# CLAY MINERALS OF A THICK SEDIMENTARY SEQUENCE IN SE PART OF THE PANNONIAN BASIN (HUNGARY)

# ISTVÁN VICZIÁN

Hungarian Geological Institute, Stefánia ut. 14, H-114 Budapest, Hungary

Abstract: Neogene pelitic rocks were investigated in a 4.2 km thick sequence of the borehole Doboz I representing a deep partial depression of the Pannonian Basin. This is the first continuously sampled sequence of the deep partial basins of the Hungarian Neogene. In the composition of the  $<2\,\mu m$  fraction mixed-layer illite/smectites and illite predominate while kaolinite and chlorite permanently appear in minor amounts. There is a systematic variation in the smectite proportion of mixed-layer illite/smectites due to diagenetic transformation of the primary clay material. Rapid changes in the smectite proportion were observed in two narrow depth intervals at 1.0 and 3.0 km ( $\approx 90$  to 50 to 25 %, respectively). Mixed-layers are of very undefined compostion as it is shown by diffuse basal reflections of ethylene glycol treated samples and by double maxima of the first order basal reflection of the untreated samples ( $\approx 14.5~\mbox{Å}$  and 11.5 ). In the clay fraction and pore spaces of Mesozoic rocks which subsided during the basin formation in the Neogene special clay minerals were formed which are indicators of a high degree of diagenetic transformation (tosudite, dickite).

Key words:

#### Introduction

Results of the study of diagenetic smectite to illite transformation in the Pannonian Basin were reported previously by Viczián (1985). In this former paper data for 14 bore-holes scattered in the whole territory of Hungary were presented. The bore-hole Doboz I is, however, the first one that represents an almost continuous sequence of the whole Neogene. The investigation of this sequence is therefore of special interest and this is the subject of the present study.

The bore-hole Doboz I is located above a relative elevation in the deep central region of the Békés Depression in SE Hungary (Kilényi et al. 1991). The samples were taken from cuttings representing 100 m intervals of a more that 4 km thick sedimentary sequence. Only the lowermost samples are core samples. The Neogene rocks are mostly clayey siltstones with variable carbonate content (see Fig. 2).

Geological and stratigraphic evaluation of the sedimentary sequence was carried out in 1984 by Jámbor and Kárpáti-Radó. According to their investigations the subsidence of this partial depression of the Pannonian Basin started in the Lower Pannonian after a long terrestrial and denudation period and lasted until the Quaternary.

### Methods of investigation

The carbonate content was dissolved by 3 % HCl and the  $<2\,\mu m$  fraction was separated by sedimentation in an Atterberg cylinder. X-ray diffraction analysis was made on oriented specimens using a Philips diffractometer and  $CuK_{\alpha}$  radiation.

Mixed-layer illite/smectites were determined in ethylene gly-

col treated specimens using theoretical diffraction patterns and determinative graphs published by Reynolds & Hower (1970), Johns & Kurzweil (1979) and Srodon (1980).

Mixes-layer illite/smectites were also characterized by two additional parameters:

IC: "illite crystallinity" of Kübler (1964) which is the width of the 10 reflection at half height in  $^{\circ}2$   $\Theta$  units of untreated samples.

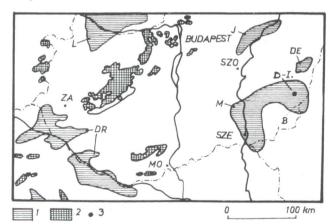
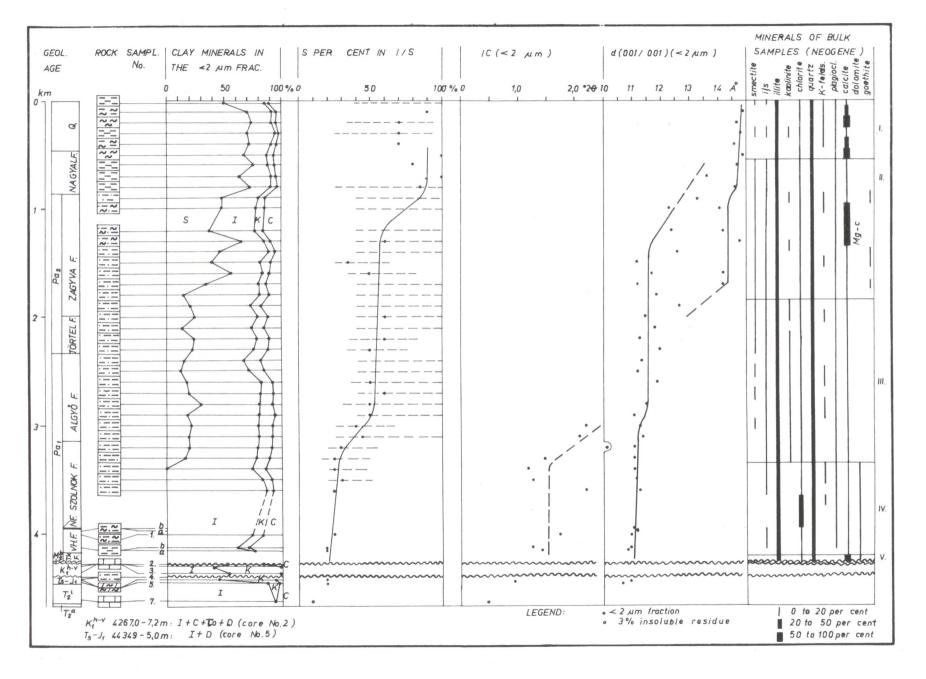


Fig. 1. Location map.

1 - partial depression of the Pannonian Basin with more than 3 km sediment thickness; 2 - areas not covered by Neogene sediments; 3 - site of the bore-hole Doboz I.

Partial depressions of the Pannonian Basin: L - Little Hungarian Plain, DR - Dráva Basin, J - Jászság, DE - Derecske Depression, M - Makó Depression, B - Békés Depression (according to Szalay & Koncz 1981).



d(001/001): d-position of the maximal intensity of the first order basal reflection, in units (see Fig. 3).

The semi-quantitative composition of the  $<2 \,\mu m$  fraction was determined by multiplying the intensities of the principal basal reflections by suitable factors. In Fig. 2 the quantity of pure smectites is shown together with highly expanded mixed-layers as layers (S and I, respectively).

Bulk composition of the samples was also determined by X-ray methods using powder specimens.

#### Results

# Variation in the bulk mineralogical composition

The bulk mineralogical composition of the pelitic rocks is remarkably constant in the whole sequence. Still it is possible to establish a mineralogical zonation on the basis of the appearance and disappearance of some minerals (e.g. kaolinite, dolomite, goethite, see Fig. 2). Calcite is permanently present but its amount varies in wide ranges.

# Variation of the clay mineralogy of the $< 2 \mu m$ fraction

The main feature in the variation of the composition of the clay-size fraction is the decrease of smectite-related phases with the increasing burial depth. The amount of illite and related phases increases with the depth. There is no appreciable variation in the kaolinite and chlorite contents which are permanently low.

# Variation in the composition of mixed-layer illite/smectites

The proportion of the smectitic component decreases with increasing depth. In a wide depth interval the composition of the individual samples is extremely variable and can not be characterized by a single value of the smectite proportion. The lower part of the transition zone can be characterized by suden decrease of the IC values of the illite-like minerals. The maximal intensity of the first basal reflection is at 15 in the uppermost  $600 \ m$  and at 11 to 12 in the lower part of the sequence. The variable smectite contents are expressed by the simultaneous appearance of two maxima in the upper part of the transition zone.

### Clay minerals in the Mesozoic basement rocks

The dominant clay mineral in the Triassic is illite. Upper Triassic to Lower Jurassic terrestrial clastic sediments and Lower Cretaceous limestones contain higher amounts of kaolinite. In voids of brecciated rocks and on the surface of fracture planes dickite and a tosudite-like mixed-layer mineral with ordered layer sequence and dioctahedral character were found (see Fig. 2).

Fig. 2. Variation of the mineralogical composition in bore-hole

Abbreviations of clay minerals in the  $< 2\mu m$  fraction: S - smectite and mixed-layer illite/smectite (smectite proportion 40 %), I - illite and mixed-layer illite/smectite (smectite portion 0 to 40 %), K - kaolinite, C - chlorite, To - tosudite-like mixed layer mineral with regular layer sequence, D - dickite, "S % in I/S" - proportion of expandable layers in mixed-layer illite/smectite, IC - illite crystallinity (see Methods of investigation).

### Discussion

Causes of vertical variations of the mineralogical composition of siltstones in the bore-hole Doboz I

The Neogene sequence represents a continuous evolution of the sedimentary environments from deep marine to alluvial (Gajdos et al. 1983). Some of the vertical variations in the bulk mineralogical composition obviously can be explained by the variation of sedimentary environments such as

- enrichment of calcite and Mg-calcite in lacustrine sediments, (in mineral zone II of the bulk samples, see Fig. 2);
- appearance of kaolinite in deltaic and near-shore facies (zone III);
- occurrence of goethite in continental clays (zone II).

Weathering may have influenced clay minerals in sediments of continental facies in zones I and II (0 to 1.7 km) and may have contributed to the inhomogeneous composition of mixed-layers in these zones. With the exception of this effect, there is no direct relationship between the bulk mineralogy of the samples and the composition of the m fraction as it was proposed by Kübler (1984) in connection with the interpretation of a Gulf Coast profile (see Ch. III.3, comments to Figs . 50 and 51). In the case of the Doboz I profile the disappearance of smectites seems to be controlled entirely by processes of diagenesis according to the classical model of the Gulf Coast Eocene (Burst 1969).

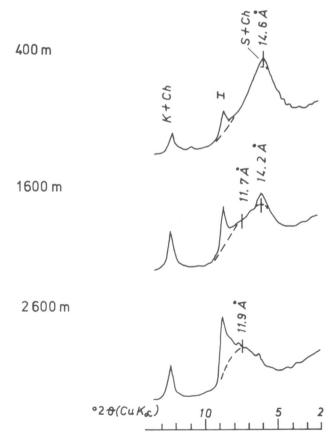


Fig. 3. Typical X-ray patterns of mixed-layer illite/smectites from different depths of the Neogene sequence (untreated specimens,  $< 2\mu m$  fraction). Note the variation of d(001/001) values and broadening of the reflection.

Abbreviations see Fig. 2.

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Comparison of bore-hole Doboz I with other areas of the Pannonian Basin and Vienna Basin

The results of the mineralogical investigation of 14 bore-holes representing various deep partial depressions of the Pannonian Basin were summarized by Viczián (1985). There is a good agreement with these sequences in respect of the composition of the m fraction and the general tendency of the diagenetic transformation of mixed-layers. There are, however, individual variations in the depth and extension of the transformation zone depending presumably on the thermal history of sediments.

Similar results were obtained also from a deep Neogene sequence of the Vienna Basin (Kurzweil & Johns 1981). The comparison with the Slovak part of the Vienna and Pannonian Basins is discussed elsewhere (see Franců et al. 1990).

# Comparison of the Molasse Basin with the Pannonian Basin

The clay mineralogy of the Swiss part of the Molasse Basin was summarized by Monnier (1982) on the basis of the analysis of 15 bore-holes. The mineral composition of the m fraction is very similar to that what was found in the Neogene of the Pannonian and Vienna Basins the only exception being the absence of kaolinite in the western part of the Molasse Basin. In some of the bore-holes the transformation of the expandable mixed-layer clay minerals occurred already at a much shallower depth  $(1.0 \ km)$  or less) than in the Pannonian and Vienna Basins. In these cases Monnier assumed the erosion of a considerable mass of sediments from the upper part of the original sequence.

In the Bavarian part of the Molasse Basin investigations were carried out in an area near Augsburg (bore-holes Scherstetten 1, Aibach, Krumbach and Marktwald, see Viczián 1984). The variation of the proportion of the smectitic component resembles the first step of transformation in the bore-hole Doboz I. The transformation takes place in the bore-hole Doboz I at a depth of approximately 1.0 km whereas in the Scherstetten profile already at about 400 metres. Lemcke (1974) assumed the erosion of about 400 to 500 metres from the upper part of the sequence of Scherstetten I. Considering this additional thickness of the sequence, the diagenetic character of the transformation seems to be acceptable. Apparently the subsidence was not deep enough to reach the lower step of transformation (drop of the smectite proportion to 20 %).

There is some similarity in appearance of correnstite- or tosudite-like phases in deeper zones both in the bore-hole Doboz I and the Swiss Molasse Basin. The investigation of the exact mineralogical nature of these phases, however, seems to be necessary in both areas.

### **Conclusions**

The clay mineral composition of Tertiary sediments of the Molasse, Vienna and Pannonian Basins is very similar. The common characteristics are:

- detrital origin;
- independence of depositional environments
- diagenetic transformation of illite/smectites.

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### References

Burst J. F., 1969: Diagenesis of Gulf Coast clayey sediments and its possible relation to petroleum migration. *Bull. AAPG*, 53, 73 - 93.

Franců J., Viczián I. & Šucha V., 1990a: Illite-smectite expandability as diagenetic temperature indicator in the Neogene basins of the Carpathian and Pannonian region. 11th Conf. Clay Miner., Petr., České Budějovice 1990, Abstracts.

Franců J., Müller P., Šucha V. & Zatkalíková V., 1990b: Organic matter and clay minerals as indicators of thermal history in the Transcarpathian Depression (East Slovakian Neogene Basin) and the Vienna Basin. Geol. Zbor. Geol. carpath. (Bratislava), 41, 5, 535-546.

Gajdos I., Papp S., Somfai A. & Völgyi L., 1983: Lithostratigraphic units of Pannonian (s. l.) formations of the Hungarian Plain. Földt. Int. Alkalmi Kiadványa. (Occasional Publ. of the Hung. Geol. Inst.). MÁFI, Budapest, 1 - 70.

Jámbor Á. & Kárpáti-Radó D., 1984: Unpublished reports, Budapest. Johns W. D. & Kurzweil H., 1979: Quantitative estimation of illite-smectite mixed phases formed during burial diagenesis. *Tschermaks Mineral. Petrogr. Mitt TMPM*, 26, 203 - 215.

Kilényi É., Kröll A., Obernauer D., Šefara P., Szabó Z. & Wessely G., 1991: Pre-Tertiary basement contour map of the Carpathian Basin beneath Austria, Czechoslovakia and Hungary. *Geophys. Trans*act., 36 1 - 2, 15 - 36.

Kurzweil H. & Johns W. D., 1981: Diagenesis of Tertiary marlstones in the Vienna Basin. Tschermaks Mineral. Petrogr. Mitt. TPMP, 29, 2, 103 - 125.

Kübler B., 1964: Les argiles indicateurs de métamorphisme. *Rev. Inst. franc. Pétrole*, 19, 1093 - 1112.

Kübler B., 1984: Les indicateurs des transformations physiques et chimiques dans la diagénse. Température et calorimétrie. In: Lagache M. (Ed.): Thermométrie et barométrie géologiques 2. Ch. 14, Soc. Franc. Miner. Crist., Paris.

Lemcke K., 1974: Vertikalbewegungen des vormesozoischen Sockels im nördlichen Alpenvorland vom Perm bis zur Gegenwart. Eclogae geol. Helv., 67, 1, 121 - 133.

Monnier f., 1982: Thermal diagenesis in the Swiss molasse basin: implications for oil generation. *Canad. J. Earth Sci.*, 19, 328 - 342.

Reynolds R. C. & Hower J., 1970: The nature of interlayering in mixedlayer illite-montmorillonites. Clays and Clay Miner, 18, 25 - 36.

Srodon J., 1980: Precise identification of illite/smectite interstratification by X-ray powder diffraction. Clays and Clay Miner., 28, 6, 401 - 441.

Szalay A. & Koncz I., 1981: Genese und Migration der Kohlenwasserstoffe in den Neogen-Depressionen des Pannonischen Beckens Ungarns. Zschr. angew. Geol., 27, 6, 266 - 272.

Viczián I., 1985: Clay mineralogy of pelitic sediments of the South German Molasse Basin. 9th Conf. Clay Miner. Petr., Zvolen 1982. Charles Univ., 101 - 105, Prague.

Viczián I., 1985: Diagenetic transformation of mixed-layer illite/smectites deep zones of the Pannonian Basin (Hungary) 5th Meeting of the European Clay Groups, Prague 1983. Charles Univ., 135 - 140, Prague.

Viczián I., 1986: Clay mineralogy of a thick sedimentary sequence in SE part of the Pannonian Basin (Hungary). 10th Conf. Clay Miner. Petr., Ostrava 1986. Abstracts.