalańska et al., 2016a, b, 2018, 2020; Sikorska et al., 2016), the Ordovician and Silurian lithofacies potentially prospective for unconventional shale-type accumulations of hydrocarbons have been characterized. The following prospectivity criteria were assumed (Wójcicki et al., 2017), accepted by the Ministry of Environment, focused on the lower Paleozoic shales (claystones with markedly shaley structure) in the Baltic–Podlasie–Lublin Basin:

1. Total organic carbon content: TOC 1–2 wt.% – minimal criterion; >2 wt.% – optimal criterion.
2. Thermal maturity of organic matter: vitrinite or vitrinite-like macerals reflectance – R_o >0.6%.
3. Effective porosity: >4%.
4. Thickness of claystones: 10 m for TOC >2 wt.%; 30 m for TOC 1–2 wt.%.
5. Clay mineral content: <60%.
6. Brittleness: silica content >10%; content of Quartz (Q) + Feldspars (Fs) + Carbonates >40%.

Based on sedimentological investigations of core material, literature, and the present-day state of knowledge, Feldman-Olszewska and Roszkowska-Remin (2016) report the rocks of the Sasino, Jantar and Pelplin formations as the potentially prospective ones on the Ordovician and Silurian.

After taking into account results of petrophysical research, as well as geochemical and petrographic analyses of organic matter, the most promising formations are the Piaśnica, Sasino, Jantar and the lower part of the Pelplin formations (Dyrka, 2016; Karcz and Janas, 2016). Petrographic-mineralogical studies indicate that the mineral composition of the clay groundmass is almost the same in all formations studied.

In addition to the dominant illite, they may contain small amounts of mixed-layered illite/smectite and chlorite and trace amounts of kaolinite. Importantly, there is a negligible proportion of swelling clay minerals; the percentage of smectite in mixed-layered illite/smectite minerals does not exceed 15% (Table 2; Sikorska et al., 2016). The content of clay minerals in the shales from the prospective formations is high and ranges from 44.8 to 62.7%, most often it is ~50–55%. This variation in the amount of clay groundmass shows no pattern.

The effect of clay mineral content on the effective porosity of the claystones is not critical. Only certain "discrete" relationships are observed, which concern the claystones of the Kociewie and Sasino formations, where an increase in clay minerals correlates with an increase in effective porosity in the samples analysed (Fig. 18A). The microporosity here is associated mainly with microspaces between clay mineral flakes in rocks in all the formations studied.

The rocks analysed contain microlaminae of silt material and dispersed carbonates, the content of which varies between individual samples. These features are observed in both the prospective and non-prospective formations. Thus, the mineral composition of the shales can be correlated with the degree of their prospectivity only in terms of susceptibility to the technological process of hydraulic fracturing. The rock component significantly impacting brittleness is the content of clay minerals (<60%; Table 2) and the low content of swelling minerals. Its quantitative indicator is the amount of detritus (quartz and feld-

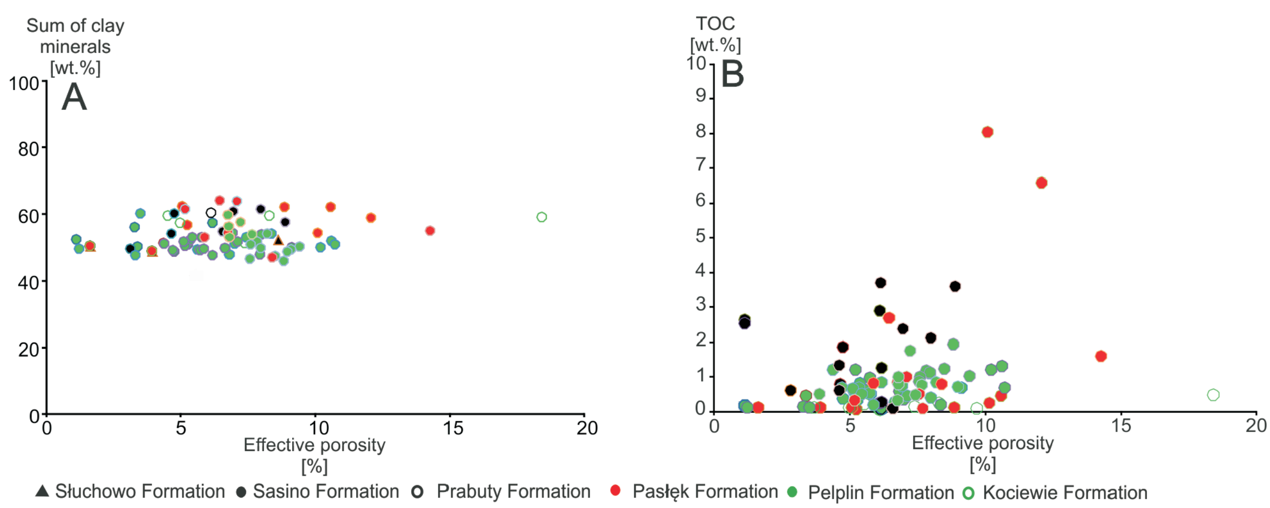


Fig. 18A – relationship between the clay mineral content and effective porosity; B – relationship between the TOC value and effective porosity

Data of TOC and effective porosity from Podhalańska et al. (2016b, 2018) spar) or the sum of detritus and carbonates. The results obtained from XRD analysis are not always reflected in microscopic observations and CL analysis. The higher content of feldspars, in particular of plagioclases, is a consequence of potassium feldspars being albitized and invisible (brown luminescence), while alkaline plagioclases are blue in CL, resembling K-feldspars. Chemical analyses of feldspars in the Pasłęk, Pelplin, Kociewie formations (Appendix 2) little difference in their composition. The SiO_2 content in feldspars is slightly lower in the Pasłęk Formation (63.8–66.9 wt.%) compared to the samples from the Pelplin and Kociewie formations (64.7–68.7 wt.%). The Al_2O_3 content is comparable among the samples from the formations analysed. The K_2O content of the potassium feldspars is ~16 wt.%. Contents of Na_2O range from 10.4 to 11.4 wt.% and CaO ranges from 0 to 1.12 wt.% in plagioclases.

An additional problem in microscopic identification is the very fine grain size. The CL images clearly show the form of feldspar accumulation, their even distribution in the groundmass, and the mode of carbonate occurrence: fine grains, pseudomorphs after feldspars, microveins, nests, etc. The analysis of these features shows no pattern in their appearance in the rocks studied. Nevertheless, in minerals such as feldspar or carbonates, secondary microporosity was noticed, resulting from diagenetic processes (mainly dissolution). Microporosity was also observed within framboidal pyrite concentrations, which is commonly found in claystones of formations known as prospective.

The effective porosity of the Ordovician rocks is 0.6–8.9%, and of the Silurian rocks it is 1.2–18.4% (Podhalańska et al., 2016b, 2018). The average effective porosity values in the prospective formations are as follows: Piaśnica – 5.0%, Sasino – 5.4%, Jantar – 4.9%, Pelplin – 6.5% (Dyrka, 2016).

The TOC content in the claystones ranges from ~0.1 to ~10 wt.% (Więcław et al., 2010; Karcz and Janas, 2016; Podhalańska et al., 2016b, 2018). The TOC values in the samples from the formations analysed are as follows: Sasino 0.24–3.7 wt.%, Prabuty 0.07–1.02 wt.%, Pasłęk 0.03–8.0 wt.%, Pelplin 0.09–1.92 wt.% and Kociewie 0.06–0.45 wt.%. The most variable TOC values were recorded in samples from the Pasłęk Formation. As can be seen from the graph of the relationship between TOC and effective porosity (Fig. 18B), a slight increase in porosity with increasing TOC content is observed here.

A more prominent relationship occurs in the case of rock samples from the Sasino Formation, although the TOC variation is smaller here than in the Pasłęk Formation. It can therefore be assumed that the increase in effective porosity in claystones is related to the increased content of organic matter, which is associated with the microporosity observed within it. The results of Słomski's (2019) research also indicate that the content of organic matter is an important factor affecting porosity, especially in the case of the Jantar bituminous claystones and the Sasino Formation rocks. The organic matter content and clay minerals greatly influence the density of the claystones. According to Jarzyna et al. (2021) density and porosity are the main petrophysical parameters of shale gas formations that are of interest. In the other formations, despite the much lower organic carbon content, there is a positive relationship between the increase in effective porosity and TOC values. The degree of maturity of organic matter in the Baltic Basin increases towards the SW, along with the increasing depth to the crystalline basement of the East European Craton (Karcz and Janas, 2016). The R_o index values (0.58–2.04%) correspond to the main phase of oil generation window and the gas generation window (Grotek, 2016). According to Papiernik et al. (2019) on the basis of a combination of thermal maturity and TOC (0.6% < R_o <2.4%, TOC >1.5 wt.%), good-quality, unconventional reservoirs can be expected in the Sasino and Jantar formations.

The organic matter underwent secondary alteration. Solid bitumens fill microfractures or block the free pore spaces. The distribution forms and nanoporosity of the organic matter require further analyses. The development of analytical methods for studying shale rocks in terms of reservoir parameters was described by Such et al. (2017), commenting on matters such as the presence of 'inaccessible' porosity in organic matter filling the pore space of rocks.

CONCLUSIONS

Petrological analysis of rocks of the Ordovician and Silurian lithostratigraphic formations in the Polish part of the Baltic Basin allowed siltstones, mudstones, limestones, marls, sandstones, as well as the most common claystones with a markedly shaley structure (shales) to be distinguished. The same type of rock in the formations analysed, both in the prospective and non- prospective formations for unconventional hydrocarbon accumulations, do not differ from each other in terms of mineralogy. In individual samples, content variations refer only to silt material and carbonates, which can affect the technological process of hydraulic fracturing and are among the determinants of prospectivity of the rocks.

Complementary results of petrological analysis, especially of clay minerals and organic matter, in association with effective porosity values, is important to assess the prospectivity of unconventional hydrocarbon accumulations in the rocks studied. Only certain "discrete" relationships are observed in the rocks studied, where an increase in clay minerals correlates with an increase in effective porosity, which concern the claystones of the Kociewie and Sasino formations. There is a slight increase in porosity with increasing TOC content in the Sasino, Jantar and Pasłęk formations.

Observations of the pore space in the rocks have allowed identification of micropores between detrital grains and within pyrite framboids, secondary micropores in grains (the effect of selective dissolution of feldspar grains and carbonates), and microporosity between the clay mineral flakes and within organic matter.

The claystones of the Sasino, Jantar, and Pelplin formations seem to be the most prospective lithofacies for unconventional shale-type hydrocarbon accumulations in the Ordovician and Silurian rocks. They are characterized by a high content of organic matter (Karcz and Janas, 2016; Papiernik et al., 2019; Podhalańska et al., 2020; Poprawa, 2020). Very important is the negligible proportion of swelling minerals in the clay fraction. The percentage of smectite in mixed-layered illite/smectite minerals does not exceed 15%. Importantly, the rocks have adequate brittleness: e.g. the content of quartz, feldspars and carbonates (here >40%) points to the suitable susceptibility of the claystones for hydraulic fracturing.

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