Model of gaseous hydrocarbon generation in the Miocene strata of the Polish part of the Carpathian Foredeep

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The TTI modelling demonstrated that the autochthonous Miocene strata located in the outer Miocene basin of the Carpathian Foredeep, north of the present edge of the Carpathian overthrust have generated only the microbial gas. In the part of the Miocene basin covered recently by the Carpathian overthrust the low-temperature thermogenic gases were generated down to 7,000 of meters depth. Finally, in the hypothetical zone of the Lower Miocene molasse located recently at depth interval 7,500–11,000 meters only the high-temperature thermogenic gases could be formed. Alternatively, at the site of the Lower Miocene molasse the depression filled with Upper Carboniferous coal-bearing formation may occur. Therefore, at depth beneath 7,500 meters, i.e. beneath the Carpathian overthrust the gas deposits can be expected. The maximum yield of the microbial methane generation within the autochthonous Miocene calculated for depth interval 900 to 1,500 meters is about 5 cubic meters per cubic meter of source rock. It is possible that the generation process of microbial methane still continues. Accumulation of microbial methane within the autochthonous Miocene strata was facilitated by high sedimentation rate and rhythmic and cyclic deposition of clays and sands.

Key words: *petroleum exploration, Miocene, basin analysis, energy sources, natural gas, genesis, methane, biogenic processes, thermal history, models, Carpathian Foredeep, Poland*

Introduction

The Carpathian Foredeep is the largest gas basin among the all foredeeps of the Alpine orogenic system in Europe. In the autochthonous Miocene strata of the Polish part of the Carpathian Foredeep only methane-dominated deposits have been discovered up to date. Since 1945 about 70 gas fields have been found of total resources around 200 billion cubic meters.

In the following paper the problem of gas generation is discussed within the autochthonous Miocene sequence of the Polish part of the Carpathian Foredeep.

The selection of analytical material for geochemical studies on the dispersed organic matter (OM) collected from the drill cores considered the spatial lithofacial development of the autochthonous Miocene molasse in the Polish part of the Carpathian Foredeep (Kotarba et al., 1998). Consequently, the four test areas representative for hydrocarbon generation conditions have been chosen in the Miocene outer basin (Fig. 1).

The two test areas A and B reflect the hydrocarbon generation conditions in the part of the Carpathian Foredeep north of the recent edge of the Carpathian orogen. The test areas C and D represent the conditions at the edge of the Carpathian orogen. Finally, the test area E includes the hydrocarbon generation conditions in that part of the inner basin which is recently covered by the overthrust (Fig. 1). For each of the test areas the TTI modelling has been carried out (e.g., Waples, 1980, 1985).

The study has been undertaken as a part of research projects of the Carpathian Foredeep financed by the State Committee for Scientific Research in Warsaw (grant No. 9 9214 92 03) and National Fund for Environmental Protection and Water Management (grant No. 2.14.0100.00.0).

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Outline of basin development

Taking into account the genetic classification of sedimentary basins (Miall, 1990), the Carpathian Foredeep belongs to the peripheral foreland basins of the Alpine-age tectonic system of the Carpathians. The Carpathian Foredeep is divided into the two basins: outer and inner (Ney et al., 1974; Oszczypko, 1996, 1997).

The formation of outer Miocene basin in the Polish part of the Carpathian Foredeep (Oszczypko, 1982; Oszczypko & Ślączka, 1985, 1989; Oszczypko & Tomaś, 1985; Kotlarczyk, 1988; Oszczypko, 1996, 1997) is closely connected with the multiphase, northward thrust of the Carpathian flysch orogen onto the foreland platform and the deposition of succeeding suites of Badenian and Lower Sarmatian molasses at the front of orogenic belt (Fig. 1). Depositional space of the Polish part of the foredeep was provided by the subsidence of polygenetic basement of the Miocene basin bordered from the south by migrating flysch nappes and from the north by stable slope of the platform. The southernmost part of this space was occupied by the Lower Miocene inner basin filled with molasse sequences ranging in age from Eggenburgian to Karpatian. These sediments were subsequently folded together with the overthrusting flysch nappes and displaced to the north where they are recently known as the Stebnik and the Zgłobice units located along the northern edge of the Flysch Carpathians (Ney, 1968; Ney et al., 1974; Oszczypko, 1997).

In the depositional space of the outer basin the Badenian transgression has invaded the foreland of just developing flysch orogen in the Lower Moravian. Tectonic migration of the Carpathians which has commenced in the Moravian (Kotlarczyk, 1988) caused folding of Lower Miocene molasse in the inner basin which resulted in: (i) formation of the Stebnik Unit in the east and (ii) displacement of the flysch orogen together with the Zgłobice Unit in the west. In the Middle Badenian (Wielician) the front of this overthrust has reached the position some 10–15 kilometers south of the recent edge of the Carpathians (Wdowiarz, 1983). At the front of this overthrust the asymmetric foredeep has been formed (outer basin) where the thicknesses of Badenian and

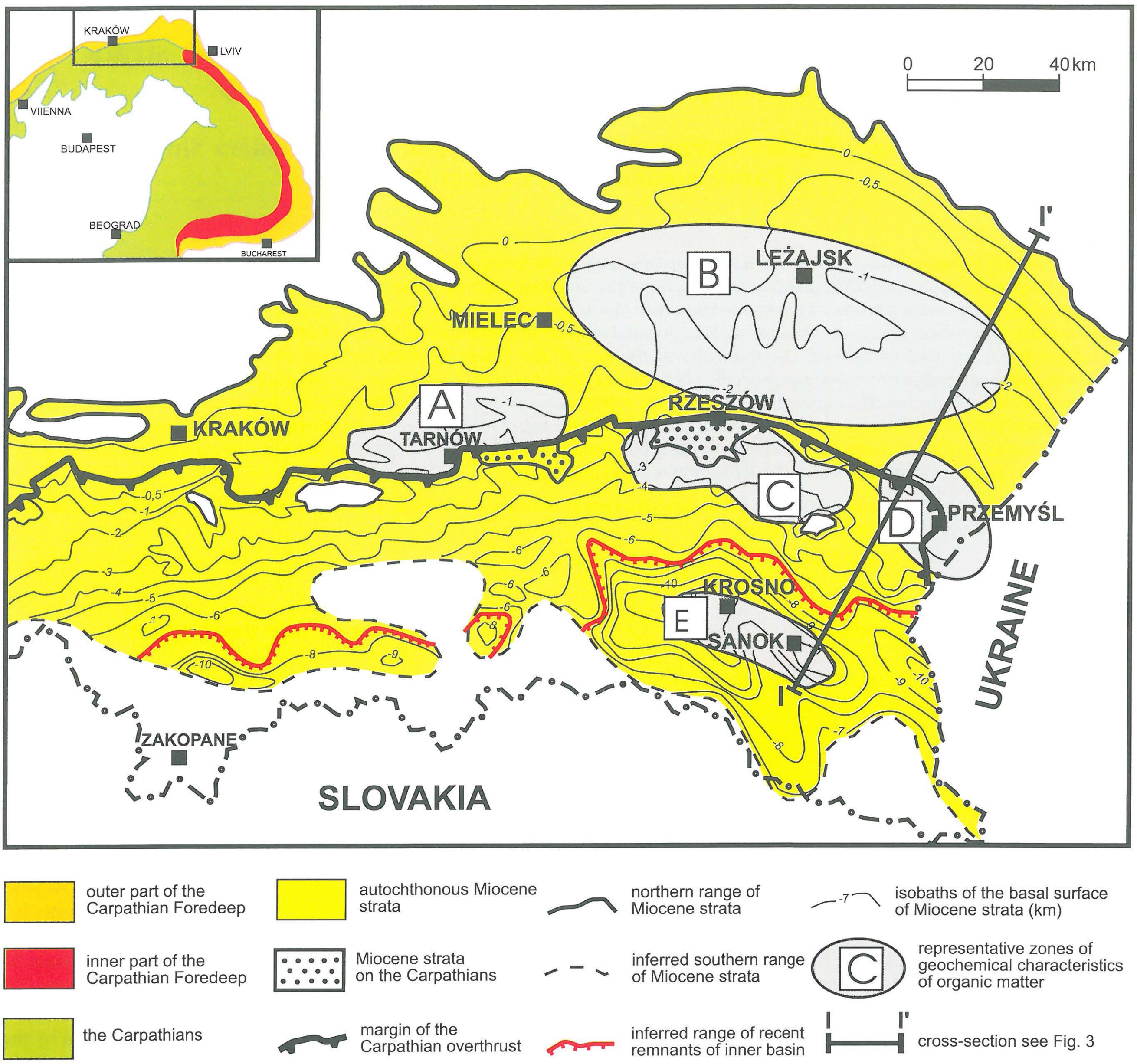


Fig. 1. Sketch map of the central and eastern parts of the Polish Carpathian Foredeep and location of studied test areas

Lower Sarmatian molasses vary from 2,500 to 3,000 meters at the edge of the Carpathian overthrust to a dozen of meters at the northern margin of the basin (Fig. 1). Thickness of autochthonous Miocene strata beneath the Carpathian overthrust ranges from some tens to some hundreds of meters (Jucha, 1985). Thickness of Lower Miocene suite (Eggenburgian–Karpatian) of the inner basin reaches maximum of 2,400 meters in the area of the Stebnik Unit (Ney et al., 1974) and amounts to some tens of meters in the area of the Zgłobice Unit (Oszczypko, 1996, 1997).

Recent depth of autochthonous Miocene strata in the outer basin, under the flysch nappes varies from 2,500–3,000 meters at the edge of orogenic belt to 7,000–8,000 meters in central part of the Flysch Carpathians. Relics of the autochthonous Lower Miocene of inner basin in Krosno–Sanok and Nowy Sącz zones are presumably located at depths interval 7,000–11,000 meters (Figs 1 and 3).

The existing genetic link between Lower Miocene molasses of the inner basin and Upper Miocene molasses of the outer basin allows the application of uniform chronostratigraphy of the Central Paratethys to lithofacial complexes of the whole Miocene sequence in the foredeep (Łuczkowska, 1967; Steiniger et al., 1990; Berggren et al., 1995) (Fig. 2).

The general lithostratigraphy includes the two main molasse formations: Lower Miocene in the inner basin and Upper Miocene in the outer basin. The Lower Miocene formation of the Stebnik and Zgłobice units comprises the Worotyszcze (Eggenburgian), Stebnik (Ottnangian), Balice (Karpatian) and Przemyśl (Lower Moravian) series of total thickness up to 2,400 meters (Ney et al., 1974). These series are barren from the point of view of petroleum exploration.

The Upper Miocene formation of outer basin which contains Badenian littoral, neritic and deltaic facies (Moravian, Wielician, Kosovian) as well as the Lower Sarmatian strata is gas-prone. Up to date 70 natural gas deposits have been discovered in these rocks and reserves of about 200 billion cubic meters have been quoted. The eastern part of the outer basin (east of Kraków) is filled with Badenian and Lower Sarmatian sediments of the following thicknesses: Lower and Middle Badenian — from 0 to 300 m, Upper Badenian — from 0 to 1,700 m, and Lower Sarmatian — from 0 to 2,900 m (Ney et al., 1974). Most of the known gas

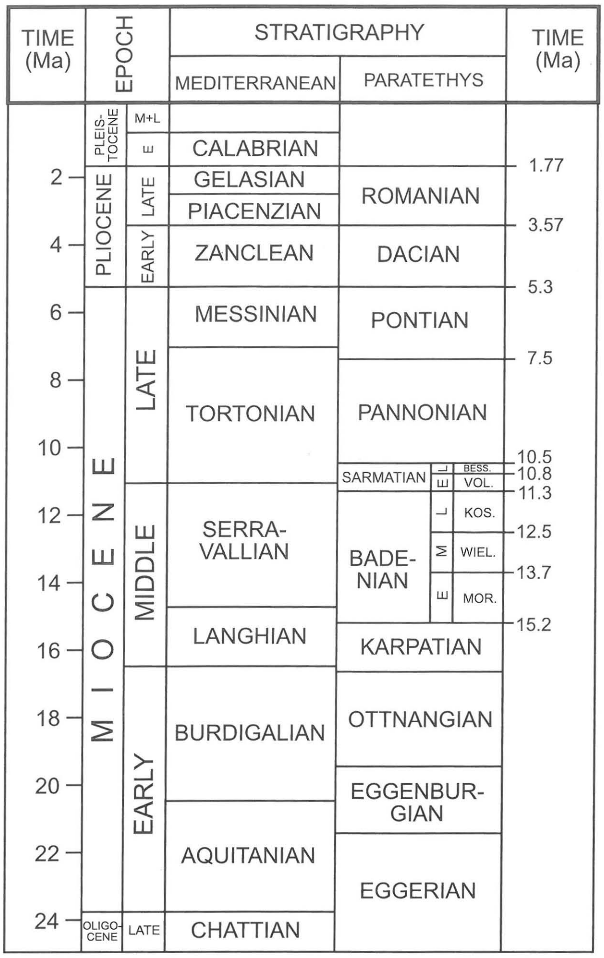


Fig. 2. Generalized stratigraphic section and regional Miocene correlation (after Berggren et al., 1995; Steininger et al., 1990; Czepiec & Kotarba, 1998)

fields are reservoired within the Upper Badenian and Lower Sarmatian strata. Both the Upper Badenian and the Lower Sarmatian are represented by clay-sandy, mainly deltaic facies (Karnkowski, 1989). Sedimentation rate of Upper Badenian sediments was maximum 1,500 m/Ma, and that of the Lower Sarmatian reached 5,000 m/Ma. On the other hand, the Lower and Middle Badenian strata comprise shallow-water psammitic, argillaceous and chemical sediments.

The maximum depth of Badenian sea was 300 meters. The warm-temperate climate during this time suggests a surficial-water temperature range of 17–20°C. In the Sarmatian the sea depth was initially about 30–50 meters. Then, progressing shallowing to about 10 meters took place. The Sarmatian sea was a warm basin with temperatures roughly corresponding to those of the Late Badenian ones (Czepiec & Kotarba, 1998).

Gaseous hydrocarbon generation system

Composition of natural gases accumulated in Miocene sequence of the Carpathian Foredeep, is dominated by methane which usually constitutes over 98 vol. %. Methane was generated during carbon dioxide reduction pathway of microbial processes (Fig. 4) (Kotarba, 1998). Higher gaseous hydrocarbons (mainly ethane and propane) which are usually minor constituents (concentrations less than 0.2 vol. %), were generated during diagenetic processes and at the initial stage of the low-temperature thermogenic processes (Kotarba, 1998).

Geochemical analyses of the OM contained in the autochthonous Miocene of the Carpathian Foredeep indicated the domination of humic, gas-prone type III kerogen in both the Upper Badenian and the Lower Sarmatian strata and only sporadic occurrence of algal, type II kerogen (Fig. 5) (Kotarba et al., 1998). The TOC contents vary from 0.02 to 3.22 wt. % (mean 0.68 wt. %). Down to the present depth 3,200 meters the OM is immature. Its transformation degree is very low and corresponds to the vitrinite reflectance R_o from 0.25 to 0.6% (dominating values around 0.4%) and to the Rock Eval T_{max} temperatures from 415 to 438°C (Kotarba et al., 1998).

The hydrocarbon generation conditions were analyzed in the 4 typical test areas. The A (Tarnów) and B (Mielec–Leżajsk) areas represent the part of the Carpathian Foredeep located north of the present edge of the Flysch Carpathians. The C (Rzeszów) and D (Przemyśl) areas are located in the autochthonous Miocene outer basin along the present edge of the Flysch Carpathians (Fig. 1). Additionally, the E (Krosno–Sanok) area was chosen as an example of hypothetical deepest levels where the relics of autochthonous Lower Miocene strata of the inner basin might have existed (Fig. 1). The hydrocarbon generation modelling applied the time-temperature (TTI) method (Waples, 1985). The time scale was based upon Berggren et al. (1995); Steininger et al. (1990), Czepiec & Kotarba (1998) (Fig. 2) whereas the geothermal gradient — $33°C/10^{-3}$ m is the average value obtained from paleotemperature calculations for the Miocene basin (Szafran, 1990) and correlated with the vitrinite reflectance values (Kotarba et al., 1998). Thicknesses of stratigraphic units were taken from the results of drillings in the analyzed parts of the outer basin. The estimated erosional reduction was 100 meters in A and B test areas and 250 meters in the remaining ones (Oszczypko, personal communication). TTI modellings in the outer basin (test areas A to D) were related to the bottom of Badenian succession as datum surface. In the test area E (inner basin) the datum surface was the top of Karpatian sequence.

The results of modelling revealed that in both the A and B test areas (north of the egde of the Carpathians) the TTI values were 0.1 and 1, respectively (Fig. 6) which proves that thermogenic gases have not been generated. At the edge of the orogen the maximum TTI values were: 21 at depth about 3,600 meters in C test area and 4 at depth about 2,600 meters in D one. These values suggest the appearance of low-temperature thermogenic processes (Fig. 6) within the autochthonous Miocene strata beneath the Carpathian overthrust (i.e. beneath 2,500 meters depth).

The modelling for the test area E indicated the possibility of generation of the higher gaseous hydrocarbon (and even liquid hydrocarbons) beneath the Carpathian overthrust, at about 7,500 meters of depth. At greater depths, within the relics of autochthonous Lower Miocene strata of the inner basin only the high-temperature methane could be produced.

The results of modelling for all the test areas proved the generation of microbial methane in almost full thickness of autochthonous Miocene of the outer basin. Taking into account the quantitative criteria for microbial gas generation (Clayton, 1992), the intensity of methane production was calculated for the generalized sedimentation conditions of the Miocene marine basin. It was found that generation processes of microbial methane were most intensive at depth

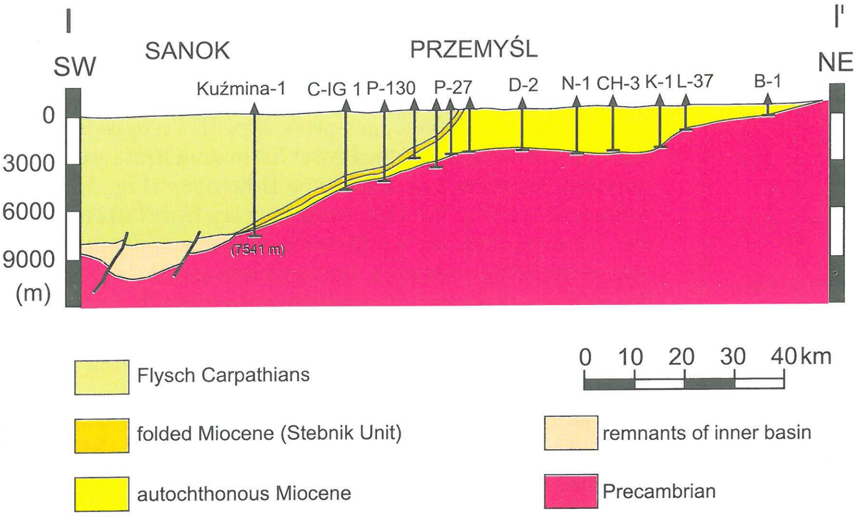


Fig. 3. Schematic cross-section through the Carpathian Foredeep

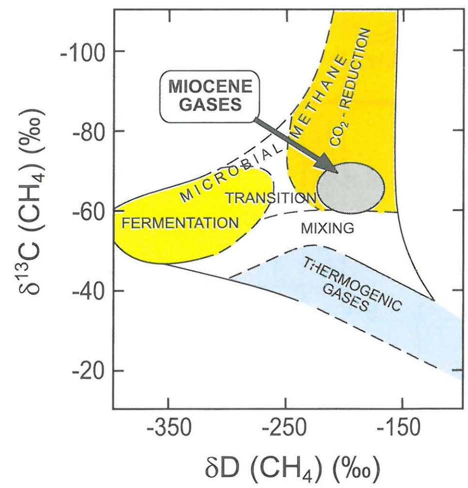


Fig. 4. Genetic characterization of natural gases accumulated in the autochthonous Miocene strata of the Carpathian Foredeep in terms of $\delta^{13}C(CH_4)$ versus $D(CH_4)$. Compositional fields from Whiticar et al. (1986)

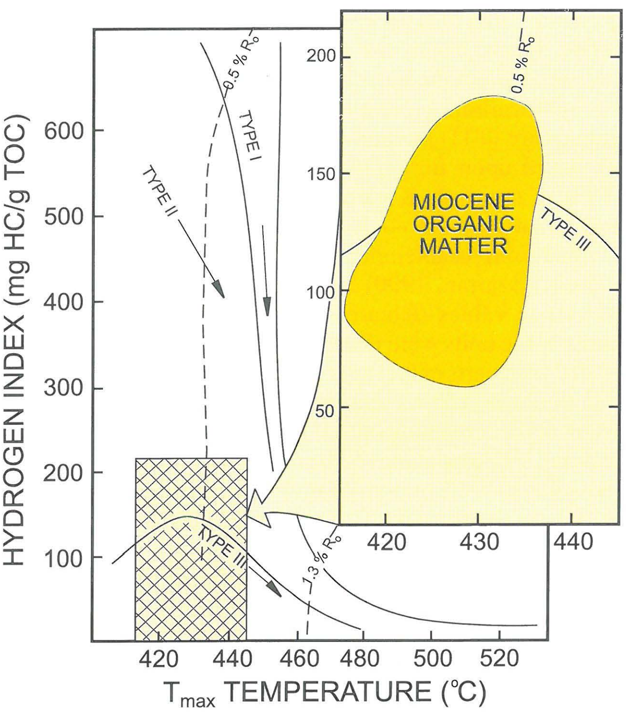
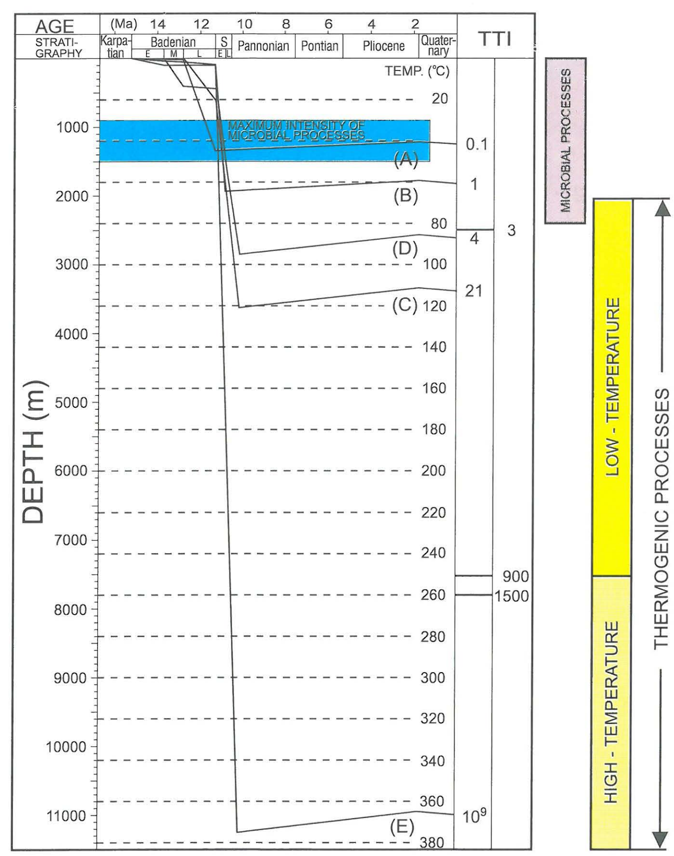


Fig. 5. Rock Eval hydrogen index versus T_{max} temperature for organic matter in the autochthonous Miocene strata of the Carpathian Foredeep after Kotarba et al. (1998)



E – Early; M – Middle; L – Late
S – Sarmatian

Fig. 6. Model of gas generation within the autochthonous Miocene basins and family of burial-history curves, subsurface temperature grid and Time-Temperature Index values for: (A) Tarnów, (B) Mielec–Leżajsk, (C) Rzeszów, (D) Przemyśl, and (E) Krosno–Sanok areas

interval 900 to 1,500 meters beneath the Miocene sea floor. Maximum yield of microbially-produced methane were about 5 cubic meters of CH_4 per cubic meter of a source rock. The volume of produced microbial methane is spatially variable within the basin and depends on the burial history. As no dramatic changes of thermal conditions have been deduced within the basin since the Sarmatian, this microbial generation processes have presumably continued until recent. Even the folding and the uplift of the Carpathian orogen, and the consequent regression of the Miocene sea at the break of Lower and Upper Sarmatian did not impede the microbial processes.

In geochemical literature the idea has been presented that microbial methane might have been accumulated as hydrate beneath the Miocene seafloor (Głogoczowski, 1980; Kotarba, 1992). However, the results of new bathymetric analyses (Czepiec & Kotarba, 1998) revealed that the Badenian sea was presumably only slightly deeper than the outer shelf, i.e. about 300 meters, at most. Depth of the Sarmatian sea was initially about 30–50 meters and has shallowed to

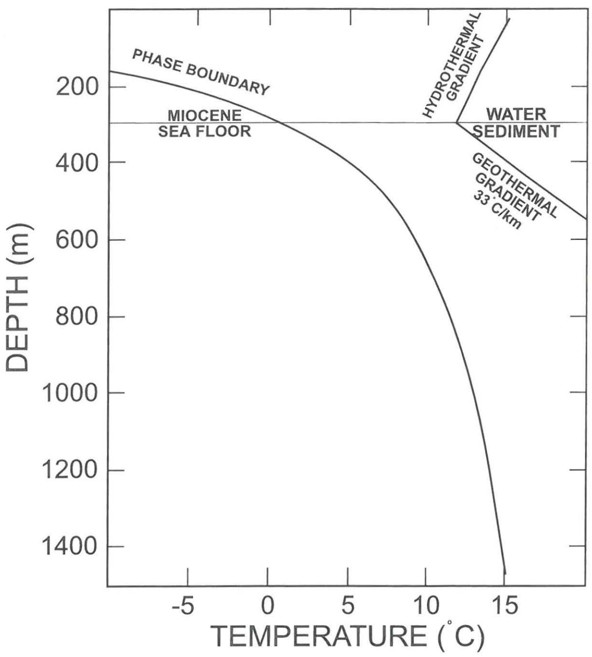


Fig. 7. Impossibility of formation of methane hydrate in Miocene sea. Phase boundary for methane hydrate after Kvenvolden & MacMenamin (1988) and Kvenvolden (1988)

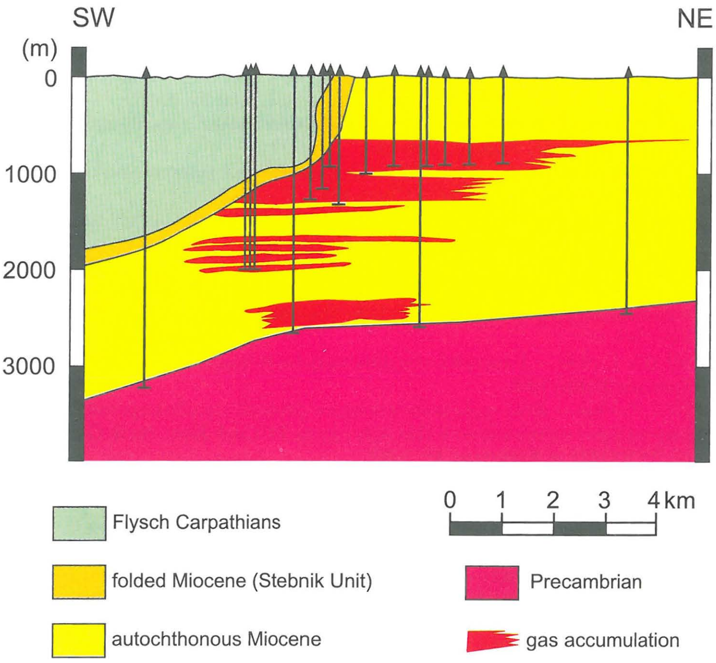


Fig. 8. Schematic cross-section through the Przemyśl gas accumulation zone after Czernicki (1977)

only about 10 meters. Both the Badenian and the Sarmatian seas were the warm basins of surface water temperatures 17–20°C (Czepiec & Kotarba, 1998). Taking into account these data, the formation of natural methane hydrate zone beneath the Miocene seafloor must be rejected (Fig. 7).

The high sedimentation rates together with rhythmic and cyclic deposition of Miocene clays and sands as well as the vigorous generation of microbial methane caused that the gas produced in claystone layers readily migrated to the sandstone beds capped, in turn, by succeeding claystones. Such generation and accumulation system of microbial gases gave rise to the formation of multi-horizontal gas fields (Fig. 8).

Conclusions

The TTI modelling proved that the autochthonous Miocene strata of the outer basin of Carpathian Foredeep located north of the orogen edge have generated only the microbial methane at depths less than about 2,400 meters below the Miocene seafloor. In the part of the Miocene outer basin covered by the Flysch Carpathians the low-temperature thermogenic gases have been generated down to about 7,500 meters of depth. The Lower Miocene molasse of the inner basin located at depths interval 7,000–11,000 meters might have generated only the high-temperature thermogenic gases. The site of hypothetical Lower Miocene strata of the inner basin may well be occupied by a depression filled with the Upper Carboniferous coal-bearing formation (Kozikowski, 1966) which seismic response is similar to that of the Miocene molasse. Nevertheless, the gas accumulations can be expected beneath 7,500 meters of depth, i.e. under the cover of the Carpathian overthrust.

Methane which is the main component of natural gas deposits accumulated within the autochthonous Miocene strata of the Carpathian Foredeep was formed during the microbial processes. Higher gaseous hydrocarbons were generated by low-temperature thermogenic reactions from the Miocene source rocks buried under the Carpathian overthrust. Small amounts of these components occur exclusively in the deepest horizons of the Miocene gas deposits (e.g., Przemyśl and Husów). Maximum yield of the microbial methane generation calculated for depth interval 900 to 1,500 meters is about 5 cubic meters per cubic meters of source rocks. It is probable that the generation process of microbial methane still continues recently. Accumulation of microbial methane within the autochthonous Miocene strata was facilitated by high sedimentation rate and rhythmic and cyclic deposition of clays and sands.

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