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ORGANIC MATTER AND CLAY MINERALS AS INDICATORS OF THERMAL HISTORY IN THE TRANSCARPATHIAN DEPRESSION (EAST SLOVAKIAN NEOGENE BASIN) AND THE VIENNA BASIN

(7 Figs.)

Abstract: Analytical characteristics of organic matter and clay minerals reveal striking differences in the rate of diagenesis and catagenesis of the Neogene rocks in two basins with high and low geothermal gradients. In the “hot” Transcarpathian depression the zone of main liquid hydrocarbon generation and smectite dehydration is relatively thin and shallow at depth from 1.7–2 km to 3–3.5 km. In the “cold” Vienna basin such a zone occurs below 3.5 km and its floor probably is not reached even in the deepest Neogene rocks at depth of 5.5 km. The organic and clay characteristics which mark the diagenetic and catagenetic stages show good relationship with subsurface temperature.

Резюме: По анализам органического вещества и глинистых минералов является, что скорость диагенетических и катагенетических процессов в неогеновых осадках резко отличается в зависимости от глубины в Закарпатской впадине (Восточно-Словацком бассейне) и Венском бассейне. В Закарпатской впадине с высоким геотермическим градиентом главная зона образования жидких углеводородов и зона дегидратации смектита находятся в относительно неглубоком и узком интервале с 1,7–2 до 3–3,5 км. В „холодном“ Венском бассейне происходят эти процессы более медленно с началом на глубине более 3,5 км.

Key words: organic matter, clay minerals, thermal history, Transcarpathian depression, Vienna basin.

Introduction

Organic matter and clay minerals undergo well measurable physical and chemical changes during postdepositional evolution of sedimentary rocks. Some of their characteristics are used as indicators of the degree of diagenetic and catagenetic alteration and of the thermal history of the sedimentary strata.

Vitrinite reflectance, Rock-Eval pyrolytic data and illite-smectite expandability are used in this study of catagenetic zonation. Earlier geochemical data and thermal models of hydrocarbon generation in the Transcarpathian depression (East Slovakian basin) are included (Franců, 1986, 1987; Franců – Šimánek, 1987; Franců – Milička, 1988; Franců et al., 1989) together with those concerning the Vienna basin (Müller, 1987; Müller – Chmelík, 1987). This paper presents a comparison of the different catagenetic rate in the course of burial and thermal history and to show the different prospects of hydrocarbon generation in these two basins with very different geothermal conditions.

Geological setting

The Transcarpathian depression (often called the East Slovakian Neogene basin on the Czechoslovak territory) and the Vienna basin are the intramountain depressions of the

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Alpine-Carpathian orogenic belt (Fig. 1). The opening of the Neogene sedimentation area was influenced by:

- last stages of North-European Platform subduction under the Carpathian–Pannonian block which became a continent-continent type of collision (Royden et al., 1982; Royden, 1988; Vass et al., 1988);
- oblique convergence of lithospheric plates resulting in final shape of the Carpathian arc (Jiříček, 1979);
- formation and evolution of the asthenolith elevation in the Pannonian region (Stegena et al., 1975).

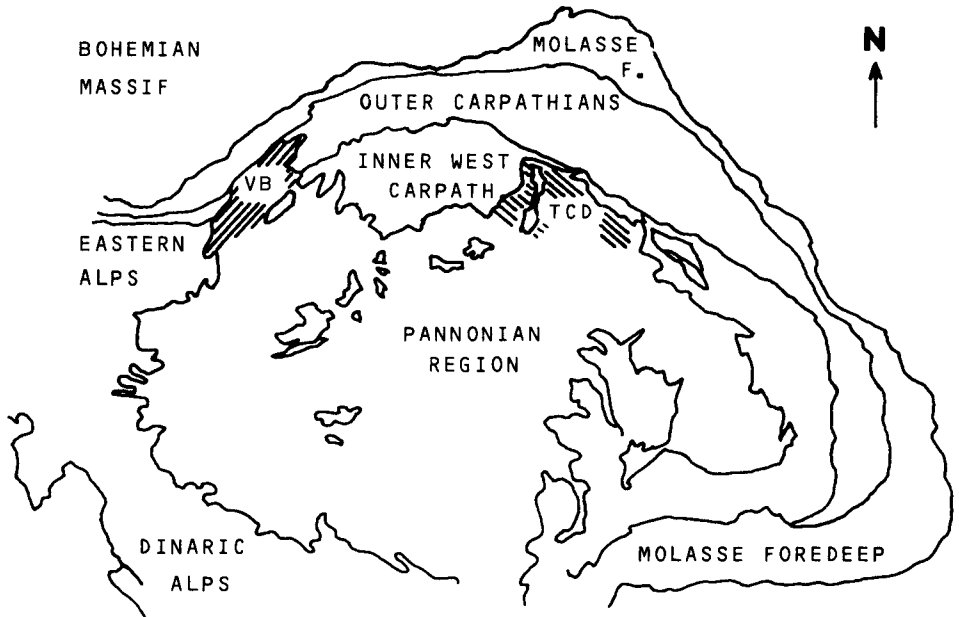


Fig. 1 Tectonic map of the Carpathian–Pannonian region (simplified after Maheř, 1974) showing the position of the Transcarpathian depression (TCD) (which is often called the East Slovakian Neogene basin on the Czechoslovak territory) and of the Vienna basin (VB).

The basement of the Transcarpathian depression was formed by consolidated paleo-Alpine structures of the Inner West Carpathians, while the basement of the Vienna basin included also the Alpine nappes of the Outer West Carpathians and the Eastern Alps.

The Neogene sedimentation in both basins is marine in the Lower Miocene and gradually changes in the Badenian upward to fresh-water facies (Jiříček – Tomek, 1981; Rudinec et al., 1981; Rudinec, 1989). The Sarmatian is brackish, the Pannonian and Pliocene are fresh-water limnic. The main differences between the two basins are as follows:

- extensive syngenetic acid to intermediate volcanism in the Transcarpathian depression which is absent in the Vienna basin;
- sedimentation in the Ottnangian in the Transcarpathian depression is not proved (Rudinec, 1978 a);

– regionally extended evaporitic strata in the base of the Upper Karpatian and in the Middle Badenian in the Transcarpathian depression;

– maximal subsidence and sedimentary accumulation are in the Transcarpathian depression during the Upper Badenian and the Sarmatian (Rudinec et al., 1981), while in the Vienna basin they take place during the Karpatian and Badenian (Jiříček – Tomek, 1981) what gives evidence of subsequent basin opening in the West Carpathian arc from the west to the east (Vass – Čech, 1983; Čech, 1988) connected with thrusting in the adjacent areas of the Outer Carpathians (Jiříček, 1979; Royden, 1988);

– the Transcarpathian depression is under influence of the Pannonian asthenolith and has a high heat-flow $Q = 82-113 \text{ mW m}^{-2}$ and high geothermal gradient (at depth of 4 km the temperature ranges from 200 to 209 °C) (Fig. 2), the Vienna basin is cooler: $Q = 41-69 \text{ mW m}^{-2}$, temperature at 4 km varies from 115 to 120 °C (Rudinec, 1978 b; Čermák, 1979; Král et al., 1985).

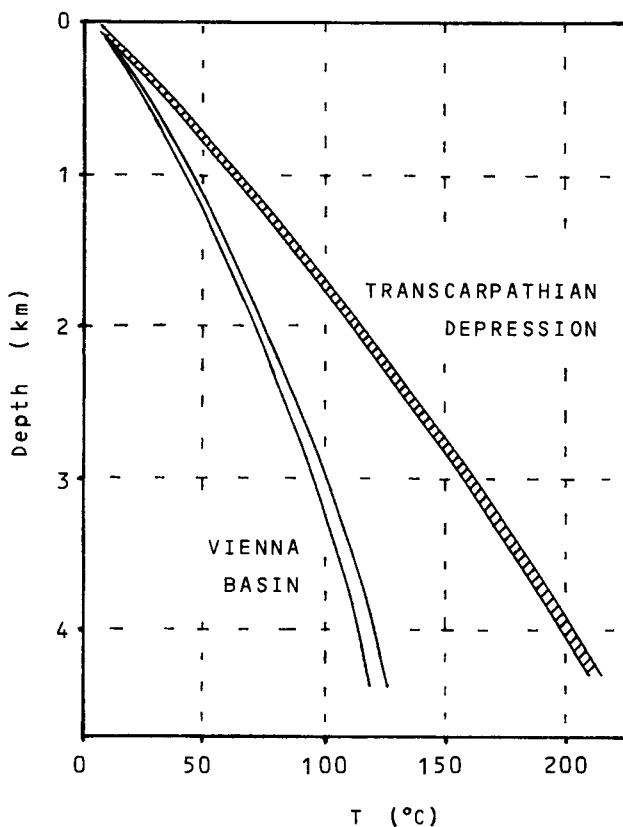


Fig. 2 Corrected borehole temperature in the studied basins (Král et al., 1985).

Methods

Shales from borehole cores were analysed. Vitrinite reflectance R_o was measured after Stach et al. (1975) (J. Franců and P. Müller analysts). From the Rock-Eval pyrolysis (Espitalié et al., 1977) and organic carbon content in rock C_{org} (determined by method of

Šmeral – Urbánek, 1986) the following characteristics were obtained: S1 – free hydrocarbons (mg HC/g rock), S2 – fixed hydrocarbons or residual source potential (mg HC/g rock), HI – hydrogen index: $HI = 100 S2/C_{org}$ (mg HC/g C_{org}), T_{max} – temperature of the maximal pyrolytic signal (°C).

Illite-smectite expandability was determined by X-ray diffraction (B. Toman and M. Plšková analysts), interpretation was based on computer diffraction simulation (Reynolds–Hower, 1970; Reynolds, 1980; Šrodoň – Eberl, 1984).

Time-temperature index TTI was calculated using the method of Lopatin (1971) developed by Waples (1980). Burial history was reconstructed using correction for compaction, corrected temperatures from borehole measurements (Král et al., 1985) were used.

Trends of catagenesis with depth

In Figs. 3 and 4 the systematic change of organic maturation indices with depth is shown. Vitrinite reflectance which is a measure of aromatization of kerogen structure, is up to depth of 1.5 km similar in the both basins and represents the diagenetic stage where kerogen is immature. From this depth on the trends of further alteration start to be divergent. While in the Transcarpathian depression at depth of 3.5–4 km the anthracitic stage is reached ($R_o > 2.2$ – 2.4 %), in the Vienna basin the catagenetic stage corresponds to the bituminous coal ($R_o = 0.7$ – 0.9 %). It is surprising that the trend of catagenesis (R_o increase) is in the Vienna basin continuing further to the underlying Upper Triassic, what means that these Mesozoic rocks (Ötscher nappe, Závod area) were not significantly deeper buried during their pre-Neogene evolution.

Pyrolytic temperature T_{max} gives independent evidence of kerogen thermal maturity (Espitalié et al., 1977, 1985, 1986). The trends of T_{max} change with depth (Fig. 3) are analogical to those of vitrinite reflectance R_o and prove the fact that in the Vienna basin the Neogene rocks at depth of about 4 km and the underlying Triassic strata (boreholes Závod 72, Kuklov 3) are at a similar catagenetic stage as the shales at depth of 2.0–2.5 km ($R_o = 0.55$ – 0.90 %, $T_{max} = 435$ – 450 °C) in the Transcarpathian depression. Geostatic pressure is at depth of 4 km in the both basins almost equal (94 MPa), the temperature is, however, in the Transcarpathian depression about 200 °C, while about 115–120 °C in the Vienna basin. The influence of temperature on kerogen maturation is, therefore, much more important than that of geostatic pressure. This phenomenon has a more general validity but the age of the strata influence the maturity as well (Tissot – Welte, 1978).

The conversion of the kerogen source potential to free hydrocarbons is documented in Fig. 4. At shallow depth where kerogen is immature, the residual source potential is of highest value and represents the initial potential of kerogen. It is quite similar in the both basins and expressed by hydrogen index HI it ranges from 150 to 200 mg HC/g C_{org} what corresponds to atomic ratio H/C of 0.7–0.8. Such characteristics are typical of humic type of kerogen, i.e. type III (Espitalié et al., 1977, 1985). As in the Neogene in the both studied basins there is the gas-prone kerogen, we avoid using the terms as “oil window” or “oil generation zone” in our further interpretations.

In Fig. 4 the zone of hydrocarbon generation from mature kerogen is characterized by decrease of residual source potential (HI) and increase of free hydrocarbons ($S1/C_{org}$). This phenomenon is observed in the Transcarpathian depression at a depth interval from 1.7–2.0 to 3.0–3.5 km. The ceiling and the floor of this zone is marked by maturity indices $R_o = 0.6$ and 1.5 %, $T_{max} = 430$ and 475 °C respectively. Generation of gas with small portion of liquid gasolinic hydrocarbons may be expected in this zone. At depth of more than 3.5 km kerogen is

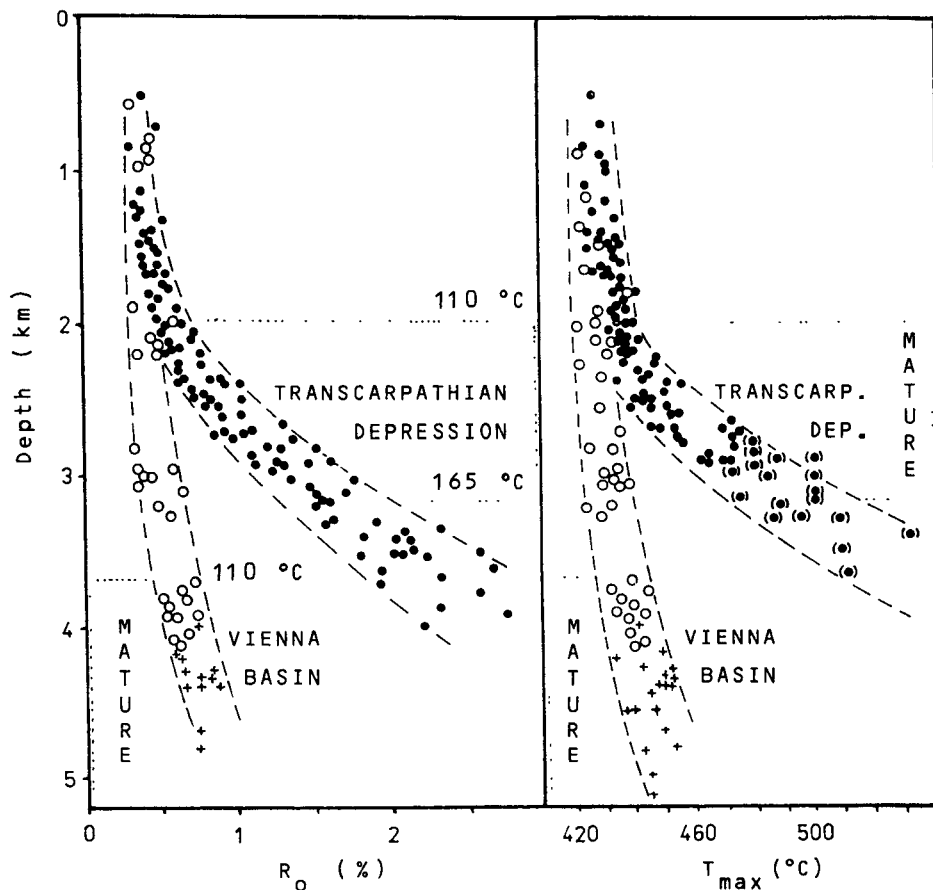


Fig. 3 Vitrinite reflectance (R_o) and maximum pyrolytic temperature (T_{max}) with depth in the Neogene of the Vienna basin and the Transcarpathian depression.

Data in parenthesis are at the detection limit; + - Triassic samples.

overmature and its source potential in all units in the central and eastern parts of the Transcarpathian depression is depleted.

In the Vienna basin the source potential (HI) slightly decreases and free hydrocarbons are formed in shales at depth of 3.5 km and more. This mature zone probably continues to the deepest parts of the Neogene in the Vienna basin (5.5 km).

Smectite illitization

The most important catagenetic alteration of the inorganic part of shales is the dehydration of the expandable clay minerals mainly of the illite-smectite group (Dunoyer de Segonzac, 1970; Perry - Hower, 1972; Drits - Kaporulin, 1973). The degree of this mineral conversion is expressed by percentage of expandable smectitic layers in

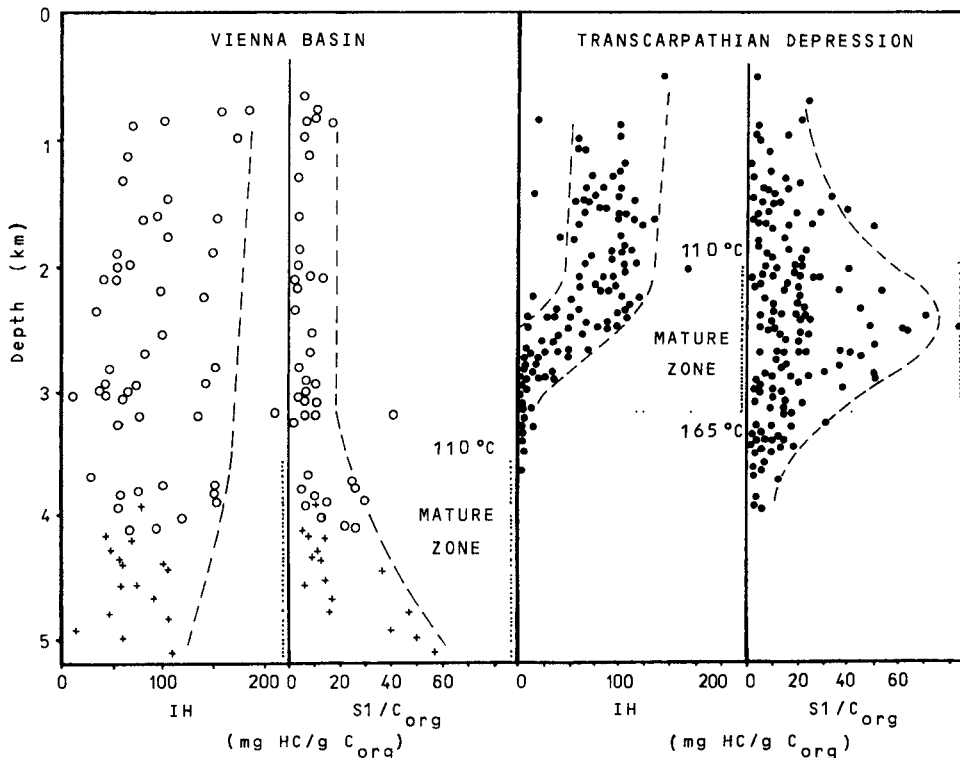


Fig. 4 Pyrolytic characteristics with depth: hydrogen index (HI) represents the fixed hydrocarbons (residual source potential of kerogen) which are during maturation converted to free hydrocarbons ($S1/C_{org}$).

mixed-layer illite-smectite and by the type of layer ordering (Reynolds – Hower, 1970; Drits – Sakharov, 1976; Reynolds, 1980; Środoń – Eberl, 1984).

In the Transcarpathian depression the randomly interstratified illite-smectite ($R = 0$) with expandability over 65 % S occurs at depth less than 1.8 km (Fig. 5) (Franců et al., 1989). This mineral type shows a distinct 1.7 nm diffraction after ethylene-glycol saturation and differs from the pure montmorillonite by the 003 diffraction shifted from $15.4^\circ 2\theta$ to higher angles. At depth ranging from 1.7 to 2.2 km this randomly ordered structure changes to illite-smectite with ordered interstratification (“IS” type with $R = 1$) and with expandability decreased to about 35 % S. The 1.7 nm diffraction disappears. This phenomenon was earlier observed by Kraus – Šamajová (1978) in a number of boreholes in the Transcarpathian depression (East Slovakian Neogene basin). They also pointed out to the differences in illitization of volcanic and of nonvolcanic smectite.

In this upper zone the expandable clay minerals undergo the first or main stage of dehydration (in terms of Perry – Hower, 1972). In the Transcarpathian depression and in many other basins it occurs at the upper boundary of the hydrocarbon generation zone. In the Vienna basin the ordered illite-smectite occurs below 3.2–3.5 km, i.e. much deeper than in the Transcarpathian depression but at similar temperatures (Fig. 5). Similar results were

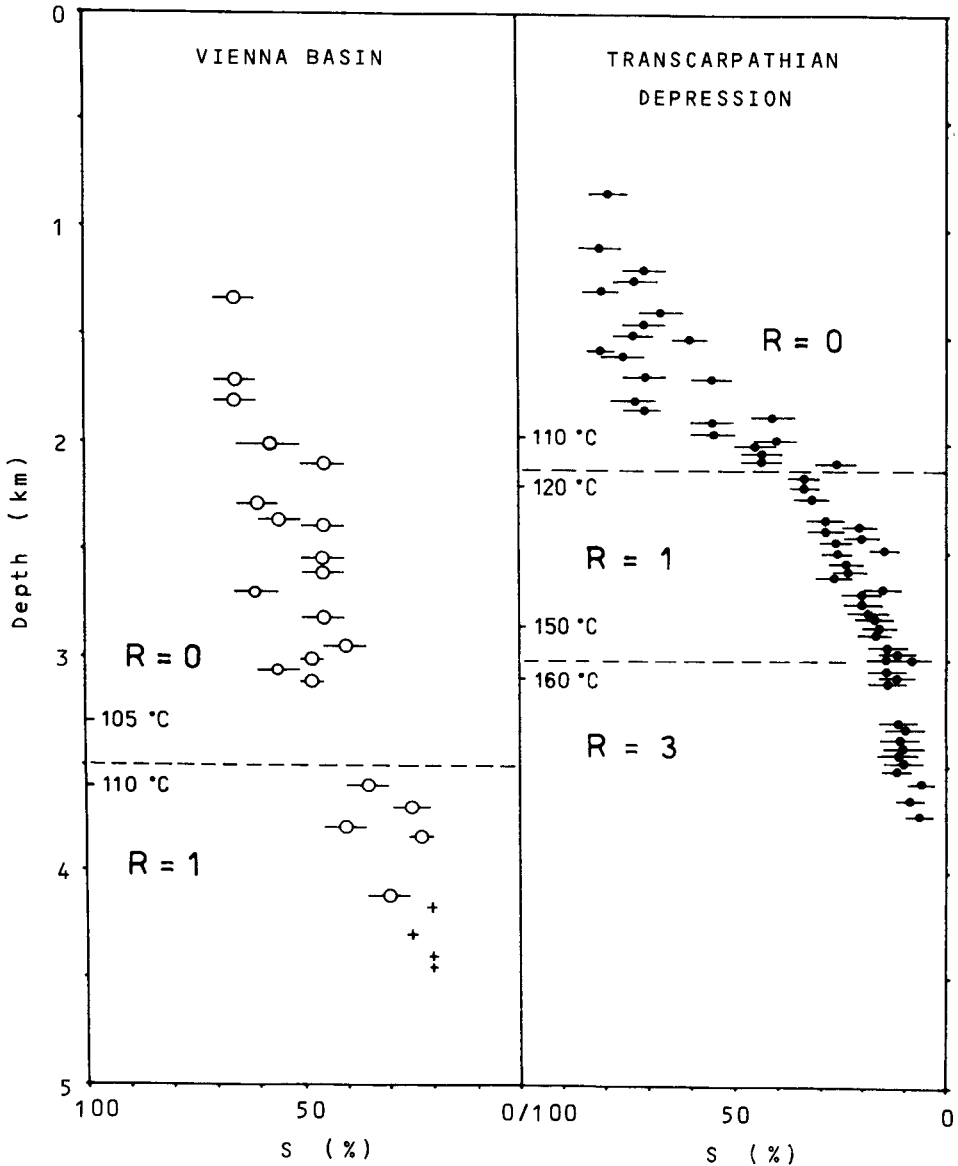


Fig. 5 Smectite-to-illite conversion with depth in the Neogene shales.

Expandability of mixed-layer illite-smectite is expressed by % S; R (Reichweite) – type of ordering; + – Triassic samples.

presented also by Johns – Kurzweil (1979) and Kurzweil – Johns (1981) from the Austrian part of the Vienna basin who observed transition to ordered illite-smectite at about 100 °C preceded by a reversal in illitization linked with higher concentration of Mg^{2+} in pore waters.

At depth of about 3 km in the Transcarpathian depression the ordered illite-smectite of the ISII type ($R = 3$) occurs with expandability of less than 15 % S. In the zone between 2.2 and 3 km the second and less intensive stage of dehydration takes place being parallel to the hydrocarbon generation (Figs. 4 and 5). At greater depth than 3–3.5 km the expandable minerals are highly illitic and dehydration is much weaker than in the upper zones. More details on the smectite-to-illite conversion and its analyses in the Transcarpathian depression are given by Franců – Milička (1988) and Franců et al. (1989).

Time-temperature model of catagenesis

The method of Lopatin (1971, 1983) is used to reconstruct the burial (depth) history and thermal exposure of the studied sedimentary sequence. Final degree of catagenesis is calculated from temperature and effective duration of heating in each $\Delta 10^\circ\text{C}$ interval (in Fig. 6 marked by full dots on the burial path of the boundary layer between the Badenian and Sarmatian) and expressed as TTI (time-temperature index).

The diagram in Figs. 6 and 7 shows the evolution of two chosen borehole sections with depth: Rebrín 1 in the Transcarpathian depression and Závod 72 in the Vienna basin. The calculated degree of thermal alteration TTI is tested by vitrinite reflectance. More details on the diagram construction and time-temperature index calculation is given in general by Waples (1980) and for one of these basins by Franců et al. (1989).

Following conclusions may be drawn from the models shown in Figs. 6 and 7:

- thickness of the zone of main hydrocarbon generation is in the Transcarpathian

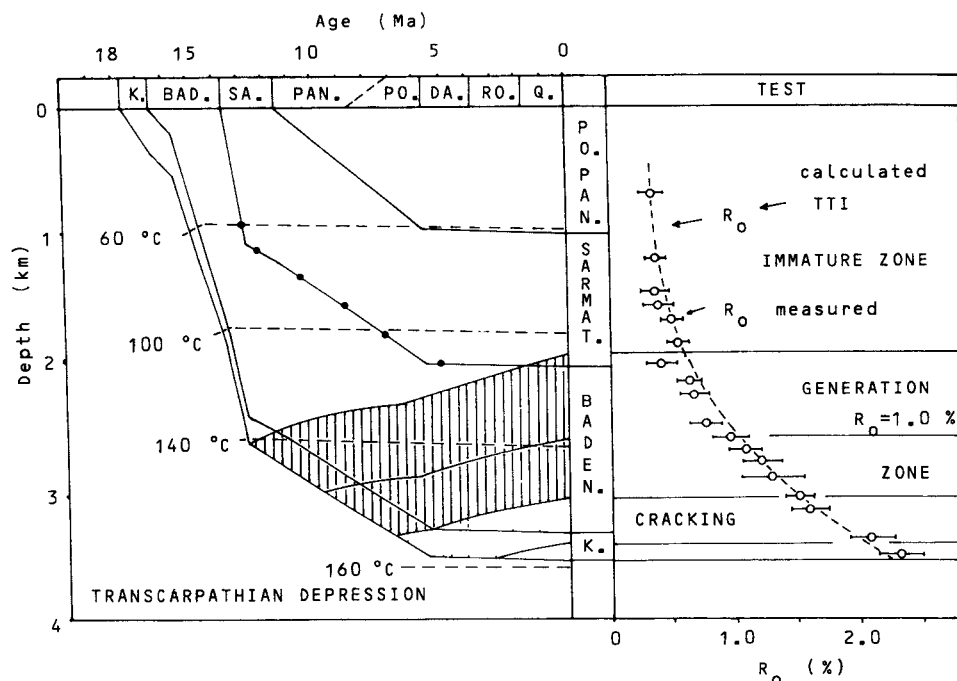


Fig. 6 Burial history of the Rebrín 1 borehole Neogene section in the Transcarpathian depression. Horizontal axis (left) is age, hatched area shows the main zone of wet gas generation, stippled area shows the cracking zone.

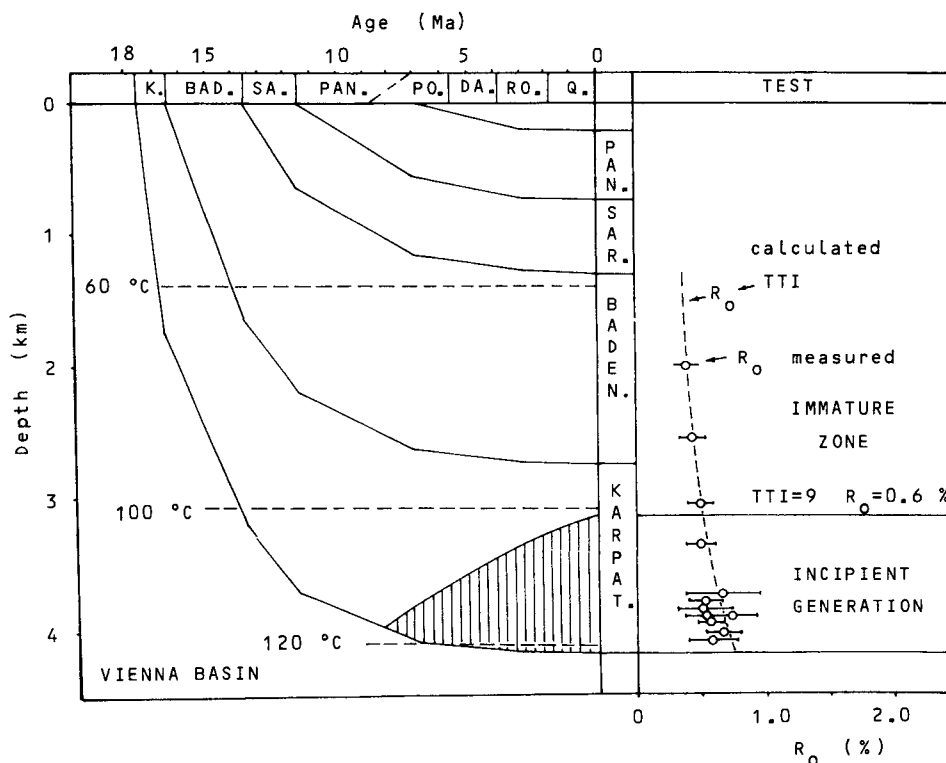


Fig. 7 Burial history of the Závod 72 borehole Neogene section in the Vienna basin.

The dashed line (right) is the theoretical vitrinite reflectance converted from the time-temperature index calculated by the model.

depression lower than 1.2 or 1.6 km. The Badenian source rocks passed through the mature stage (marked by R_o from 0.6 to 1.5 %) within 7–9 mil. years. The maturity gradient with depth is up to $0.8 \% R_o \text{ km}^{-1}$;

– diagenesis and catagenesis proceeds much weaker with depth in the Vienna basin. Onset of hydrocarbon generation ($R_o = 0.6 \%$) is very diffuse at depth ranging from 3 to 4 km. In the deepest Karpatian sediments the maturation gradient with depth is about $0.15 \% R_o \text{ km}^{-1}$ and the maturation rate is $0.18 \% R_o (10 \text{ mil. years})^{-1}$.

Conclusions

In the both studied basins the humic, i.e. gas-prone type III kerogen is disseminated in the Neogene rocks and gas with minor portion of liquid hydrocarbons may be their maturation product.

Geochemical analyses and time-temperature modeling give evidence of strong catagenetic alteration of the Neogene rocks with depth in the Transcarpathian depression. In a relatively narrow mature zone with ceiling at depth of 1.7–2.0 km and floor at 3.0–3.5 km conditions are favourable to wet gas generation. At greater depth cracking should dominate and below 3.5 km only dry gas (methane) may survive the high temperature.

Smectite illitization and dehydration slightly anticipate the hydrocarbon generation but overwhelmingly the both processes occur paralelly and are spatially linked with overpressured formation fluids.

In the Vienna basin with much lower geothermal gradient all diagenetic and catagenetic alterations are much weaker and shifted to greater depth. In this "cold" basin the incipient hydrocarbon generation is observed at depth over 3.5 km, while the sediments in the "hot" basin plunge at this depth from the cracking to dry gas zone. The maximum hydrocarbon generation at vitrinite reflectance $R_o = 1\%$ probably does not occur even in the deepest parts of the Neogene in the Vienna basin at 5.5 km, while in the Transcarpathian depression it occurs at 2.5 km.

Intensive smectite-to-illite conversion within a relatively narrow depth interval in the Transcarpathian depression ought to result in a significant sandstone cementation below the depth of 2 km, this problem being worth of further study. In the Vienna basin these processes should be observed below 3.5 km.

Modeling gives evidence that in the Vienna basin the organic maturation in strata buried to 3.5 km has been roughly 6 times slower than in the Transcarpathian depression.

From these facts it may be concluded that the catagenetic conditions for generation of economic amount of hydrocarbons (with chance not to be dispersed too much) are more favourable in the Neogene shales in the Transcarpathian depression than in the Vienna basin.

The pre-Tertiary rocks in the Transcarpathian depression are mostly overmature or metamorphosed to meta-anthracitic or higher stages and, therefore, lost their source potential in pre-Neogene time.

In the Vienna basin the pre-Neogene rocks are often in mature stage and cracking may be dominant at depth of 6–7 km (160–170 °C). Biogenic methane at shallow depth and early catagenetic gas probably contribute to the total hydrocarbon reserves in the Neogene rocks. The oil which forms economic accumulations in this basin should, however, be derived from some pre-Neogene source rocks either of autochthonous position on the slopes of the Bohemian Massif, or of the Alpine-Carpathian nappes at depth ranging from 4 to 6 km. Gas generation which plays important role in oil migration probably occurred in recent geological history even deeper than 6 km.

In the Austrian part of the Vienna basin the oils are proved to be of one family (with a single exception) originating from the marls of the autochthonous Malmian buried below the Flysch nappes, the Alpine nappes and the Neogene to depth of 4–6 km (W e l t e et al., 1982; K r a t o c h v i l – L a d w e i n, 1984; L a d w e i n, 1988).

The present geochemical study needs to be complemented by oil-oil, oil+ gas-source rock correlations using biomarkers to understand better the migration history of hydrocarbons in the both basins. Next step in modeling should apply kinetic equations for hydrocarbon formation, include the quantitative estimation of source potential in a basin-scale and compare it with the known reserves. Clay mineral study should be extended to porosity reduction linked with mineral neoformation in pore space of the reservoir rocks.

This paper collected evidence of different diagenetic and catagenetic zonality in two selected basins and documented the important evolution stages by indices which show good mutual correlation and good relationship with subsurface temperature.

Acknowledgements: We express our thanks to the management of Oil and Gas Industry (NPP) for permission to publish this paper. We greatly acknowledge the suggestions of Prof. I. K r a u s, Prof. W. D. J o h n s, and Dr. J. Š r o d o ň concerning clay mineralogy, Ing. V. Š i m á n e k in organic geochemistry, Ing. R. R u d i n e c, Dr. F. C h m e l í k and Dr. M. K o v á č in geology. We thank Dr. B. T o m a n and M. P l š k o v á for X-ray diffraction analyses.

APPENDIX

List of boreholes studied in this paper:

Transcarpathian depression: Čičarovce 8, Lastomír 1, Ložín 1, Pavlovce 1, Ptrukša 22, Rebrín 1, Stretava 21, Trhovište 26. *Vienna basin*: Borský Jur 14, Břeclav 25, 26, Hrušky 3, 8, 12, 33, 34, 35, 45, 186, 188, 228, Kuklov 3, Lakšárská Nová Ves 7, Týnec 1, Závod 72, 74, 75, and 78.

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