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## HYDROCARBON GENERATION ZONE IN THE EAST SLOVAKIAN NEOGENE BASIN: MODEL AND GEOCHEMICAL EVIDENCE

(19 Figs., 1 Tab.)



**Abstract:** Kerogen maturation and smectite illitization in shales from deep boreholes are studied using geochemical analyses and Lopatin's method of burial and thermal history reconstruction. The boundaries between the immature, hydrocarbon generation, cracking, and dry gas zones are traced in regional sections and maps. Kerogen maturation and smectite dehydration are confronted with the zone of overpressured fluids. The area of relatively better conditions for hydrocarbon generation throughout the basin evolution is shown.

**Резюме:** На основе моделирования термической истории методом Лопатина, анализом созревания органического вещества и трансформации смектита в иллит определена катагенетическая зональность в неогеновых осадках Восточнословацкого бассейна в региональных разрезах и картах. Характеризована область с относительно лучшими условиями для образования углеводородов в рамках развития бассейна. Образование углеводородов и дегидратация смектита оказывают связь с аномально высокими пластовыми давлениями.

### Introduction

The East Slovakian basin has been explored for more than 30 years for petroleum and several pools are producing economical amounts of gas. Drilling to greater depth brings technical problems because of high temperature (200 °C at 4 km depth) and anomalously high fluid pressure (Rudinec, 1969, 1974, 1976, 1978b). In this paper hydrocarbon prospects of the deeper parts of the basin and of different areas are evaluated on the basis of characterization of diagenesis and catagenesis of rocks, their hydrocarbon source potential and delineation of the space in profiles and maps where the relatively most intensive hydrocarbon generation may be expected.

Organic geochemical methods are used to study maturation of kerogen (insoluble organic matter dispersed in rocks) and its conversion to hydrocarbons and residual kerogen. This concept of Vassoevich et al. (1969), Tissot—Welte (1978), Hunt (1979) and Durand (1980) searching where and when the "principal phase of hydrocarbon formation" took place in a basin is followed in this paper.

Clay mineral transformations during diagenesis and catagenesis are attended by dehydration which was often believed to play an active role in hydrocarbon

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migration from source rock (Burst, 1969; Perry—Hower, 1972; Klubova, 1973; Vassoevich et al., 1975; Bruce, 1984). Our study of smectite illitization shows another catagenetic indicator of the stage of postdepositional evolution of pelites.

The present stage of rocks in borehole section is characterized by analyses. At attempt to show a dynamic image of processes of alterations within basin evolution which led to the present stage, the time-temperature modeling of Lopatin (1971) and Waples (1980) is applied.

### Geological setting

The East Slovakian basin (Fig. 1.) is the western part of the Transcarpathian depression which is a promontory of the Pannonian basin in NE direction to the Carpathian arc. The present shape was formed mainly during a fast subsidence in the Late Badenian until Pannonian with associated formation of syndimentary faults and extensive volcanic activity. Paleogeographic and

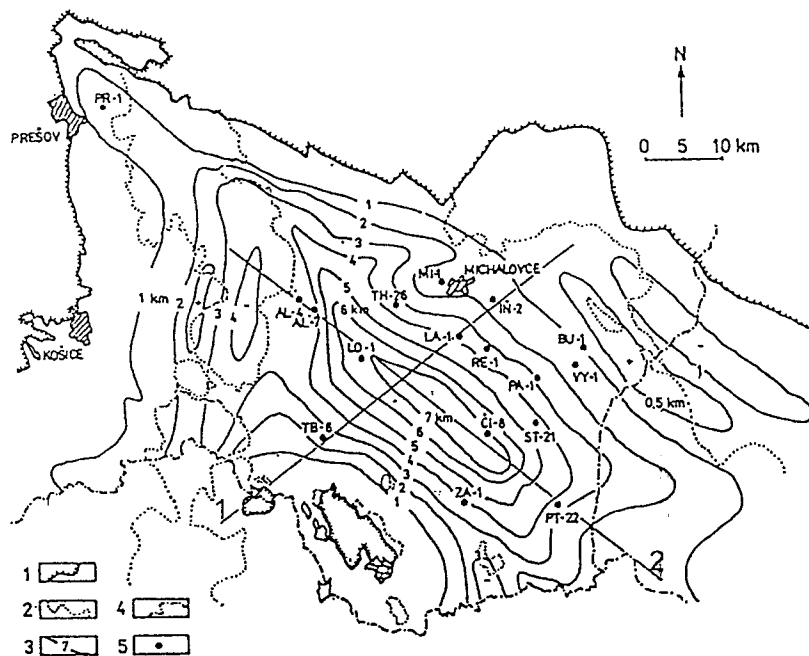


Fig. 1. Morphology of the basement in km below sea level of the East Slovakian Neogene basin (Czechoslovak part of the Transcarpathian depression) after Rudinec (1986) and the studied boreholes. Lines 1 and 2 are sections in Figs. 17. and 18. Legend: 1 — basement outcrops; 2 — contours of volcanic mountains; 3 — depth below sea level; (+), (—) — local elevations or depressions; 4 — border-line of CSSR; 5 — boreholes: AL — Albinov, BU — Bunkovce, CI — Čičarovce, IN — Iňačovce, LA — Lastomír, LO — Ložín, MI — Michalovce, PA — Pavlovce, PR — Prešov, PT — Ptruksa, Re — Rebrin, ST — Stretava, TB — Trebišov, TH — Trhovište, VY — Vysoká, ZA — Zátin.

structural evolution was described by Rudinec—Slávik (1973), Čverčko (1977), Rudinec (1978a), Gašparik (1979), Rudinec et al. (1981). The lithostratigraphic section of the Neogene in this basin shown in Fig. 2. is modified after Rudinec et al. (1981), Rudinec (1983) and includes the latest radiometric ages (Vass et al., 1987).

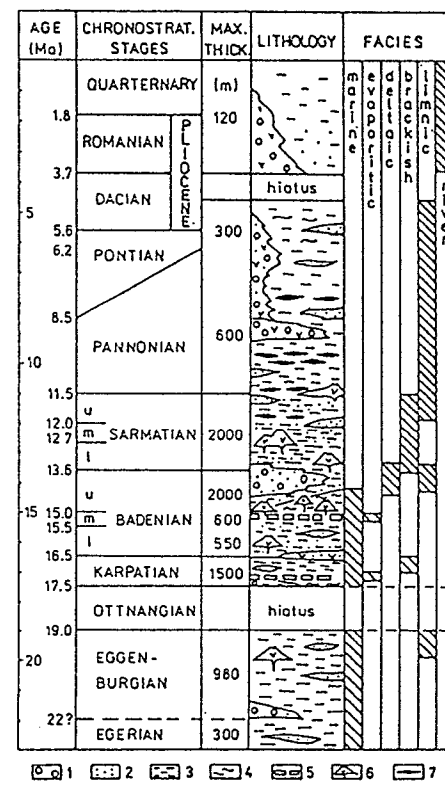


Fig. 2. Lithology, stratigraphy and facial evolution of the Neogene in the East Slovakian basin (Rudinec, 1983). Absolute ages are in mil. years (Vass et al., 1987).

Lithology: 1 — conglomerates; 2 — sandstones; 3 — shales; 4 — marls; 5 — evaporites; 6 — stratovolcanoes; 7 — coal seams.

Following features are typical of the East Slovakian basin:  
 — regional evaporitic strata on the base of the Upper Karpatian and in the Middle Badenian;  
 — extensive volcanic activity which was from the Eggenburgian till the Sarmatian mostly acid and from the Badenian till the Pannonian mostly intermediate;  
 — oxidizing facies of the Upper Karpatian.

Sedimentation rate reached in the East Slovakian basin maximum later than in other molasse basins of the West Carpathians (Vass—Čech, 1983; Vass et al., 1988). Maximal thickness of the Neogene filling is of about 7–8 km (Fig. 1.) (Mořkovský et al., 1986).

The most distinct attributes of the East Slovakian basin are very high heat flow (82–113 mW m<sup>-2</sup>) (Čermák, 1983; Kráľ et al., 1985) and average

geothermal gradient (40—54 mK m<sup>-1</sup>). In the central and SE part of the basin there is temperature of 200—209 °C at depth of 4 km (Rudinec, 1976, 1984a, b, 1986).

Đurica et al. (1979) and Đurica (1982) deduce from mineralogical analyses on recent metamorphism of zeolite facies at depth of 3—4 km.

In a more detailed study of clay minerals Kraus—Šamajová (1973, 1978) distinguished volcanic and nonvolcanic type of montmorillonite according to different ability to fix potassium irreversibly. Using this characteristic together with mineral association and trace element content they reconstructed material input from different source areas (volcanites, flysch belt, Mesozoic of Humenné, Paleozoic) in the sedimentation cycles of Neogene. In the whole East Slovakian basin they observed transformation of montmorillonite (smectite) to illite in a critical depth interval of 1.5—1.8 km. In evaporitic strata they found mixed-layer minerals illite-montmorillonite as products of diagenetic alteration of montmorillonite from volcanic glass.

Francú—Milička (1988) studied smectite illitization and organic maturation in four selected boreholes in the East Slovakian basin and concluded that in immature and early mature stages of source rocks diagenesis and catagenesis, the index of crystallinity of illite (Kübler, 1967) characterizes the detrital inherited illite or mica rather than degree of postdepositional alteration. Also, this index strongly depends upon the relative amount of detrital illite and newly formed illite/smectite in the analyzed sample. Consequently, the expandability of illite/smectite is studied in the present paper as a more reliable catagenetic indicator.

#### Previous petroleum geochemical studies

Hydrocarbon accumulations of economic importance are situated mainly in the E and NE part of the East Slovakian basin (Fig. 19). Hydrocarbons are gaseous with a minor content of condensate up to 120 g/Norm. m<sup>3</sup> of gas (gas field P tr u k š a, Fig. 1.) or 250 g/Norm. m<sup>3</sup> of gas (Stretava) (Rudinec, 1976b; Đurica, 1982).

Decrease in mineralization of formation waters in the East Slovakian basin was presented as an evidence of change of paleosalinity from marine facies through brackish to freshwater facies (Fig. 2.) by Michaliček (1970). Paleosalinity changes were proved also by Kraus—Šamajová (1978) on the basis of B and Sr content in clay minerals. Šimánek (1963, 1965, 1966, 1968, 1975) and Đurica et al. (1980) concluded following information from organic geochemical research:

- the Badenian and Lower Sarmatian strata have relatively the best source properties, in global scale, however, they are not very rich source and may produce rather gas than oil;
- coalification of organic matter attains the stage of anthracite at depth less than 4 km (Koch in Đurica et al., 1980);
- at contacts with volcanic bodies coalification is anomalously increased.

These authors pointed out the close linkage of thermal alteration of kerogen, hydrocarbon formation and parallel mineral transformations. This orientation is followed also in the present study.

#### Aims of this study

Both model and analyses were used to study hydrocarbon generation in the East Slovakian basin (Fig. 3.). First, the genetic type of kerogen and starting source potential of fine-grained rocks are characterized using chemical analyses. Then, degree of catagenetic alteration is estimated from maturation indices as vitrinite reflectance  $R_o$  and temperature  $T_{max}$  of Rock-Eval pyrolysis. Stage of catagenesis is also determined by model including burial history and time-temperature index (TTI) calculation. Analytical and calculated indices are compared and if they fit within a general correlation, or are acceptably close to it, we consider the determination of maturity to be quite reliable. In such case the modeled reconstruction of burial and thermal history is used as a part of geochemical interpretation indicating when the favourable thermal conditions to hydrocarbon generation were reached and what was the maturation rate.

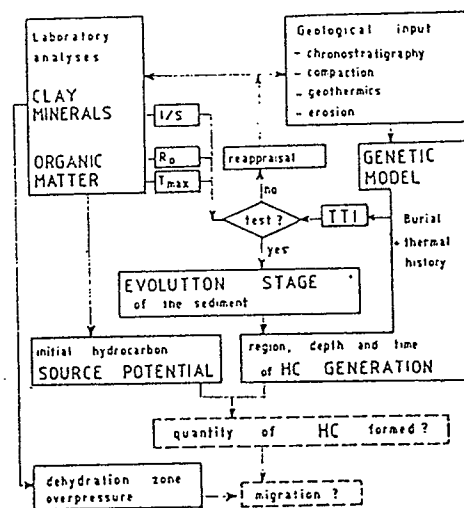


Fig. 3. Geochemical analyses and modeling used in this study of hydrocarbon generation in the sedimentary basin.

If the numerical result of model (TTI) does not correspond to analytical indices, we check possible errors either in analyses or in some of the geological data entering the model. Erosion and changes in geothermal gradient throughout geological history are often weaker points in modeling. We try to change the entering data within admissible alternatives until we obtain agreement of model with analyses.

Expandability of illite/smectite is believed to be an index of mineral catagenesis. We examine its relation to time-temperature index, i.e. to the total thermal exposure of rock. By comparing of the critical depths of smectite dehydration (illitization) and hydrocarbon generation, we aim at better understanding of possible interaction of these catagenetic processes and their links to fluid overpressure and migration.

### Methods

From cores of 17 boreholes shown in Fig. 1. fine-grained rocks were sampled. Vitrinite reflectance  $R_o$  was measured distinguishing different maceral groups and recycled organic matter. Internationally accepted conditions were followed (Stach et al., 1975): monochromatic nonpolarized light of 546 nm, oil immersion, photometric field  $2 \times 2 \mu\text{m}$ , Leitz MPV 2 (J. Francú analyst). Some details about vitrinite population determination were mentioned in the previous works of Francú — Müller (1983) and Francú (1986).

Organic and mineral carbon in rock ( $C_{org}$  and  $C_{min}$ ) were determined by method of Šmeral — Urbánek (1986). Rock-Eval pyrolysis (Espitalié et al., 1977) was used to determine the concentration of free hydrocarbons: S1 (mg HC/g rock) or FHC =  $S1/C_{org}$  (mg HC/g  $C_{org}$ ), fixed hydrocarbons: S2 (mg HC/g rock) or expressed as hydrogen index  $HI = S2/C_{org}$  (mg HC/g  $C_{org}$ ), and organic  $CO_2$ : S3 (mg  $CO_2$ /g rock) or oxygen index  $OI = S3/C_{org}$  (mg  $CO_2$ /g  $C_{org}$ ). Maximum pyrolysis temperature  $T_{max}$  (°C) is the peak of the signal S2 and is used as indicator of stage of maturation of kerogen in source rock. As a standard the menilite shale was used (Strnad et al., 1981).

Clay mineral fraction (less than  $2 \mu\text{m}$ ) was separated by sedimentation after ultrasonic disintegration. Oriented samples on glass slides (air-dried, saturated with ethylene-glycol by vapour method, and heated at  $550^\circ\text{C}$  for 2 hours) were analysed by X-ray diffraction under following conditions:  $CuK_\alpha$  irradiation, Ni filtered, 40 kV, 20 mA, slits 1—0 and 2—1, time constant 4, goniometer  $2^\circ 2\theta \text{ min}^{-1}$  from  $35$  to  $3^\circ 2\theta$ , chart speed  $20 \text{ mm min}^{-1}$ , diffractometer Philips PW 1150/25 (B. Toman and M. Plšková analysts). Minerals were determined after Brindley — Brown (1980), expandability of mixed-layer minerals illite/smectite (I/S) were determined by comparison of measured diffraction patterns with computer-modeled diffraction profiles of Reynolds — Hower (1970), Drits — Sakharov (1976), Reynolds (1980), Šrodoň (1980, 1981, 1984) and Šrodoň — Eberl (1984). This method was explained also by Johns — Kurzweil (1979). Positions of diffractions at  $5.21$ — $6.8$ ,  $10.4$ — $8.7$ , and  $15.8$ — $17.7^\circ 2\theta$  shown in Fig. 12. were taken as decisive.

Selected clay fraction samples were analysed by thermogravimetry (TG, heating rate  $20^\circ\text{C min}^{-1}$ , nitrogen, Du Pont 990, I. Horváth analyst) to quantitatively characterize the amount of water released during dehydration up to  $150^\circ\text{C}$  from expandable clay minerals (Fig. 11.).

Time-temperature modeling of catagenetic alterations is based on assumption that coalification and kerogen maturation is a complex physicochemical process which may be described by kinetic equations of first order mononucleous reactions (Karweil, 1956). Resulting product (e.g. vitrinite reflectance) depends on thermal exposure of the reacting system (rock) which is a linear function of time and exponential function of temperature. Lopatin (1971) proposed a simple method showing how series of strata were buried to depth and passed through subsequent temperature intervals (Fig. 14.) and received partial "impulses of heat". Total thermal exposure of rock called time-temperature index (TTI) is sum of these partial impulses.

In our burial history diagrams we use simplified relationship of Perrier —

Quiblier (1974) to correct the present sedimentary layer thickness for compaction.

The Lopatin's method is explained by Waples (1980, 1984 and 1985) who tested it in different geological setting and presented correlations of TTI with different catagenetic indicators.

### Source rock quality

Source rock richness is controlled by primary accumulation of organic matter in sediment and type of kerogen. Generally, the lower limit of total organic carbon  $C_{org}$  for an effective source rock is  $0.5\%$  Dow, 1977; Tissot —

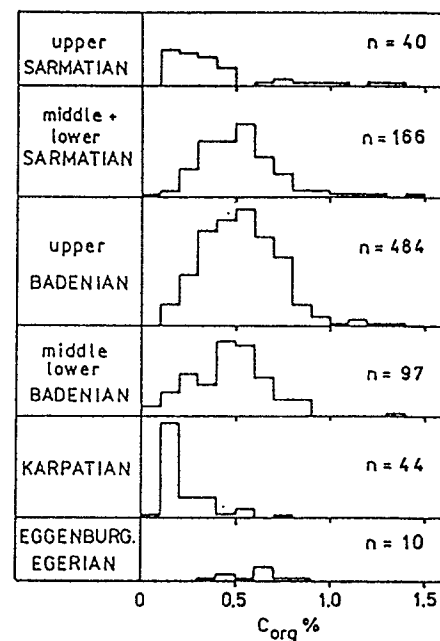


Fig. 4. Histograms of concentration of organic matter in pelites of the Neogene units in the East Slovakian basin:  $C_{org}$  — total organic carbon in rock in mass percentage,  $n$  — number of samples in a population, vertical axis has variable scale.

Weltje, 1978). Below this critical value hydrocarbons, if any generated, are so diluted that being adsorbed on clay mineral surface they cannot migrate from their host rock.

Histograms of total organic carbon content in shales and siltstones in main Neogene stratigraphic units of the East Slovakian basin are shown in Fig. 4., comprising latest data as well as those of Šimánek, (1963, 1965). Badenian and Sarmatian pelites are relatively more favourable, although they are not very rich source rocks. Sediments of the Upper Karpatian are often of oxidizing facies,  $C_{org}$  is generally less than  $0.2\%$  and as a source of hydrocarbons they are the least promising.

Total organic carbon content itself does not guarantee good source potential. Rocks containing high amount of inertinite (primarily oxidized), recycled or

graphitic organic matter may have no source potential (Tissot—Welte, 1978). Decisive property is hydrogen content in the macromolecule of kerogen. Tissot et al. (1974) characterized three essential types of kerogen according to their atomic ratios H/C and O/C. Similar diagram is shown in Fig. 5. (after Espitalié et al., 1977) where hydrogen and oxygen indices (HI and OI) are used from Rock-Eval pyrolysis. Arrows show direction of chemical changes in course of maturation of each type from immature to overmature stages. Because of lower reliability of oxygen index of samples with  $C_{org}$  less than 1%, diagram HI— $T_{max}$  (Espitalié et al., 1985) is used (Fig. 6.) to determine the kerogen type. Good oil-source rocks contain kerogen type I or II (lacustrine algal or planktonic marine). Neogene rocks from the East Slovakian basin contain humic type (III) of kerogen which is poorer in hydrogen (H/C less than 0.8 and HI less than 150, Figs. 5 and 6), and is derived from continental plant debris without considerable microbial alteration. As observed

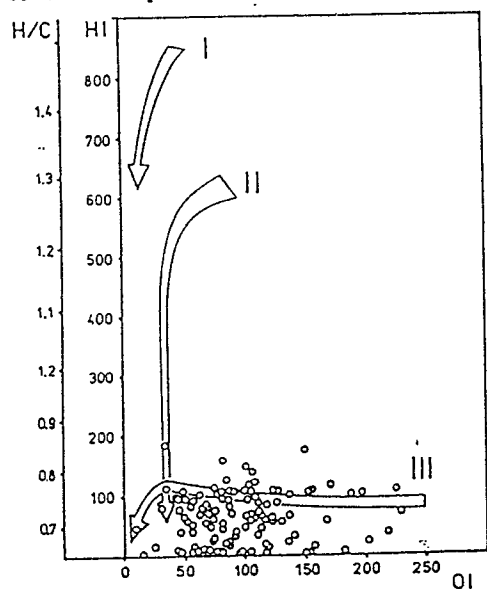


Fig. 5. Evolution paths of three essential types of kerogen in a modified diagram of van Krevelen: type I — algal, sapropelic, rich oil-prone, type II — marine, planktonic or mixed and rich in lipitine, major oil- and gas-prone, type III — humic, continental flora, gas-prone (Tissot et al., 1974; Espitalié et al., 1977).

Notes: HI — hydrogen index (mg HC/g  $C_{org}$ ), H/C — atomic ratio transformed from HI after Espitalié et al. (1977), OI — oxygen index (mg  $CO_2/g C_{org}$ ). Circles denote samples from the East Slovakian Neogene basin.

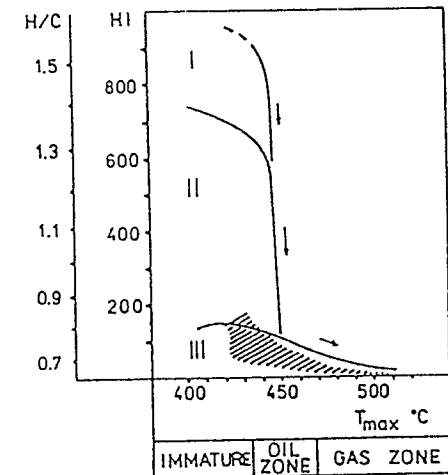


Fig. 6. Evolution paths of essential types of kerogen as decrease of hydrogen content (HI, H/C, see Fig. 5) during increasing maturation (arrows) indicated by pyrolytic temperature  $T_{max}$  (Espitalié et al., 1985). Studied samples lie in the hatched area.

in many basins all over the world, this type of kerogen is not able to generate economic amounts of oil but it is a good source of gas and condensate (wet gas) (Tissot—Welte, 1978).

Presence of humic kerogen in the East Slovakian basin is confirmed also by microscopical observations. In the Upper Karpatian shales and siltstones kerogen is of inertinitic character and has hydrogen content below detection limit of Rock-Eval pyrolysis. This unit has therefore poor source properties not only because of low organic carbon content but an unfavourable type of kerogen as well, what supports earlier conclusions of Šimánek (1975) and Durica et al. (1980). The Lower Karpatian, however, shows according to preliminary evaluation more promising character and should be given more attention in future studies.

#### Maturation indices

Catagenesis (or thermal diagenesis, Robert, 1988) of sedimentary rocks commences at deeper burial below younger units when under influence of higher temperature (over 50—60 °C) organic matter and clay minerals undergo substantial physicochemical alterations (Drits—Koporulin, 1973; Tissot—Welte, 1978; Pelet, 1980). Kerogen becomes mature and the principal phase of hydrocarbon generation is reached. Zone of maximal liquid hydrocarbon formation is often called "oil window" (Pusey, 1973). In a simplified scheme, with further maturation and thermal exposure, the rocks pass from "oil zone" to "gas zone" where liquid hydrocarbons are cracked to gaseous ones (Fig. 6.).

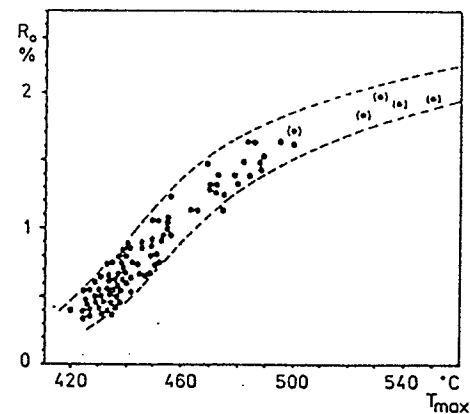


Fig. 7. Correlation of independent maturation indices:  $R_o$  — vitrinite reflectance,  $T_{max}$  — maximum pyrolytic temperature (points in parentheses are at detection limit).

It is necessary to determine the degree of catagenetic alteration to be able to answer whether the source rock reached conditions favourable for hydrocarbon generation. Héroux et al. (1979) presented a review of mostly used catagenetic or rather maturation indices. In our study vitrinite reflectance  $R_o$  and  $T_{max}$  of Rock-Eval pyrolysis are determined and mutually correlated. The relationship shown in Fig. 7. is very similar to a correlation belt of  $R_o$  —  $T_{max}$  for a genetic sequence of humic coals presented by Teichmüller—Durand (1983) with a slight difference that for  $R_o < 0.4\%$  their  $T_{max}$  values are by 5—10 °C lower than the ours. This shift of maximum pyrolysis

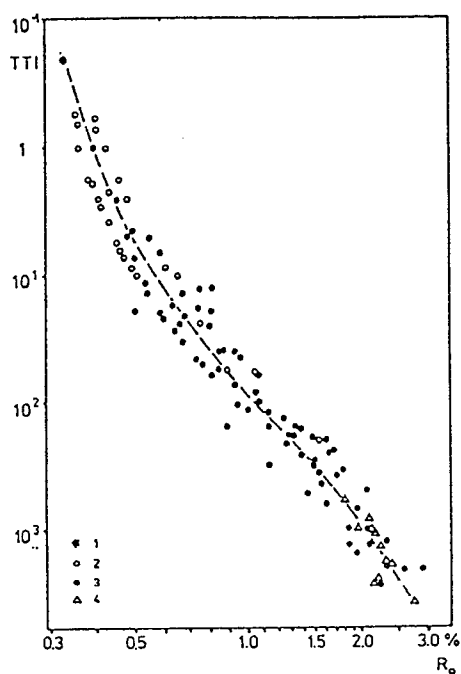


Fig. 8. Correlation of measured vitrinite reflectance  $R_0$  and calculated time-temperature index (TTI) representing thermal exposure of rock. This plot is used to transfer calculated TTI to theoretical vitrinite reflectance  $R_{TI}$  which is compared with measured  $R_0$  in tests of maturation models (Figs. 14. and 15).  
Stratigraphy of samples: 1 — Pannonian; 2 — Sarmatian; 3 — Badenian; 4 — Karpatian.

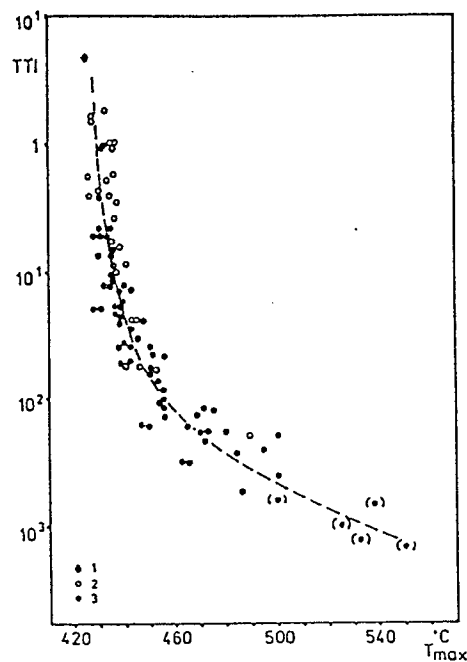


Fig. 9. Correlation of measured  $T_{max}$  of pyrolysis and TTI used by testing of models similarly as Fig. 8.  
Stratigraphy of samples: 1 — Pannonian; 2 — Sarmatian; 3 — Badenian (points in parantheses at detection limit).

temperature in whole rock analysis towards higher values may be attributed to mineral matrix effect. We use the correlation in Fig. 7. for mutual conversion of  $R_0$  and  $T_{max}$  if one of these indices is not available, sometimes also for checking-up on true vitrinite population identification in debatable histograms (under condition that kerogen is of humic type).

For each sample the time-temperature index is calculated. TTI does not have physical units, its values are just proportional to total thermal exposure of rock and is therefore used as a scale for catagenetic evolution from very early to very late stages. We correlate theoretical TTI with real analytical properties — vitrinite reflectance  $R_0$  and  $T_{max}$  of pyrolysis, and expandability of illite/smectite (which is discussed below). Figs. 8, 9 and 11 and Tab. 1 are based on the most reliable set of data and are used for mutual conversions and cross-checking of independent catagenetic indices.

Relationship  $R_0$  — TTI (Fig. 8.) is very similar to that of Waples (1980) except when TTI is less than 10. Than the  $R_0$  values are by less than 0.1% lower. This may be caused by subjective influence on  $R_0$  determination. The similarity of our relationship with that of Waples (1980, tabled data) is surprising, as some authors (e.g. Issler, 1984) obtained different correlations of  $R_0$  — TTI in different basins what caused decrease of confidence of geologists in the TTI method. We use Fig. 8. (sometimes also 9 and 11) to transfer TTI calculated from model to theoretical vitrinite reflectance  $R_{TI}$  (or other catagenetic indices). The modeled reconstruction of maturation history in each borehole section is then verified by comparing  $R_{TI}$  with measured vitrinite reflectance  $R_0$  (Figs. 14. and 15.).

Table 1

Correlation of hydrocarbon generation and cracking with smectite dehydration and of different catagenetic indicators:  $R_0$  — vitrinite reflectance,  $T_{max}$  — peak pyrolysis temperature, TTI — time-temperature index, %S in I/S — expandable layers in illite smectite.

Hydrocarbon zones	$R_0$ %	$T_{max}$ °C	TTI	%S in I/S	Ordering	DEHYDRATION
IMMATURE	0.4	428	1	75 ± 10	R=0	
EARLY GAS	0.5	431	5	65 ± 15	Ran-	DEHYDRATION
CONDENSATE	0.6	435	13	40 ± 15	dom	
	0.7	440	22	33 ± 10		
WET GAS	0.8	445	38	29 ± 8		
	0.9	450	58	26 ± 8	R 1	
CONDENSATE	1.0	455	80	23 ± 8	(IS)	
	1.1	461	115	21 ± 7		
GAS	1.2	465	145	19 ± 7		
	1.3	472	200	17 ± 6		
CONDENSATE	1.4	480	250	15 ± 5		
	1.5	485	300	15 ± 5	R=3	
CONDENSATE	1.6	493	360	14 ± 5	ISII	
	1.7	(500)	440	12 ± 5		
CONDENSATE	1.8	(510)	550	12 ± 5		
	1.9	(525)	750	11 ± 5		
CONDENSATE	2.0	(540)	900	11 ± 5		
	2.2	-	1400	10 ± 5		
DRY GAS	2.4	-	2000	8 ± 3		
	2.6	-	3000	8 ± 3		

#### Main phase of hydrocarbon generation

Numerous studies have gathered evidence of organic origin of oil and gas in sedimentary basins. Several generalized schemes were proposed to characterize the main phase of petroleum formation (oil window) and that of gas formation as stages of postdepositional evolution of sediments controlled by favourable thermal conditions (Philippi, 1965; Vassoevich et al., 1969;

Teichmüller, 1971; Pussey, 1973; Tissot et al., 1974; Vassoevich et al., 1974; Durand, 1980). Powell—Snowdon (1983) have shown great diversity in the onset, maximum, and end of generation of liquid and gaseous hydrocarbons according to the type of kerogen, if their maturation is compared using an universal scale (e.g. vitrinite reflectance).

Published data on critical values of catagenetic indicators serve us a guideline to characterization of boundaries between subsequent immature, mature = generation, cracking, and dry gas stages (zones). We, however, do not follow such numbers strictly and prefer to use boundaries based on observations in the East Slovakian basin.

The scheme in Fig. 10. is based on the trend of decrease of hydrogen index (HI) which illustrates the depletion of residual source potential of kerogen from the immature through mature to overmature stages. Hydrocarbon characteristics of Rock-Eval pyrolysis are plotted as a function of thermal exposure of rock (TTI).

In the immature stage the source potential (amount of fixed hydrocarbons) is maximal and represents a starting value 50—150 mg HC/g  $C_{org}$  typical

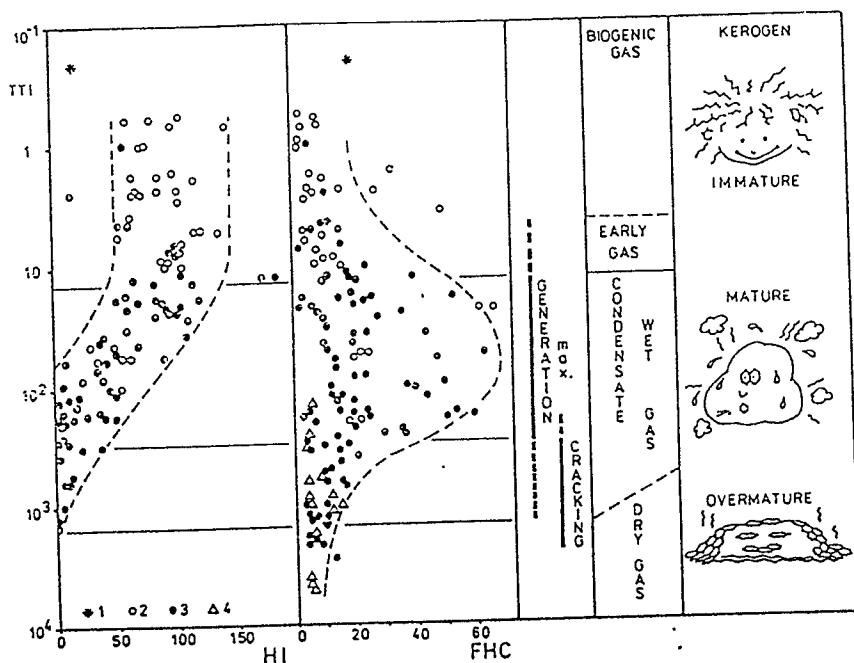


Fig. 10. Scheme of essential stages of generation and cracking of hydrocarbons during kerogen maturation as a function of thermal exposure expressed by TTI. In mature stage the source potential of kerogen is depleted as deduced from hydrogen index HI (mg HC/g  $C_{org}$ ) decrease, while the content of generated free hydrocarbons FHC (mg HC/g  $C_{org}$ ) reaches maximal values. Subsequent decrease of FHC is believed to be result of cracking of "wet" hydrocarbons to dry gas (methane) and of migration.

of gas-prone kerogen. Free hydrocarbons are mostly inherited. This stage occurs at shallow depth and in its upper part formation of microbial methane may be expected (Tissot—Welte, 1978).

Economical amount of hydrocarbons are generated during catagenesis by thermal cracking of maturing kerogen (Fig. 10.). Hydrocarbon generation is the most intensive where the residual potential decreases most sharply, and where the maximum of free hydrocarbon content (FHC) is reached. This conversion of fixed hydrocarbons to free ones may be also expressed by transformation ratio (Espitalié et al., 1985) which equals to relative decrease of residual potential (IH) compared with the starting value. According to analogy with other basins (Tissot—Welte, 1978; Powell—Snowdon, 1983), we may expect generation of early gas at the beginning of the mature zone in the East Slovakian basin (TTI = 6—15,  $R_0$  = 0.50—0.65,  $T_{max}$  = 430—435 °C). In the main phase of hydrocarbon generation we cannot expect formation of economic amount of oil from humic kerogen, therefore, we do not use the term "oil generation zone". Gas with small amount of light liquid hydrocarbons (condensate — wet gas) may be expected to be generated in the mature zone demarked by catagenetic indices TTI = 15—300,  $R_0$  = 0.65—1.50 ‰,  $T_{max}$  = 435—485 °C.

After the peak generation is reached, thermal cracking of already formed liquid hydrocarbon fraction (condensate) to gaseous hydrocarbons becomes more important process than the fading generation of hydrocarbons from kerogen (Tissot—Welte, 1978). In the East Slovakian basin cracking and migration of hydrocarbons from source rock is believed to be indicated by decrease of free hydrocarbon content (FHC) in fine grained rocks in a stage demarked by catagenetic indicators TTI = 300—1400,  $R_0$  = 1.5—2.2 ‰ (Fig. 10., Tab. 1.).

In the deepest and hottest parts of sedimentary basins dry gas (methane) occurs as a final product of cracking of liquid and gaseous hydrocarbons. All samples with TTI > 1400 and  $R_0$  > 2.2 ‰ have their residual source potential below detection limit (hydrogen index HI < 5 mg HC/g  $C_{org}$ , S2 < 0.01 kg HC/metric ton of rock) and a very low free hydrocarbon content (Fig. 10.).

#### Smectite illitization

Transformation of smectite (S) to illite (I) in mixed-layer minerals illite-smectite (I/S) results in gradual decrease of expandability of the starting material. This process is observed in many basins and is controlled mainly by burial, increasing temperature and time (Perry—Hower, 1972; Drits—Koporulin, 1973; Heling—Teichmüller, 1974). At present AIPEA recommends to use the term smectite for the whole group of expandable 2:1 minerals, while montmorillonite only for one mineral of this group (e.g. Čičel et al., 1981).

It is assumed that in pelites, in contrary to psammities, the catagenetic alterations are predominantly isochemical and that the pore water chemistry (influencing the smectite to illite transformation) is controlled more by source material and sedimentary facies than by deep fluid migration. Starting point of the transformation trend, characteristic of shallow depth (Fig. 13.) and low TTI (Fig. 11.), depends on degree of supergene illitization due to wetting

and drying (Šrodoň—Eberl, 1984). This is stronger when the source materials of smectite are micas or feldspars weathered and transported for a long distance and weaker in case of volcanic glass weathered at place. This is the reason why in basinal fine-grained sediments instead of pure smectite (montmorillonite) at least partially illitized mixed-layer I/S minerals occur.

Degree of illitization is expressed by relative content of expandable, smectite-type (S) layers (with 1.7 nm spacing after glycolation) and type of their interstratification with illite-type layers (I) in I/S structures (Reynolds—Hower, 1970). Our results show how expandability decrease with increasing thermal exposure of rock (TTI) (Fig. 11.) and depth (Figs. 12. and 13.) in the East Slovakian Neogene basin. Key points of transformation are boundaries between essential interstratification types:

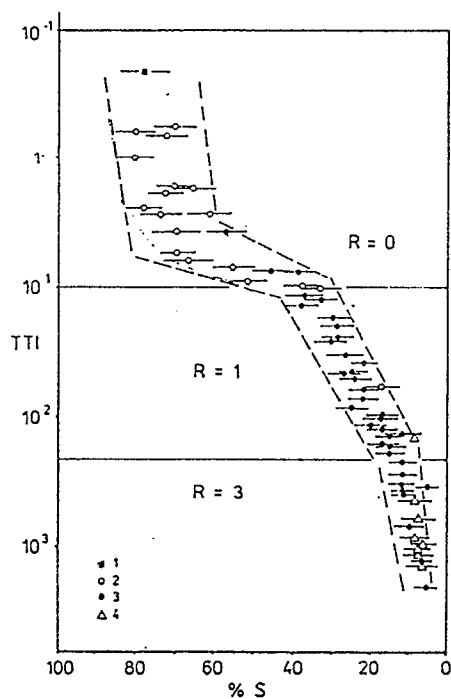


Fig. 11. Correlation of smectite to illite transformation expressed by content of expandable smectitic S layers in mixed-layer illite-smectite (I/S) and calculated TTI. Interstratification type: R=0 random; R=1 (IS) and R=3 (ISII) ordered. For stratigraphy see Figs. 8. and 9..

First type: Random interstratification of I and S layers, R = 0. It is typical of I/S minerals with 90—35 (40) % S (expandable layers). R means "reichweite" and represents type of interstratification (Reynolds, 1980). This early diagenetic type of I/S minerals with distinct diffraction peak of 1.7 nm (Fig. 12.) used to be described as montmorillonite (e.g. Heling—Teichmüller, 1974). This type changes to the second one in the East Slovakian basin at depth of about 2 km, TTI of 8—15,  $R_0$  of 0.6 %, pyrolytic  $T_{max}$  of 435—440 °C and subsurface temperature of 100—110 °C.

Second type: Ordered IS interstratification, R = 1, expandability from 35 to 15 % S: observed occurrence up to depth of 3 km, TTI 200,  $R_0$  1.3 %, pyrolytic  $T_{max}$  470 °C, and subsurface temperature 150—160 °C.

Third type: Ordered ISII interstratification, R = 3, expandability of less than 15 % S.

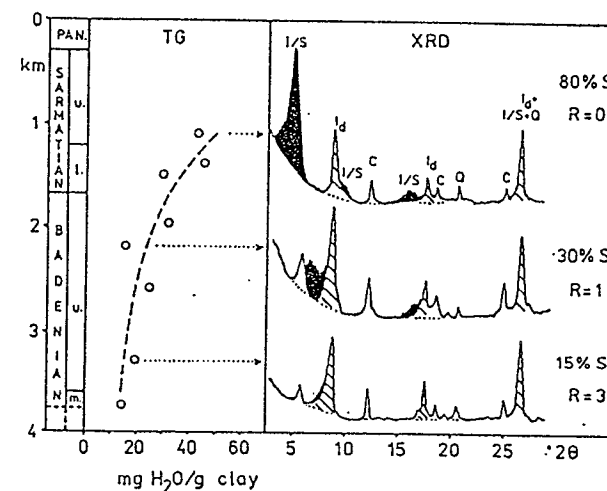


Fig. 12. Transformation of smectite to illite in borehole section Ložín 1. Content of water mostly bound in interlayers of illite/smectite is determined by thermogravimetry (TG, dehydration up to 150 °C), analyses provided by courtesy of I. Horváth, ÚACh SAV. Decreasing expandability (% S) of I/S minerals is determined from the character of diffraction patterns: I/S — illite/smectite (black), Id — discrete illite (hatched), C — chlorite, Q — quartz.

Diffraction patterns of these essential types of illite-smectite (oriented ethylene-glycol saturated samples) in the borehole Ložín 1 section are in Fig. 12.

During smectite illitization expandable interlayers collapse and highly hydrated cations (e.g. Ca, Mg) are expelled (Burst, 1969). Decrease in water content bound in interlayers during catagenesis is documented on a series of samples from borehole Ložín 1 using thermogravimetric analysis — weight loss by dehydration up to 150 °C (Fig. 11.). Many authors believed that water released by dehydration of clay minerals is flushing hydrocarbons out from the source rock (Weaver, 1960; Karpova, 1972). Recently, more attention is given to migration of inorganic gels (as by-products of smectite illitization) from shales to adjacent sandstones where they precipitate as newly formed clay minerals and secondary quartz rims cementing carrier rock pores (Hower et al., 1976; Vassoevich et al., 1975; Foscolos, 1984; Pollastro, 1985). These problems will be dealt with in our future studies. At present we use expandability as mineral indicator of catagenetic stage and thermal exposure (Fig. 10.) and compare smectite dehydration and hydrocarbon generation with overpressured zones.



*Zone of overpressured fluids*

Formation fluids are often overpressured in the East Slovakian basin. Rudinec (1978b, 1986) studied the trends of formation fluid pressure with depth and characterized four zones with different pressure gradient:

1st zone is at depth 1100—1600 m with pressure gradient 10.47—12.06 kPa m<sup>-1</sup>.

2nd zone - 1600—2000 m, gradient 13.9—16.5 kPa m<sup>-1</sup>.

3rd zone - 2000—3000 m, gradient 15.9—19.1 kPa m<sup>-1</sup>.

4th zone - 3000—4000 m, gradient 14.9—16.8 kPa m<sup>-1</sup>.

The same author showed the distribution of formation pressure gradients in a map of the East Slovakian basin and found that overpressure occurs in the SE and central parts of the basin with maximal subsidence during the Badenian, Sarmatian and Pannonian. He considered deformation and compression of undercompacted sediments during fast subsidence along synsedimentary faults to play decisive role in overpressure formation.

Mechanism of fluid overpressure formation is complex and may be explained in general terms as follows (e.g. Hunt, 1979). If a sedimentary column comprises alternating pelites and psammities, the fluids released from pelites may be drained through coarser-grained rocks and fluid pressure is near to hydrostatic. Important precondition for overpressure is presence of thick shale sequence and closed system.

It is generally accepted that the fluid movement is pressure-driven: Different opinions are on the origin and material character of fluids which are overpressured. Some authors emphasize the gas generation in good source rocks while the influence of smectite dehydration they consider negligible. They give the following arguments:

— in some oil-producing source rocks there is almost no smectite and still the migration of oil in direction of pressure decrease occurs (Tissot — Welte, 1978);

— overpressure occurs not in all shales but close to rich source rocks in active stage of hydrocarbon generation (Law, 1984; Spencer, 1987);

— from overpressured horizons in some regions (e.g. Rocky Mountains) only drilling mud and gas without formation water are recovered (Spencer, 1987);

— migration of hydrocarbons from the source rock cannot be driven by smectite dehydration as the latter takes place at an earlier stage of burial than the maximum of oil generation (Perry — Hower, 1972; Waples, 1980).

On the other hand, some of the following arguments used to be and some still are given in favour of the active role of clay dehydration in overpressure formation and primary hydrocarbon migration:

— quantitative proportion of shales in total sedimentary filling of many basins is substantial;

— productive oil-bearing horizons are situated in some regions at depth by less than 500 m above the zone of smectite dehydration (Mexican Gulf coast region, Burst, 1969);

— overpressured fluids occur in sequences rich in pelites at similar depth where smectite-to-illite transformation takes place in many basins (Vískovskiv, 1974; Bruce, 1984; Polster et al., 1984);

— smectite dehydration results in mineral volume decrease what may cause opening of migration paths (Powers, 1967). Water is released from smectite interlayers and surface area of clay minerals decrease. If hydrocarbons were formed in shale and adsorbed on surface of clay minerals, smectite dehydration should cause hydrocarbon desorption and their movement with other fluids from pelites to psammities (Weaver, 1960; Karpova, 1972; Klubova, 1973; Vassoevich et al., 1975; Dypvik, 1983);

— even though the first dehydration of illite-smectite anticipates the onset of hydrocarbon generation, the second phase of dehydration (during transformation of ordered mixed-layer illite-smectite to highly illitic material) takes place during the main phase of oil generation (Perry — Hower, 1972; Powell et al., 1978; Dypvik, 1983);

— in low permeability shale-rich complexes, even after smectite dehydration, the released water may be retained closed in the source rock and escape only in later stage at higher temperature when overpressure opens microfractures — pathway for water and hydrocarbon migration (Bruce, 1984).

These ideas are sometimes criticized, their rationale, however, is worth of further research. To answer the question of possible material source of overpressured fluids, we compare the depth zones of hydrocarbon generation, smectite dehydration, and formation fluid overpressure in the East Slovakian basin (Fig. 13.). Pressure data are from Rudinec (1978b), temperature from Král et al. (1985).

The onset of formation pressure higher than hydrostatic is in the central and SE parts of the East Slovakian basin at depth of 1.6 km, subsurface temperature of 90 °C, vitrinite reflectance  $R_o = 0.45\%$ , pyrolytic temperature index  $T_{max} \leq 430$  °C, TTI of about 3, and expandability of illite-smectite 60—70 % S. Strong increase of fluid pressure is typical of following 400 m (until 2 km) where the most intensive dehydration may be expected, as the expandability of I/S decreases to 40—50 % S (Fig. 13.). Illitization jump at depth (o 1.6—1.8 km in this basin was already observed by Kraus — Šamajová (1978). Hydrocarbon generation is, however, at this depth only incipient and mild.

Maximal gradient of “over-hydrostatic” fluid pressure is in the zone of maximal hydrocarbon generation and concomitant second stage of dehydration (from 40—50 to 10—15 % S in partly random but mostly ordered I/S). In the cracking zone the overpressure is quite high but the difference over hydrostatic pressure gradient remains almost constant.

Decline of overpressure (4th zone of Rudinec, 1978b) coincides with the end of quantitatively important hydrocarbon generation and smectite dehydration in dry gas zone. Organic matter enters the anthracitic stage of coalification ( $R_o > 2.2$ — $2.4\%$ ).

Slight anticipation of hydrocarbon generation by smectite-to-illite transformation may be seen in Fig. 13. Zone of dehydration lies at depth interval from 1.8 to 3.0 km marked by subsurface temperature 100—155 °C, vitrinite reflectance  $R_o = 0.5$ — $1.4\%$ , pyrolytic index  $T_{max} = 430$ — $480$  °C, and TTI = 5—250 (Fig. 10., Tab. 1.). On the contrary, the main zone of hydrocarbon formation lies at depth of 2—3.2 (3.5) km, subsurface temperature 110—165 (175) °C, and is marked by  $R_o = 0.65$ — $1.5\%$ ,  $T_{max} = 430$  (435)— $485$  (540) °C and TTI = 15—300 (1400). Such anticipation of organic maturation by smectite dehydration

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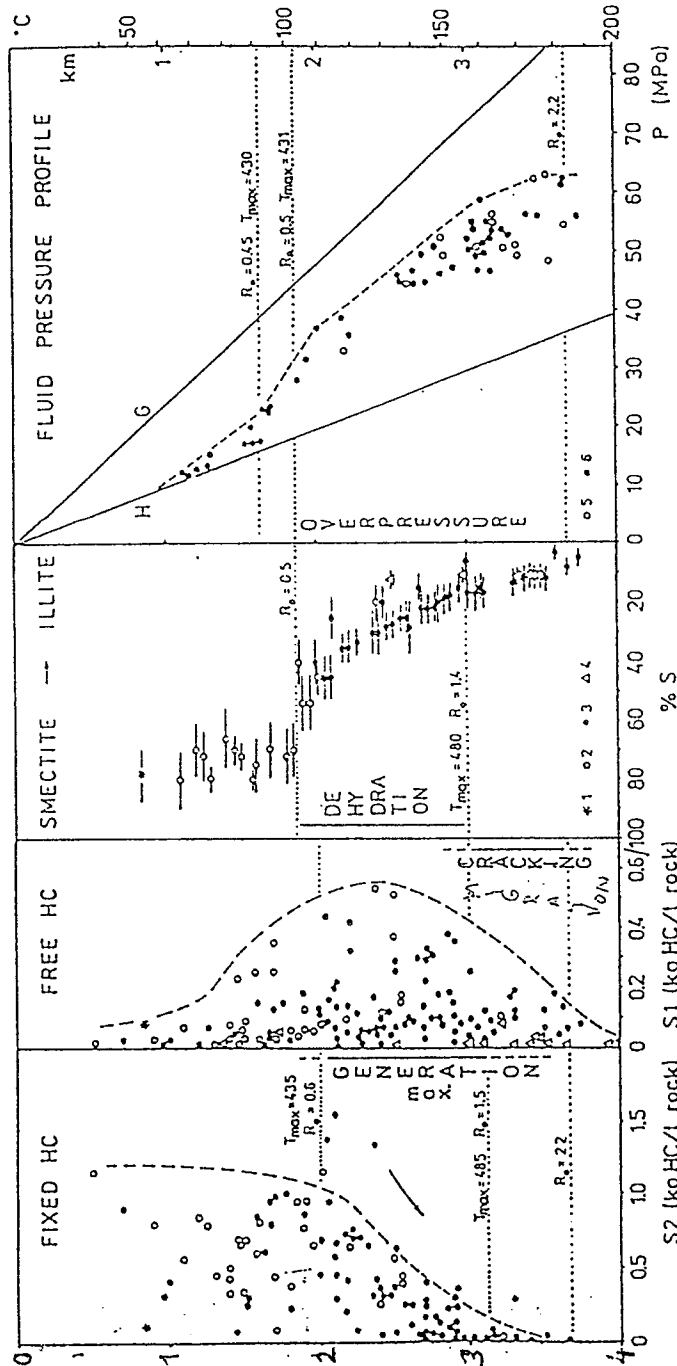


Fig. 13. Comparison of depth zones of fixed hydrocarbon potential depletion (S2), "sweating out" free hydrocarbons (S1) and their cracking in source-rock, dehydration of expandable clay minerals and of occurrence of formation fluid overpressure in the East Slovakian basin (latter data from Rudinec, 1978b).  
 Legend: 1 — Pannonian; 2 — Sarmatian; 3 — Badenian; 4 — Karpatian; 5 — gas field Lastomir; 6 — Siretava; H — hydrostatic pressure gradient; G — geostatic pressure gradient.

was described also by Bruce (1984) and Burtner — Warner (1986). It is, however, important that the major parts of these two processes occur parallelly at the same stage of postdepositional evolution.

From data obtained from the East Slovakian basin we may conclude that in the upper part of the overpressured zone the formation fluid pressure increase may be linked more with the smectite dehydration, while in the lower part more with hydrocarbon generation. These two processes may produce fluids, but thermal expansion of "free" fluids should be understood as the driving mechanism of overpressure build-up.

In the dehydration zone we may expect consequent sandstone pore cementation by authigenic minerals derived from by-products of smectite illitization in adjacent shales. The lowered permeability of carrier rocks may play even more important role in overpressure rise than the mere water expulsion from clay minerals.

*Model of catagenetic zonality and its evolution*

In each borehole section marked boundaries of subsequent zones represent main stages of diagenetic and catagenetic alteration of sedimentary rocks. The burial and thermal history is a graphical reconstruction of catagenetic evolution of the present section showing when and at what depth the strata passed through the conditions favourable to hydrocarbon generation.

In the middle of Fig. 14 the section of the borehole Siretava 21 is shown. On the left side there is a diagram of accumulation of sediments in geological past, the vertical thickness of sedimentary layers being corrected for compaction. Broken lines represent simplified history of layers burial at boundaries between stratigraphic units. The steeper the slope of the line, the faster the sedimentary accumulation during the respective period. The rationale of the method of Lopatin (1971) is shown on an example of boundary layer between the Sarmatian and Badenian (Fig. 14). The layer is passing through subsequent 10 °C temperature intervals. In each one it is exposed to a specific temperature (arithmetic mean of the interval is used) for a certain effective time  $\Delta t$  (in Ma = million years) from which the thermal exposure is calculated. Total thermal exposure (time-temperature index TTI) is a sum of partial exposures throughout the whole geological past (Lopatin, 1971; Waples, 1980).

TTI is a theoretical value proportional to the degree of catagenetic alteration of organic matter. Imperfections of this index (simplification of kinetic equations and taking some variables as constants) are reviewed e.g. by Waples (1984), Yüklér and Kokes (1984), Robert (1988). They may be reduced, however, by a good calibration of TTI with a real measured parameter, e.g. vitrinite reflectance, as shown in Figs. 8 or 9 and 11.

Theoretical vitrinite reflectance RTI is a numerical result of modeling and is shown in Fig. 14 (right) as a function of depth (dashed curve). Modeled RTI is quite consistent with the measured vitrinite reflectance  $R_v$ , the differences are mostly within a standard deviation. The model reconstruction (Fig. 14, left side) is therefore accepted for deductions on the time and depth of maturation.

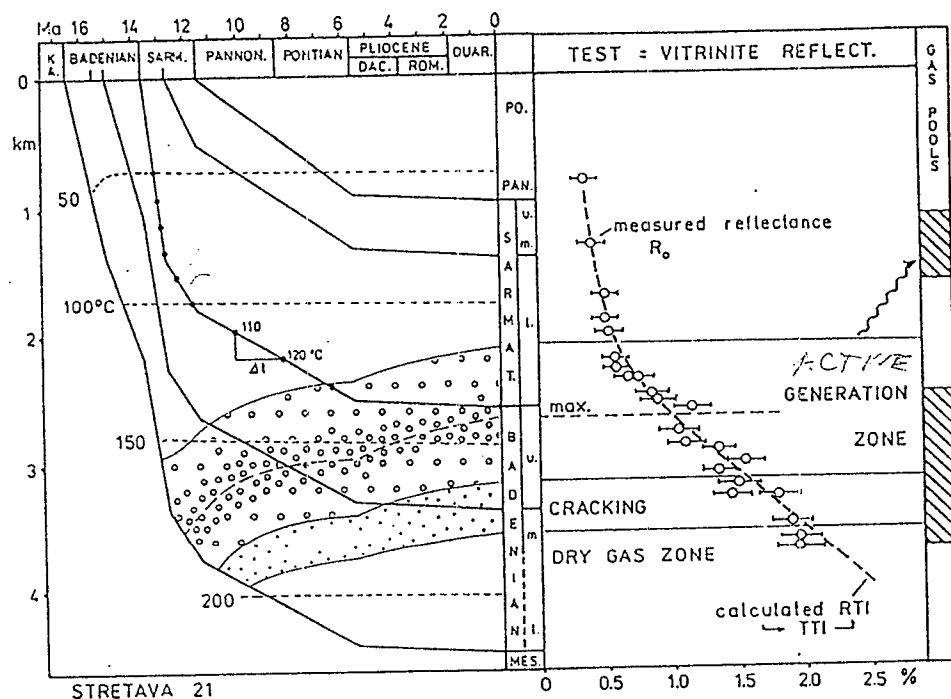


Fig. 14. Burial history of sedimentary section Stretava 21 (borehole bottom 3738 m). Each layer is exposed to subsequent 10°C temperature intervals for characteristic residence time  $\Delta t$  (Ma). From partial increments the total thermal exposure (time-temperature index TTI) is calculated and transferred to theoretical vitrinite reflectance RTI (using Fig. 8). Modeled catagenetic zonation (right) is tested using measured vitrinite reflectance  $R_o$ .

From geochemical characteristics we conclude that the Badenian and Sarmatian shales have relatively good source potential. From reconstruction in Fig. 14. (left) it is evident that these units entered the favourable conditions to hydrocarbon generation in the Sarmatian. Each layer passed through the generation zone ( $R_o = 0.6-1.5\%$ ) within 7-9 million years and then entered cracking zone. The Lower and Middle Badenian reached to take place at depth of 2 to 3.2 km where also fluid overpressure exists. Gas pools are situated at similar depth interval and also 400-900 meters above (1.1-1.6 km). We assume that the upper reservoirs were connected with the generation zone by migration paths.

In a similar manner to Fig. 14. all other borehole sections shown in Fig. 1. are studied. Some of them differ from the general trend of maturation with depth and show discrepancy between model and analytical measurements of maturity indicators. These anomalous cases are connected with oxidizing facies, volcanism, erosion, and salt-bearing strata.

### Anomalous catagenetic phenomena

Upper Karpatian sediments are mostly developed in oxidizing facies. Organic matter is fusinitic with nil hydrocarbon potential and yields no usable pyrolytic data. Vitrinite reflectance is difficult or impossible to obtain because of lack of vitrinite. Expandable minerals are difficult to study, unless special sample treatment is used, because of high iron-oxide content. It is therefore uneasy to characterize the catagenetic stage of this type of sediments using geochemical or optical analyses. In section of boreholes Trhovište (TH-26), Michalovce (MI-1, Fig. 1.) instead of continuous change of catagenetic indicators and even of porosity abrupt jump is observed in the depth relation at the transition from the Badenian to Upper Karpatian. Catagenetic alteration cannot be studied in oxidizing facies shales in the same way as in black or dark grey shales because of their different nature.

In borehole Zatin (ZA-1) at depth of 2.1 km, kerogen is altered to anthracitic rank at a contact with an andesite lava body. At greater depth the rank (maturity) is lower and corresponds to "normal" burial catagenesis. Our results confirm the earlier ones of Ďurica et al. (1980). The contact alteration of volcanites does not reach more than few tenths of meter, though.

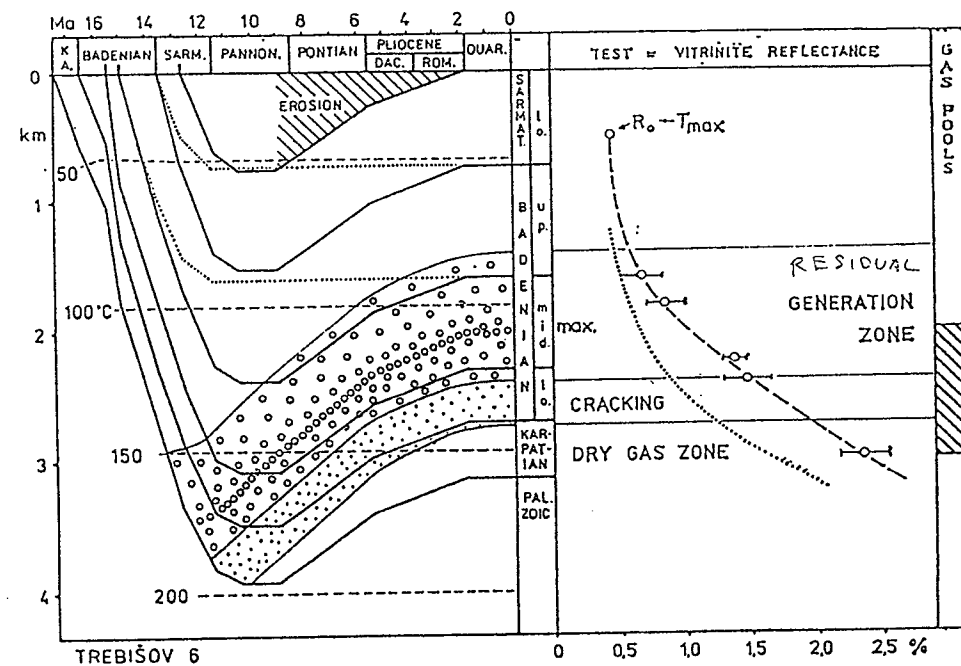


Fig. 15. Burial/erosional history of borehole section Trebišov 6. Reconstruction of history without erosion (depth of layer — dotted line, left) gives calculated catagenetic stage (dotted curve, right) much lower than the measured one by  $R_o$ . Modeling some additional accumulation in the Sarmatian and subsequent erosion brings agreement in modeled and measured maturity.

In borehole Trebišov 6 maturation indices and porosity show depth trend parallel to the majority of other boreholes but shifted to shallower depth. This indicates a possible erosion of overlying strata missing in the present section. To estimate the thickness of erosion, modeling was applied (Fig. 15.). In the first solution the diagram of burial history without erosion was constructed (dotted lines for the Sarmatian and Upper Badenian base, Fig. 15. left). The calculated catagenetic trend with depth is shown as a dotted curve (Fig. 15. right) of RTI. It is distinctly far from the measured vitrinite reflectance  $R_o$ . The first solution is, therefore, unacceptable. If higher geothermal gradient was effective in past, the trends of calculated and measured data would not be parallel. Erosion seems to be more probable. The Paleozoic of Zemplincium which forms the basement in this part of the basin is supposed to be, according to paleogeographical studies of Rudinec (1978a), emerged from sea since the Pannonian (Fig. 1., 10 km to S from TB-6). Till the Pannonian a sedimentary column should have been accumulated and after the Pannonian eroded. We do not assume a vehement uplift of Zemplincium and fast erosion. With this input information we increase the thickness of accumulation (Sarmatian) and erosion in the model until calculated RTI (dashed curve) does not fit the measured  $R_o$ . The reconstruction shown in Fig. 15. with erosion of about 750 m is acceptable under the given rules of modeling.

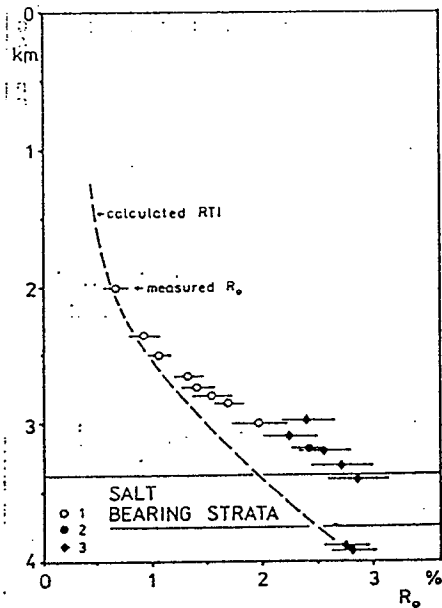


Fig. 16. Anomalous trend of vitrinite reflectance  $R_o$  above salt-bearing strata in the Albinov 4 and 7 section. Measured  $R_o$  shows higher gradient than the predicted one by the model based on recent temperature gradient. Albinov 4: 1 — Badenian, 2 — Karpatian; Albinov 7: 3 — Karpatian.

One important conclusion may be drawn from the model in Fig. 15. The hydrocarbon generation in course of maturation took place only until the end of the Pannonian. After erosion the strata were exposed to lower temperatures and the catagenetic alteration should have ceased. The present state

is "frozen" result of processes which are not already active now. Prospects of economic hydrocarbon accumulation occurrence in such a structure are from this point of view lower than in those without erosion (e.g. Stretava, Fig. 14.), as the earlier generated gas could diffuse for 8 mill. years being not replenished as the maturation faded away.

A particular trend of catagenesis is observed in boreholes Albinov 4 and 7, which are about 2—4 km far from the volcanites of the Slánske vrchy mountains (Figs. 1 and 18). The maturation trend ( $R_o$ ) is steeper above the salt-bearing strata of the Upper Karpatian (Fig. 16.). Below the salt the measured vitrinite reflectance  $R_o$  is equivalent to RTI calculated by model using unchanged temperatures throughout the geological history. On the contrary to eroded section of Trebišov 6 (Fig. 15.), in case of Albinov the curves of  $R_o$  and RTI are not parallel but from the depth of 2.5 km they diverge with different slope (Fig. 16.). The reason may be in temperature redistribution event due to good thermal conductivity of salt-bearing strata, their thickness (400 m) and direct connection with neighbouring volcanites. During the Sarmatian volcanic activity the salt may have acted as lateral thermal conductor and caused elevated temperatures in the overlying strata, the underlying strata being not strongly affected. Sealing properties of salt may have also retarded the maturation products diffusion and thus the maturation itself.

#### Catagenetic zonation in basin scale

In Figs. 17. and 18. the boundaries of subsequent catagenetic zones in basin profiles are shown (situation is in Fig. 1.). In the shallow zone to depth of 1.7 km kerogen is immature and we do not expect an important hydrocarbon formation. Clay minerals of illite-smectite type are highly expandable.

In the zone between 1.7 (2) and 3.2 (3.5) km the rocks reached the main

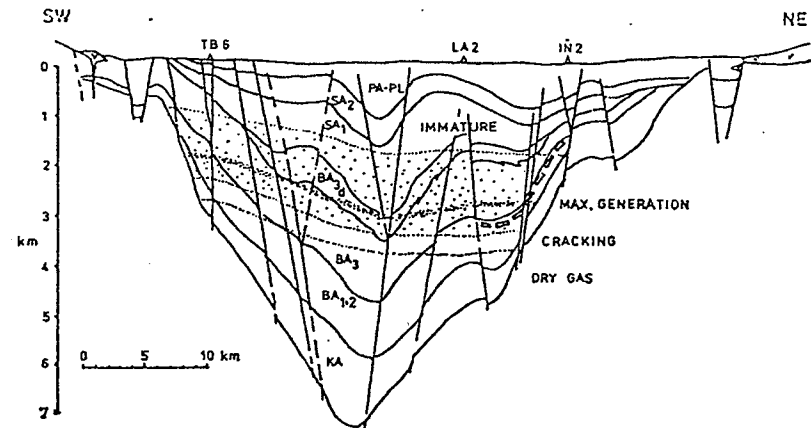


Fig. 17. Zones of hydrocarbon generation, cracking and stability of dry gas in NE — SW section of the East Slovakian basin (Fig. 1 — line 1). Depth related to sea level. Stratigraphy: KA — Karpatian, BA — Badenian (3d — Upper detrital), SA — Sarmatian, PA — Pannonian, PL — Pliocene.

stage of hydrocarbon generation from mature kerogen. Its maximum ( $R_{11} = 1.0\%$ ) is at depth of about 2.3–2.7 km. Wet gas is expected to be generated as the kerogen in the Neogene strata is of humic type. In the cracking zone the conversion of liquid hydrocarbons (condensate) to methane is supposed to occur together with decreasing gas generation from kerogen.

The transition from cracking to the dry gas zone is assumed to be at 2.2% vitrinite reflectance at depth of 3.5–3.8 km. At greater depth methane is stable. At present we are not able to predict the maximum depth where methane may "survive". It is, however, important to realize that methane is exploited from dry gas zone in several basins in the world (Tissot — Welte, 1978). Application of advanced drilling and exploiting technology may reveal new gas reserves at depth greater than 3 km.

The depth boundaries of catagenetic zones are not equal in the whole basin. In the central part the mature zone is relatively deeper than at the margins (Figs. 17. and 18.), in the southern and NW parts it lies at smaller depth (Třebišov 6, Albínov 4, 7).

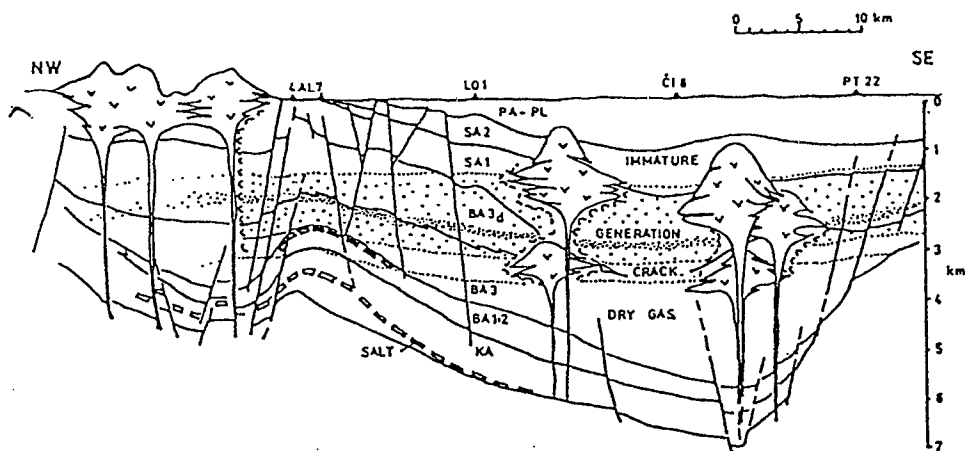


Fig. 18. Catagenetic zonation in the NW — SE section of the East Slovakian basin (Fig. 1 — line 2): v — volcanic bodies, rectangles — salt-bearing strata in NW part in the Upper Karpatian and Middle Badenian, for other details see Fig. 17.

Paleogeographic maps of the Sarmatian and Upper Badenian after Rudinec (1978a) are in Fig. 19. showing where these units entered the generation and cracking zones. Intensive wet gas generation and concomitant second stage of smectite dehydration takes place in the central and SE parts of the East Slovakian basin. These are characteristic by the highest sedimentary accumulation rate in the Badenian and Sarmatian and by the highest geothermal gradient. It is this region where formation fluid overpressure is maximal (Rudinec, 1978b; 1986) and in its northern and eastern parts the main gas and gas-condensate fields are situated. The southern-central part of the basin may be partially depreciated by buried volcanites (Čičarovce, Malčice, Zátin) which could thermally influence the lateral migration in the NW, N, NE and E directions.

Sedimentary rocks of the Upper Karpatian contain unfavourable type of kerogen with almost no source potential and even under favourable catagenetic conditions they could not generate hydrocarbons. The mature zone in Figs. 17. and 19., therefore, does not concern the Upper Karpatian. According to geochemical analyses and modeling, the relatively optimal prospects may have the mature sequences of the Lower Sarmatian and Upper Badenian (Fig. 19.) or structures connected with these by migration paths, even if they would be some of the basement units in the marginal areas.

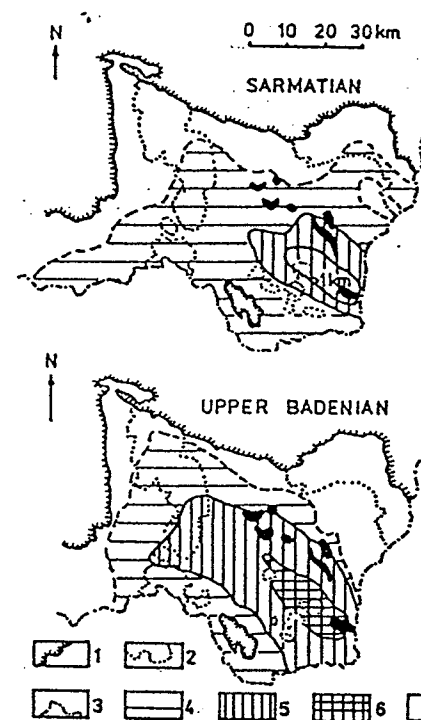


Fig. 19. Present extent of mature source rocks ( $R_o > 0.6\%$ ) in paleogeographic maps (Rudinec, 1978a) of the Sarmatian and Upper Badenian in the East Slovakian basin.

Legend: 1 — Neogene basin margins, 2 — volcanic mountain contours, 3 — state borderlines of Czechoslovakia, 4 — immature stage, 5 — mature stage where hydrocarbon generation has started, 6 — stage of hydrocarbon cracking ( $R_o > 1.5\%$ ), 7 — gas fields (in NW mostly in the Upper Badenian and in SE situated mostly in the Lower Sarmatian reservoirs, Rudinec, 1976b).

The source potential of the Lower Karpatian and Mesozoic basement in the NW part of the basin is not definitely clear. The Eggenburgian and Egerian, as well as Paleogene in the Prešov 1 borehole (Fig. 1.) reached only early mature stage and have very low catagenetic gradient. This fact gives evidence that in the NW part of the East Slovakian basin some of the pre-Badenian units may be in the mature stage favourable to hydrocarbon generation.

#### Conclusions

Organic geochemical data give evidence that in the East Slovakian basin catagenetic alteration of sedimentary rocks increase with depth showing a high gradient. Maximal degree of catagenesis is reached in the deepest central

and SE parts of the basin and seems to be closely spatially related to the overpressured formations and gas fields.

Several catagenetic indicators ( $R_o$ ,  $T_{max}$ , % S, and TTI) give a fairly good mutual correlation and are used as a scale for stages of postdepositional alterations. The hydrocarbon generation zone is characterized on the basis of Rock-Eval pyrolysis as an interval where the source potential of kerogen (expressed by fixed hydrocarbon content) systematically decreases and is converted to free hydrocarbons. This zone is in the central and SE parts of the basin at depth ranging from 1.7–2 to 3.2–3.5 km and is relatively thin if compared with other young basins, e.g. the Vienna basin (Franců — Müller, 1989). Burial and thermal history reconstruction reveals that the sedimentary rocks passed through the mature stage (covering the interval of vitrinite reflectance from 0.6 to 1.5 %) within 5–10 mil. years. The maximal genetic potential of the Lower Sarmatian and Badenian shales (S1 + S2 of the Rock-Eval 1 pyrolysis) is ranging from 0.5 to 1.2 kg HC/m<sup>3</sup> metr. ton of rock or 1.2–2.9 10<sup>6</sup> metr. ton HC/km<sup>3</sup>. More detailed quantitative assessment of hydrocarbons generated in different parts of the East Slovakian basin throughout subsequent stages of geological history will be dealt with in the forthcoming paper.

The zone of smectite dehydration lies slightly above but, for the major part at the same depth as hydrocarbon (wet gas) generation. Both processes coincide spatially with fluid overpressure. The water released from expandable clay minerals may not be the decisive agent in hydrocarbon generation but its thermal expansion may contribute to the fluid movement from pelites to psammites what should result in carrier rock cementation by newly formed minerals.

In areas where erosion seems to have taken place (southern margin) the model reveals that the present stage of catagenesis is residual and hydrocarbon generation has faded since the uplift and erosion in the Pannonian.

At present it is difficult to say if methane may "survive" depth of 6 or 7 km or if it is displaced by CO<sub>2</sub>, H<sub>2</sub>S and N<sub>2</sub>. Methane is, however, stable at greater depths than the present exploited formations in the East Slovakian basin.

Further study should be focused at possible migration paths from the central part towards marginal structures in the Neogene filling or in the basement. The regional extension, maturity and source potential of the lower ("grey") Carpathian should be studied in the future as there are some hints that this unit may have favourable source properties and hydrocarbon generation conditions in the NW part of the basin between the Albinov and Prešov boreholes.

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