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GEOLOGICAL, GEOCHEMICAL AND ISOTOPIC MODELS OF METHANE CONCENTRATION IN THE UKRAINIAN COAL BASINS

Problems of methane origin in coal bearing sequences determine possibility utilization of the methane and creation of safety conditions during underground mining. There is no general agreement about the nature of methane from coal bearing sequences of the Donbas and Lviv-Volyn coal basins. According to the first ideas methane deposits here have been formed due to transformation of initial organic material and its modern resources are limited by total amount of previous organic sources. It is shown that these ideas need in improving because there are evidences on depth origin and permanent addition of the methane as consequence of unique gas deposits located under coal bearing sequences. Geochemical and isotopic peculiarities of some chemical elements and hydrocarbon and joint gases evolution in different zones of the coal bearing sequences in the Donbas and Lviv-Volyn coal basins during the Hercynian and Mesozoic tectonic epochs have been used to create a new model of methane origin and migration. Main temperature and pressure limits of hydrocarbon concentrations within these zones are established to determine main methane deposits for future utilization.

Keywords: *methane, coal deposits, Ukraine, stable isotopes.*

1. Introduction and statement of the problem

We analyzed situation with own traditional fuel resources in Ukraine and estimated possible supplying periods of these sources according to native industrial demands. Beside of oil, gas, coal and uranium ores Ukraine has big additional fuel resources such as methane of coal-bearing suits, gas-hydrates, and geothermal energy. Most realistic additional energetic resources at present time the authors connected with methane of the coal beds as well as using of gasification of coal by new proposed methods [3, 5].

Ukraine produces a variety of types of coal. Coal reserves in Ukraine amount to around 47,1 billion tons (metric tons). The largest reserves of anthracitic coal (92 %) are located in the Donets'k Basin in the southeast of the country; most of the remainder is in the Lviv-Volyn Basin in the West. A total of 244 operating mines were reported by IEA for the last time. Of these 239 were underground, and only five surface pits.

During last decades methane of coal deposits becomes more popular energetic source among other domestic fuels. For the Ukrainian coal basins methane resources have been estimated as 2,5 Tm^3, according to relatively realistic calculations, and up to 25,0 Tm^3 from very optimistic experts. Total recourses of the methane have similar scales if compare it to traditional natural gas deposits of Ukraine.

At the same time the methane is dangerous component during mining of coal at the depth levels. Very often it is difficult to ensure the fulfillment of necessary arrange-

ments because of technical problems and geological factors. Solving these problems is very important for any country, which hopes to improve supply of energetic resources via domestic fuels. Very significant it is also for Ukraine as a result of methane and sands explosions in the coal mines, which happen very often.

To support safety of the underground mining there is a necessity to clean up and reduce contents of methane in the underground. The methane should be utilized during this process. On this way there is a big problem. Nature of methane from coal bearing sequences of the coal basins of Ukraine traditionally regards as local, dominantly organic in origin. According to this first idea methane deposits here have been formed due to transformation of initial organic materials and its modern resources are limited by total amount of organic sources. It is basement point of previous and modern strategy of providing with normal conditions of coal mining. The most common and effective methods of achieving this goal are ventilation with degassing and pre-gassing of separate blocks of the mine fields. But that previous idea about limited volumes of methane in coal bodies needs improving because there is evidence on deep origin of methane and its continuous addition. So, purposes of our study are:

1) Analysis of traditional hypotheses on methane nature and possible its volumes in coal seams. 2) Estimation of evidences on methane origin in coal-bearing sequences. It gives a chance to determine a valid strategy for safety of underground mining and methane utilization.

3) Provide facts on continuous addition of methane to underground mines and its depth nature.

4) Create a joint geological, geochemical and isotopic model of methane concentration in the Ukrainian coal basins.

5) Determine the most perspective areas for discovery of deep methane deposits, which are a source for methane within upper seams of coal.

2. Main initial statements

2.1. Spatial and temporary distribution of methane and its isotopic features

To create a new model of origin, migration and concentration of methane we used such main initial statements. In the same area, oil and gas deposits of industrial scale as well as coal deposits are enlarged upon the same area. There is a coincidence in spatial distributions in the same area of oil, gas, and coal deposits, which have often industrial scales. In the underground coal mines, oil and gas are common. Appearance of oil and gas directly in the underground coal mines (Far East – [18]; Japan – [9]; etc.). Very often we can distinguish renewals oil and gas deposits with time or appearance of gas and oil in structures which suggested as having no prospects [3, 4, 21]. Very often we can see new portions of gas and oil in old deposits. Scale of hydrocarbons inflows is similar to the scale of industrial extractions. Practical consequence of this phenomenon is recalculation of the gas and oil resources estimated early [3]. Isotopic compositions of the C from the methane support ideas about its deep source. Entering of methane and coal-dust in underground mines is repeated even after big scale ventilation with degassing and reducing of methane concentrations to safe levels.

2.2. Main geological and compositional features of coal-bearing sequences

Investigations of heat flows [11, 15] show complicated pictures of isotherms location in different geological structures of Ukraine. The isotherm of 600°C is indicator which suggests upper limits of methane generation.

We revalued previous data on distribution of different sorts of coal in two main coal basins of Ukraine. Zoned localization of high and low quality coals is very clear for the Donbas region (Figure 1). Good example for illustration of spatial coincidence is distribution of oil and gas deposits in the Northern Donbas (Figure 2). Different in scale gas and oil deposits are disseminated among coal mines. In similar situation gas deposits (for example, Velyki Mosty and Le-

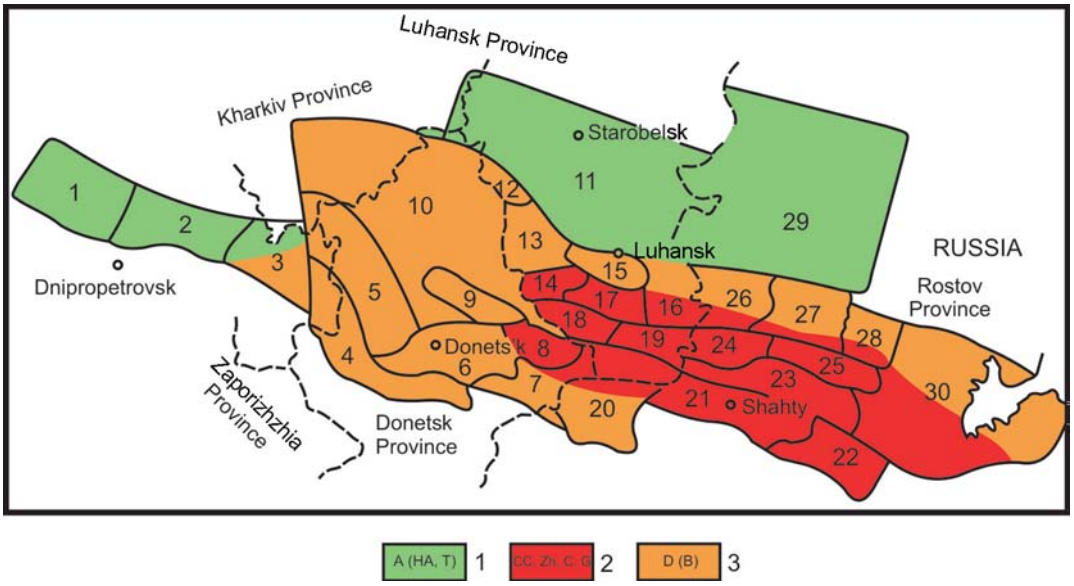

Fig. 1. Methane perspectives of the Donbas coal bearing districts
Legend: 1 – Distribution of the A (HA, T) coal sorts – Central Anthracite Massif. The possible main gas deposit is located in C_{1v} rocks. 2 – Distribution of the CC, Zh, C, G coal sorts. Gas columns are possible in these areas. 3 – Big areas of dominant distribution of the D (B) coal sorts. No perspective districts

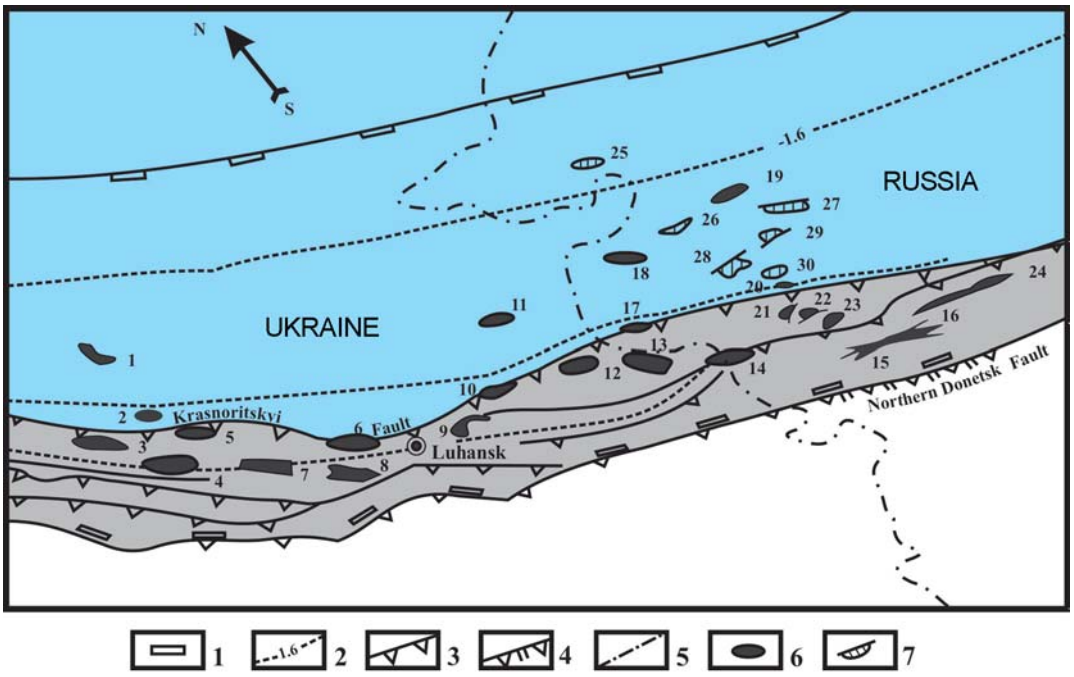

Fig. 2. Distribution of oil and gas deposits in the Northern Donbas
Legend: 1 – boundary of the oil-gas bearing district of the Northern side of the Dnipro-Donetsk Depression; 2 – isohypse, km (surface of the crystalline basement which coincides with boundaries of the structural-tectonic zones of the Northern side of the Dnipro-Donetsk Depression); 3–4 – faults; 5 – state border; 6 – deposits of Ukraine (1– 13) and Russia (14–24); 7 – small scale gas deposits (25–30)

lykivske gas deposits) and gas manifestations are distributed in the Lviv-Volyn Coal Basin (Figure 3).

3. Dispersion and secondary halos as main indicators of methane migration

3.1. Main features of dispersion and secondary halos

Geological and geochemical investigations [14, 15] show that oil and gas deposits and manifestations in coal-bearing sequences are accompanied by bitumen of different in compositions as well as numbers of secondary alterations. We use these facts as a basement of geological model [13, 15] of methane concentrations regarding ideas about primary (Figure 4) and secondary (Figure 5) halos.

Source of primary or dispersion halo (Figure 4) is an independent gas deposit containing large resource. It is located in the low part of coal strata [13]. Three zones are determined in primary halo (Figure 4): 1) zone of gas (composition – mainly methane, with

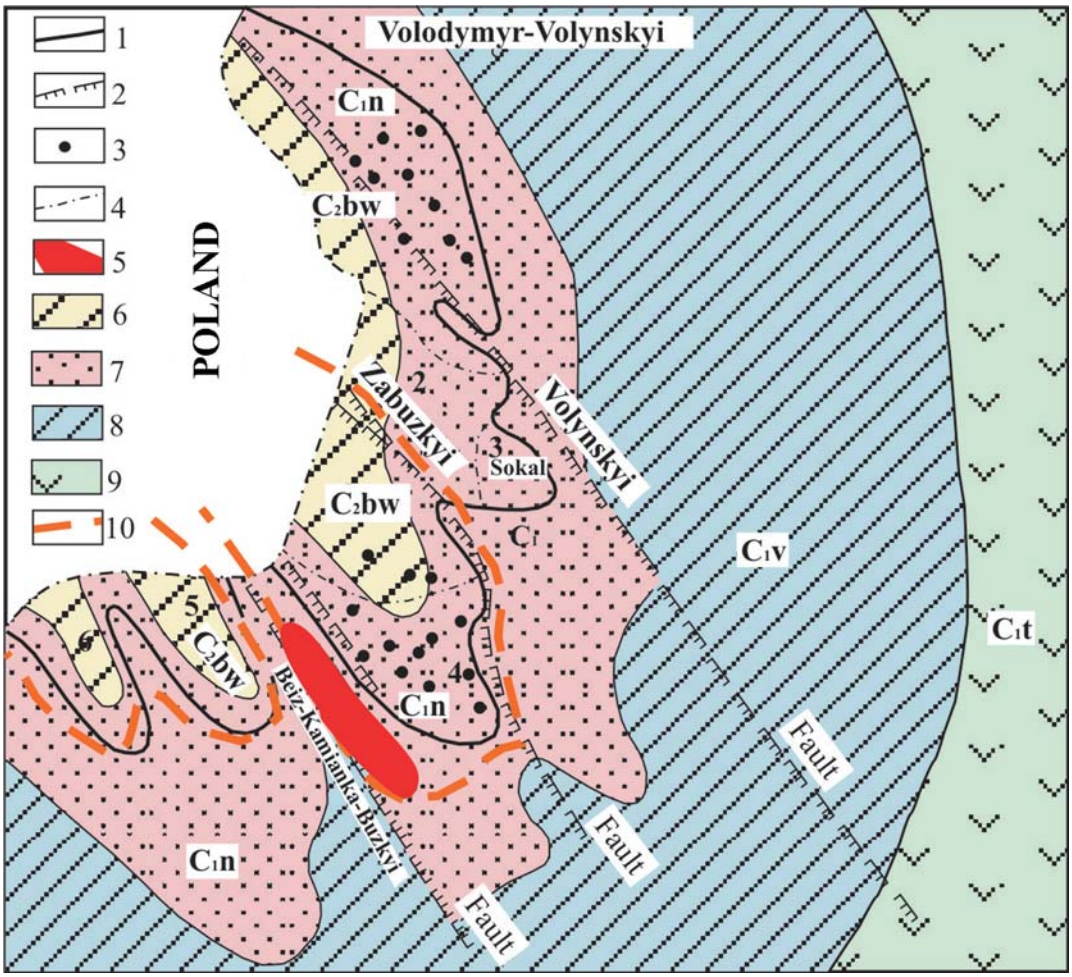

Fig. 3. Geological map of the Lviv-Volyn Coal Basin (modified after Kryvtsov, [10])
Legend: 1 – coal seam n_7^n; 2 – faults; 3 – coal mines; 4 – contour of deposits; 5 – Velyki Mosty gas deposit. Layers: *6* – Bashkirian; *7* – Namurian; *8* – Visean; *9* – Tournaisian. Hard coal deposits: *1* – Volynske; *2* – Zabuzke; *3* – Sokalske; *4* – Mezhyrichenske; *5* – Tiahlivske; *6* – Karivske; 10 – contour of possible gas columns

heavy isotopes of H, He, N, Hg), 2) zone of coal (^{12}C:^{13}C=99,0; 1H/D˜8300), 3) zone of bitumen (^{12}C:^{13}C=90,0–91,0; mainly light isotopes of N, S, Hg). Upper part of coal-bearing sequence will contain more disperse carbohydrates deposits or manifestations. Namely they can be the main source of gas components migration to coal mines.

Secondary halo or gas column has more wide distribution within rocks of coal-bearing sequence. It is possible distinguish the gas columns by indicator characteristics (Figure 5), which include geochemical investigation of gas compositions, isotopic search of C and D from methane on separated depth levels sampling, etc. Most fine indicator which can mark limits of the gas column is Hg (Figure 5). In this sense there is a big similarity between secondary alteration of ore deposits and gas column for oil, gas, and coal deposits.

3.2. Isotopic signatures of the halos in coal-bearing sequences

Generalizing of collected and published the $\delta^{13}C$ data from methane gives temperature limits for methane generation (Figure 6). Limits of data from 242 gas deposits

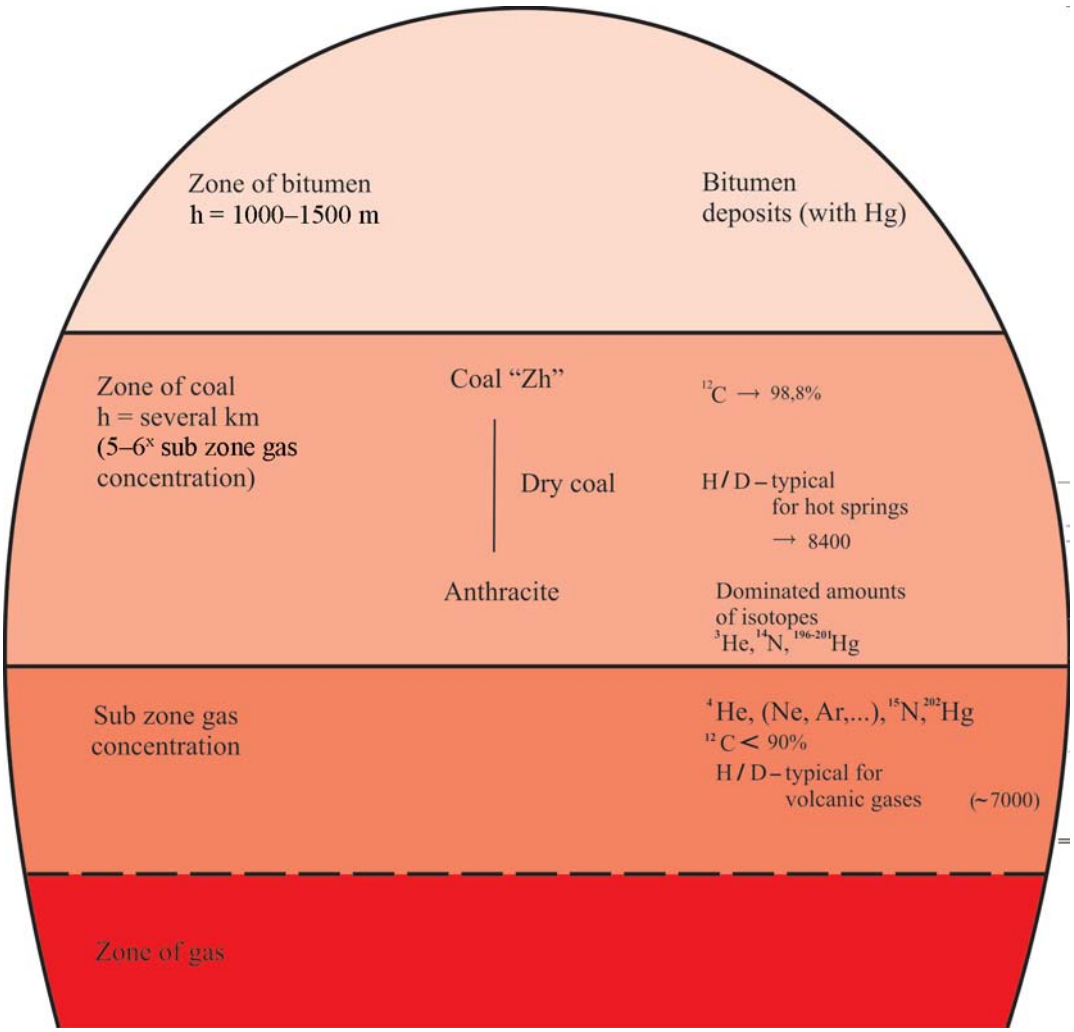

Fig. 4. Separate parts and characteristics of the dispersion halo

of the former USSR and theoretical curve in connection with data from main gigantic gas deposits of the world determine field for $\delta^{13}C$ values, which are typical for mantle methane. We found more isotopically heavy composition of the methane carbon at the more depth levels. It is in accordance with tendency which determined for other regions [4, 7, 8, 19]. General nature of oil and gas according these data is finding as deep originated under its migration along faults and crushed zones.

4. Results and discussion

4.1. Main peculiarities of methane migration in the Ukrainian Coal Basins

We used these noted indicators to show possible mechanism of hydrocarbons migration and concentration in sedimentary sequences of the Donbas and Lviv-Volyn Coal Basins. We created a number of geological, geochemical, and isotopic models on possible origin and migration of methane to determine main zones of the Earth crust and local structures, which can indicate general source of the gas deposits within coal bearing sequences. Geochemical and isotopic peculiarities of evolution of hydrocarbon and accessory gases and some indicator chemical elements in different zones of the Earth for the Donbas region are established (Table 1, 2). Main temperature and pressure limits of hydrocarbon concentrations within these zones as well as inside primary and secondary dispersion halos are determined. As a result we suggested separate levels within coal-bearing sequences for studied regions. Possible stratigraphic levels of gas column

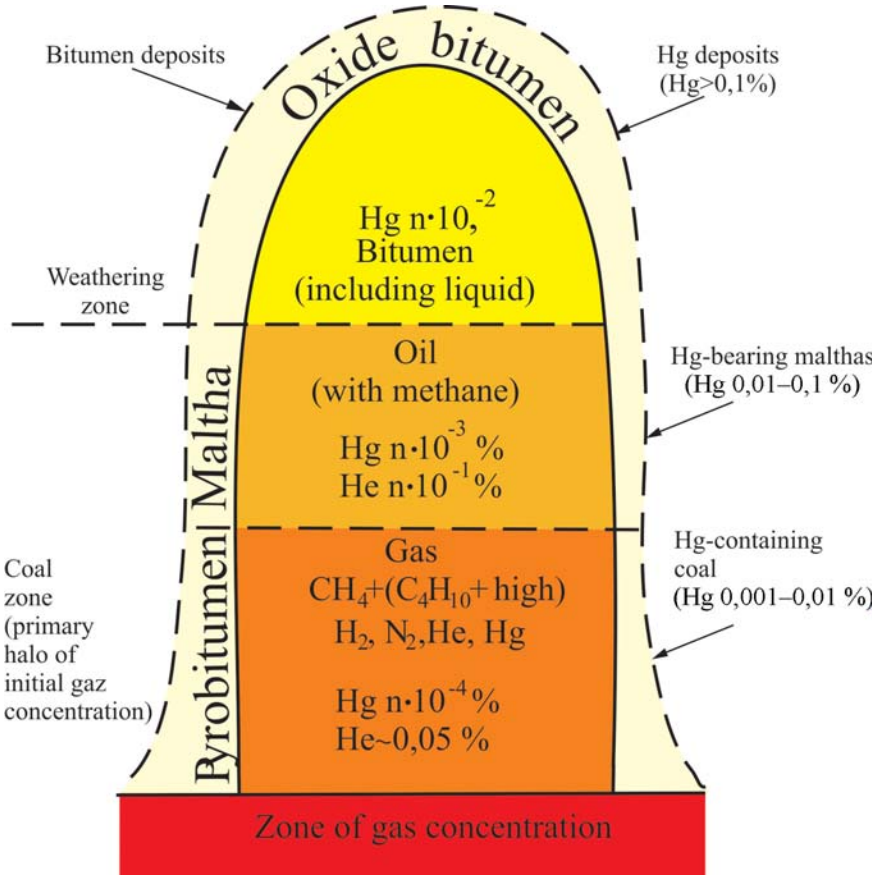

Fig. 5. Indicator parts and characteristics of the secondary halo (gas column)

locations have been found for two kinds of orogenies in the Donbas (Table 1, 2).

4.2. Gas columns and determining of methane origin in the Donbas and Lviv-Volyn Coal Basins

We examined the Donbas Coal Basin and discovered at least two areas with clear evidences on presence of the gas columns (Figure 7). They are located in the most dangerous parts of the Donbas, where methane explosions in coal mines happened very often. Due to more detailed studies on these areas we can determine relationships between structures, coal sorts, gas columns and find possible way to the primary gas deposits. For the western part of the Donetsk-Makiivka coal district we created a map (Figure 8) to determine a main tendency of gas columns development.

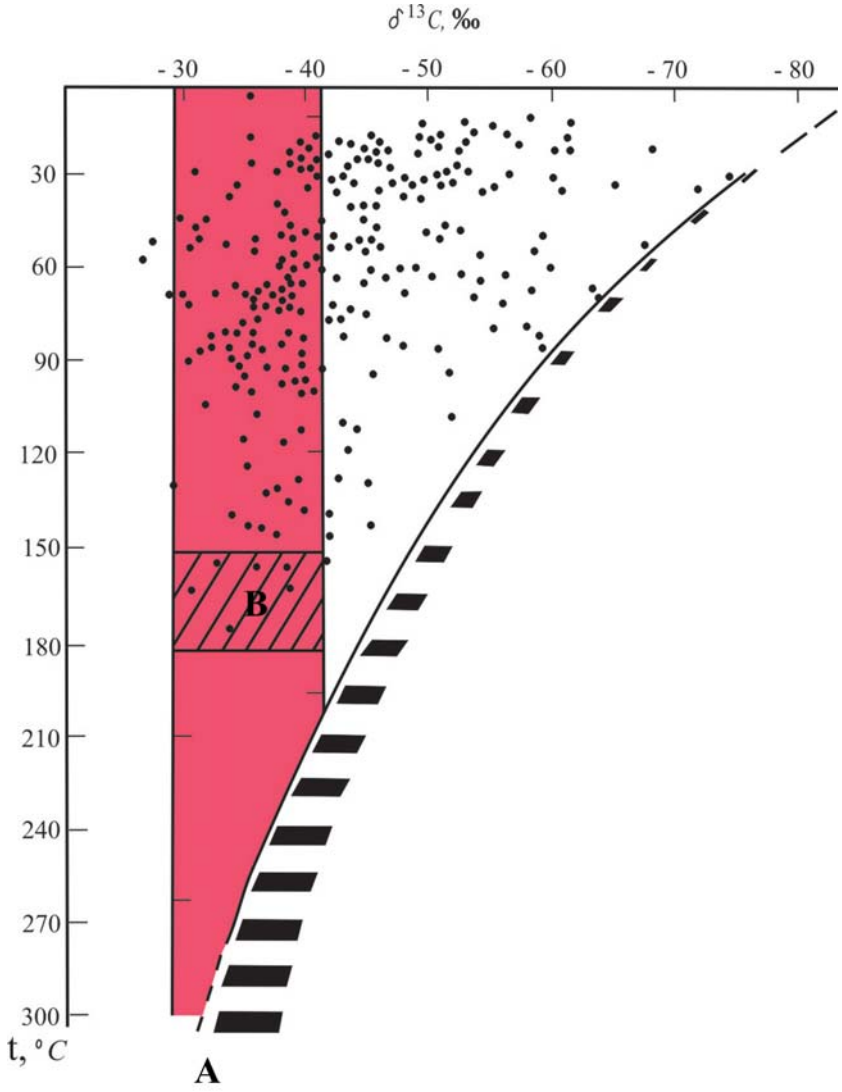

Fig. 6. $\delta^{13}C$ values vs. temperatures of methane generation (modified after [16, 22, 24, 25])
A – Limits of data from 242 gas deposits of the former USSR and theoretical curve; B – Data from main gigantic gas deposits (hosting rocks of the C – P ages); red lines are limits for $\delta^{13}C$ values, which are typical for mantle methane

Table 1. Migration and concentration of gas in sedimentary sequence of the Donbas in Hercynian orogeny

<table>
<tr><th colspan="2">Geospheres</th><th colspan="2">Processes of formation and migration of gas components</th><th>PT conditions of processes</th><th>Isotopic composition gas components (averages)</th></tr>
<tr><td rowspan="5">Crust</td><td rowspan="3">$C_1v - C_3ks$</td><td rowspan="2">Halo</td><td>Coal (sorts A (HA, T), argillites, limestones. Small gas deposits</td><td>P – 500 MPa
t° – 230–300 °C</td><td rowspan="5">$^3He/^4He$ = $n\cdot10^{-5} - n\cdot10^{-8}$

$\delta C^{13} \approx -35$ ‰</td></tr>
<tr><td>Argillites, sandstones C_1v</td><td rowspan="2">P ~ 700 MPa
t° – 300 °C</td></tr>
<tr><td colspan="2">Limestones C1v.
Maine gas deposit
(m ~ 800 m)

CH_4 – up to 90 %,
N_2~5 % HHC – 3 %,
CO_2 – 2 % (He,Hg))
Depth 7,5 km</td></tr>
<tr><td>Riphean – C_1t</td><td colspan="2">↑ ↑ ↑
↑ ↑
CH_4+HHC N_2
CO_2 He… Hg
Effusives, salt, sandstones, limestones
Formation of heavy homologs of methane
($CO+CH_2 \rightarrow C_mH_n+H_2O$)
Depth 22 km</td><td>P ~ 800 MPa

t° – 500 °C</td></tr>
<tr><td>Pre-cambrian</td><td colspan="2">Granite part
(basement)
Depth 33 km</td><td>P ~ 900 MPa
t° – 520–570 °C</td></tr>
<tr><td colspan="2">Mantle</td><td colspan="2">↑ ↑ ↑
CH_4 N_2 et al. He...

Zone of gas concentration</td><td>P ~ 1000 MPa
t°~600 °C</td><td>$^3He/^4He = n\cdot10^{-5}$
$\delta C^{13} \approx -15$ ‰</td></tr>
</table>

Note. Created with additional data after [1, 2, 6, 17, 20, 23, 25]
* Thickness of Carboniferous sequence – 7,5 km (average).

Similar methods and indicators we used for analysis of the Lviv-Volyn Coal Basin. In addition to the Velyki Mosty gas deposit clear gas column has been detected within Mezhyrichenskyi coal district (Figure 9). From geological map (Figure 3) we can see other possible gas columns. The northern-eastern end of these columns is located, probably, in the Lublin Hard Coal Basin, which is the Polish analogous of the Lviv-Volyn Coal Basin.

5. Conclusions

Analysis of problems of methane origin in coal bearing sequences gives a chance to determine possibility of methane utilization and creation of safety conditions during un-

Table 2. Hydrocarbon concentrations in the Donbas in Mesozoic and Cenozoic orogenies

Sequences	Hydrocarbon deposits and hosting rocks of gas column	PT conditions of deposits formation
Coal sequence of Carboniferous	Sendstones, limestones, Seam coal : Sorts $B_1 - B_2$ Gas appearance Halo (thermolithic gas zone) G Gas deposits CC, C, Zh Oil deposits A, HA, T Condensate-gas deposits Gas. Main deposit Depth 5,5 km	P~100 MPa t°~ 50–70 °C ↑ P ~300 MPa t° ~ 200 °C
Effusive-sedimentary sequence Rephean –Tournaisian Crystalline basement	↑ ↑ ↑	
Mantle	↑ ↑ ↑ CH_4+HHC N_2 He, Hg	

Note. Created with additional data after [2, 12, 17, 23].

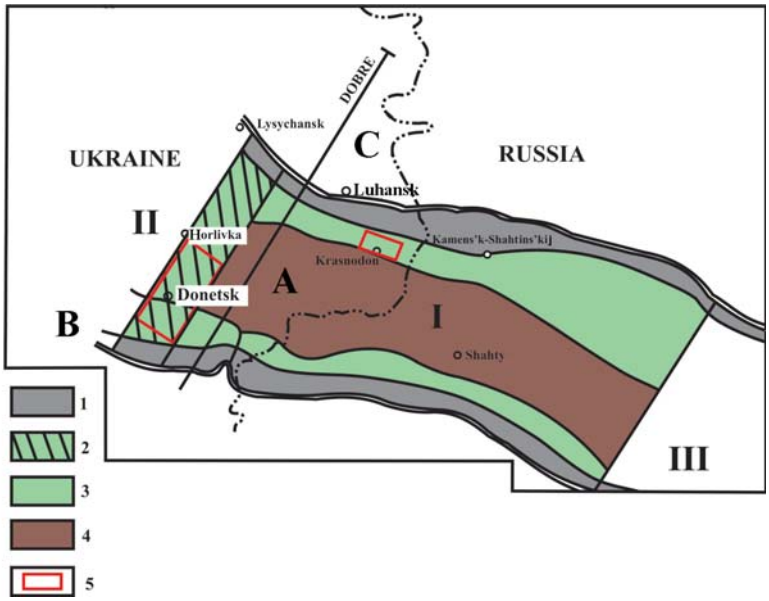

Fig. 7. Main tectonic structures of the Donbas
A – Don-Dnipro Sag (part of the Karpinskyi Lineament), B – Ukrainian Precambrian Shield, C – Voronezh Anteclise. I – Donetsk Folding Structure; II – Donetsk-Dnipro Depression; III – Kalmyk Uplift. Legend: 1 – zone of faults of the Karpinskyi Lineament; 2 – zone of the Sologub-Chekunov Mantle Fault; 3 – rocks of the Mz-Kz tectonics; 4 – Anthracite Massif; 5 – areas of possible gas columns

derground mining of coal. It is shown that previous idea about limited volumes of methane in coal bodies needs improving because there is evidence on deep origin of methane and its continuous addition.

Several important geological, geochemical and isotopic facts support a nontraditional point of view for the Ukrainian Coal Basins about necessity to accept a new method to determine an origin of methane with receiving new precise data at local levels.

Searching gas deposits in the Donbas and Lviv-Volyn Coal Basins has lately become the problem of current importance as a result of methane explosions in coal mines, which happen very often. It's used to be believed that methane accumulations were only because of coal strata degasification, but now this concept is inefficient to take preventive measures in underground mining. Other processes leading to high gas concentration and first of all its influx into mines from depths should be reviewed. One of the variants could be detection of an independent deposit containing large gas resource with big scale desorption from more disperse hydrocarbon deposits below the coal strata. Namely they can be the main source of gas migration to the coal mines. Discovery of main gas deposits, which are the main source for new portions of methane within coal-bearing sequences, will improve supplying by domestic fuels and safety of coal mining.

As we see, suggested way with exploring of gas columns is convenient instrument in struggle against the methane pollution out of mining areas.

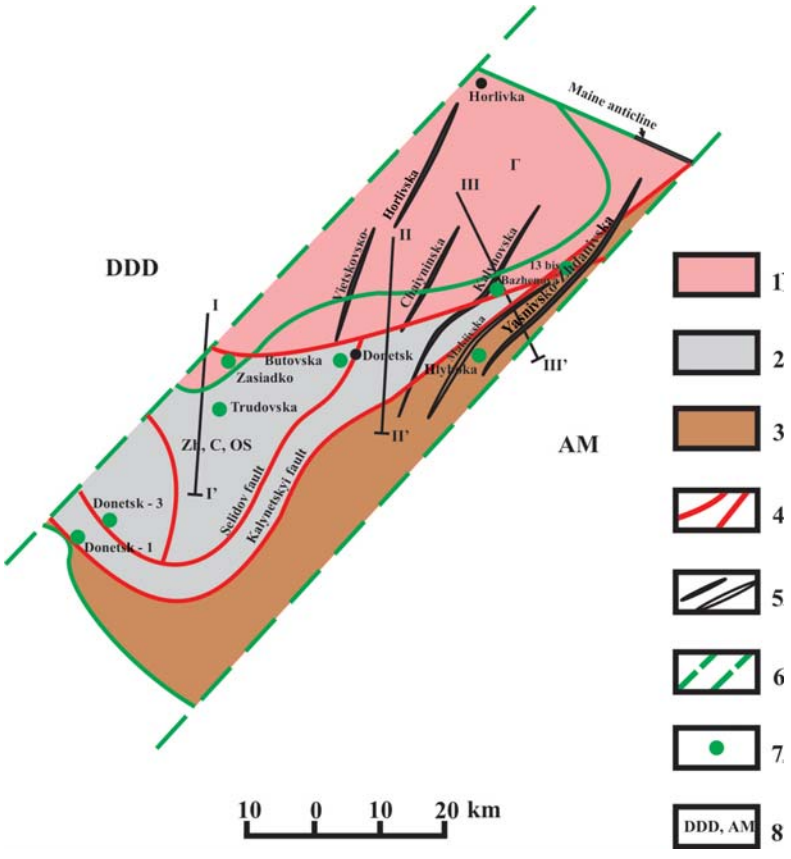

Fig. 8. Structural sketch of the Western part of the Donetsk-Makiivka Coal District
Legend: 1–3 – distribution of different sorts of coal; 4 – faults; 5 – axis of folds (a – anticline, b – syncline); 6 – limits of the Sologub-Chekunov Mantle Fault; 7 – coal mines; 8 – fragments of the Karpinskyi Lineament: DDD – Donetsk-Dnipro Depression, AM – Anthracite Massif

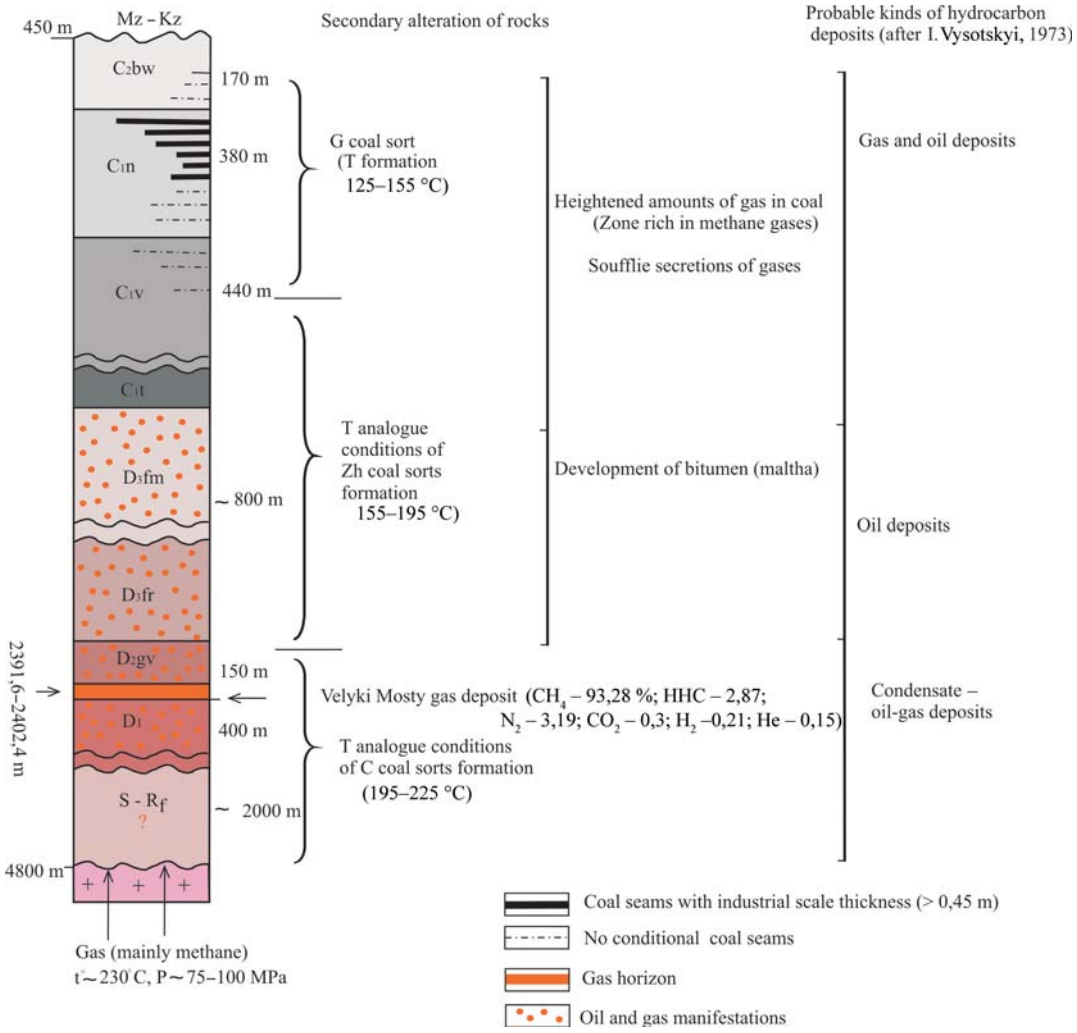

Fig. 9. General picture of the gas column within sedimentary sequence of the Mezhyrichenskyi coal bearing district (Lviv-Volyn Coal Basin)

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ГЕОЛОГІЧНІ, ГЕОХІМІЧНІ ТА ІЗОТОПНІ МОДЕЛІ КОНЦЕНТРАЦІЙ МЕТАНУ В УКРАЇНСЬКИХ ВУГІЛЬНИХ БАСЕЙНАХ

Проблеми походження метану в багатих на вугілля геологічних утвореннях визначають можливості утилізації метану і створення безпечних умов підземного вуглевидобутку. На сьогодні немає єдиної думки щодо природи метану у вугленосних товщах Донецького і Львівського вугільних басейнів. Згідно з популярними ідеями метанові поклади утворилися внаслідок трансформації первинного органічного матеріалу, а тому їхні сучасні ресурси визначаються загальною кількістю вихідного органічного матеріалу. Роз'яснено, що цей погляд потребує вдосконалення, оскільки є докази первинного глибинного походження метану і постійного поповнення його ресурсів, можливо у вигляді унікальних за об'ємами покладів, що розміщені під вугленосними товщами. Геохімічні індикатори та особливості ізотопного складу деяких елементів і вуглеводнів, а також загальна еволюція газів у різних зонах підрозділів з вугіллям Донбасу та Львівсько-Волинського басейнів під час герцинської й мезозойської тектонічних епох використано для створення нової моделі походження метану та його міграції. Визначено головні обмеження температур і

тисків для концентрацій вуглеводнів усередині цих зон, які визначають метанові поклади та можливості їхньої майбутньої утилізації і створення безпечних умов вугледобування.

Ключові слова: *метан, вугільні родовища, Україна, стабільні ізотопи.*

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ГЕОЛОГИЧЕСКИЕ, ГЕОХИМИЧЕСКИЕ И ИЗОТОПНЫЕ МОДЕЛИ КОНЦЕНТРАЦИЙ МЕТАНА В УКРАИНСКИХ УГОЛЬНЫХ БАССЕЙНАХ

Проблемы происхождения метана в богатых углем геологических образованиях определяют возможности утилизации метана и создания безопасных условий подземной угледобычи. На сегодня нет единого взгляда на природу метана в угленосных толщах Донецкого и Львовского угольных бассейнов. Согласно популярным идеям метановые скопления образовались в результате трансформации первичного органического материала, а поэтому их современные ресурсы определяются общим количеством исходного органического материала. Показано, что эта точка зрения нуждается в усовершенствовании, поскольку имеются доказательства первичного глубинного происхождения метана и постоянного пополнения его ресурсов, возможно в виде уникальных за объемами залежей, которые расположены под угленосными толщами. Геохимические индикаторы и особенности изотопного состава некоторых элементов и углеводородов, а также общая эволюция газов в разных зонах толщ с углем Донбасса и Львовско-Волынского бассейнов во время герцинской и мезозойской тектонических эпох использованы для создания новой модели происхождения метана и его миграции. Определены основные ограничения температур и давлений для концентраций углеводородов внутри этих зон, которые определяют метановые залежи и возможности их будущей утилизации и создания безопасных условий добычи угля.

Ключевые слова: *метан, угольные месторождения, Украина, стабильные изотопы.*