



Research article

## On peculiarities of composition and properties of ancient hydrocarbon source rocks

Mariya A. Bolshakova✉, Kseniya A. Sitar, Dmitrii D. Kozhanov

Lomonosov Moscow State University, Moscow, Russia

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**Abstract.** Precambrian rocks are widespread within all continents of the Earth; that said, sedimentary associations of these deposits are of special interest in search for oil and gas fields. A wide range of paleontological, lithological and geochemical methods is utilized for conducting integrated geological-geochemical analysis and evaluating the initial hydrocarbon generating potential of organic matter of Precambrian source rocks. Investigated were peculiarities of depositional environments of the organic matter, specific features of its composition in sedimentary rocks and its generation characteristics. Own research efforts were performed in combination with generalization of other authors' publications focused on Precambrian sequences enriched in organic matter – their occurrence, isotopic and biomarker characteristics and realization schemes of the hydrocarbon generation potential of Precambrian organic matter in the process of catagenesis. Geochemical peculiarities of initial organic matter are illustrated on various examples, type of the organic matter is determined together with the character of evolution of realization of its initial generation potential.

**Keywords:** Proterozoic; petroleum; hydrocarbon source rocks; organic matter; petroleum basins

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**Introduction.** Hydrocarbon source properties or petroleum generation potential of sedimentary rocks is an integral value that depends first of all on amount of organic matter (OM) in a rock and of quality thereof, i.e. on the genetic type of kerogen. Organic matter of Proterozoic rocks has a cognate initial biocenotic composition. Notwithstanding that seabed stromatolite-forming and brown algae appeared as early as in Early Riphean, the main role in forming the OM of Proterozoic rocks belongs to remains of planktonic cyanobacteria (Gloeocapsomorpha) and acritarchs (Acritarcha) and bacteria [1]. This is the reason for a rather undiversified composition of the OM of ancient deposits – it is represented by sapropelites and oxidized varieties thereof – oxypropelites. Sapropelic OM under various grades of oxidation is characteristic of practically all subdivisions of Precambrian. Hydrocarbon source rocks are present at various stratigraphic levels of Riphean. They are detected in Lower, Middle and Upper Riphean deposits in sedimentary and petroleum-bearing basins of practically all continents.

Some of the oldest hydrocarbon source rocks were found in Lower Riphean. These rock masses are represented by dark gray mudstone, mudstone-siltstone, siltstone-limestone, and clayey-dolomitic associations. Maximum concentrations of organic carbon in them vary within a wide range, reaching 6-7 % (as an example of source rocks of East Siberia [2, 3]). Early Riphean is characterized by dominance of prokaryotes, mainly cyanobacteria – seabed stromatolite-forming and planktonic ones, and other bacteria. There are encountered representatives of planktonic algae ascribed to Acritarcha, cyanobacteria Gloeocapsomorpha. In addition, in Lower Riphean deposits there are found remains of bottom algae looking similar to brown (?) algae Vendotaenia. It is probable that eukaryotic and even multicellular algae Phaeophita appeared as early as in Early Riphean [4]. Phytomyces (fungi) are encountered in OM concentrates of Early Riphean. Remains of brown (?) along with blue-green algae



are encountered in Middle and Upper Riphean deposits as well. In Middle and especially in Late Riphean, planktonic algae *Acritarcha* are widely developed, a part of them (unicellular with a nucleus) probably belong to the *Chlorophita* group [5].

In Middle Riphean sections preserved up to the present time, interval containing increased OM concentrations are also identified, they are characterized by high variability of not only organic carbon contents but also of degree of organic matter transformation. For example, in East Siberia to hydrocarbon source rocks belong dark-colored mudstones and marls of the *Strelnaya-Gora*, *Derevnino* and *Nizhnyaya-Tunguska* (*Burovaya*) formations encountered in *Turukhansk* district; those of the *Shuntar* formation of the *Yenisei Ridge*, and black clayey limestones and dolomites of the *Malga* formation in the *Aldan-Maya* trough in the east of the *East Siberian platform*. The highest OM contents are found in the *Malga* formation (organic carbon up to 5-8 % [5]). At that, carbon of kerogen of the *Shuntar* formation is the isotopic-lightest, while  $\delta^{13}\text{C}_{\text{HOM}}$  values decrease in the westward direction from  $-26.0$  to  $-26.3$  ‰ (the *Gorbilok-Glushikha* zone) to  $-31.0$  ‰ (the *Kamenka* zone), that reflecting a higher degree of OM catagenetic transformation in the first two zones [6]. In addition, in the *Kuyumba* district, in a number of wells there are penetrated dark gray clayey limestones, dolomites and marls of the *Taiga* formation in which organic matter concentration sometimes reaches 4.5 % (with average concentration of 0.25 %). In the southeast of the *Siberian platform*, rocks of the *Khaiverga* formation of Middle Riphean are represented by mud shales in the lower part and average concentration of organic carbon is 1.7, reaching 3.95 % in the area of the *Khaiverga* river.

Upper Riphean-Vendian hydrocarbon source rocks are also detected in the Proterozoic part of the section. Notwithstanding active manifestation of erosion events at postdepositional stages in basins within practically all continents, OM-enriched intervals are identified in remaining sections (parts of sections) of the said age. As compared with more ancient intervals, they are studied better (for example sedimentary basins of *East Siberia* [7], the *Volga-Ural*, *Bohai*, *Amadeus*, *Carpentaria* [8, 9], *North Oman – Haushi* and *Shuaiba*, *South Oman – Rajasthan*, *Vindhyan* [10], *Kohat – Potwar*, *MacKenzie*, etc.). In Upper Riphean in various parts of the *Siberian platform*, there are encountered terrigenous and less frequently carbonate rocks containing increased organic carbon concentrations in the *Turukhansh* district, *Patom* highland, within the *Baikit* antecline (over there, the *Iremeken* formation is identified that is represented by black mudstones with a brownish hue in which organic carbon concentrations reach 8-9 %).

In Vendian, diversity of the organic world dramatically increases [11]. The Vendian biota includes a wide diversity of algae, non-skeletal and primitive skeletal Metazoa, represented mainly by tabular organisms. Non-skeletal Metazoa appeared as early as in Riphean, though neither remains nor casts thereof have been found up until now. There were found only trace fossils thereof that is explained by fragility of their protective and integumental membranes [4]. However, in Vendian concentrates of dispersed organic matter, especially from rocks that are relatively enriched, those same cyanobacteria and to a lesser extent brown algae are prevalent [5].

By lithofacies depositional environments, development of various forms of hydrobionts and degree of their diagenetic transformation, two regions are identified in Vendian-Cambrian deposits of the *Tunguska* basin. Organic matter of mainly marine facies zone (the *Igarka-Norilsk* area) was formed largely from non-mineralizable algae, while in environments of salt-producing lagoons (most part of the *Tunguska* syncline) – from mineralizable planktonic algae of the same classes. Presence of organic-matter enriched rocks in the Vendian section is most probably associated with the “*Ediacaran*” explosive development of biocenoses. Analysis of vertical zonation of organic-carbon enriched sequences reveals several intervals with increased OM content. At that, organic carbon concentrations are much higher than those recorded in Lower and Middle Riphean rocks. By data of [6], in the *Baikit* syncline of *East Siberia*, organic carbon concentrations in black mudstones of the *Tira* formation reach 14 % (though they vary within a very wide range). In *China*, in rocks of the *Datango*



formation (the Sichuan basin) organic carbon concentrations in some places reach 21 % [12, 13]. Such enriched levels of organic matter accumulation are presumably associated with planetary events, in particular with active manifestation of geodynamic processes [14].

The purpose of this paper is to demonstrate peculiarities of composition and petroleum-generating properties of the most ancient organic matter and to somewhat enlarge the available understanding in the matter under consideration.

**Results and discussion thereof.** *Peculiar features of chemical composition of Precambrian organic matter.* The main bio-producers in the Precambrian were prokaryotes represented by bacteria, cyanobacteria and Archaea. Their development started approximately 3.8-3.7 billion years ago. The principal events that affected geochemical composition of that time organic matter are associated with development of the photosynthesis in prokaryotes [15-17], due to which fractioning and selective accumulation of light carbon isotope  $^{12}\text{C}$  in organisms and metabolites thereof took place. That is why lighted isotopic composition of carbon is observed in Precambrian deposits – 32-34 ‰ [18]. The appearance of eukaryotes approximately 2.7 billion years ago also affected individual geochemical

composition of Precambrian organic matter. In addition to lighted isotopic composition of carbon, ancient organic matter is also characteristic of specific biomarker features, e.g. values of content ratios of steranes [19, 20] (tetracyclic hydrocarbons evolved from sterols produced by eukaryotes) and hopanes [21] (pentacyclic hydrocarbons built from lipids of cellular membranes synthesized by prokaryotes).

In the composition of organic matter of Precambrian sedimentary rocks of the earth (the McArthur basin, the Transvaal group, the East Siberian basin, etc.) hopanes prevail over steranes (Fig.1). This reflects wide development of microbial communities as well as the domination role thereof in the formation of initial organic matter of hydrocarbon source rocks.

Prevalence of steranes  $\text{C}_{29}$  over steranes  $\text{C}_{27}$  and  $\text{C}_{28}$  noted in many works [22-24] is also an attribute of Precambrian OM (Fig.2). In this case, such a feature that is usually characteristic of Type III organic matter is usually considered as associated with domination of green algae in the community of eukaryotic phytoplankton [25, 26]. Note that Type III organic matter is also determined for some Precambrian rocks by results of pyrolysis or elemental analysis of kerogen [27]. According to investigations in [28], this is associated with the fact that the initial organic matter contained a small amount of lipids, while deposition took place in oxidative marine and shallow-water marine environments. However, a part of organic matter (Types I, II, IIS) was accumulated in reducing marine environments. Thus,

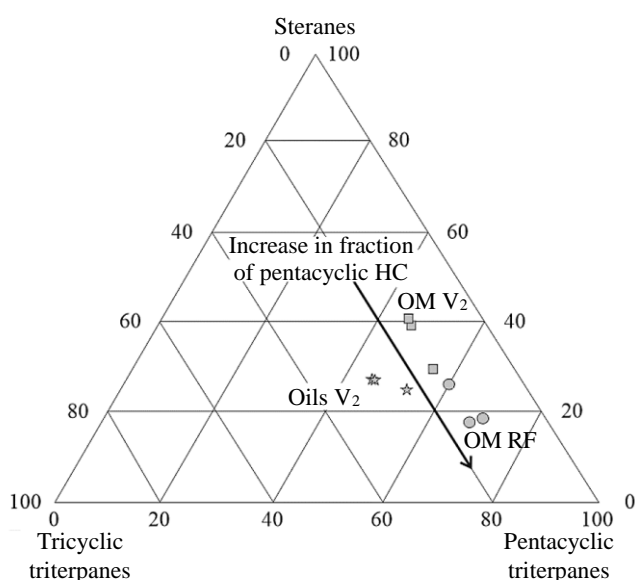


Fig. 1. Trigonogram of distribution of tri-, tetra- and pentacyclic hydrocarbons

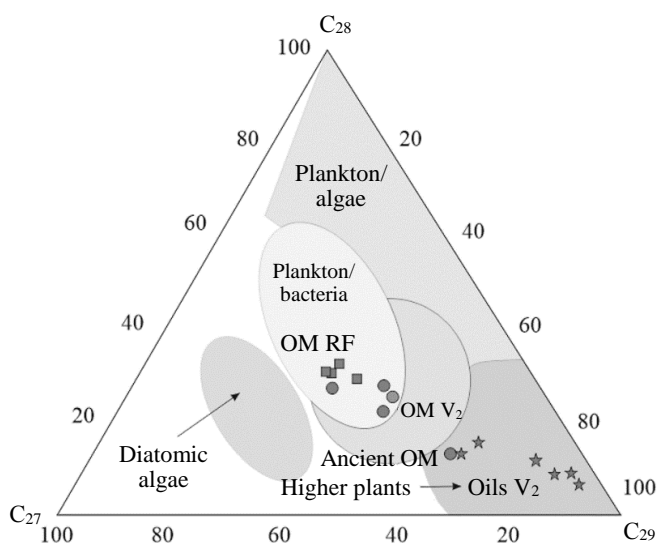


Fig. 2. Trigonogram of distribution of sterane hydrocarbons in Riphean-Vendian deposits



determining the type of initial organic matter of these ancient deposits is rather problematic. This phenomenon can be illustrated by results of studies (Fig.3, 4).

Specific alkanes characteristic of Precambrian organic matter are some methyl-alkanes. For example, 12- and 13-monomethyl-alkanes identified [29] in OM and oils of Precambrian deposits of East Siberia.

Such individual compounds were identified [30] also in oils of the Arah group (Oman). The hydrocarbons under consideration form two homologous series: 12-methyl-alkanes of the C<sub>24</sub>-C<sub>30</sub> composition and 13-methyl-alkanes of the C<sub>26</sub>-C<sub>30</sub> composition. One of the potential Precambrian markers is presence of branched mono- and dimethyl-alkanes [3] in the composition of saturated hydrocarbon fraction of organic matter whose appearance is apparently associated with specific bacteria of Early Ediacaran. In addition to the above-listed features of Precambrian organic matter, other attributes characteristic of Precambrian organic matter are discussed in publications (see Table). Thus, organic matter of Precambrian source rocks has specific characteristics of its composition: prevalence of steranes C<sub>29</sub>, presence of branched alkanes, prevalence of pentacyclic hydrocarbons over tricyclic and tetracyclic ones. Appearance of these features is in turn associated with a considerable contribution of bacteria in the composition of initial organic matter.

It is demonstrated that evaluation of the organic matter type in ancient deposits by pyrolysis is problematic. The Type III of organic matter determined by this method is probably associated first of all with postdepositional oxidation degree of the organic matter. At the same time, the initial organic matter according to data of chromatography and isotopic analysis of carbon had Type II of organic matter (marine, sapropelic).

*Fundamental regularity in realization of hydrocarbon-generation potential of Precambrian organic matter.* Ancient organic matter is often hydrogen-depleted in its elemental composition as compared with a typical Phanerozoic organic matter and is ascribed to Type III kerogen (see Fig.3).

With the purpose of digital modeling of the formation of petroleum potential, software programs for basinal (geological-geochemical) modeling are utilized [31]. In such software programs, properties of organic matter of source rocks are described by means of three characteristics inherent with catagenetically unaltered rocks:

- initial Total Organic Carbon content (TOC, wt.%) – measure of organic matter content in a rock;

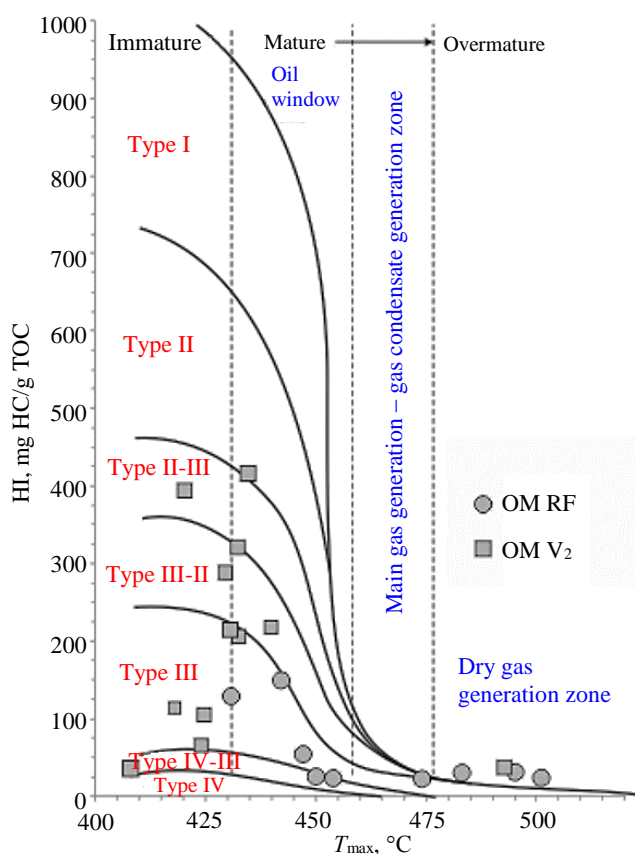


Fig.3. Pyrolytic characteristics of Riphean-Vendian deposits on the modified Van Krevelen diagram

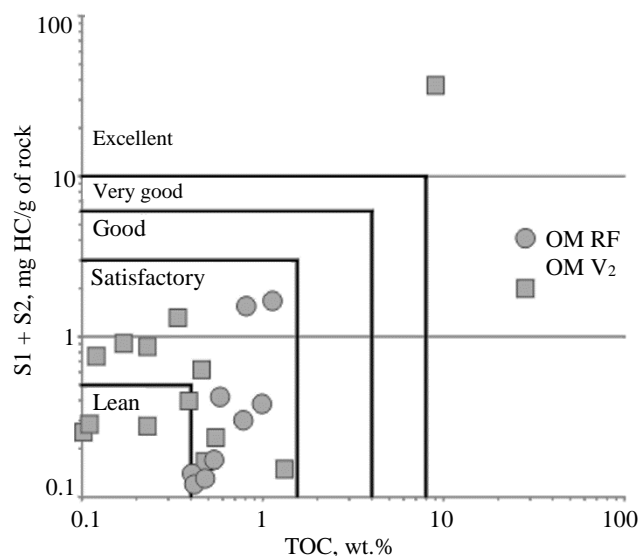


Fig.4. Pyrolytic characteristics of Riphean-Vendian deposits on the generation potential diagram



- initial Hydrogen Index (HI, mgHC/gTOC) of organic matter in hydrocarbon source rock – hydrocarbon generation potential of organic matter of a rock;
- kinetic spectrum of kerogen destruction (the law describing generation of oil and gas by source rocks in catagenesis).

**Molecular markers of Precambrian organic matter**

Key characteristic	Source of organic matter	Source
Dominance of ethyl-cholestanes in the composition of steranes	Algae, cyanobacteria	[22]
High 24-isopropyl/n-propyl-cholestane ratio	Archaeocyatha	[32-34]
Prevalence of regular structures over regrouped ones, $\alpha\beta\beta$ -isosteranes over $\alpha\alpha\alpha$ -steranes	–	[35]
Domination of C <sub>29</sub> homologs in the composition of $\alpha\beta\beta$ -isosteranes	–	[35]
Prevalence of 18 $\alpha$ (H)-neohopanes over 17 $\alpha$ (H)-21 $\beta$ (H)-hopanes	Bacteria	[36]
Tricyclanes ( $\Sigma$ T) dominate in the composition of terpanes	–	[37]
Tricyclane index 2T <sub>19-20</sub> /T <sub>23-26</sub> < 1	–	[37]
Prevalence of Hh <sub>35</sub> homohopanes over Hh <sub>34</sub>	–	[37]
Domination of 2 $\alpha$ -methyl series	Methylotrophic bacteria and cyanobacteria	[38]
Domination of 3 $\beta$ -methyl series	Bacteria of Acetobacter type	[38]
Presence of 2- and 3-methyl-substituted alkanes	Bacteria	[39]

One of the objectives of this research was to clear out whether or not the organic matter of Precambrian source rocks has similar generation characteristics; how does such organic matter realize its potential in the course of time in the capacity of classical sapropelic organic matter; whether or not hydrogen depletion affects the mechanism of realization of the potential of such organic matter in the process of catagenesis. This question is of utmost importance in the assessment of hydrocarbon potential of basins in which Proterozoic hydrocarbon systems are present, while publications answering this question are practically unavailable. In the libraries of commercial software packages for basinal modeling, there are no kinetic spectra of kerogen destruction obtained on samples of Proterozoic rocks. In the process of modeling, researchers have to use the available possibilities to substitute data on such organic matter for “standard” kinetic spectra and it is a rather difficult solution in favor of which organic matter type one is to decide.

Proterozoic organic matter has already been attracting attention of geochemists for more than 50 years. During this time, researchers have shown that organic matter of this age generated oil and gas in various petroleum basins throughout the duration of geological history of our planet. There are next to none publications, which reflect kinetics of realization of Proterozoic organic matter. Such

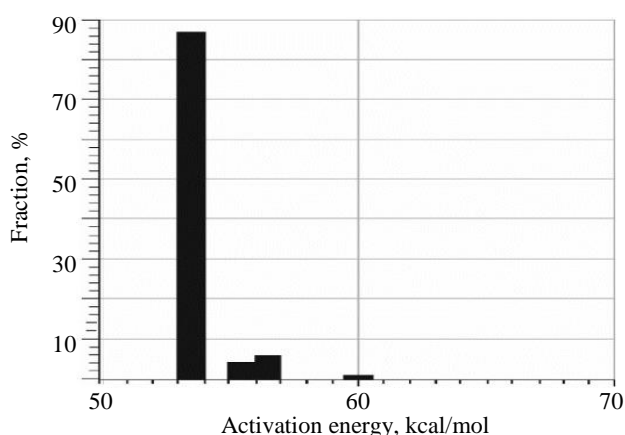


Fig.5. Kinetic spectrum of transformation of Late Riphean organic matter of the Iremeken formation, East Siberia, A = 5.3E + 13 [40]

publications may be conditionally divided into two inadequate parts: the first – works in which researchers-geologists (dealing mainly in basinal geological-geochemical modeling) “synthesize” kinetics to model generation by Riphean-Vendian rocks relying upon common sense, results of standard pyrolytic assays and information on how the potential of such or another type of organic matter is realized. Most researchers have to use an approach of this kind. The second type of works is publications presenting results of author’s kinetic studies of organic matter-enriched Precambrian rocks [30, 40, 41].

For example, the paper [40] reflects results of experimental studies over a Riphean mudstone (the Iremeken formation of Upper Riphean, the



Yurubcheskaya 104 well, 2182-2186 m, East Siberia) enriched in organic matter. The authors made a lot of interesting studies on this specimen – from hydrolysis in various versions to analyzing isotopic composition of carbon. The single-component kinetic spectrum was restored (Fig.5). The sample under study contains 12.6 % of organic carbon, hydrogen index is 463 mgHC/gTOC,  $T_{\max} = 445 \text{ }^{\circ}\text{C}$  [40], i.e. the rock under study has already partly realized its generation potential that somewhat reduces the significance of this kinetics for utilizing thereof in digital modeling.

The obtained spectrum is characterized by maximum output of hydrocarbon fluids at activation energy of 54 kcal/mol. The pre-exponential factor ( $A$ ) equals  $5.3 \cdot 10^{13} \text{ s}^{-1}$ . In the work [30], a sample of a catagenetically immature Neoproterozoic-Early Cambrian source rock from the South Oman basin was studied. The sample in question is characterized by a high organic matter potential, hydrogen index is 714.7 mgHC/gTOC. Organic matter in it is significantly enriched in hydrogen and belongs to Type I. The kinetic spectrum obtained in the paper [30] is presented in Fig.6.

The lower activation energy for the maximum output of hydrocarbon fluids (50 kcal/mole for the Oman sample) may be associated both with the difference in maturity of the studied materials and with the fact that organic matter of the Siberian sample might be partly oxidized in the process of sediment diagenesis. The wide spectrum of energies at which transformation of organic matter takes place is probably associated with sulfur content of this organic matter that is mentioned by the authors of this research. Sulfur in the organic matter composition results in the fact that it becomes more chemically reactive and its transformation starts at earlier stages of catagenesis [42].

Regularities in organic matter transformation of Proterozoic source rocks are studied on rocks of the basins of Australia [41], studies are undertaken on numerous weakly altered catagenetically samples of Proterozoic age. Comparison is performed of variation of transformation coefficient of Proterozoic organic matter with the behavior of “standard” organic matter types (the types were selected by organofacies from the work [43]). In Fig.7, dotted line shows the change in transformation coefficient for source rocks from the basins of Australia and solid lines show transformation of “standard” organofacies of Oman (green line [30]) and of East Siberia (orange line [40]). It is seen in Fig.7 that notwithstanding the fact that a part of samples from Australia is enriched in Type III organic matter, all of the samples demonstrate transformation inherent in organic matter of sapropelic genesis. All of the variation trends of transformation coefficient of Precambrian organic matter are similar to the trends of sapropelic (or mixed) organic matter of organofacies A, B, and C.

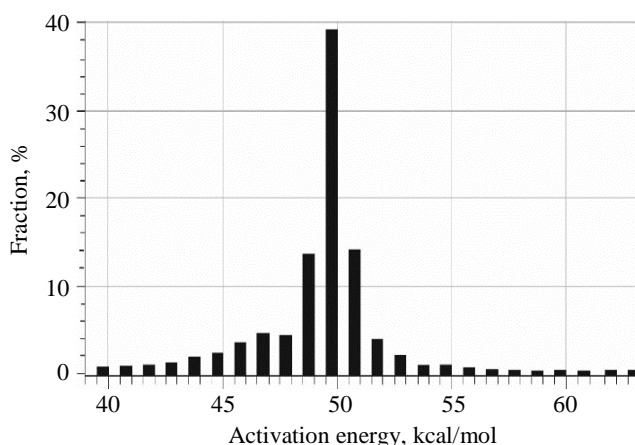


Fig.6. Kinetic spectrum of organic matter transformation of Neoproterozoic – Early Cambrian rocks of South Oman,  $A = 1.1E + 13 \text{ s}^{-1}$  [30]

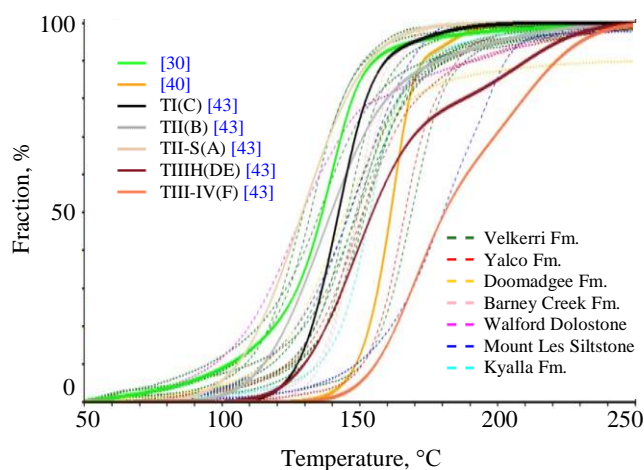


Fig.7. Transformation coefficient versus temperature for Proterozoic hydrocarbon source rocks and for organic matter of the “classical” organofacies



**Conclusion.** The literature review and the own research do show that hydrocarbon source rocks are rather widespread in Proterozoic rocks. They were identified in various sedimentary basins of the world and may be confined to various stratigraphic subdivisions of Proterozoic.

Organic matter of Precambrian has the specificity of molecular-isotopic composition – in the soluble part of organic matter, specific methyl-alkanes, steranes C<sub>29</sub> are identified, pentacyclic compounds dominate among cycloalkanes, isotopic composition of carbon is light as a rule.

Pyrolytic studies of Precambrian organic matter often lead to the fact that it is ascribed to the Type III (pseudohumic).

Generalization of the small in number investigations in the kinetics of Precambrian organic matter transformation makes it possible to conclude that the trend of its transformation in the process of catagenesis corresponds to the transformation trend of sapropelic organic matter (even for the oxysapropelic pseudohumic organic matter).

Upon the whole, Precambrian organic matter is characteristic of the scheme of transformation in the process of catagenesis similar to the regularity described in [43] for Type I organic matter (organofacies C).

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**Autrors:** **Mariya A. Bolshakova**, Candidate of Geological and Mineralogical Sciences, Leading Researcher, [m.bolshakova@oilmsu.ru](mailto:m.bolshakova@oilmsu.ru), <https://orcid.org/0000-0001-9240-291X> (Lomonosov Moscow State University, Moscow, Russia), **Kseniya A. Sitar**, Candidate of Geological and Mineralogical Sciences, Senior Researcher, <https://orcid.org/0000-0003-1386-8442> (Lomonosov Moscow State University, Moscow, Russia), **Dmitrii D. Kozhanov**, Postgraduate Student, <https://orcid.org/0000-0002-2641-918X> (Lomonosov Moscow State University, Moscow, Russia).

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