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GEOLOGICAL PRECONDITIONS FOR IDENTIFYING OF PERSPECTIVE AQUIFER STRUCTURES FOR NATURAL GAS AND CO_2 STORAGE IN NORTH-EASTERN BULGARIA

Summary. The methodical approaches for identifying of perspective aquifer structures for natural gas and CO_2 storage indicated by the authors in previous works are applied for a part of North-Eastern Bulgaria. They are based on the main requirements for the suitability of the geological sites and are connected to preconditions ensuring lithologic, structural-tectonic, seismotectonic, hydrogeological; petrophysical, termobaric and other requirements. The regional reservoir systems defined for Northern Bulgaria are examined in the Phanerozoic section of the studied territory. They are characterized by specific lithological features and development and are of great interest for the natural gas and CO_2 storage. The structural-tectonic conditions in the Moesian Platform (The Varna Monocline, The North Bulgarian Uplift and The Alexandria Depression) and the Dolna Kamchia Depression determine the presence of different geological structures. Different types of natural traps are bounded to them. Some of these traps correspond to the requirements for natural gas and CO_2 storage. The seismic and neotectonic situation is variable (basically low to middle and in rare cases higher), which determines different earthquake activity and behavior of the fault breaks in the neotectonic development of the territory. The natural reservoir connected to the Upper Jurassic – Lower Cretaceous permeable complex, in spite of its high capacity and filtration potential, contains fresh water with active hydrodynamic regime and this fact makes it unsuitable for natural gas and CO_2 storage. The defined reservoir systems and sealing formations are characterized by too different structure and petrophysical properties and this influences their suitability of the natural traps natural gas and CO_2 storage. Perspective structures for natural gas and CO_2 storage, connected to Devonian, Triassic, Middle Jurassic and Paleogene reservoir systems, are registered on the basis of the geological conditions' analysis and the criteria parameters in the Phanerozoic section of the studied territory.

GEOLOGICZNE UWARUNKOWANIA IDENTYFIKACJI PERSPEKTYWICZNYCH STRUKTUR WODONOŚNYCH DLA SKŁADOWANIA GAZU ZIEMNEGO I CO_2 W N-E BUŁGARII

Streszczenie. Metodykę oparto na ocenie kryteriów litologicznych, strukturalno-tektonicznych, sejsmotektonicznych, hydrogeologicncyh, petrofizycznych, termicznych i ciśnieniowych. Zastosowano ją dla identyfikacji struktur wodonośnych perspektywicznych z punktu widzenia składowania gazu ziemnego i CO_2, na terenie północno-wschodniej Bułgarii. Zidentyfikowano i oceniono warunki geologiczne składowania dla poziomów wodonośnych, reprezentujących struktury

od dewonu po górny eocen, leżących w zasięgu Monokliny Warny, obniżenia Dolnej Kamczy, wyniesienia północnobułgarskiego i obniżenia Aleksandrii. Głębokość występowania stwierdzonych poziomów waha sie od 500 do 1700 m.

1. Introduction

The methodical approaches worked out by the authors in previous studies for identifying of perspective aquifer structures for storage of natural gas and CO_2 (Balinov et al., 2007, 2008a, 2008b) are applied for a part of North-Eastern Bulgaria (The North Bulgarian Uplift, the Alexandria Depression, the Varna Monocline and the Dolna Kamchia Depression). They are based on the main requirements for suitability of the geological sites and are connected to the concrete geological preconditions: lithological, structural-tectonic, seismotectonic, lithological-physical (petrophysical), hydrogeological and thermobaric ones.

The lithological and the petrophysical preconditions determine the presence in the sediment sections of permeable (reservoir) and hard-permeable (sealing) rocks (formations, complexes). They form different type of natural reservoirs at definite conditions between them and favorable petrophysical properties. The reservoir systems from the depth interval 500-2500 m. are of a great interest from the point of view of the natural gas and CO_2 storage. It corresponds to the requirements for the minimal depth of the objects for CO_2 storage (about 800 m), the recommended maximal depth (2500 m) and to the world experience regarding the optimal interval for natural gas storage (500-1500 m). Special attention is paid to the reservoir systems containing water with increased mineralization that is not interesting for domestic and other purposes.

The role of the structural-tectonic preconditions is connected to the formation of different types of local structures and natural traps that at favorable combination of other factors are of a great interest as local sites for storage of natural gas and CO_2.

The seismotectonic preconditions are in direct relation with the selection of perspective structures, as well as with the problems of the geological risk, connected to possible wholeness break of the already built storages of natural gas and CO_2 in result of the neotectonic development of the territories where the local sites are situated.

The hydrogeological preconditions are connected to the hydrochemical parameters of water that define their suitability for different purposes and testify in indirect way for their regime. They are also connected to the hydrodynamical parameters that play an important role in the selection of perspective structures and conditions for storage of natural gas and CO_2.

The thermobaric preconditions are mainly related with the conditions for CO_2 storage. In accordance with the regulated requirements (Metz et al., 2005; Chadwick et al., 2007) CO_2 should be at over-critical conditions (p_{cr}=71.9 bar, t_{cr}=34.3°C). Such conditions exist at depths more than 800 m. The thermobaric conditions are also directly connected to the amount of the natural gas and CO_2 stored in the local structures and are examined at their prognostic evaluations.

2. Analysis and evaluation of the geological preconditions Natural Reservoir Systems

The reservoir systems are examined on the background of the complexes in the Phanerozoic section of Northern Bulgaria that are predominantly permeable and hard-permeable regional complexes (RPC and RHC): Devon-Carboniferous RPC, Permian RHC, Lower Triassic RPC, Lower Triassic RHC, Middle Triassic RPC, Upper Triassic – Middle Jurassic RHC, Upper Jurassic – Lower Cretaceous RPC, Lower Cretaceous RHC (Balinov, 1975, Kalinko, 1976, Bokov, Chemberski, 1987). They form four regional natural reservoirs (RNR) connected to the eponymous RPC: Devonian-Carboniferous, Lower Triassic, Middle Triassic and Upper Jurassic – Lower Cretaceous. In the North-Eastern Bulgaria they are characterized by a number of specific features regarding their development, stratigraphic range, structure and lithological-physical parameters. They are situated at different depth because of the variable tectonic conditions. Significant hiatuses in the sediment section are sometimes the reason for the absence of some permeable and hard-permeable complexes. Because of this a part of the separated natural reservoirs are not present in separate areas of the studied territory and others are with heterogeneous stratigraphic range.

In the Paleogene section of the North-Eastern Bulgaria predominantly permeable and hard-permeable lithologic-physical bodies (LPB) are separated. They are connected to the reservoir systems of the Upper Paleocene – Eocene and the Oligocene section (Balinov, 1975, Deshev, 1976).

In *the Paleozoic section* the Devonian-Carboniferous permeable complex is of a great interest. It is developed in the North-Eastern part of the North Bulgarian Uplift at depth from 800 to 1000 m. It is represented by variable carbonate rocks, differentiated in several formations. The Dolomite Formation and the Stripe Limestones Formation are with wide spread. The Interclastic and Kernel Limestones Formation (IKLF) and the Organogenic

Limestones Formation (OLF – Lower Carboniferous) are with restricted distribution. In some places the wash out of the Paleozoic sediments is significant and in result of this only the Dolomite Formation (DF) is present. The carbonate rocks are represented by micro fine-grained, interclastic and kernel limestones, dolomites and dolomitized limestones. The total depth of the permeable complex is more than 280 m. The reservoirs are of mixed – porous-cavernous-fracture type. They are characterized by low capacity and favorable filtration properties. The wide development of the macro and micro-fractures creates a hydrodynamic relation in the whole section of the complex and in some places with the above-laying Jurassic terrigenous rocks.

The Middle Jurassic hard-permeable formation is in the role of a cover. Predominantly hard-permeable Lower Permian sediments join it in a narrow stripe in the north-eastern part of the Tolbuhin-Vetrino Block. The cover is built of non-limy, differently silty argillites in irregular alternation with clayey siltstones. They contain interbeds and layers of sandstones. They are fine to middle-grained, cemented in different degree. The thickness of the predominantly hard-permeable complex varies in the limits from 70 to 270 m. Its sealing properties are variable due to the presence of permeable sandstone layers which in some places is significant.

In *the Mesozoic section* the natural traps connected to the Lower Triassic Middle Triassic and Middle Jurassic permeable sediments are of a great interest.

The Lower Triassic permeable complex is interesting in the western part of the North Bulgarian Uplift, the southern part of the Alexandria Depression and the southern part of the Varna Monocline. It is situated at depth of about 1700 m. It is represented by the Red-colored Sandstone Formation (RSF). It is built of varicolored and varigrained sandstones with often interbeds of siltstones and argillites. Its thickness in the western part of the North Bulgarian Uplift reaches up to 260 m. and in the southern part of the Varna Monocline – 100 m. The reservoirs are of porous type and are characterized with favorable capacity and filtration properties (class III) (Balinov and others, 1977). Volcanogenic-terrigenous rocks from the Lower Permian complex, containing in this region permeable sandstone layers, are bounded to the permeable complex in the southern part of the Varna Monocline.

The Lower Triassic Hard-permeable Complex, developed in the western part of the North Bulgarian Uplift, is in the role of a cover. It is mainly built of argillites. They are varicolored, irregularly silty and non-limy. Non-limy fine-grained siltstones and sandstones are present as thin interbeds. Its thickness varies from 219 to 411 m. The hard-permeable complex is with comparatively homogenous lithological structure. According to the data from laboratory

analysis the rocks possess relatively favorable sealing properties (D and E classes) (Yordanov and others, 1985). In the southern part of the Varna Monocline the Middle Jurassic Hard-permeable Formation is in the role of a cover. Its structure is complicated, because of the sediments variable in age and lithological composition included in its structure. In the section there are argillites, clear to weakly clayey aphanitic limestones and argillites, non-limy to limy, slightly silty and in some places passing into marls. The total depth rarely exceeds 100 m. The sealing properties of the formation are variable, because of the presence of permeable layers in some places of the section.

The Middle Triassic Permeable Complex is of a great interest in the western part of the North Bulgarian Uplift and the southern part of the Alexandria Depression, where it is situated at depth about 1200 m. It is represented by carbonate sediments – limestones and dolomites. The limestones are micro to fine-crystalline, partially oolitic, partially pseudo breccia-like. The dolomites are microcrystalline ones. Very thin interbeds or inclusions of anhydrite occur. The limestones take the biggest part of the section. The thickness of the complex varies from 145 to 455 m. The permeable sediments from the basis of the Middle Jurassic Predominantly Hard-permeable Complex join the complex in some places. They develop its volume and its stratigraphic range. The reservoirs possess variable capacity and filtration properties. They are significantly fractured. Its open capacity varies from units to 16%. They are of fracture or porous-fracture type. According to their filtration properties they are referred to III to V classes.

The Middle Jurassic Hard-permeable Formation, which thickness reaches 105 m is in the role of a cover. It is represented by argillites that are interbeded by siltstones and sandstones. The siltstones are sandy and limy. The sandstones are fine to middle-grained, quartzy, and non-limy. In some places the section is mainly represented by siltstones.

In the range of the Middle Jurassic Hard-permeable Formation there are permeable sandstone bodies that take part in the formation of reservoir systems with zonal and local development. They take different parts of the section. The sandstone bodies presenting in the lower and upper part are in contact at some places with the below-laying Middle Triassic Permeable Complex and the above-situated Upper Jurassic – Lower Cretaceous Complex and form unified reservoir systems.

In the middle part of the section the permeable sandstone body forms a natural reservoir with zonal development. It is situated at depth of about 1100 m. It is built of sandstones in alternation with siltstones and argillites. Its thickness reaches to 48 m. Its capacity properties are variable. Its open porosity varies from 10 to 25%, 17 % average. The permeability varies

from 1 to 4000 mD. The cover is built of irregularly alternation of argillites, clay siltstones and clay. Its thickness varies from 27 to 70 m.

In *the Tertiary section* the Paleogene and Oligocene permeable sediments from the Varna Monocline and the Dolna Kamchia Depresion are of a great interest. They form reservoir systems with complicated structure which is predetermined by an irregular alternation of permeable (sands, sandstones, conglomerates, limestones) and hard-permeable (clay, clay siltstones, marls, argillites and others) layers and interbeds with different thickness and restricted distribution (because of the pinching out or facial displacement) and complex spatial relationships. The thickness of the permeable bodies varies in wide limits – from units to several tens of meters, as they grow in the southern part of the Lower Kamchia Depression. They are characterized by too variable reservoir parameters (I-V class).

The predominantly permeable and predominantly hard-permeable lithologic-physical bodies (formations) build reservoir systems of layer and combined (layer-massive) type (Deshev, 1976). In the Oligocene sediments there are reservoirs mainly of lithological type, connected to zones with pinching out of the sandstone horizons. The indicated special features in the structure of the reservoir systems determine the presence of "lithological windows", that realize a hydraulic relation between the stage situated systems and sub-systems. In separate regions the permeable carbonate sediments of the Paleocene are in contact with Upper Cretaceous sandy-carbonate rocks forming a unified natural reservoir. The Galata gas deposit is bounded to such a reservoir. The gas accumulation near Staro Oriahovo village is connected to a natural reservoir of the lithologically restricted type.

3. Structural-tectonic preconditions

On the basis of the seismic survey and the drilling in the studied territory of North-Eastern Bulgaria about 25 local structures are separated (Bokov, Chemberski, 1987). They are bounded to different stratigraphic complexes – Devonian, Permian, Triassic, Upper Jurassic – Valanginian, Lower Cretaceous, Upper Cretaceous and Paleogene (Upper Eocene and Oligocene). Significant parts of them are with unclear depth structure.

A part of the above-mentioned structures are confirmed and new structures with wider range are registered in result of the additional researches carried out by the authors (reinterpretation of the seismic sections, structural tectonic buildings and others). The

selection of perspective structures for storage of natural gas and CO_2 is made on the basis of the structural analysis at different stratigraphic levels compared to the maps of the natural reservoirs.

In the Paleozoic and the Mesozoic section in the areas with presence of perspective natural reservoirs the local positive structures are well-expressed over the structural plan on the Lower part of the Middle Jurassic Complex. In the Paleogene section of the Varna Monocline and the Dolna Kamchia Depression the presence of such structures is well-expressed over the structural plan on the top of the Eocene sediments.

The separated seven perspective structures are situated in different areas of the North Bulgarian Uplift, the southern part of the Dolna Kamchia Depression and the southern part of the Varna Monocline. In some of them there are more than one potentially perspective natural traps.

4. Seismotectonic preconditions

Their role is directly connected to the behavior of the fault breaks in the tectonic evolution of the North-Eastern Bulgaria. Tectonic blocks of different size and displacement are differentiated on them in result of the Old Cimmerian tectonic movements (Bokov, Chemberski, 1987). The examined structures, formed in the Paleozoic and in the lower part of the Mesozoic section are complicated by additional faults and fractures with displacement. In the Jurassic-Cretaceous and the Paleogene section the biggest part of this breaks are buried. The data from the tectonic break and the physical anisotropy of the different type of rocks in the interval Early Creations – Late Paleocene show a stable tendency for rotation of the axes of compression in the tectonic field of North-Eastern Bulgaria (Shanov, 1990). They are a good basis for identifying of tectonic breaks with characteristics of contemporary active faults and evaluation of their possible influence over the local structures.

On the basis of the seismotectonic studies in the North-Eastern Bulgaria three zones with increased potential seismic activity are separated: Gorna Oriahovica, Shumen and Kaliakra (Kostadinov and others, 1992). They are characterized by strong break of the earth's crust and parts of the fault dislocations are with neotectonic activity. There are zones with increased tangential tensions that could cause new faults, generating centers of earthquakes. The biggest part of the studied territory is in these seismic zones, except the north-eastern parts of the North Bulgarian Uplift.

5. Hydrogeological preconditions

Devonain, Triassic, Middle Jurassic and Paleogene aquifer complexes are of a great interest in the studied territory of the North-Eastern Bulgaria. They are closely connected to the development of the separated perspective natural reservoirs.

The Devonian aquifer complex is in the central and in the eastern part of the North Bulgarian Uplift, which is built of carbonate rocks and is connected to the eponymous reservoir systems. The water is with mineralization from 2 to 30 g/l. It is under pressure and of fracture-karst type. The piezometric levels referred to fresh water regularly vary from + 54.8 m to +16.5 m. The water is with low flow rate and of chlorine-sodium type. The coefficient of filtration is low (about 10^{-2} m/d).

The Lower and Middle Triassic aquifer complexes from the western part of the North Bulgarian Uplift and the Alexandria Depression are with similar hydrodynamic characteristics. The sandstones (Lower Triassic) and the limestones and the dolomites (Middle Triassic) are with low hydroconductivity (from 0.3 to 5.6 d cm/cP). The flow rates vary in the limits from 2.4 to 50 m^3/d. The piezometric levels are from -30 to -47 m. The Lower Triassic water is salty and mineralized (from 30 to 65 g/l) by chemical composition. The water is of chlorine-calcium type. The content of strontium, iron, bromine, iodine and ammonium is high. This is a proof that the aquifer bodies are well-covered. In the Middle Triassic Complex there is a well-expressed vertical hydrodynamic zoning (the mineralization increases from 21 to 77 g/l).

The Middle Jurassic complex contains aquifer horizons in the western part of the North Bulgarian Uplift and the Alexandria Depression, but in the Varna Monocline it is hard-permeable. Inflows with variable flow rate from 3.4 to 47 m^3/d are obtained from aquifer sandstones. The piezometric levels are from -4.8 to 52 m. The water in the Middle Jurassic section is mineralized – from 15.9 to 26.3 g/l. It is of chlorine-calcium and chlorine-magnesium type. It contains the micro-components iodine, bromine, boron, ammonium and others and this is a proof that the aquifer layers are covered. In the Upper part of the complex the water is with parameters close to these of the Upper Jurassic - Lower Cretaceous carbonate complex due to the hydrodynamical relation between them.

The Aquifer bodies and horizons in the Eocene section of the Dolna Kamchia Depression are interesting for their capacity and productive possibilities. The terrigenous and terrigenous-

carbonate rocks that build them are with relatively good conductivity. The rough values are in the limits from 5–10 to 30–40 m^2/d. There are no representative data for the piezometric levels. The expected values in conditions of water–pressure regime are of the rate of tens meters over the sea level. The chemical composition of the water is salty with mineralization from 8-10 до 40 -50 g/l. The main components are the chlorides and the sodium. The sulphate reduction processes are the reason for the low content of sulphates and the comparatively higher content of bicarbonates. The content of iodine (from 10-12 to 35-50 mg/l) and bromine (to 50-60 mg/l) is high. The water is characterized with increased to high temperature. According to the depth of situation of the aquifer bodies and the influence of the convectional transfer of heat from the deepest levels, the layer temperatures of water vary in the limits from 30 to 65^{0}C. The high and comparatively stable in time mineralization in the southern and central parts of the depression is a proof of good sealing of the aquifer bodies and the structures from the atmospheric-infiltration water exchange.

In the Lower levels of the Oligocene section aquifer bodies with small volume are identified. They are included in the clay sediments dominating in the section.

The Lower Eocene aquifer horizon in the Varna Monocline is characterized by relatively high capacity and filtration properties of the terrigenous and carbon rocks that build them. In its whole range the aquifer horizon is in regime of active water exchange and infiltrational atmospheric supply. This is the reason for the low mineralization of water and the decreased presence of chlorine compounds. In the range of the southern part of the Avren Step the aquifer horizons are not touched by the fault dislocations and are with reliable upper and lower water-retentions.

The Oligocene aquifer bodies from the Varna Monocline are bounded to the locally developed permeable bodies and are with good hydrogeological coverage.

6. Basic results

On the basis of the analysis and evaluation of the geological conditions for storage of natural gas and CO_2 in aquifer formations in regions of North-Eastern Bulgaria (the Varna Monocline, the Dolna Kamchia Depression, the North Bulgarian Uplift and the Alexandria Depression), perspective aquifer horizons are identified. They represent potential storages of

natural gas and CO_2. They are bounded to different stratigraphic levels – from Devonian to Upper Eocene. Their depth varies in limits from 500 to 1700 m.

The local structures and the natural traps connected to them are of different type – structural (anticlinal or brachyanticlinal) or lithologically restricted. According to the preliminary evaluations they correspond to the requirements for storage of natural gas and/or CO_2 in aquifer structures. In the limits of the identified perspective structures in a number of cases more than one natural trap with different stratigraphic adherence are defined.

The reservoir formations that are bounded to perspective natural traps are built of terrigenous or carbonate rocks. The terrigenous reservoirs are of porous type. According to the prognostic evaluations their quantitative parameters are in the limits of the basic criteria requirements. The significant lithological changeability defines the variations of their capacity and filtration properties. The porosity is in the limits from 5 to 30% as the predominant values are between 15 and 20%. The permeability varies from several units to several hundreds of mD. The effective thickness of the reservoir formations in separate natural traps is from 10 to 70 m. The prognostic capacity varies in wide limits – from several tens to several hundreds of millions t regarding the CO_2 and several hundreds of millions m^3 to several billions of m^3 regarding the natural gas.

The carbonate reservoirs that are connected to a part of the perspective traps are of mixed type, most often porous-cavernous-fracture type. They are characterized by low capacity parameters (the fracture-cavernous capacity does not exceed 1%) and its filtration properties are favorable. Due to the significant thickness of the carbonate reservoir formations the prognostic capacity of the natural traps reaches up to several hundreds of millions t regarding the CO_2. These structures are not recommended for natural gas storage.

In the Paleozoic section two perspective structures are separated. They are situated in the north-eastern part of the North Bulgarian Uplift. They are connected to the Middle and Upper Devonian carbonate reservoirs.

In the Mesozoic section two perspective structures are identified. One of them is situated in the southern part of the Varna Monocline and is connected to the Permian-Triassic clastic permeable sediments. The other one comprises areas from the western part of the North Bulgarian Uplift and the southern part of the Alexandria Depression. Three natural traps are registered in it. They are connected to the clastic and carbonate Lower Triassic, Middle Triassic and Middle Jurassic reservoir formations.

In the Tertiary section three perspective structures are separated. One of them is situated in the southern part of the Varna Monocline and the clastic reservoirs in it are with Lower

Eocene age. The other two are situated in the southern part of the Lower Kamchia Depression. The reservoirs in them are represented by sandstones and limestones of the Middle Eocene - Upper Cretaceous sediments and of the sandstones from the Upper Eocene.

The perspective aquifer structures in the Paleozoic and Mesozoic section are mainly recommended for CO_2 storage and these from the Tertiary section are recommended for natural gas storage. The main prognostic parameters of the potential storages of natural gas and CO_2 are evaluated for each one of the perspective aquifer structures and the natural traps connected to them.

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