

Geochemistry, composition, and structure of protoasphaltenes in organic matter of recent lacustrine sediments

L.S. Borisova^{a,b,*}

^a A.A. Trofimuk Institute of Petroleum Geology and Geophysics, Siberian Branch of the Russian Academy of Sciences,
pr. Akademika Koptuyuga 3, Novosibirsk, 630090, Russia

^b Novosibirsk State University, ul. Pirogova 2, Novosibirsk, 630090, Russia

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Abstract

Asphaltenes and asphaltene acids of organic matter (OM) of lacustrine sediments in the Novosibirsk and Tomsk Regions of West Siberia have been investigated by applying physical and physicochemical methods (elemental and X-ray diffraction analyses, IR, NMR, and EPR spectroscopy, Rock Eval pyrolysis, and electron microscopy). Compared with asphaltenes of dispersed OM at the early catagenesis stages, asphaltenes of OM at the diagenesis stage have a high number of hydrogen atoms and heteroatoms, a low degree of aromaticity, a low concentration of paramagnetic centers, a large number of carbon atoms in the long paraffin chains and oxygen-containing groups. Tetravalent vanadium is absent from asphaltenes of recent lacustrine sediments, and only complex compounds of copper have been identified. The sediments contain not asphaltenes but their precursors, protoasphaltenes. These are less condensed compounds with a looser nongraphite structure and a rather high petroleum potential. At the diagenesis stage, protoasphaltenes are immature protokerogen blocks.

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Introduction

Geochemical studies of the composition and structure of asphaltenes of recent sediments are not numerous (Castex, 1977; Chernova and Shishenina, 1979; Danilova et al., 1983; Galimov and Kodina, 1982), although in the 1950–1980s many researchers (N.B. Vassoevich, V.S. Veselovskii, A.E. Kontorovich, E.A. Romankevich, B. Tissot, V.A. Uspenskii, J. Hunt, etc.) considered the mechanism and trends of diagenesis of organic matter (OM). In the last years, laboratory modeling in anaerobic environments has given an insight into catagenesis of OM of recent lacustrine sediments (Rogozina, 2012; and others). Bazhenova (1990) focused the main attention on the formation of hydrocarbons (HCs) from resin–asphalt components of OM at the early stages of its catagenesis.

In this paper, we continue studying heterocyclic compounds of bitumens of dispersed organic matter (DOM) in West Siberia (Borisova, 2012; Kontorovich and Borisova, 1989a, 1989b, 1994). The goal of our research is to elucidate the composition and structure of asphaltene components of OM

bitumens at the diagenesis stage. Asphaltene components of DOM of fossil sediments are usually high-molecular heteroatomic compounds. During diagenesis, copolymers, such as asphaltenes or asphaltene-like compounds, form in the sediments. As shown below, these components insoluble in petroleum ether differ considerably in composition and structure from asphaltenes of fossil sediments. We called them protoasphaltenes.

The objects of our study were asphaltenes and asphaltene acids of OM of West Siberian lacustrine sediments. A selection of 17 samples from bottom sediments and water was studied. Sapropels were sampled from the lakes of the Kulunda halogenic zone (Topol'noe, Bezymyanoe, Azhbulat, Chebakly, Chany, Kankul', Minzelinskoe, Taraskul', Ubinskoe, and Negunaevskoe in the Altai Territory and Tomsk and Novosibirsk Regions) (Table 1)¹.

Chloroform-extracted bitumen, asphaltenes, and asphaltene acids from bottom sediments were separated by the schemes admitted at the Institute of Petroleum Geology and Geophysics, Novosibirsk, and at the Siberian Research Insti-

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* E-mail address: BorisovaLS@ipgg.sbras.ru (L.S. Borisova)

Table 1. Bottom sediments of lakes of southern West Siberia

Sampling zone	Sampling locality	Depth, m	Brief description
Steppe	Altai Territory, Lake Topol'noe, 200 m from the shore	From the surface	Gray-brown sapropel
	Altai Territory, Lake Bezymyanoe (east of Lake Bol'shoi Azhbulat)	0–0.05	Reddish sediments with H ₂ S odor
		0.1–0.15	Black viscous sediments
		From the surface	Reddish sediments, sodium chloride
	Altai Territory, Lake Bol'shoi Azhbulat, 200 m from the shore	From the surface	Red sediments
	Altai Territory, Lake Bol'shoi Azhbulat (brine sample)	0.15	Black-and-white sediments
	Novosibirsk Region, Lake Chebakly (southwest of Lake Chany)	From the surface	Sapropel
	Novosibirsk Region, Lake Chany, Near Yudino Village	From the surface	Upper sediment layer with algae and mucus
		0.1	Sediments with black sandstone and with H ₂ S odor
Novosibirsk Region, extinct lake near (west of) Lake Chany	0.3	Sapropel	
Novosibirsk Region, Kargat region, Lake Kankul'	From the surface	Black mud with H ₂ S odor	
Forest-steppe	Novosibirsk Region, Kolyvan' region, Lake Minzelinskoe	0.2	Dark sediments
		0.5–0.6	Dark gray soapy sediments
Taiga	Tomsk Region, Lake Taraskul'	From the surface	Black mud-like sapropel
	Novosibirsk Region, Ubinskoe region, Lake Ubinskoe	0.1–0.15	Sediments with bluish-gray clay
	Novosibirsk Region, Suzun region, Lake Negunaevskoe, 10 m from the shore	From the surface	Loose mucous bottom sediments

tute of Geology, Geophysics and Mineral Raw Materials, Novosibirsk (Kontorovich, 1973; Kontorovich et al., 1976). The composition and structure of asphaltene components were studied by applying physical and physicochemical methods: elemental and X-ray diffraction analyses, IR, NMR, and EPR spectroscopy, Rock Eval pyrolysis, and electron microscopy.

The lacustrine sediments are silt-sandy and sand-silty muds and sapropels. The geochemical environment of accumulation and initial OM transformation makes impact on the content and, partly, the composition and structure of asphaltenes. Study of the sediments from different zones has shown that the amount of asphaltenes in the steppe lakes is lower than that in the taiga and forest-steppe lakes, where the TOC content is also higher (on the average, 7.18% against 0.71%) (Table 2). During diagenesis, accumulation of resin–asphaltene components is closely related to the composition of sediments and the sampling depth (redox environment): The content of asphaltenes increases with depth, whereas the contents of asphaltogene acids and alcohol–benzene resins decrease. In

addition, the content of hydrogen in asphaltenes increases and the content of oxygen decreases (Table 2).

The average elemental compositions of asphaltene components of the studied bitumens and average data are presented in Table 3. Asphaltenes and asphaltogene acids of sapropels are richer in hydrogen and heteroatoms but poorer in carbon than asphaltene components of fossil sediments formed at the early stages of catagenesis. Correspondingly, their H/C_{at} values are higher (1.72–1.92 against 1.00–1.20 in asphaltenes of the fossil sediments), which indicates their more aliphatic structure.

The NMR-spectroscopic study of asphaltenes of the lacustrine-sediment bitumens showed their low aromaticity (f_a) (on average, 0.15) (Table 3) and a high degree of substitution of hydrogen of their aromatic ring by aliphatic groups (K_{subst}). Asphaltogene acids of OM of the studied samples are, in turn, more aromatic and less substituted than asphaltenes.

As seen from the IR spectra of asphaltogenes of the studied sediments (Fig. 1), aromatic groups are minor (the weak band

Table 2. Content of polar fractions and elemental composition of asphaltenes of sapropels

Sampling zone	Sampling depth, m	TOC, %	Benzene resins	Alcohol–benzene resins	Asphaltenes	Asphaltogene acids	Elemental composition of asphaltenes components, %			
							C	H	O	N + O + S
Steppe	From the surface	0.7	12.5	46.5	5.7	26.3	70.6	10.6	16.5	18.8
	0.1–0.3		16.4	32.5	16.3	25.5	73.1	12.1	12.7	14.8
Forest-steppe	0.5–0.6	8.5	17.4	22.4	42.3	13.8	79.7	13.3	5.5	7.0
Taiga	From the surface	5.9	14.6	33.9	15.5	32.7	77.3	12.9	8.0	9.8

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