GEOLOGIJA. 2008. Vol. 50. No. 2(62). P. 88–93
DOI: 10.2478/v10056-008-0012-7

Chemical composition of oils from recently discovered fields in West Lithuania

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Zdanavičiūtė O., Dakhnova M., Kleinas A. Chemical composition of oils from recently discovered fields in West Lithuania. *Geologija*. Vilnius. 2008. Vol. 50. No. 2(62). P. 88–93. ISSN 1392-110X

Four minor oil discoveries have been made in West Lithuania in recent years. Studies of the oil composition show that its physical and chemical properties (density, viscosity, petrol content, etc.) and the group composition of hydrocarbons (content of saturated and aromatic hydrocarbons, tars and asphaltenes) mainly depend on the formation conditions and distances of migration between the kitchen and accumulation areas. According to the distribution patterns of *n*-alkanes and isoprenoids, the examined oils are comparable and generated from sapropel organic matter. There are certain differences in biomarker and carbon isotope data, indicating oil generation from different source rocks containing organic matter of different catagenesis.

Key words: West Lithuania, oilfield, oil composition

Received 7 March 2008, accepted 8 April 2008

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INTRODUCTION

The majority of oil fields in West Lithuania were discovered during the Soviet era more than 30 years ago. In recent years, only minor oil discoveries have been made, all located in a close vicinity to producing fields (Fig. 1). Two small oilfields, Agluonėnai and Uoksai, were discovered in the license block operated by the JSC "Minijos nafta". Two others, Šiaurės Vėžaičiai and Žadeikiai, were found within the license block of the JSC "Manifoldas".

This paper discusses the physical properties, chemical composition, and carbon isotope analyses of oils obtained from the newly discovered fields. Oil samples for laboratory analyses were collected under surface conditions and supplemented with the analysis of a downhole oil sample from the Antkoptis-1 well.

GEOLOGICAL SETTING

The area is situated in the eastern part of the Baltic Syneclise, within the prominent tectonic zone referred to as the Gargždai Zone of Elevations (Stirpeika, 1999). Its structural pattern comprises a NNE-SSW trending fault system and related anticlines serving as traps for hydrocarbon accumulations. Oilfields discovered so far produce from low porosity Cambrian sandstone (and occasionally from Lower Ordovician sandstone) at a depth of around 2000 m.

Recent oil discoveries in the area are associated with small structural traps that were found by detailed seismic surveys near the producing fields. Their location is shown in Fig. 1.

The Agluonėnai oilfield was discovered in 2005, approximately 2 km west of the Šiūpariai and Petų Šiūpariai oilfields.

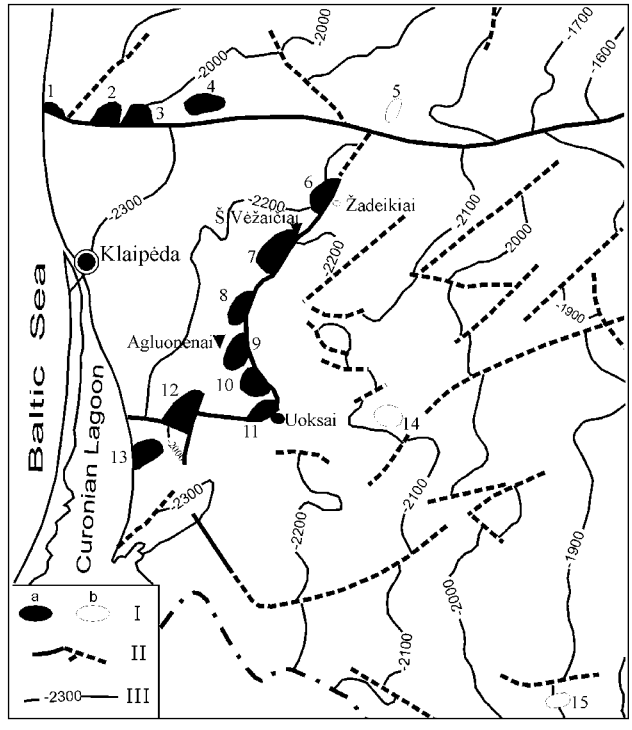

Fig. 1. Sketch map of the distribution of oilfields in the Early Palaeosoic deposits of Lithuania. I – oilfields: a) producing; b) non-producing; *1* – Girkaliai, *2* – Genčiai, *3* – Kretinga, *4* – Nausodis, *5* – Plungė, *6* – Ablinga, *7* – Vėžaičiai, *8* – Šiūpariai, *9* – Pietų Šiūpariai, *10* – Degliai, *11* – Pociai, *12* – Vilkyčiai, *13* – Sakučiai, *14* – Šilalė, *15* – Lauksargiai; II – faults; III – isolines of basement

1 pav. Naftos telkinių paplitimo Lietuvos apatinio paleozojaus dariniuose Lietuvoje schema. I – naftos telkiniai: a) eksploatuojami; b) neeksploatuojami; *1* – Girkaliai, *2* – Genčiai, *3* – Kretinga, *4* – Nausodis, *5* – Plungė, *6* – Ablinga, *7* – Vėžaičiai, *8* – Šiūpariai, *9* – Pietų Šiūpariai, *10* – Degliai, *11* – Pociai, *12* – Vilkyčiai, *13* – Sakučiai, *14* – Šilalė, *15* – Lauksargiai; II – tektoniniai lūžiai; III – kristalinio pamato izolinijos

Oil accumulation occurs in a horst-like structure within the Middle Cambrian and Lower Ordovician successions (Fig. 2). The reservoir is sealed by faults laterally and by overlying shales. The productive interval is composed of inter-bedding sandstone and siltstone with effective porosities up to 7.6% and permeability reaching 37 mD.

The Uoksai oilfield was discovered in 2004 close to the Pociai oilfield. It is associated with a small compactional anticline formed above the basement high (Fig. 2). The productive interval is of poor reservoir properties. Only the uppermost sandstone layer within the Middle Cambrian sequence is considered as a reservoir. Its thickness is around 6 m and average porosity 6.1%.

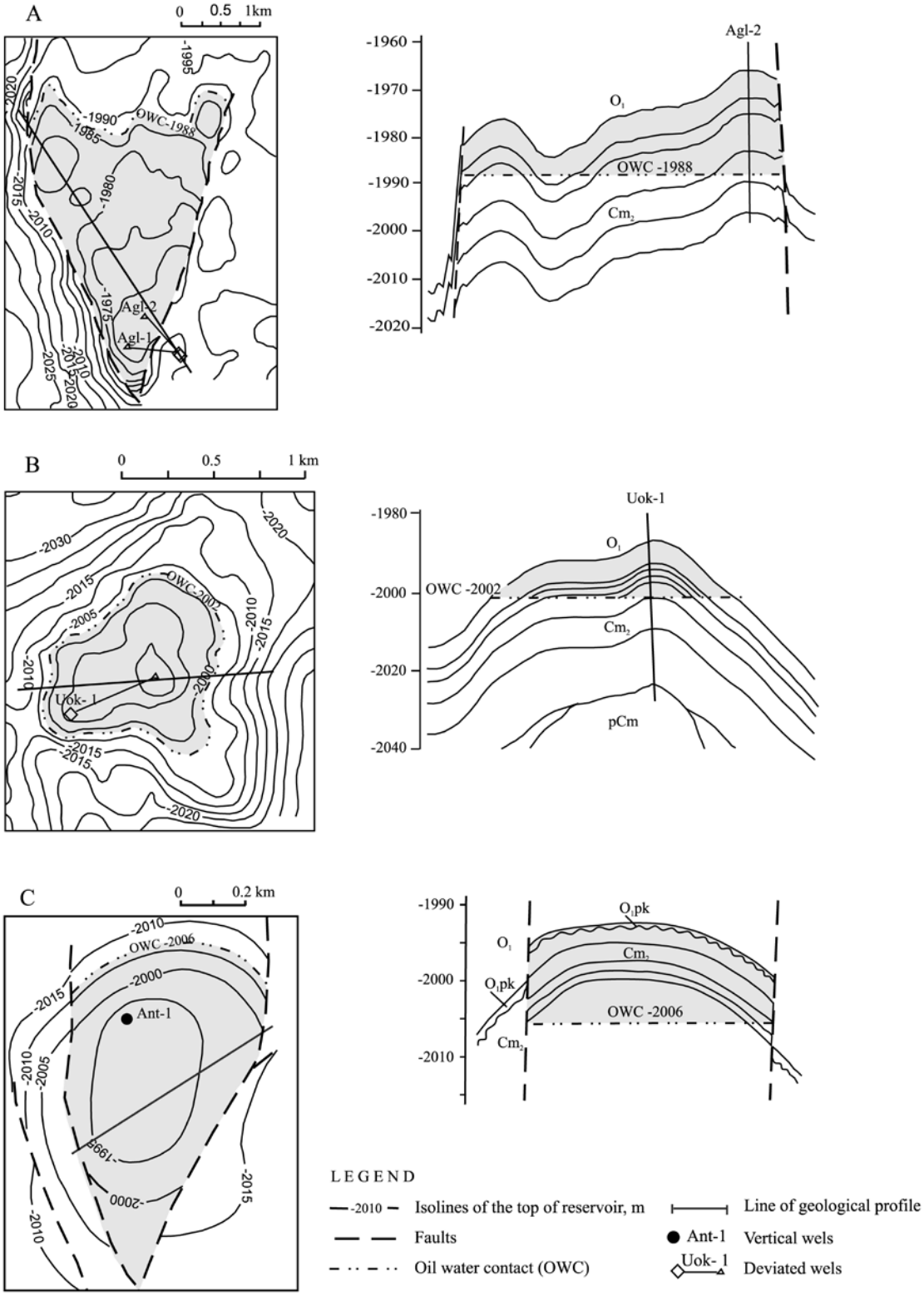

Fig. 2. Sketch structural maps of the top of the reservoir and geological profiles of Agluonėnai (*A*), Uoksai (*B*) and Šiaurės Vėžaičiai (*C*) oilfields

2 pav. Agluonėnų (*A*), Uoksų (*B*) ir Šaurės Vėžaičių (*C*) naftos telkinių kambro kraigo schematiniai struktūriniai žemėlapiai ir geologiniai profiliai

The Šiaurės Vėžaičiai oilfield was discovered in 2005. It is associated with a small structural trap located to the north of the Vėžaičiai oilfield. The reservoir properties of Cambrian sandstone and siltstone show a wide range, with porosities measured from 4.45% to 9.05% and permeabilities ranging from a few mD to 116 mD. The average porosity of the pay zone is 6.26% and the average permeability 13.8 mD (Žvirblis et al., 2006).

The Žadeikiai oil discovery is most recent in the region (2007). Oil is identified in the top of the Žadeikiai structure located between the Vėžaičiai and Ablinga oilfields. The reservoir comprises Cambrian and Lover Ordovician sandstone with a low porosity and the netpay thickness of only 0.8 m.

MATERIALS AND METHODS

Core samples from the Agluonėnai-2, Antkoptis-1, Uoksai-1 and Žadeikiai-1 wells were analysed at All-Russian Research Geological Oil Institute (VNIGNI): the physical and chemical properties of oil and the group composition of hydrocarbons were determined. Analysis of crude oil, petrol (b.p.–150 °C) and saturated oil fractions was carried out by the method of gas chromatography. Two samples were examined for relict hydrocarbons (biomarkers) and for carbon isotope composition.

Gas chromatography (of crude oil and saturated hydrocarbons) was performed using a Hewlett Packard chromatograph of HP-5890 series with a flame-ionization detector (FID) and a high performance quartz capillary column (60 m long and 0.32 mm i.d).

Gas chromatography and mass spectrometry (GS-MS, analysis of biomarkers) analyses of saturated and aromatic petroleum hydrocarbons were carried out using a coupled CLARUS 500 chromatograph–mass-spectrometer (Perkin Elmer). The analysis conditions were the following: the insertion of samples was performed using a sampler; helium was used as a gas carrier; injection pressure was 30 psi; helium flow velocity was 2 ml/min at 100 ºC. The HP-1701 column was 60 m long, 0.32 mm i.d., 25 µm film thicknesses. The temperature regime for analysis used the following heating steps in the thermostat column: 2 min up to 100 °C, 12.5 °C/min from 100 °C to 150 °C, 3 °C/min from 150 °C to 300 °C, at 300 °C isotherm 14 min, the operating regime of the injector "split / splitless". The column of the gas chromatographer was directly inserted into the MS ion source. The SIR mode was used for mass spectrometry.

The parameters of biomarkers were determined; m/z 191 (tricyclic and pentacyclic triterpanes), m/z 217 (steranes), m/z 218 (isosteranes), m/z 259 (diasteranes), m/z 231 (triaromatic steranes), m/z 253 (monoaromatic steranes). The peak areas of mass fragmentograms were calculated using Perkin-Elmer Total Chrom Client / Server program.

Carbon isotope composition was determined with a VG OPTIMA isotope mass-spectrometer (Company Fisons). Accuracy to size was ±0.5‰.

Two reservoir oil samples from the well Antkoptis-1 were examined at the laboratories of the Lvov branch of the Ukrainian State Geologic Exploration Institute (UkrNIGRI).

RESULTS AND DISCUSSION

The Cambrian oils within Lithuania have moderate gravities (800–830 kg/m^3), low viscosity (5–16 µm^2/s), contain a high amount of petrol fractions (more than 30% volume) and a low content of tars (max up to 15%), asphaltenes (below 3%) and sulphur (less than 0.25%). The variations of the mentioned parameters are predetermined by the present depth of reservoir occurrence. The density, viscosity, content of tars and asphaltenes in oils increase with shallow depths (Zdanaviciutė et al., 1997, 2004, 2007). The data show that the oil from the newly discovered oilfields is also rather light (803.6–814.8 kg/m^3) and of low viscosity (6–7.41 µm^2/s) except oil from the well Žadeikiai-1 (Table 1). The boiling point of oil from the wells Uoksai-1 and Antkoptis-1 is very high – 85 and 78 °C, respectively, instead of the common 50 °C, showing a lower contain of petrol fractions in these oils. It is not typical of the Cambrian oils of West Lithuania. Perhaps it is related with sampling, transportation, storing and examination conditions of the oil samples. The oil from well Žadeikiai-1 is heavier. Its density reaches 853.2 kg/m^3 (kinematic viscosity 20.03 µm^2/s) what is not characteristic of the Cambrian and usual of the Ordovician oils of Lithuania (except oil from the well Gondingas-1, Plungė oilfield, where the density reaches 870.0 kg/m^3). The boiling point of oil from the well Žadeikiai-1 is 66 °C; the content of petrol fraction (b. p.–200 °C) is low only 20%.

Maltene fractions by were dominated saturated compounds (57–72.9%). Commonly, their concentrations are higher in lighter oils, therefore saturated hydrocarbons in the considerably heavier oil from the Žadeikiai oilfield reach up to 43.4%. The concentration of aromatic hydrocarbons varies negligibly, except oil from the well Žadeikiai-1 here the concentration of saturated hydrocarbons reaches up to 39.0%. The rate of saturated aromatic hydrocarbons is 1.11–3.26. Asphaltene contents were moderate and ranged from 1.2 to 4% (Table 1).

The *n*-alkane distribution was unimodal, i. e. dominated by n-C_{11}–n-C_{15} with the maxima at n-C_{15} and a market reduced abundances of n-alkanes in the range C_{20}–C_{35} (Fig. 3). The distribution of *n*-alkanes in oil from the well Žadeikiai-1 has the maximum at C_{13} (the common maximum for oils from other oilfields is C_{15}) and a markedly lower concentration of heavy hydrocarbons, indicating a shorter migration path. The proportions of acyclic isoprenoids relative to *n*-alkanes were generally low, and their ratio was stable (0.14–0.22). The pristine and phytane ratio is very stable and varies from 2.37 to 2.67, perhaps indicating a more oxic depositional environment (Zdanaviciute and Bojesen-Koefoed, 1997). The ratio of odd and even *n*-alkanes (CPI) calculated for C_{23}–C_{33} *n*-alkanes is close to unity. The higher pristane/n-C_{17} and phytane/n-C_{18} ratios in oil from well Žadeikiai-1 indicate a lower maturity (Table 2).

The Lithuanian oils, like many Palaeozoic oils, are characterised by very low concentrations of sterane and triterpane with a similar distribution. In general, they are characterised by prominent tricycle triterpanes, comparatively low hopane to sterane ratios and low proportions of extended hopanes.

The concentration of steranes and triterpanes in the examined samples is also very low. The distribution of regular steranes is

Table 1. **Physical properties and chemical composition of oils**
1 lentelė. **Fizinės ir cheminės naftos savybės**

Well	Sampling interval, m	Age	Gravity, kg/m^3	Kinematic viscosity, 10^{-6} m^2/s	Saturated vol. %*	Arom. vol. %*	Polars vol. %*	Saturated / Arom.	Asphaltene, %	Boiling point, C° (b. p.)	Fraction b. p.-150 C°, volume %	Fraction b. p.-200 C°, volume %
Agluonėnai-2	1996–2030	Є$_2$dm	803.6	6.0	57.0	29.0	10.0	1.96	4.0	43	30	45
Uoksai-1	2187–2200	Є$_2$dm	814.8	7.09	65.6	23.9	8.3	2.74	2.2	85	18	30
Antkoptis-1	1900–2003	Є$_2$dm	814	7.41	72.9	22.3	5.1	3.26	–	78	16	24
Žadeikiai-1	1985–1992	O$_1$pk	853.2	20.03	43.4	39.0	13.4	1.11	1.2	66	14	20

* Calculated for petrol-free fractions (b. p. 200 °C).

Table 2. **The ratios of *n*-alkanes and isoprenoides for saturated fractions**
2 lentelė. **N-alkanų ir izoprenoidų sočiosiose naftos frakcijose sudėties geocheminiai rodikliai**

Well	Sampling interval, m	PR / PH	PR / nC$_{17}$	PH / nC$_{18}$	Isoprenoides / n-alkanes, %	Isoprenoides, %	N-alkanes, %	CPI nC$_{23}$-nC$_{33}$
Agluonėnai-2	1996–2030	2.67	0.55	0.24	0.14	7.08	50.71	1.04
Uoksai-1	2187–2200	2.64	0.63	0.27	0.18	9.19	51.09	1.0
Antkoptis-1	1900–2003	2.59	0.58	0.25	0.17	8.35	49.56	1.0
Žadeikiai-1	1985–1992	2.37	0.82	0.38	0.22	9.94	45.02	1.0

PR/PH – pristane/phytane ratio;
PR/n-C$_{17}$ – pristane/*n*-heptadecane ratio;
PH/n-C$_{18}$ – phytane/*n*-octodecane ratio;
CPI – Carbon Preference Index calculated as $2 \times \text{sum}\ (n\text{-}C_{23-33})/2 \times \text{sum}\ (n\text{-}C_{24-30}) + n\text{-}C_{22} + n\text{-}C_{32}$.

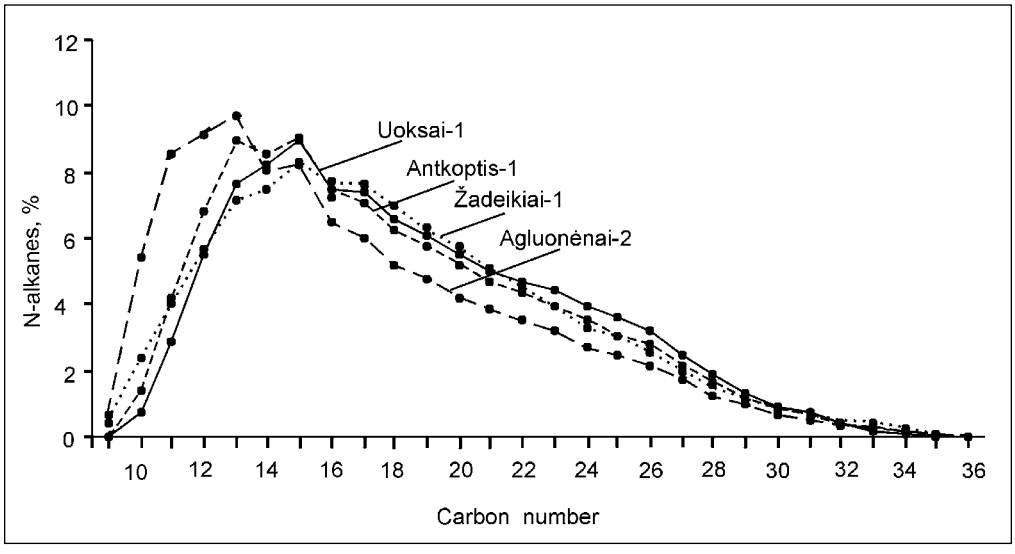

Fig. 3. Plot of the distribution of *n*-alkanes in Cambrian oils
3 pav. N-alkanų paplitimo kambro naftoje grafikas

very similar for both samples, showing approximately equal proportions of C_{28} and C_{29} sterane species and large proportions of C_{27} (Table 3). The hopane / sterane ratios are low (0.3–1.7). The data obtained allow an assumption that the oil was generated by type II organic matter of composed of plankton, algae and detrital bacterial material.

According to carbon isotopic composition, the analysed oils are rather comparable: enriched with the light isotope $\delta^{13}C$ whose concentrations reaches –30.57‰ in the oil from well Agluonėnai-2, –31.22‰ in the oil from well Uoksai-1 and –29.34‰ in the oil from well Žadeikiai-1. This is a characteristic feature of the Lower Palaeozoic oils.

Data of investigation of reservoir oil samples from the well Antkoptis-1 were presented in Table 4. The data show that according to physical properties, oil from the Antkoptis-1 well is comparable with oils from other Lithuanian oilfields. Its density at a pressure of 21.3 MPa is 764.0–766.6 kg/m^3 (for oil of the zone of Gargždai elevations this parameter is 727–781 kg/m^3) and gas / oil ratio (GOR) 42.8–43.9 m^3/t (for the oils of the Gargždai elevations zone 42.5–63.8 m^3/t).

As implied by the data on pristane and phitane, diasterane C_{27} and regular sterane C_{27}, moretane and hopane, tricyclic hydrocarbon T_{23} and hopane ratios, the properties of Cambrian oil are more similar to those of bitumoides of the

Table 3. **Sterane and triterpane biomarker ratio**
3 lentelė. **Steranų ir triterpanų biožymenų santykiai**

Well	DIA27 / RST27	RST29 (S/S + R)	RST29 (ββ/ββ +αα)	RST27, (%)	RST28, (%)	RST29, (%)	X/H30	H29 / H30	Ts / Tm	M30 / H30	H32 S / (S + R)
Agluonėnai-2	2.31	0.52	0.6	24.8	24.8	49.4	0.77	0.55	3.08	0.07	0.48
Žadeikiai-1	2.21	0.54	0.6	21.4	23.3	55.2	0.69	0.36	2.53	0.19	0.52

Sterane biomarker ratios:
DIA27 / RST27 – (sum C_{27} diasteranes) / (sum C_{27} regular steranes);
RST29 (S/S + R) – regular sterane C_{29} epimerization ratio;
RST29 (ββ / ββ + αα) – regular steranes C_{29} ratio of ββ-epimers to total;
RST27, RST28, RST29 – reguliar steranes (sum of regular steranes, calculated from ββ epimers in m/z 218 ion fragmentograms).

Triterpane biomarker ratios:
X / H30 – triciklane / hopane;
H29 / H30 – norhopane / hopane;
Ts / Tm – trisnorneohopane / trisnorhopane;
M30 / H30 – moretane / hopane ratio;
H32 S / (S + R) – bishomohopane 22S / (22S + 22R) epimerization ratio.

Table 4. **Main properties of reservoir oil from the Antkoptis-1 well**
4 lentelė. **Antkopčio-1 gręžinio sluoksnio naftos pagrindinės savybės**

Parameter	Sample 1	Sample 2
Bubble point, MPa	3.4	3.4
Specific gravity, kg/m³, at P reservoir	766.6	764
Density at 15°C or 20°C, kg/m³	828.6	827.6
Viscosity, MPa s at P reservoir	1.90	1.78
Formation volume, Bo	1.140	1.144
Shrinkage, %	12.28	12.59
Gas / oil ratio (GOR) : m³/t m³/m³	42.8 35.5	43.9 36.3

Sampling interval 1993–2001 m (two samples); sampling depth 1800 m; reservoir pressure 21.3 MPa; reservoir temperature 72 °C.

Silurian source rocks. Complicated conditions for primary migration are characteristic of the Silurian section; therefore this correlation is far from simple. Establishing the correlation between oil and organic matter of source rocks is a rather difficult task in the Lithuania. The organic matter of the source rocks has a highly similar composition (according to the data of Rock-Eval analysis: sapropelic, type II) and it contains very small amounts of relict hydrocarbons. Considering that the formation of oilfields took a long geological time, the accumulated oil here could have been generated from various source rocks. Source rocks exist within the Cambrian, Ordovician and Silurian successions in Lithuania. The start of oil generation in Lithuania is related to the late Silurian, while the basic oil generation phase is thought to be in the Devonian and Permian times. The low rate of sedimentation in the whole area of the Baltic Syneclise and different movements of the Earth's crust have created conditions for a slow formation or even destruction of previously formed oil fields. The oil fields are well preserved because of favourable hydrological conditions and good sealing conditions of the overlying strata (Sakalauskas et al., 1999; Suveizdis, 1979).

CONCLUSIONS

Detailed geochemical studies of crude oils from the newly discovered oilfields in Lithuania lead to the following generalizations.

The physical and chemical parameters (density, viscosity, content of gasoline fractions) of Cambrian oils and the group composition of hydrocarbons (content of saturated and aromatic hydrocarbons, tars and asphaltenes) mainly depend on the depth of occurrence and the geological situation of the oilfields and on the migration distances of hydrocarbons from the source rocks to the traps and their entrapment.

Gas chromatograms of the alkanes are unimodal, dominated by C_{13}–C_{19} homologs (with the peak maximum at C_{15}), and low amounts of C_{20} and heavier hydrocarbons. According to the distribution patterns of *n*-alkanes and isoprenoides, the examined oils are comparable and generated by sapropel organic mater.

The study oils, like all oils of Baltic Syneclise, are charakterised by very low concentrations of steranes and triterpanes. There are certain differences in biomarker and carbon isotope data, indicating oil generation by different source rocks containing organic matter of different maturity.

The existence of light solid bitumen of various types also indicates a stepwise character of the oil field formation. The low sedimentation rates together with low tectonic activity in the Baltic Syneclise ensured conditions for the slow formation of the oil fields and even a subsequent destruction of already formed ones. The main reservoirs for petroleum accumulations are Cambrian Deimena Formation sandstones which were brought into hydraulic communication with various source rocks occurring in different stratigraphic intervals. During the long time of their formation, the oilfields have been continuously replenished; therefore, the oil traps in the Cambrian rocks could accumulate oil generated not only from the Cambrian, but also from the Ordovician and Silurian source rocks.

ACKNOWLEDGEMENTS

The authors are grateful to the staff of the geochemical laboratory of the All-Russia Research Geological Institute of Oil (VNIGNI) for investigating oil samples, and to the heads of the JSC "Minijos nafta" and "Manifoldas" for kind permission to use their data. We thank Sergei Kanev for providing data and Elena Vikšraitienė for technical support. The reviewer Povilas Suveizdis improved the manuscript and is acknowledged with thanks.

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VAKARŲ LIETUVOJE NAUJAI ATRASTŲ TELKINIŲ NAFTOS CHEMINĖ SUDĖTIS

Santrauka

Naujų naftos paieškos darbų dėka buvo surasti trys nedideli naftos telkiniai – Agluonėnų, Šiaurės Vėžaičių, Uoksų ir nedidelė naftos sankaupa Žadeikių struktūroje. Atrastų naujų telkinių naftos ištekliai yra maži, tačiau aukšta žalios naftos kaina pasaulyje daro ir mažų telkinių eksploataciją pakankamai rentabilią. Be to, naftos gavyba Lietuvoje smarkiai mažėja, todėl atrasti naftos telkiniai palaikys išgaunamos naftos kiekį. Agluonėnų ir Šiaurės Vėžaičių naftos telkiniai išsidėstę Gargždų pakilumų zonoje, Uoksų naftos telkinys ir Žadeikių naftos sankaupa – į rytus nuo Gargždų regioninio lūžio

Nefrakcionuotos („žalios“) naftos ir sočiųjų naftos frakcijų tyrimai buvo atlikti dujų chromatografu. Dviejuose mėginiuose ištirti reliktinių angliavandenilių vadinamieji biožymenys bei nustatyti anglies izotopų duomenys. Be šių duomenų, kurie nustatomi atmosferos sąlygomis (naftos bandiniai atrinkti gręžinio žiotyse), Antkopčio-1 gręžinio nafta buvo ištirta ir sluoksnio sąlygomis (giluminiai naftos bandiniai). Straipsnyje yra pateiktos naujų telkinių naftos fizinės ir cheminės savybės.

Remdamiesi Lietuvoje naujai atrastų telkinių naftos tyrimų duomenimis galime teigti, kad kambro uolienų naftos fizinės ir cheminės savybės (tankis, klampumas, benzino kiekis), grupinė angliavandenilių sudėtis (sočiųjų, aromatinių angliavandenilių, dervų ir asfaltenų kiekis) daugiausia priklauso nuo slūgsojimo sąlygų ir nuo migracijos iš generacijos židinių iki telkinio atstumų. Pagal n-alkanų ir izoprenoidų pasiskirstymą tirtų plotų nafta yra labai panašios sudėties ir generuota sapropelinės organinės medžiagos. Biožymenų ir anglies izotopų tyrimo duomenimis, kai kurie skirtumai leidžia manyti, kad naftą generavo įvairios naftos motininės uolienos su skirtingos katagenezės organine medžiaga.

Giluminės naftos ėminių tyrimų duomenys rodo, kad Antkopčio-1 gręžinio nafta sluoksnio sąlygomis pagal fizines savybes yra panaši į kitų Lietuvoje atrastų telkinių naftą.

Tirtos naftos anglies izotopinė sudėtis taip pat panaši, praturtinta lengvuoju izotopu ($\delta^{13}C$): Agluonėnų-2 gręžinio naftoje –30,57‰, Uoksų-1 –31,22‰ Žadeikių-1 –29,34‰, ir tai būdinga apatinio paleozojaus naftai.

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ХИМИЧЕСКИЙ СОСТАВ НЕФТИ НОВЫХ ОТКРЫТЫХ МЕСТОРОЖДЕНИЙ В ЗАПАДНОЙ ЛИТВЕ

Резюме

В результате новых поисковых работ были обнаружены три небольших месторождения нефти: Аглуоненай, Северо-Вежайчяй, Уоксай, а также небольшое скопление нефти в Жадейкской структуре. Запасы нефти в обнаруженных месторождениях невелики, но высокая мировая цена на сырую нефть делает эксплуатацию даже небольших месторождений достаточно рентабельной. Кроме того, добыча нефти в Литве значительно падает, поэтому даже небольшие нефтяные месторождения увеличат количество добываемой нефти. Нефтяные месторождения Аглуоненай и Северо-Вежайчяй расположены в Гаргждайской зоне поднятий, а Уоксайское месторождение и Жадейкское скопление нефти – восточнее регионального Гаргждайского разлома.

Описаны физико-химические свойства и групповой углеводородный состав нефти вышеназванных месторождений. Проведен газохроматографический анализ нефракционированной („сырой“) нефти, а также насыщенной фракциями нефти. Выполнен анализ реликтовых углеводородов (биомаркерный анализ) двух образцов и исследован изотопный состав углерода нефти. Кроме этих исследований, проведенных в атмосферных условиях (пробы нефти отобраны в устье скважины), изучена глубинная проба нефти из скважины Анткоптис-1 (в пластовых условиях).

На основании проведенных исследований новых месторождений нефти в Литве можно утверждать, что физико-химические свойства (плотность, вязкость, количество бензина), групповой углеводородный состав (количество насыщенных и ароматических углеводородов, смол и асфальтенов) кембрийских нефтей в основном зависят от условий залегания, а также от расстояния миграции нефти от очагов генерации до месторождения. По данным распределения н-алканов и изопреноидов на исследованных площадях нефть сходна и генерирована сапропелевым органическим веществом. А результаты исследований биомаркеров и изотопного состава углеродов показали некоторые различия, что позволяет предположить, что нефть генерирована органическим веществом различного катагенезиса. По данным изотопного состава углерода исследованные нефти имеют сходство: они обогащены легким изотопом $\delta^{13}C$, величина которого в нефти скважины Аглуоненай-2 составляет –30,57‰, в Уоксай-1 –31,22‰, в Жадейкяй-1 –29,34‰, что характерно для нижнепалеозойской нефти.