Hydrocarbon potential of Northern promontories of the Pannonian Basin System in Slovakia

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Abstract

The geodynamic evolution of four Western Carpathian Neogene basins in Slovakia is presented with regard to organic maturation and hydrocarbon generation. Core samples were studied from depths up to 5 300 m in the Vienna Basin, up to 3 200 m in the Danube Basin and up to 4 100 m in the East Slovakian part of the Trans-Carpathian Depression, and also samples from shallower wells in the South Slovakian Basin.

As geothermal conditions in these basins differ, so also does the trend in the depth maturation of organic matter. While the maturation of organic matter and corresponding conversion of kerogen to hydrocarbons takes place at the shallowest depth in the East Slovakian Neogene Basin, it occurs more deeply in the Danube Basin and at the deepest conditions in the Vienna Basin. The local geothermal conditions in individual basins exhibit differences in sedimentary organic matter maturation dependent on depth differences, mainly between their northern and southern parts. The kerogen of the South Slovakian Neogene Basin is currently in the passive maturation stage.

Different geodynamic evolution in individual basins influenced the intensity of kerogen to hydrocarbon conversion, and also the volume of generated and entrapped hydrocarbons.

Key words: hydrocarbon potential, organic maturation zonality, Northern promontories of Pannonian Basin system, Slovakia

Introduction

Recently, the hydrocarbon exploration in Slovakia has mainly focused on the sedimentary fill of our three largest Neogene basins, i.e. the Vienna, Danube and East Slovakian basins. In addition, newly obtained geochemical results also allow the assessment of the South Slovakian Basin sediments. The comparison of the source of hydrocarbon potential and the maturation depth zonality, as well as the kerogen to hydrocarbons conversion is significant mainly for obtaining knowledge concerning: (1) the source and origin of the sedimentary organic matter, (2) calculation of the volume of potential source rocks entering the active generation zones and (3) the calculation and comparison of generated hydrocarbons. Based on these results and the geo-tectonic evolution of the individual basins, consideration can be given to the hydrocarbon volume that can be entrapped into accumulations.

General geological outline

In the last century, the most important regions with hydrocarbon deposits in the Western Carpathians were the

Carpathian Foreland and the Neogene basins. In Poland, these were the Carpathian Foreland with the fore-deep and the Outer West Carpathian Flysch nappes, while in the Czech Republic they consisted of the Carpathian Foreland with the fore-deep and the northern part of the Vienna Basin. In Slovakia, these regions are represented by the north-eastern part of the Vienna Basin, by the northern part of the Danube Basin and by the East Slovakian Neogene Basin. These areas are often referred to as Northern promontories of the Pannonian basin system (Fig. 1).

The origin and the evolution of these basins were controlled by the geo-dynamical processes connected with subduction of the lithospheric fundament of the Outer-Carpathian Flysch Belt below the Carpathian-Pannonian lithospheric plate and the rise of the Pannonian asthenolith.

The classification of the West Carpathian Neogene has been introduced in several publications (e.g. in Royden et al., 1983; Čech, 1988; Kováč, 2000; Vass, 2002; Vass and Čverčko, 1985; Jiříček, 2002 and in Janočko et al., 2006). These explained the origin of basins from classical and global tectonic viewpoints. Many of the suggestions recently published are certainly interesting and original,

however these differ substantially and they are continually developed, due to the quite complex geological structure of the Western Carpathians.

Jiříček (2002) considered the Carpathian Foredeep and the Neogene basins as a single evolutionary unit. According to his suggestion, their formation and evolution is related to the oblique collision of the African and European lithospheric plates, the sinistral rotation of the African plate and successive closing of the Tethyde Ocean, and to the formation of foredeep and shifting of Flysch nappes from the west to the east. According to the discrepancy rule the following formations sank beneath the Flysch nappes; (1) the formations of the Eggenburgian and Ottnangian units in Lower Austria, (2) the Karpatian sediments in South and Middle Moravia, (3) the Lower Badenian deposits in Northern Moravia, (4) the entire Badenian sedimentary packet in Poland (5) the Lower Sarmatian sediments in Ukraine and (6) the complete Sarmatian sedimentary packet in Romania (Jiříček, 1979). The formation and evolution of the Central Western Carpathian Neogene basins were closely connected with these processes acting in the front of Carpathian arc.

The so-called intra-Carpathian channel (Jiříček, 1978) was formed in the Peri-Klippen area during the Early Miocene time. The southern-originating deltas entered into this channel from the Vienna Basin to the East Slovakian Basin. During this entire period, the Lower Miocene sediments on the Carpathian-Pannonian lithospheric plate maintained a "piggy-back" basin position (Jiříček, I.c.).

At the change of the Lower Miocene period to the Middle Miocene, the subducted slope of the northern European and the Carpathian-Pannonian lithospheric plates collided. The Magura Flysch gradually rose, relief inversion took place and the depositional centres of the Neogene basins shifted to the south (Jiříček, 2002). The related shifting of the Flysch nappes to the east has produced the following inversions; (1) in the Vienna Basin during the Karpatian-Early Badenian periods, (2) in the Danube Basin at the beginning of the Early Badenian period and in (3) the East Slovakian Basin this occurred until the Late Badenian period. The rearrangement of the Neogene basins was connected with the initiation of

striking tectonic activity, mainly in syn-sedimentary radial faults, which, together with the accompanying inversion, transformed the Neogene basins contours to their current form (Jiříček, I.c.).

In one of the most recent synthetic works, Janočko et al. (2006) divide the Western Carpathian Neogene basins, according to the mechanism of their origin, into several classes:

- (1) Basins of the foredeep, formed as a consequence of the North European lithospheric plate-flexure induced by the weight of accretionary wedges (Flysch nappes) and sub-surface load (the "slab-pull" mechanism):
- (2) Piggy-back basins on the Flysch accretionary wedge, originating from the crustal extension at the trailing-edge of the wedge due to the tension generated by the wedge-thrust process;
- (3) Wrench fault furrow basins formed at active convergent margins of the convergent overriding Carpathian-Pannonian lithospheric plate;
- (4) Transtensional forearc and intraarc basins which originated synchronously with the formation and growth of the Miocene volcanic arc, and which opened due to oblique convergence of the North European lithospheric plate and Carpathian-Pannonian lithospheric plates, utilizing the "strike-slip pull-apart" mechanism;
- (5) Backarc basins which originated from the mechanism of back-arc extension and from thermal lithospheric activity due to increased heat flow from the rising Pannonian asthenolith.

The first-mentioned type of Neogene basin is absent in Slovakia; this is the one in the frontal part of the West Carpathian and below the front of the Outer Flysch Belt with the older autochthonous units of the North-European lithospheric plate in the basement of the Neogene sediments.

The piggy-back basin type is represented by the Lower Miocene formation in the western part of the Vienna Basin, which lies on the Western Carpathian Flysch nappes accretionary wedge.

The relics of wrench fault furrow basins are evident in the Lower Miocene formations of the north-eastern part of the Vienna Basin, together with the partial Vadovce and

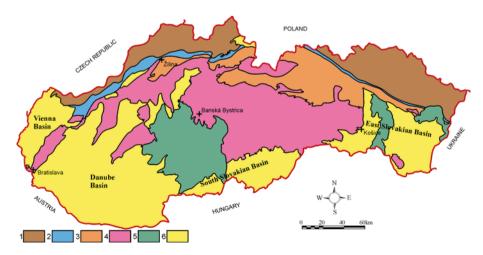


Fig. 1. Schematic geological map of the West Carpathian Neogene Basins in Slovakia. 1 – Flysch Belt; 2 – Klippen Belt; 3 – Central Carpathian Paleogene; 4 – Inner West Carpathian units; 5 – Late Cenozoic volcanics; 6 – Neogene basins.

Dobrá Voda depressions and the Lower Miocene sediments of the northern part of the East Slovakian Basin.

The transtensional forearc basin is represented by the Middle to Upper Miocene sediments of the Vienna Basin, while the transtensional intraarc basin is represented by the Middle to Upper Miocene sediments in the East Slovakian Neogene Basin.

The South Slovakian Basin is one of the back-arc basins, and the thermal extensional back-arc basin is represented by the Danube Basin.

The explicit determination of the genetic type of the Neogene basins in the Western Carpathians is quite difficult. This problem was documented by Royden (1985) in the Vienna Basin example. Although the opening mechanism was mainly pull-apart, in this case the opening was controlled by the relatively shallow crustal faults and not by typical transcurrent or transform faults, which originate more often in connection with the ocean floor. Consequently, there are several differences in the Vienna Basin compared to other typical pull-apart basins. It is relatively cold, it does not have its own volcanic centres and the only foreign units are in its surrounds, and not within the basement itself. Therefore, the Vienna Basin is described as a specific thin-skinned pull-apart basin (Royden, I.c.).

It is obvious that the West Carpathian Neogene basins represent one evolutionary unit and they can not be considered in isolation. The differing formative mechanisms in the individual evolution of basins, together with other specific phenomena, caused hydrocarbon generation, migration and accumulation to occur in each basin at various intensities and at different times and depths. The result is a different hydrocarbon potential in each individual basin. It can generally be stated that, in the past, the junction area of the West Carpathians and the North European platform formed relatively fair conditions for hydrocarbon generation, migration and accumulation. The reasons for the different hydrocarbon potentials can be found in the specific features, which can be summarized in several points. The basin position within the Carpathian arc is closely related to the different basement of the Neogene sedimentary fill.

The Vienna Basin is closest to the front of the Carpathian arc and it comprises two floors in the basement of the Neogene sediments. The higher floor in the western and northern part is composed of Upper Cretaceousand Paleogene sediments of Outer Western Carpathian Flysch nappes. The southern and eastern part contains the Mesozoic and Paleogene sediments of the Klippen Belt, Mesozoic sediments of the Northern Calcareous Alps and the Central Western Carpathian nappes. These latter contain folded post-orogenic sediments of the Upper Cretaceous Gossau type and also Central Carpathian Paleogene sediments. The lower floor consists of Paleozoic to Tertiary sediments of the dipped slopes of the Northern European Platform.

The sediment thickness of the basement together with Neogene sediments can exceed 10 km. This accumulation caused that the heat flow is a relatively very low in the Vienna Basin and dispersed organic matter in Neogene sediment in the deepest parts of the basin attained only the early generation stage. Thus, the Neogene source rocks contributed only a small amount to the entire basin hydrocarbon potential. On the other hand, mainly the Paleozoic to Tertiary sediments of dipped slopes of the Northern Platform with their fairly good source-rocks occur at a depth of 4–8 km in the main oil generation zones. The main portion of the hydrocarbon potential of the Vienna Basin is regarded as being derived from these sediments. This also confirms the biomarker study, as in Welte et al. (1982), Ladwein (1988) or Franců et al. (1996).

The Danube and the East Slovakian Neogene basins are significantly more distant from the Carpathian arc front. The basement of these basins comprises only the remnants of the Mesozoic sedimentary cover units and nappes of the Central Western Carpathians (East Slovakian Neogene Basin) and the remnants of the central Carpathian Paleogene sediments or pre-Cambrian or Paleozoic crystalline complex of the Danube Basin.

The Pre-Neogene sediments in the basement of both these basins have relatively small thicknesses, and these include practically no hydrocarbon-source rocks except for central Carpathian sediments. The reason the contribution of hydrocarbon potential of pre-Neogene rocks is negligible, in comparison to the Vienna Basin there can be found in this the absence of very good source-rocks.

The Danube and the East Slovakian Neogene Basins are situated on the margin of the Pannonian asthenolith and they are crossed by a neo-volcanic belt which caused increased heat flow. Consequently, Neogene sediments in of the deep central parts of these basins entered the gas-generation phase.

The Neogene sediments in the South Slovakian Basin have very small thicknesses of less than 1 km, and therefore this Slovak territory has negligible hydrocarbon potential.

From a lithological point of view, Western Carpathian Neogene basins are composed of comparable sediments within individual stratigraphic stages. Sedimentation in marine environments ended in the Vienna, Danube and East Slovakian basins in the Late Badenian. Brackish sedimentation ended in the East Slovakian Neogene Basin in the Early Pannonian time, in the Danube Basin during the Middle Pannonian time and in the Vienna Basin during the Late Pannonian time. Sedimentation in brackish environments changed until the end of the Pontian stage in all basins within lacustrine environments, and then lacustrine sedimentation changed to the fluvial system during the Dacian to Romanian stages.

The hydrocarbon generation zones within the Vienna Basin were reached only by sediments older than Upper Badenian. In the East Slovakian Neogene Basin these zones were reached only by sediments older than Upper Sarmatian and in the Danube Basin only by those older than Middle Pannonian. This is again connected with the heat flow intensity and with the geological evolution. Post-rifting sediments and sediments connected with the thermal subsidence in the Danube and East Slovakian

Neogene basins are developed in much greater thicknesses than those in the Vienna Basin. The final result was that marine sediments which were relatively rich in dispersed organic matter subsided to greater depths and to higher temperatures.

Methods and results

Basic organic-geochemical analysis

Organic geochemical characteristics of the investigated well cores are primarily based on the total organic carbon (TOC) and the total inorganic carbon (TIC). The quality of source rocks gave the hydrocarbon potential and the kerogen type, and the interpretation of the thermal maturity was based on the results of Rock-Eval pyrolysis. The chemical composition of the rock extracts and their correlations were evaluated on their aliphatic, aromatic NSO compounds relationships, and also on the n-alkane, isoprenoid and aromatic hydrocarbon distribution. These analyses were carried out according to standard methods used in accredited Brno laboratories in the Czech Geological Survey Prague (CGS). Microphotometric measurements were instituted by the first author in the same institution

Tab. 1 Number of basic organic-geochemical analyses carried out in individual basins

Region	TOC/TIC	Rock-Eval umber of analy	microphotometry ysis
Vienna Basin	273	199	110
Danube Basin	318	319	132
South Slovakian Basin	289	257	21
East Slovakian Basin	411	232	177

Explanations for tables 1 - 5: TOC - total organic carbon; S₁, S₂, HI, T_{max} - Rock-Eval pyrolysis parameters; R₀ - vitrinite reflectance in non-polarized light, nm - non measured

(CGS) Brno on the Leitz Wetzlar MPV II microphotometer under the following conditions: Monochromatic light (λ = 546 nm), circular micro-photometric field (r = 1 mm) and calibration standard – glass prism (Ro = 1.24 %). Measurements were carried out in oil immersion on polished sections from the well cores.

The overview from the basic organic geochemical analysis in individual basins is presented in Tab. 1. The kerogen quality and its thermal maturation stage in the Neogene sedimentary fill and its basement in the investigated areas are presented in Tabs. 2–5. All analyses with their entire characteristics of well name, depth position, geological unit and detailed stratigraphy are listed in Pereszlényi et al. (1996).

Lacustrine organic matter (OM) of alginites was also present with the "classical" oil source rocks in the South Slovakian Basin maar structures. This OM type was studied in 99 samples in the Lučenec Depression at Pinciná at depths spanning 7–48 m. The hydrogen index from the Rock Eval pyrolysis was up to 962 mg HC/TOC at the maximum, and this indicates the II – I kerogen type of algal origin. These sediments are thermally immature, and according to their high TOC values of 3.8 to 28.6 wt.%, they correspond to the oil shales, and therefore these were not evaluated with "classical" oil source rocks. A more detailed organic geochemical description can be found in Vass et al. (1997) and also in Milička (2000).

The increased TOC and S2 residual hydrocarbon potential values with low HI values for the hydrogene index shown in Tab. 5 correspond to the coal seams in the Plachtince and Pôtor beds (Vass et al., 2005).

Modelling

It is possible to reconstruct the actual temporal and spatial processes of the hydrocarbon origin, migration and accumulation, which occurred in the basin formation period. According to the geological structure it is necessary to model several points, such as wells.

Tab. 2
Basic organic-geochemical characteristics of Neogene sedimentary fill and the basement in the Slovak part of the Vienna Basin

Geological unit	TOC %	S ₁ mg/g	S ₂ mg/g	HI mg/g	T _{max} °C	R _o %
Neogene	0.30 - 2.50	0.0X - 0.85	0.0X - 4.50	11 – 225	420 – 445	0.37 - 1.08
Paleogene	0.40 - 0.95	0.0X	0.0X - 0.58	32 - 83	427 - 447	0.37 - 1.26
Mesozoic	0.0X - 0.89	0.0X	0.0X - 0.78	36 – 211	435 – 446	1.14 - 1.65

Tab. 3
Basic organic-geochemical characteristics of Neogene sedimentary fill and the basement of the Danube Basin

Geological	TOC	S ₁	S ₂	HI	T _{max}	R _o
unit	%	mg/g	mg/g	mg/g	°C	%
Neogene	0.0X - 1.94	0.0X - 0.30	0.10 - 4.80	20 - 409	425 – 446	0.24 - 0.82
Paleogene	0.0X - 1.45	0.0X - 0.23	0.12 - 5.86	62 - 404	427 – 444	0.33 - 0.80
Pre-Tertiary basement	0.0X - 0.40	0.0X - 0.42	0.0X - 1.78	10 – 110	440 – 530	0.78 - 1.80

Tab. 4
Basic organic-geochemical characteristics of Neogene sedimentary fill and the basement of the South Slovakian Basin

Geological unit	TOC %	S₁ mg/g	S ₂ mg/g	HI mg/g	T _{max} oC	R _o %
Neogene	0.10 - 1.80	0.0X - 0.54	0.0X - 2.03	1 – 185	425 – 538	0.65 - 2.86
Mesozoic	0.24 - 5.18	0.0X	0.0	_	_	2.51
Paleozoic	0.10 - 1.20	0.03 - 0.18	0.0	_	_	3.36 - 4.32

Tab. 5
Basic organic-geochemical characteristics of Neogene sedimentary fill and the basement of the East Slovakian Basin

Geological unit	TOC %	S₁ mg/g	S ₂ mg/g	HI mg/g	T _{max} °C	R _o %
Lower Miocene	1.42 – 47.0	0.0X - 2.80	1.60 - 48.14	39 – 226	370 – 471	0.32 - 0.52
Paleogene	0.0X - 1.34	0.X - 0.55	0.0X - 1.28	13 - 203	415 - 438	nm
Mesozoic	0.20 - 1.17	0.0X	0.0	-	_	nm

The model of basin formation is created on the input optimizing parameters of sediment burial history, in the form of subsidence curves and hydrocarbon generation zones. These optimizing parameters include (1) the type of event, such as deposition, hiatus, erosion and thrusting, (2) the duration time, (3) the lithology and initial sediments thicknesses, (4) the initial porosities, (5) the water depth during deposition and (6) the paleo-temperatures and paleo-heat flow.

Modelling proceeds consecutively in solving geological tasks, so that the geological parameters describing the evolutionary basin sequences are recorded "step by step." Time is the main factor to which all parameters processes and products are related. The above-mentioned optimizing parameters, together with the corresponding algorithms, simultaneously represent the basis for calculations of theoretical vitrinite reflectance, of actual steady state temperatures, of steranes or hopanes isomerization, or of other parameters.

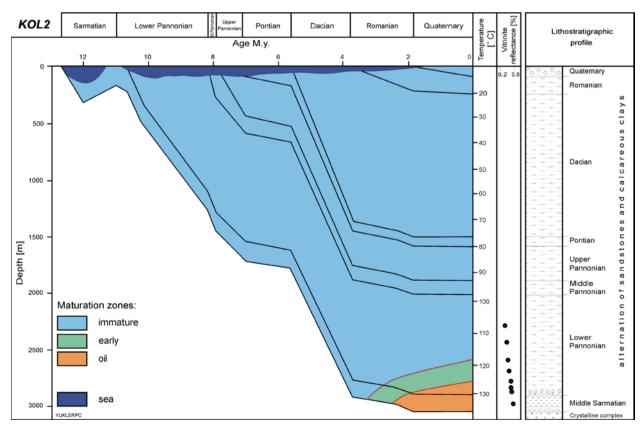


Fig. 2. Burial history plot and hydrocarbon generation windows in Kolárovo-2 well (Danube Basin).

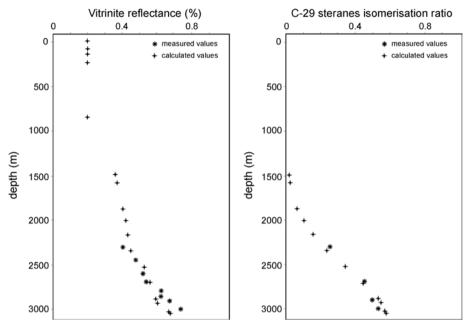


Fig. 3. Comparison of calculated (modeled) and measured parameters in Kolárovo-2 well (Danube Basin).

Theoretical calculations are compared with the control parameters, such as measured vitrinite reflectance, biomarkers analysed by GC-MS, steady state temperatures measured in wells and also the laboratory-estimated porosity in the well cores. Consequently, the optimizing parameters are modeled as long as an acceptable match within the theoretically calculated and control-measured parameters is reached. An example of modelling in the Kolárovo-2 well in the southern part of the Danube Basin is presented in Figs. 2 and 3.

Method of hydrocarbon resources calculation

The hydrocarbon potential of the basins was estimated on the basis of interpretations of the organic-geochemical analyses, and on the kinetic modelling of hydrocarbon generation during their geological history. The calculation of hydrocarbons generated in a given region is based on the division of the hydrocarbon deposit originating process into the following phases: generation, expulsion, migration, entrapment and preservation. For the quantitative expression of the amount of generated hydrocarbons, we used Waples' (1985) method which mainly concentrates on the basic organic geo-chemical parameters.

Since this method is used in the American oil industry (originally Moshier and Waples, 1985), calculation results are expressed in imperial units, and these are transformed into SI units using conventional conversion factors.

a) Input parameters

The hydrocarbon generation equation is based on the following three basic organic-geochemical data; synsedimentary TOC values, hydrogen index and vitrinite reflectance.

Hydrocarbon volume (HC) = (k) \cdot (TOC) \cdot (HI) \cdot (f), where:

HC – hydrocarbon volume in million of barrels/cubic mile; and when transformed into SI units: million of barrels/cubic mile \cdot 0.0381427 = million m³/km³

k – conversion constant (0.7 of a shale of approx. 2 300 – 2 400 kg/m³ specific weight gives a hydrocarbon equivalent of approximately 900 kg/m³ oil

TOC – average content of the total organic carbon in wt.% HI – average hydrogen index in mg of hydrocarbons/g TOC

f – fractional conversion (0) – immature organic matter and (1) – mature OM; the considered values are based on the measured vitrinite reflectance

b) Calculation of oil generation

general formula:

volume of generated oil = $(k) \cdot (TOC) \cdot (HI) \cdot (f)$

where: the generated oil volume is expressed in millions of barrels/cubic mile, and converted into SI units: million of barrels/cubic mile \cdot 0.0381427 = million m³/km³

c) Calculation of gas generation

general formula:

volume of generated gas = $(k) \cdot (TOC) \cdot (HI) \cdot (f) \cdot (a)$

where: (a) is the conversion constant. When we want to express the gas volume in billion cubic feet/cubic mile of the source rock (a) = 6. When we want to express it in millions of m^3/km^3 , we must multiply the referred value by the coefficient of 0.0067935.

Remark: If we want to express the gas volume as an oil equivalent, we do not use (a) above; and we can then calculate the oil generation instead.

d) Hydrocarbon expulsion during the primary migration

For expulsion to take place, the threshold value of the generation of approximately 50 million barrels of hydrocarbons must be reached, and the oil, gas or oil equivalent for gas generation from 1 $\rm km^3$ of source rock is attained. When this threshold value is reached, the expulsion efficiency of oil is approximately 80 % and around 50 % for gas.

General formula:

The volume of expelled hydrocarbons = $(A) \cdot (B)$,

where: (A) – volume of generated hydrocarbons; (B) – expulsion efficiency

e) Efficiency of secondary migration and accumulation

Geologically, the efficiency of secondary migration and accumulation is generally accepted to be in the range of 10 to 20 %.

General formula:

The volume of accumulated hydrocarbons = the volume of expelled hydrocarbons multiplied by the efficiency of the secondary migration and accumulation.

It is most convenient to use imperial units up to this stage and then transform the results into SI units.

f) Total source volume in individual generation zones

General formula:

The volume of accumulated hydrocarbons in million m^3/km^3 multiplied by the volume of potential source rocks in km^3 = the total source volume.

Individual calculations must be carried out for each individual generation zone in a given region (for example for early oil, condensate and dry gas) because the fractional conversion for each generation zone is usually calculated

from vitrinite reflectance, and the volume of potential source rocks is different in individual generation zones.

The final resource volumes represent the addition of source volumes in the particular generation zones. The following values are normally used by conversion to equivalents:

1 m³ of oil corresponds approximately to 0.88 metric ton (t) 1 m³ of oil corresponds approximately to 1,000 m³ of gas 1 t of oil corresponds approximately to 1,222 m³ of gas

Kerogen maturation zonality

The Tertiary, or Neogene, basins developed in the final stages of orogenetic processes forming the Western Carpathians. These cover approximately 40 % of the territory of Slovak Republic. Their sedimentary basin fill reaches a thickness up to several km and it often lies discordantly on the basement. The molasses sediments of Neogene basins mainly comprise clays, sands and gravel, denoting shale, sandstone and conglomerates, plus evaporates and coaly and volcaniclastic formations deposited in the marine and brackish, but also in lacustrine-fluvial and terrestrial environments. The sources of the organic matter in these sediments originate from decomposition products of mostly continental but also of marine and lacustrine organic matter.

Regarding the total organic mater (TOC) amounts, these are rather poor to fairly good, so from this viewpoint the sediments of the Danube Basin are relatively poorer than those in the East Slovakian and Vienna basins. The Neogene sediments of the South Slovakian Basin are reduced to the Lower Miocene depositions, with mainly Ottnangian and Eggenburgian coal samples analysed here. Egerian sediments of the Lučenec Formation (Lučenec

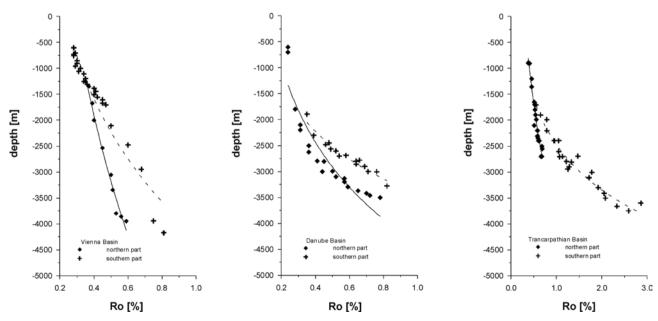


Fig. 4. Kerogen maturation trends based on vitrinite reflectance in the northern and southern part of the Vienna, Danube and East Slovakian Neogene basins.

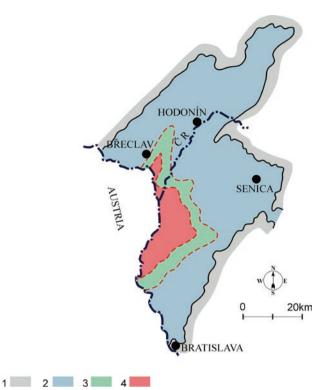


Fig. 5. Vienna Basin – hydrocarbon generation zones at the base of Neogene sediments. 1 – pre-Neogene Inner West Carpathian Units; Generation zones: 2 – immature; 3 – early oil; 4 – oil.

Depression) represent average and rather poor source rocks when compared with the average source rocks in the Danube Basin. The most dominant kerogen type in all three basins is the terrestrial type III. This is followed by the mixed terrestrial-marine type III-II, with coal lignite seams and coaly shales present in some places.

The depth maturation trends of organic matter depend on the geothermal gradient in individual basins. These differ considerably, and this fact is supported by microphotometric measurements, where the structural ordering of synsedimentary vitrinite reflects temperature

alterations at the given depths (Fig. 4). The kerogen maturation trends related to differing depths concurrently exhibit different alternative gradients in the northern and southern parts of the individual basins. This corresponds to the actual heat flow, or geothermal gradient distribution (Fig. 4).

Hydrocarbon generation zones at the base of the Neogene sediments are presented in Figs. 5, 6, 7 and 8. In cross section, the generation zones are shown in approximately NW–SE geological orientation in Figs. 9, 10 and 11. According to the Rock Evaluation pyrolysis results, vitrinite reflectance measurements, n-alkane distribution, and also in some cases, the steranes and pentacyclic triterpane distribution of Neogene sedimentary fill in the investigated basins can be summarized as follows:

The average hydrocarbon potential of Neogene sediments varies depending on the organic matter type by around 0.5–2.0 kgHC/t rock, and in the best locations it is 2–5 kgHC/t rock. Hence, active hydrocarbon generation is expected in the deepest buried Lower- and partly Middle Miocene sediments in the Vienna Basin and also in the Middle to Upper Miocene sediments in the central part of the Danube Basin (Figs. 9 and 10).

The processes of kerogen maturation and its conversion to hydrocarbons in the East Slovakian Neogene Basin take place at the shallowest depths of 1.8–2.7 km (see also Franců et al., 1989), while, in the Danube Basin this is deeper at 2.3–3.5 km, and it occurs at the greatest depths of about 3 to 5 km in the Vienna Basin (Fig. 12).

Modelling of kerogen alteration based on sediment burial history and on geothermal conditions indicates that the differences in the basins are mainly due to differential geothermal gradient distribution. These different thermal conditions and the relative kerogen conversion rate also influence the depth interval of the individual hydrocarbon generation zones (Fig. 12). Differences in the maturation stages of the organic matter also depend on local geothermal conditions, and these are particularly evident between the northern and southern parts of the basins. This is connected with sedimentation inversion, resulting in a shift of the depositional centres to the south, and

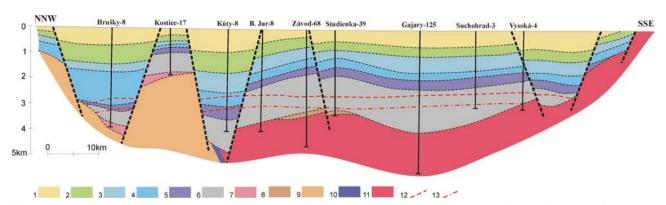
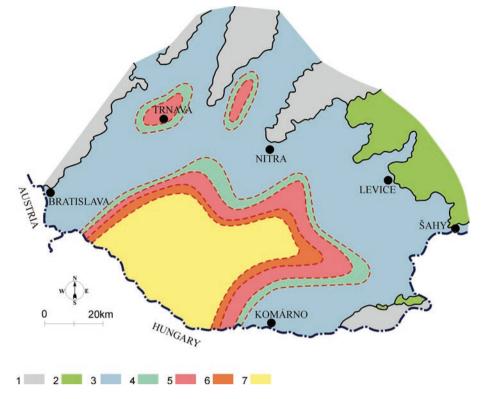


Fig. 6. Vienna Basin: NNW—SSE schematic geological cross section with hydrocarbon generation zones. 1 – Dacian, Pontian and Pannonian; 2 – Sarmatian; 3 – Upper Badenian; 4 – Middle Badenian; 5 – Lower Badenian; 6 – Karpatian; 7 – Ottnangian; 8 – Eggenburgian; 9 – Flysch nappes; 10 – Klippen Belt; 11 – Mesozoic rocks of Alpine and Central Carpathian nappes; Top generation zones: 12 – early oil, 13 – oil.

Fig. 7. Danube basin – hydrocarbon generation zones at the base of Neogene sediments.1–pre-Neogene Inner West Carpathian units; 2 – Late Cenozoic volcanics; 3 – immature, 4 – early oil; 5 – oil; 6 – condensate; 7 – dry gas (methane).



also with the change in tectonics to the thermal phase of subsidence.

Hydrocarbon resources of Neogene sediments calculated by the Waples (1985) method

The different geodynamic evolution in individual Neogene basins has influenced the conversion intensity of kerogen to hydrocarbons, and thus also the volume of generated and entrapped hydrocarbons.

Vienna Basin (Slovak part)

The kerogen type II - III, in the sense of Espitalié et al. (1986), was considered for the calculation of hydrocarbon resources in the Slovak part of the Vienna Basin. This resulted in 40 % oil and 60 % gas (40 wt.% for oil and 60 wt.% for gas) being generated from the whole hydrocarbon volume. The threshold value for the active early oil generation stage was not exceeded in the Neogene sediments of the Vienna Basin, and it was reached only

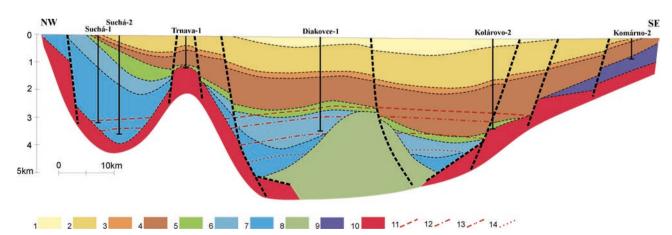


Fig. 8. Danube Basin: NW–SE schematic geological cross section with hydrocarbon generation zones. 1 – Romanian and Quaternary; 2 – Dacian; 3 – Pontian; 4 – Pannonian; 5 – Sarmatian; 6 – Upper Badenian; 7 – Middle Badenian; 8 – Lower and Middle Badenian volcanics; 9 – Mesozoic and Paleozoic rocks of Transdanubian Central Range; 10 – crystalline rocks of Inner West Carpathians; Top of generation zones: 11 – early oil; 12 – oil; 13 – condensate; 14 – dry gas.

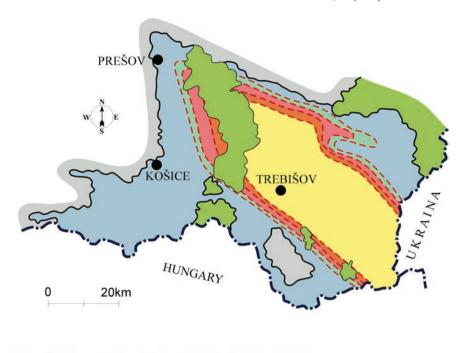


Fig. 9. East Slovakian Neogene Basin – hydrocarbon generation zones at the base of Neogene sediments. 1 – pre-Neogene Inner West Carpathian units; 2 – Late Cenozoic volcanics; Generation zones: 3 – immature; 4 – early oil; 5 – oil; 6 – condensate; 7 – dry gas (methane).

for the main oil generation stage. The threshold value for primary migration in the Vienna basin was not exceeded in the early generation stage, and this was only reached for the main oil generation stage. Regarding the efficiency of secondary migration within the individual generation stages that were reached, the following amounts of hydrocarbons were generated:

1 2 3 4 5 6 7

Oil generation stage:

oil: 17.7 million t gas: 7.9 billion m³

Total amount of generated hydrocarbons calculated from all generation zones:

 Σ oil: 17.7 million t Σ gas: 7.9 billion m³

Remark: With regard to its geothermal gradient, the Vienna Basin is relatively cold. When we consider an active bacterial activity up to a depth with 70 °C rock temperature (Rice and Claypool, 1981), this corresponds to depths of up to 2 km in various parts of the Vienna Basin. Mainly Lower Miocene to Lower Pannonian marine and brackish sediments occur at these depths and they comprise immature hydrocarbon source rocks. Therefore, a contribution in the order of billion m³ of bacterial (biogene) methane can be expected, as seen in e.g. the Lower

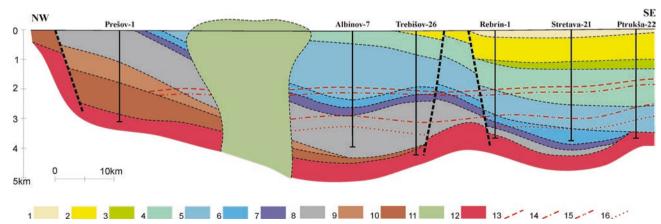
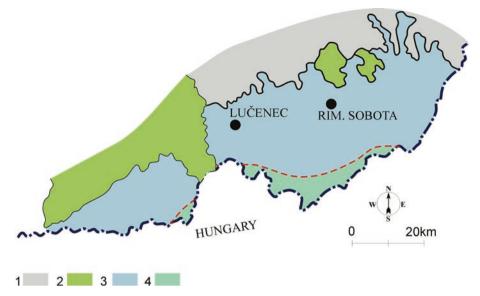


Fig. 10. East Slovakian Basin: NW–SE schematic geological cross-section with hydrocarbon generation zones. 1 – Pontian; 2 – Pannonian; 3 – Upper Sarmatian; 4 – Middle and Lower Sarmatian; 5 – Upper Badenian; 6 – Middle Badenian; 7 – Lower Badenian; 8 – Karpatian; 9 – Egger and Eggenburgian; 10 – Central Carpathian Paleogene; 11 – Miocene volcanics; 12 – Mesozoic and Paleozoic basement rocks; Top of generation zones: 13 – early oil; 14 – oil; 15 – condensate; 16 – dry gas.

Fig. 11. South Slovakian Neogene Basin – hydrocarbon generation zones at the base of Neogene sediments. 1 – pre-Neogene Inner West Carpathian units; 2 – Late Cenozoic volcanics; Generation zones: 3 – immature; 4 – early oil.



Pannonian deposit of Suchohrad and in the Sarmatian sediments in Malacky. However, in "hotter" basins, such as the Danube and East Slovakian Neogene basins, the temperature of 70 °C is already attained, especially in fresh water sediments with poor organic matter content, and therefore the contribution of bacterial methane is considerable lower.

Danube Basin

Here, the kerogen type III was considered in the hydrocarbon resource calculation. The threshold value for expulsion in the early and oil generation stages was not reached. It was attained only for the condensate and gas generation stage.

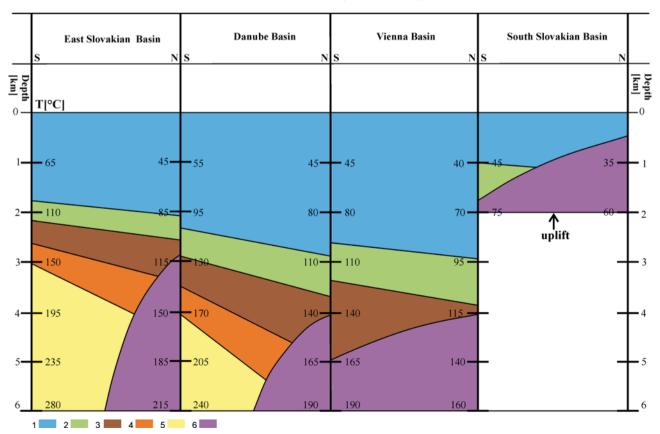


Fig. 12. Comparison of hydrocarbon generation zones in Neogene sediments of Vienna, Danube, East Slovakian and South Slovakian basins. Temperature values according to Franko et al. (1995). Generation zones: 1 – immature; 2 – early oil; 3 – oil; 4 – condensate; 5 – dry gas; 6 – undivided pre-Neogene rocks.

Regarding primary migration efficiency, the following hydrocarbon volumes were generated in particular generation stages:

Condensate generation stage

oil: 12.6 mil. t gas: 30.3 mld. m³

Dry gas generation stage

oil*: 17.2 million m³ gas: 38.8 billion m³

Total amount of generated hydrocarbons calculated from all generation zones:

 Σ oil: 12.6 million t Σ gas: 86.3 billion m³

The new 2D seismic profile interpretation clearly indicated the presence of buried Badenian stratovolcanoes, and it showed that the original source rock index of 0.55 decreased to 0.40. Consequently, the hydrocarbon generated volumes decreased as follows:

Condensate generation stage

oil: 9.2 million t gas: 22.0 billion m³

Dry gas generation stage

oil*: 12.5 million m³ gas: 28.2 billion m³

Total amount of generated hydrocarbons calculated from all generation zones:

 Σ oil: 9.2 million t Σ gas: 62.8 billion $\rm m^3$

*oil volume thermally degraded to gas within the dry gas generation zone

East Slovakian Basin

Here, the kerogen type III was considered for hydrocarbon resource calculation. The threshold value for expulsion in the early and oil generation stages was not reached. It was attained only for the condensate and gas generation stage.

Regarding primary migration efficiency, the following hydrocarbon volumes were generated in particular generation stages:

Condensate generation stage

oil: 12.8 million t (light oil, condensate)

gas: 30.8 billion m³

Dry gas generation stage

oil*: 32.6 million m³ gas: 73.4 billion m³

Total amount of generated hydrocarbons calculated from all generation zones:

 Σ oil: 12.8 million t Σ gas: 136.8 billion m³

*oil volume thermally degraded to gas within the dry gas generation zone

South Slovakian Basin

Here, the kerogen type III was considered for hydrocarbon resource calculation i.e. that from the total amount of generated hydrocarbons corresponds 25 % of them to oil and 75 % to gas (0.15 wt.% for oil and 0.75 wt.% for gas). Neogene sediments (mainly Lower Miocene sediments) reached practically only the initial stage of early generation zone and, moreover, regarding their depth position they occur actually in a passive (relict) generation stage.

Conclusions

With respect to the sedimentation environment type, the total amount of organic matter varies considerably. Although the volume of total dispersed organic matter in the investigated basins was generally rather poor at 0.5–1.0 wt.%, in some places it was relatively good at 2–5 wt.%. The most prospective source rocks are represented by the Lower Miocene sediments in the Vienna Basin, by the Badenian and Sarmatian sediments in the East Slovakian Basin and by Lower Pannonian sediments in the Danube Basin. Neogene sediments in the South Slovakian Neogene Basin consist of reduced Lower Miocene deposits, and for TOC content they represent only average, but most often, rather poor source-rocks.

The most dominant kerogen type in all three basins is the terrestrial type III (HI = 100–250 mgHC/g TOC). However, in some places there are marine and lacustrine sediments representing the mixed terrestrial-marine type III-II (HI = 300–400 mgHC/g TOC).

The vertical organic matter thermal maturation trend differs in each basin. Kerogen maturation occurs at shallower depths in the East Slovakian Neogene Basin and it is deepest in the Vienna Basin. The amounts of hydrocarbons generated in the Danube Basin are reduced due to buried Neogene volcanics. Sediments in the South Slovakian Neogene Basin do not actually produce hydrocarbons. This is mainly due to their depth position and geological history, and they only reached the early hydrocarbon generation window (Fig. 12).

The large Neogene basins in Slovakia are still considered to be the oldest "traditional" oil prospecting areas and these remain the most prospective hydrocarbon areas in our country due to their high exploration potential. The most prospective areas are mainly concentrated in the central deepest parts of the basins, which have not yet been sufficiently explored by the deep boreholes.

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References

ČECH, F., 1988: Dynamics of Neogene Carpathian basins in relation to deep structure, crustal type and fuel deposits. Bratislava, GÚDŠ, 293 p.

- ESPITALIÉ, J., DEROO, G. & MARQUIS, F., 1986: La pyrolyse Rock Eval et ses application. Partie II. Rev. Inst. franç. Pétrole, 40, 6, 755 – 784.
- FRANCŮ, J., RUDINEC, R. & ŠIMÁNEK, V., 1989: Hydrocarbon generation zone in the East Slovakian Neogene Basin: Model and geochemical evidence. *Geol. Carpath.*, 40, 3, 355 384.
- Franců, J., Radke, M., Schaefer, R. G., Poelchau, H. S., Čáslavský, J. & Boháček, Z., 1996: Oil-oil and oil-source rock correlations in the northern Vienna Basin and adjacent Carpathian Flysch Zone (Czech and Slovak area). In: Oil and gas in Alpidic thrust belts and Basins of Central and East Europe. Wessely, G. & Liebl, W. (eds.). Special publication of the European association of geoscientist and engineers, No. 5, Geological Society, London, 343 354.
- FRANKO, Ö., REMŠÍK, A. & FENDEK, M. (eds.), 1995: Atlas of geothermal energy of Slovakia. *Bratislava*, *GÚDŠ*, *230 p.*
- Janočko, J., Pereszlényi, M., Vass, D., Bezák, V., Kohút, M., Polák, J., Mello, J., Jacko, S., jr. & Jacko, S., 2006: Geology and hydrocarbon resources of the Inner West Carpathians, Slovakia and Poland. *In: Golonka, J. and Picha, F. (eds.): The Carpathian and their Foreland: Geology and hydrocarbon resources: AAPG Memoir, 84, 569 603.*
- Jıňíčeк, R., 1978: Paleogeography of Lower Miocene in West Carpathians. Zem. Plyn Nafta, 23, 1, 21 28. (In Czech.)
- JIŘÍČEK, R., 1979: Discrepant evolution of the northern branch of Alpine orogene. Zem. Plyn Nafta, 24, 4, 555 580. (In Czech.)
- Jıříček, R., 2002: The evolution of the Molasse in the Alpine--Carpathian Foredeep and the Vienna Basin. *Exploration Geophysics, Remote sensing and Environment, Prague*, 9, 1 – 2, 179. (In Czech.)
- Kováč, M., 2000: Geodynamic, paleogeographical and structural evolution of Carpathian – Pannonian region during Miocene: New regard to the Slovak Neogene basins. *Bratislava, Veda,* 202 p. (In Slovak.)
- Ladwein, H. W., 1988: Organic geochemistry of the Vienna Basin; Model for hydrocarbon generation in overthrusts belts. *AAPG Bull.*, 72, 5, 586 – 589.
- MILIČKA, J., 2000: Organic geochemical evaluation of sedimentary organic matter and natural hydrocarbons occurrences in selected regions of West Carpathians, Habilitation Thesis. Comenius University, Faculty of Natural Sciences, Bratislava, 167 p. (In Slovak.)

- Moshier, S. O. & Waples, D. W., 1985: Quantitative evaluation of Lower Cretaceous Mannville Group as source for Alberta's oil sands. *AAPG Bull.*, 69, 161 172.
- PERESZLÉNYI, M., MILIČKA, J. & VITÁLOŠ, R., 1996: Hydrocarbon prospection in selected regions of Western Carpathians, Geochemistry (in Slovak). *Manuskript. Bratislava, VVNP, 67 p.*
- RICE, D. D. & CLAYPOOL, G. E., 1981: Generation, accumulation and resource potential of biogenic gas. *AAPG Bull.*, 65, 1, 5 25.
- ROYDEN, L. H., 1985: Vienna Basin: A thin-skinned pull-apart basin. In: Biddle, K. T. and Christie-Blick, N. (eds.): Strike-slip deformation, basin formation and sedimentation. The SEPM, Special Publications, 37,319 – 339.
- ROYDEN, L., HORVÁTH, F. & RUMPLER, J., 1983: Evolution of the Pannonian Basin System. 1. Tectonics. *Tectonics*, *2*, *1*, *63 90*.
- VASS, D., KONEČNÝ, V., ELEČKO, M., MILIČKA, J., SNOPKOVÁ, P., ŠUCHA, V., KOZÁČ, J. & ŠKRABANA, R., 1997: Alginite a new resource of the Slovak industrial minerals potential. *Bratislava, Miner. Slov.*, 29, 1 39. (In Slovak.)
- VASS, D. & ČVERČKO, J., 1985: Lithostratigraphic units of the Neogene in the East Slovakian Lowland. Geol. Práce, Spr., 82, 111 – 121. (In Slovak.)
- Vass, D., 2002: Litostratigraphy of Western Carpathians: Neogene and Buda Paleogene. *Bratislava*, *GÚDŠ*, 202 p.
- Vass, D., Milička, J., Pereszlényi, M. & Elečko, M., 2005: Lower Miocene coal measures buried by the Krupinská planina Plateau volcanics (Southern Slovakia). *Bull. Geosci., 80, 1, 105 112.*
- Welte, D. H., Kratochvil, H., Rullkötter, J., Ladwein, H. W. & Schaefer, R. G., 1982: Organic geochemistry of crude oils from the Vienna Basin and an assessment of their origin. *Chem. Geol.*, 35, 33 68.
- Waples, D. W., 1985: Geochemistry in Petroleum Exploration. D. Reidel Publishing CO., IHROC, Boston, 231 p.

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Uhľovodíkový potenciál severných výbežkov panónskeho panvového systému na Slovensku

Prieskum zameraný na uhľovodíky bol na území našej republiky donedávna sústredený predovšetkým do oblasti troch najrozsiahlejších neogénnych panví – viedenskej, východoslovenskej a podunajskej (obr. 1). V posledných rokoch pribudlo množstvo údajov z organicko-geochemického výskumu sedimentov, umožňujúce porovnať ich uhľovodíkový potenciál a do tohto porovnania zahrnúť aj sedimenty juhoslovenskej panvy.

Organicko-geochemická charakteristika skúmaných vzoriek je založená na stanovení obsahu celkového organického (TOC) a celkového anorganického uhlíka (TIC). Kvalita zdrojových hornín, uhľovodíkový potenciál, typ kerogénu a čiastočne aj miera jeho tepelnej premeny boli interpretované na základe výsledkov pyrolýzy Rock-Eval. Kvalitatívne zloženie horninových extraktov a ich

vzájomné korelácie boli zhodnotené na základe pomeru frakcií alifatických, aromatických a NSO-zložiek a distribúcie n-alkánov, izoprenoidov a aromátov metódami plynovej chromatografie (GC) a hmotnostnej spektrometrie (GC-MS). Prehľad základných organicko-geochemických analýz vykonaných v jednotlivých panvách je uvedený v tab. 1. Typ a kvalita kerogénu a stupeň jeho termálnej premeny v neogénnej výplni jednotlivých panví a v ich podloží je sumárne uvedený v tab. 2 – 5.

Proces vzniku, migrácie a akumulácie uhľovodíkov je možné pomocou modelov rekonštruovať v čase a priestore formovania panvy. Čas je hlavným faktorom, na ktorý sa vzťahujú všetky parametre, procesy a produkty. Teoretické výpočty sú porovnávané s kontrolnými meranými, resp. analytickými parametrami, ako je odraznosť vitrinitu,

izomerizácia steránov a triterpánov, ustálené teploty merané vo vrtoch a laboratórne stanovená pórovitosť vo vrtných jadrách. Ako príklad je uvedené modelovanie generačných okien uhľovodíkov vo vrte Kolárovo-2 na obr. 2 a 3. Hĺbkový trend tepelnej premeny sedimentárnej organickej hmoty v závislosti od geotermálneho gradientu bol posudzovaný najmä na základe merania odraznosti vitrinitu (obr. 4). Generačné zóny uhľovodíkov na báze neogénnych sedimentov sú znázornené na obr. 5, 6 a 7. V geologických rezoch približne SZ - JV orientácie sú generačné zóny znázornené na obr. 9, 10 a 11. Uhľovodíkový potenciál jednotlivých neogénnych panví bol stanovený na základe interpretácií organicko-geochemických analýz a kinetického modelovania tvorby uhľovodíkov počas geologickej histórie. Výpočet objemu uhľovodíkov v danom geologickom regióne je založený na rozdelení procesu vzniku ložísk uhľovodíkov do viacerých fáz: generovanie, expulzia (vytesňovanie), migrácia, zachytenie v pasciach a ich uchovanie. Na kvantitatívne vyjadrenie vygenerovaného množstva uhľovodíkov sme použili metódu výpočtu podľa Waplesa (1985), ktorá zohľadňuje najmä základné organicko-geochemické parametre.

Poznatky získané na základe interpretácie analytických údajov, modelovania vzniku uhľovodíkov a výpočtu potenciálne vygenerovaných uhľovodíkov možno zhrnúť nasledovne: Celkové množstvo organickej hmoty do značnej miery kolíše v závislosti od typu sedimentačného prostredia. Napriek tomu, že celkové množstvo rozptýlenej organickej hmoty je v študovaných panvách väčšinou nízke (0,5 – 1,0 hm. %), existujú aj relatívne bohaté polohy

s obsahom 2 – 5 hm. %. Najperspektívnejšími potenciálnymi zdrojovými horninami sú spodnomiocénne sedimenty viedenskej panvy, bádenské a sarmatské sedimenty východoslovenskej panvy a sedimenty spodného panónu v podunajskej panve. Neogénne sedimenty juhoslovenskej panvy sú redukované na spodnomiocénne uloženiny a predstavujú len priemerné až chudobné potenciálne zdrojové horniny.

Dominantným typom kerogénu je terestrický typ III (HI = 100 – 250 mgHC/g TOC), v morských a lakustrinných sedimentoch sa však lokálne nachádza tiež zmiešaný terestricko-morský typ III-II (HI = 300 – 400 mgHC/g TOC).

Hĺbkový trend premeny organickej hmoty a tvorby uhľovodíkov je v každej z týchto panví odlišný. Vo východoslovenskej panve prebiehajú procesy zrenia kerogénu v najmenších hĺbkach, v podunajskej vo väčších a vo viedenskej panve najhlbšie (obr. 12). Sedimenty juhoslovenskej panvy v súčasnosti uhľovodíky neprodukujú, najmä pre ich aktuálne malú hĺbku pochovania. Z pohľadu geologickej histórie dosiahla organická hmota týchto sedimentov len počiatočné štádium produkcie uhľovodíkov, ktoré je v súčasnosti naviac pasívne.

Aj keď veľké neogénne panvy Slovenska možno považovať za tradičné naftárske oblasti, resp. oblasti s relatívne vysokým stupňom preskúmanosti, z hľadiska vyhľadávania a prieskumu ložísk uhľovodíkov ostávajú naďalej najperspektívnejšími. Ide najmä o centrálne časti panví, ktoré zatiaľ neboli dostatočne overené hlbokými vrtmi.