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TYPICAL CLAY MINERAL ASSOCIATIONS FROM GEOLOGICAL FORMATIONS IN HUNGARY A REVIEW OF RECENT INVESTIGATIONS

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Abstract: In this review some typical formations are listed according to tectonic units and in a stratigraphic order. Results achieved in the last years are specially emphasized. In the Mecsek Mts Middle Triassic marly dolomites contain corrensite. There is only illite and some illite/smectite depending on diagenetic grade in the Liassic limestones. In the Toarcian manganese ores at Úrkút (Trandanubian Range) typical chemical sedimentation and diagenesis produced an assemblage of celadonite and nontronite. In the Lower Cretaceous sandstones and marls of Gerecse Mts a corrensite-rich association could be derived from ophiolitic sources. In thick clastic detrital sequences of Paleogene and Neogene basins mixed-layer illite/smectites display diagenetic transition toward illite depending on depth, temperature, rate of subsidence and subsequent tectonism. In Pliocene and Quaternary basaltic craters of W Transdanubia bentonites consist of iron-rich beidellites. Red clays contain disordered kaolinite-bearing and a smectite(+vermiculite)-bearing units. At Visonta high charge beidellites were found. Paleontological and mineralogical records agree well in the fresh and paleosol layers of loess. In the clay fraction of Recent soils, chernozems are characterized by illite, brown forest soils, humic gleys and salt affected soils by more smectite-rich associations.

Key words: Hungary, diagenesis, paleosols, loess, carbonate rocks, claystones, red clays, clay minerals.

Introduction

Clay minerals are important and characteristic components of most geological formations of Hungary. Systematic investigations carried out in the last 30–40 years have shown that sedimentary and volcanogenic formations bear typical clay minerals depending on their conditions of formation and secondary modifications such as diagenesis or hydrothermal alteration. The actual state of knowledge on clay minerals in Hungarian sedimentary rocks was summarized by the present author several times (Viczián 1975, 1987, 1995). For details of the classification of clay minerals in Hungarian rocks we refer to these former review articles. The present review mainly considers the results of the last few years. The most important new data will be arranged according to regional geological units, in a stratigraphic order. The geological units and localities mentioned in the paper are shown on Fig. 1. The present paper does not review the clay mineral researches carried out on incipient (or very low-grade) metamorphic formations of Hungary. In this respect the author refers to the recent overviews of Lelkes-Felvári et al. (1996) and Árkai (2001). Because of the wealth of the new investigations only very brief descriptions will be given.

Materials and methods

In most cases clay mineralogical analysis was carried out as part of a normal petrographic analysis of rock samples collected in connection with geological mapping or other geological research projects. The most frequently used analytical methods were X-ray diffraction, thermal and chemical analysis. In some cases petrographic and electron microscopy, IR spectroscopy and determination of K/Ar ages were also used.

Paleozoic and Mesozoic formations

Mecsek Mts (southern Transdanubia)

Lower Carboniferous granitoid rocks as possible candidates for repository site of low- to medium-level radioactive waste in the Eastern Mecsek area are intersected by various types of fissures filled by carbonate and clay minerals (*vermiculite-like Mg>Ca montmorillonite, palygorskite* and *illite*). The minerals were formed in a Mg-rich hydrothermal environment (Kovács-Pálffy et al. 2000a). K/Ar ages measured on *illites* indicate that formation of the fissure filling took place during the Cretaceous.

In the last years a formerly virtually unknown Upper Permian formation, the Boda Claystone Fm attracted much interest as a possible repository site of high-level radioactive waste (Árkai et al. 2000). Its main clay minerals proved to be much *illite-muscovite* and less *chlorite*. The peculiarity of the formation is the low *quartz* and extremely high authigenic *albite* and high *hematite* contents, which are the result of sedimentation in highly alkaline and oxidative lakes under semi-arid to arid climatic conditions. The degree of diagenesis is high, close to the boundary of the anchizone due to thermal effects culminating in the Lower Jurassic period. Epigenetic alteration in fractured rocks may have produced minor amounts of expandable phases. Hydraulic properties

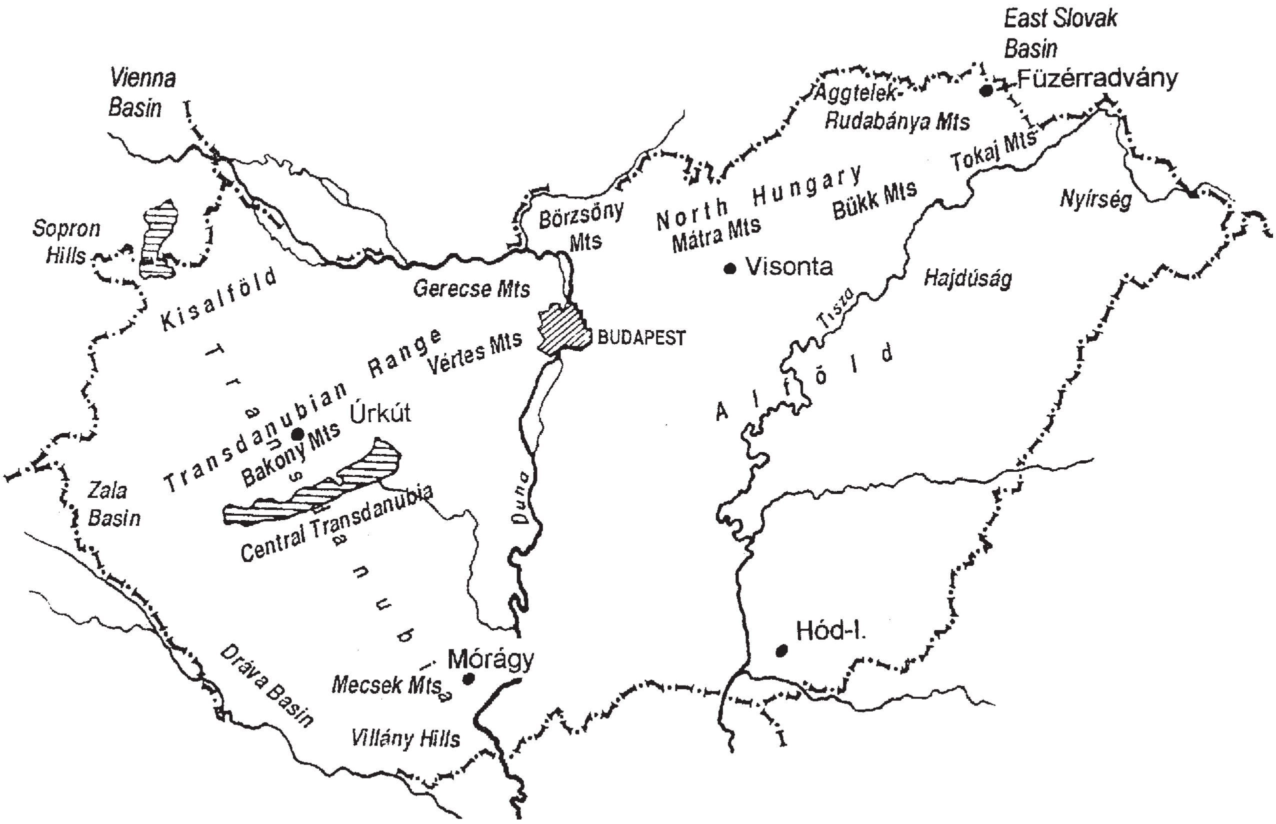

Fig. 1. Sketch map showing the geographical names occurring in the paper (after Császár 1997, modified).

are highly affected by the quality of clay minerals (Kesserű 1998).

Former mineralogical investigations (Viczián 1992, 1993a) have shown that Middle Triassic rocks of the Mecsek Mts. bear clay minerals similar to analogous formations in the German Triassic Basin. Formations comparable to the Röt Member contain *Mg-chlorite* and *corrensite* and formations similar to the Muschelkalk contain only *illite* and authigenic *K-feldspar*. Recently the study of another bore hole profile confirmed the repartition of the clay minerals and the existence of the mineralogical correlation between the Mecsek Mts and the German-type Triassic (Viczián 2000a).

Substantial development has been achieved in the study of Lower and Middle Jurassic marly carbonate rocks overlying the Liassic black coal deposits. In barren rocks of the coal highly diagenetic *illite* and *kaolinite* are the dominant phases. Grading upwards *kaolinite* decreases and the expandability of *illite/smectite* increases as a combined effect of paleogeographical conditions of deposition in a deepening sea and decreasing overburden pressure during postdepositional alteration. An excellent analysis of the details of the paleogeographical factors, cyclic deposition and interrelation with organic matter was given by Raucsik (1999) and Raucsik & Merényi (2000).

Bakony and Gerecse Mts (Transdanubian Range)

In the year 2000 a monographic work has been published by Polgári et al. discussing the conditions of formation of the Toarcian manganese ores at Úrkút, Southern Bakony Mts. From the point of view of clay mineralogy the most important question is the formation of the so-called Radiolarian marl member in which typical chemical sedimentation and diagenesis produced an assemblage of *celadonite* and *nontronite*. These minerals were correctly identified for the first time by Kaeding et al. (1983). The *celadonite* was formerly regarded as *glauconite*. X-ray and IR analyses carried out by Pápay in 1985 are published in the book of Polgári et al. (2000) proving the real celadonitic nature of this mineral. Samples of the Úrkút material were also investigated by B. A. Sakharov, who has found *celadonite-1M* with 20 % nontronite type interlayers and *nontronite* (published by Varentsov et al. 1988).

Recently Weiszburg and his students were able to make exact chemical determination of the Úrkút *celadonite* on carefully separated samples. The ordered structure was proved by Dódony by electron diffraction (Mizák et al. 2000; Weiszburg et al. 2001). K/Ar ages indicate the diagenetic formation of the *celadonite* in the Middle to Upper Jurassic (Polgári et al. 2000).

In the Lower Cretaceous marl and sandstone series of the Gerecse Mts substantial components are *mixed-layer chlorite/smectite/vermiculite* minerals (Viczián & Kovács-Pálffy 1997). The mineralogy of these formations has interesting geotectonic implications. In accordance with petrographic analyses (Árgyelán 1996) these minerals may be attributed to redeposited ophiolitic material. In the Bersek Marl and Lábatlan Sandstone Fms *regular interstratifications (corrensites)* occur. In the overlying conglomerate and marl formations, however, only *irregular interstratifications* may be found. *Regular interstratifications* point to heating up to slightly above 100 °C, which seems to be restricted only to the two formations mentioned.

Tertiary basins

Until the first half of the '90s, in connection with the hydrocarbon prospection, extensive studies were carried out on the diagenetic transition of *mixed-layer illite/smectites* towards *illite* in clastic detrital sediments of the Neogene Pannonian Basin. The smectite proportion in I/S proved to be an important diagenetic indicator. Summarizing publications have shown the effect of depth, temperature, rate of subsidence and subsequent tectonism as well as the importance of source material on the transition. Correlation with the vitrinite reflection was established. Comparison with the East Slovak and Vienna Basins was done (Hámor-Vidó & Viczián 1993; Franců et al. 1993; Viczián 1994; Mátyás 1994; Tanács & Viczián 1995; Hillier et al. 1995).

The studies were later extended to the thick pelitic sequences of the North Hungarian Paleogene Basin (Viczián 1993b, 1996, 1997), however, a detailed publication on this subject is still missing.

Recently an excellent analysis of *illite/smectite* diagenesis was carried out on the Pannonian sequence of the deepest Hungarian hydrocarbon exploration well Hód-I in a M.Sc. Thesis of the Eötvös University, Budapest (Judik 2001).

In Hungary, clay mineralogy began with the study of the various products of alteration of the Tertiary volcanic rocks. One of those classical minerals was *"sarospatite", a mixed-layer illite/smectite* from the locality Füzérradvány, Tokaj Mts (Viczián 1997, 2000b). After a period of stagnation in the study of hydrothermal clays, some new progress seems to be starting. Molnár (2000) developed a new model of the hydrothermal alteration in the Tokaj Mts based mainly on fluid inclusion studies. New mineralogical results of the study of *bentonite* deposits were included into the Ph.D. Thesis of Kovács-Pálffy (1998).

Pliocene and Quaternary

So called basaltic bentonites together with sediments rich in Algal remnants were detected in lacustrine deposits formed in crater lakes of Pliocene basaltic volcanoes in western Transdanubia (Viczián et al. 1992). Bentonites may contain up to 80–90 % *smectite* phases. The mineralogical nature of the *smectite* has been determined as *iron-rich beidellite* (Juhász 1989). Partial data on the mineralogical properties of this *Fe-rich beidellite* were published by Barna & Földvári (1992, 1996) and by Földvári (2000), however, a detailed mineralogical analysis is still missing.

In SE Transdanubia extensive areas are covered by the red clay formation of Upper Pliocene–Lower Pleistocene age (Tengelic Fm). *Disordered kaolinites* found in red clay deposits on the top of limestone surfaces in the Villány Hills were studied by Bidló (1985). Recently by means of mineralogical and geomorphological investigations the formation was subdivided into a lower, *kaolinite*-rich part, formed in a warm, humid climate and an upper, *smectite*-rich part, which may be attributed to climatic conditions at the boundary of savannah and arid zones (Schweitzer & Szöőr 1997). On the surface of the Mórágy Granite, *biotites* may have been weathered to *mixed-layer biotite/vermiculites* during this latter period of red clay formation (Kovács-Pálffy et al. 2000b).

Another Quaternary red clay formation is found in the southern foothills of the Mátra Mts. *Smectites* in these red clays found in the open pit works at Visonta have been determined by various treatments and Green-Kelly test as *high charge beidellites*. The underlying grey clays are characterized by a mixture of *low charge beidellite* and *montmorillonite* (Németh et al. 1999). It is supposed that the material of the red clay is derived from the surface of the volcanic rocks of the Mátra Mts, where similar red clays rich in *smectites* were found (Berényi-Üveges et al. 2000).

The mineralogical composition of Quaternary loess and its paleosol horizons has been the subject of many former studies. In a recent study mineralogy, geochemistry and paleontology were compared in a loess profile in SE-Transdanubia. Unweathered loess contains well crystallized detrital clay minerals, mainly *illite* and *chlorite*. In paleosol horizons clay material is enriched due to dissolution of carbonates and is dominated by *smectite* and, occasionally, *vermiculite*. Conclusions relating to paleoenvironmental conditions obtained by various methods are in good agreement (Hum & Fényes 1995; Hum 2000).

Recent soils

The clay mineral associations of the basic soil types of Hungary were determined by Varjú & Stefanovits (1979). Clay mineralogy of salt-affected and meadow soils was studied by Kapoor et al. (1986). According to these investigations, in the clay fraction of chernozems, *illite*-dominated associations, while in brown forest soils, humic gleys and salt affected soils, assemblages enriched in *smectites* are typical. Salt affected soils may contain high amounts of *amorphous silica* (Szabolcs & Szendrei 1980). In Holocene alluvial sediments of the SE Alföld area Kalmár et al. (1997) differentiated between inherited *smectites* and *smectites* of pedogenic origin. A map of the repartition of clay mineral associations in Hungarian soils was published by Stefanovits & Dombóvári (1985) and Stefanovits (1985).

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