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Hungarian contribution to the mineralogy and geology of clays

Commemorating the 50th anniversary of the Hungarian Clay Minerals Group founded in 1960

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Magyar eredmények az agyagásványtan és -földtan terén

Az 1960-ban alapított MFT Agyagásványtani Szakosztály 50. évfordulója alkalmából

Összefoglalás

A cikk a magyar földtudományi agyagkutatás főbb eredményeit foglalja össze témák szerint csoportosítva.

Általános művek. A Magyarhoni Földtani Társulat Agyagásványtani Szakosztályának alapítója, NEMECZ írta az első magyar általános szakkönyvet az agyagásványokról. Tágabb értelemben BÁRDOSSY alapvető műveit a bauxitokról szintén az agyagkutatás körébe sorolhatjuk. A talajok agyagásványaival STEFANOVITS, SZENDREI és NEMECZ, újabban pedig NAGY és KÓNYA könyvei foglalkoztak.

Egyes agyagásványok vizsgálata. A Füzérradványon bányászott híres illites ásványt MAEGDEFRAU és HOFMANN írta le még 1937-ben. Ezt nevezhetjük „a világ második illitjének" (az illinoisi után). Az ásvány további kutatásáról NEMECZ és VARJÚ számolt be. A vastartalmú agyagásványok, mint a szeladonit, glaukonit és a „Fe-dús montmorillonit csoport" kémiai rendszerezését lényegesen előrevitték WEISZBURG és TÓTH E. munkái. ERDÉLYI részletesen tanulmányozott számos szerpentinásványt. Újabban DÓDONY végzett szerkezeti vizsgálatokat szerpentinásványokon (pl. antigorit, poligonális szerpentin).

Agyagásványok meghatározási módszerei. Új, és ma is használható röntgendiffrakciós mennyiségi határozási módszert NÁRAY-SZABÓ vezetett be. A PAULIK testvérpár termikus meghatározó készüléket fejlesztett ki, amelyet „derivatográf"-nak neveztek. Az első termikus határozó atlaszt az agyagásványokra vonatkozólag FÖLDVÁRI-VOGL állította össze. A termikus módszert jelenleg is magas szinten alkalmazza FÖLDVÁRI.

Talajok és üledékek. Magyarország talaj-agyagásvány térképét STEFANOVITS és DOMBÓVÁRI készítették el. BIDLÓ részletesen vizsgálta a vörös agyagokat, amelyek legtöbbször fosszilis talajokból származnak. Bazaltkráterekben fosszilis bentonitos tavi üledékeket ismert fel SOLTI. Ezek anyagát FÖLDVÁRI, Zs. BARNA és mások vizsgálták. A Kárpát-medence főleg riolitos eredetű bentonittelepeit KOVÁCS-PÁLFFY vizsgálja rendszeresen.

Kőzetrétegtani alkalmazások. A magyarországi üledékes formációkat agyagásvány-tartalmuk szempontjából VICZIÁN jellemezte. Jelenleg ezt a kutatási irányt RAUCSIK, VARGA A. és SZAKMÁNY folytatják tovább. VICZIÁN, MÁTYÁS J. és mások a szmektit–illit diagenetikus átalakulást alkalmazták a szénhidrogén-prognózisban. Az igen kisfokú metamorfózis rétegszilikátjainak fő kutatója ÁRKAI, munkatársai TÓTH M., JUDIK és mások. Ő vezette be a klorit „kristályossági" fokot (Árkai-index), munkatársaival vizsgálta az indexásványokat, a kőzetek szövetét stb. Kutatásaikat Magyarország határain túlra, a svájci Alpokra és DK-Európára is kiterjesztették.

A hidrotermális elváltozási öveket a Tokaji-hegységben elsőnek SZÉKYNÉ FUX ismerte fel. Később NEMECZ és VARJÚ, újabban MOLNÁR részletesen vizsgálták az elváltozással létrejött agyagásványokat. A nemesagyag-telepek kutatásával évtizedekig MÁTYÁS E. és ZELENKA foglalkozott, alkalmazhatóságukat és kolloid tulajdonságaikat JUHÁSZ, SZÁNTÓ és tanítványaik vizsgálták. A nehézfémek által okozott környezeti szennyezés modellezését NÉMETH végzi. SZABÓ egyik fő szakértője az agyagos szigetelőréteg kialakításának városi hulladéklerakók körül. Agyagos mellékkőzetben kívánják elhelyezni a nagy intenzitású radioaktív hulladékot a Mecsek hegységben.

Tárgyszavak: agyagásványok, szedimentológia, talajtan, igen kisfokú metamorfózis, környezeti ásványtan

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Abstract

The main results of the Hungarian clay research are reviewed in the field of earth sciences in a thematic order.

A *general textbook* on clay minerals was published by the founder of the Clay Minerals Group, Professor NEMECZ. In a broader sense, the important books on bauxites by BÁRDOSSY belong to the field of clay research. Soil clay minerals were discussed in the textbooks of STEFANOVITS, SZENDREI and NEMECZ, and recently by NAGY and KÓNYA.

Specific clay minerals. The famous illite mineral of Füzérradvány — the "second illite in the world" (after the illite of

Illinois) — was described by MAEGDEFRAU & HOFMANN as early as 1937 and later reviewed by NEMECZ & VARJÚ. WEISZBURG and E. TÓTH contributed much to the chemical systematics of celadonite, glauconite and the "Fe-rich montmorillonite" series. Earlier, serpentine minerals were extensively studied by ERDÉLYI and in recent years by DÓDONY (structural analyses of antigorite and polygonal serpentine).

Determinative methods in clay mineralogy. A method of quantitative analysis by X-ray diffraction was introduced by NÁRAY-SZABÓ. A special apparatus for thermal analysis — called the "Derivatograph" —was developed by PAULIK et al. An atlas of thermal analysis was first compiled by FÖLDVÁRI-VOGL. The application of this method was later continued at a higher level by FÖLDVÁRI.

Soils and sediments. STEFANOVITS and DOMBÓVÁRI published a map of clay minerals in Hungarian soils. Red clays, mostly relict fossil soils were studied by BIDLÓ. FEKETE published a book on recent tropical soils. Fossil bentonitic lake sediments of basaltic craters were recognised by SOLTI and analysed by FÖLDVÁRI, ZS. BARNA and others. KOVÁCS-PÁLFFY focused his systematic studies on rhyolite-related bentonite deposits of the Carpathian Basin.

Lithostratigraphic applications. Clay minerals of Hungarian sedimentary formations were systematically characterised by VICZIÁN. Today this line has been continued by RAUCSIK, A. VARGA and SZAKMÁNY. The diagenetic transformation of smectite to illite was widely applied for CH prospecting by VICZIÁN, J. MÁTYÁS and others. Layer silicates of the very lowgrade metamorphic stage have been studied by ÁRKAI and his co-workers M. TÓTH, JUDIK etc. He introduced the parameter "chlorite crystallinity" (Árkai index), studied index minerals, and also applied textural analysis. His studies extended to Hungary, the Swiss Alps, and to South East Europe.

Zones of hydrothermal alteration in the Tokaj Mts were first recognised by SZÉKY-FUX and later intensively studied by NEMECZ and VARJÚ and recently by MOLNÁR. Clay deposits of economic value were studied for several decades by E. MÁTYÁS and ZELENKA, their application and colloidal properties were analysed by JUHÁSZ, SZÁNTÓ and their students. Model experiments of environmental pollution caused by heavy metals were carried out by NÉMETH. Clay liners for communal waste depositories were developed by SZABÓ. Pelitic host rocks for high intensity radioactive waste depositories have been studied in Mecsek Mts.

Keywords: clay mineralogy, sedimentology, soil science, very low-grade metamorphism, environmental mineralogy

Introduction

The Hungarian Clay Minerals Group was founded in 1960. In this review, on the occasion of the 50th anniversary of this Group, the main results of Hungarian clay research in the field of earth sciences are presented. With respect to the references in given here, only a few publications have been selected. The main considerations were that the work is typical for the author or the subject. This historical account follows the example of the review made by VOGL (1982) on the development of the analytical methods during the first 20 years of the Clay Group (1960–1980). A valuable source of data regarding the activity in the early periods can be found in the bibliography compiled by SZENDREI (1979). This bibliography is available in manuscript form and contains nearly 500 items. A similar bibliographic compilation, with brief historical notes, was written by VICZIÁN in 1999. Recently valuable bibliographical and biographical data were collected by SZENDREI (2010) for the purposes of a home page of the Hungarian Geological Society.

Books on Clay Minerals

A general textbook on clay minerals was published by the founder of the Clay Mineral Group, Professor NEMECZ (1981). In a broader sense the important books of BÁRDOSSY (1982, BÁRDOSSY & ALEVA 1990) on bauxites and the bauxite sedimentological studies of MINDSZENTY (1999) belong to the clay research. Soil clay minerals were discussed in the textbooks of STEFANOVITS (1981), SZENDREI (1994) and NEMECZ (2006). FEKETE (1988) published a book on tropical soils. It is not intended here to review the colloid chemistry of clay minerals in detail. Only a few university courses and textbooks on colloids which are closely related to clay minerals are mentioned. Such are the university courses by JUHÁSZ (1995–1996), the book by SZÁNTÓ (1987) and the recently published work by M. NAGY & KÓNYA (2009). JUHÁSZ & OPOCZKY (1982, 1990) have become international-renowned specialists on the mechanochemistry of silicate minerals.

Specific clay minerals

The famous illite mineral of the locality of Füzérradvány, — also called sarospatakite and more recently "Zempleni illite" — can be regarded as the "second illite of the world" because it was described by MAEGDEFRAU & HOFMANN as early as 1937 (after the Illinois illite: see VICZIÁN 2000). Later the mineral was reviewed by NEMECZ & VARJÚ (1970) and was identified as mixed-layer illite/smectite-1M. Hungarian investigations on Füzérradvány illite in many respects preceded the corresponding American studies but whatever, they are in accordance with them (see: VICZIÁN 1996, 1997).

ERDÉLYI et al. (1957a, b) described a high temperature hydrothermal illite-2M variety which he called "hydro-muscovite".

From the Mecsek uranium deposits Cr-bearing mica was identified by KISS (1960) and further specified by SZTRÓKAY & PUSKÁS (GÁL-SÓLYMOS et al. 1994).

TOKODY (1962) described a smectite variety as an alteration product of andesite and he called it "mauritzite". This mineral was later characterized by KÁKAY SZABÓ (1983) and identified as saponite with a high iron content by WEISZBURG et al. (1993).

J. BARNA (1983) described it as a natural — clay-humic substance complex originating from brown coals. It was especially the presumed hydrous varieties in the serpentine group that were extensively studied by ERDÉLYI et al. (1957a, b) and ERDÉLYI & VENIALE (1970).

In a later period PAPP (1993) studied the intergrowth of serpentine minerals and DÓDONY (1997), DÓDONY et al. (2001) published electron microscopic structural analyses of the serpentine varieties antigorite and polygonal serpentine.

The regular mixed-layer mineral "allevardite" (i.e. rectorite) was recognised by NEMECZ et al. (1963) from hydrothermal deposits at Király-hegy in the Tokaj Mts. Another regular mixed-layer clay mineral — corrensite — was first described from Hungary by VICZIÁN (1993),); its occurrence was in the German-type Triassic formations of the Mecsek Mts.

Classification and nomenclature

Immediately before the formation of the Hungarian Clay Group a proposition on classification was submitted jointly by Hungarian specialists for the AIPEA and published by FÖLDVÁRI-VOGL (1958). Already this classification contained the term "couches intercalées" (approximately identical with "interstratified") and "sárospatakite" was included into this group.

WEISZBURG et al. (2004, 2008) contributed much to the chemical systematics of Fe-bearing members of the mica and smectite groups (e.g. celadonite, glauconite, and with recognition of the "Fe-rich montmorillonite" series).

Determinative methods in clay mineralogy

A so-called "direct" method of quantitative analysis by X-ray diffraction was developed by NÁRAY-SZABÓ & PÉTER (1967). This was improved later by several authors including BÁRDOSSY et al. (1980), RISCHÁK (KOMKOV et al. 1989, SIDORENKO et al. 1992) and SAJÓ (FERET et al. 1997). A special apparatus for thermal analysis — called the "Derivatograph" — was constructed by PAULIK et al.(1986). The apparatus was manufactured in Hungary for several years. An early Atlas of thermal analysis was compiled by FÖLDVÁRI-VOGL (1958).

Later this method was developed to a high standard on the international level by FÖLDVÁRI (1986, 1991). SZÖŐR & BOHÁTKA (1985) combined derivatographic analysis with the mass spectrometry of escaping gases. SZENDREI (2001) applied the analysis of specially prepared thin sections to describe the micro-morphology and genetic processes of soils. The radioactive age of clay substances is determined mainly by the K-Ar method devised by BALOGH (ÁRKAI et al. 1995, 2003), PÉCSKAY et al. (2005, 2006) and their co-workers.

Soils

STEFANOVITS & DOMBÓVÁRINÉ (1985) published a map of clay minerals in Hungarian soils. Typical Hungarian saline soils were studied by SZENDREI (1985). Red clays, which proved to be mostly relicts or redeposited palaeosoils, were studied by BIDLÓ (1980, 1985), FEKETE & STEFANOVITS (1998) and BERÉNYI ÜVEGES et al. (2003). SCHWEITZER & SZÖŐR (1997) correlated mineralogy and the age of red clays. NEMECZ & CSIKÓS-HARTYÁNI (1995) carried out detailed mineralogical analyses on finely-separated grain fractions of soils.

Lacustrine clay sediments, bentonites

Sediments from Lake Balaton were studied by a research team led by CSERNY (2002). Specific fossil lake sediments of the craters of basalt volcanoes were recognised by SOLTI and analysed by Zs. BARNA & FÖLDVÁRI (1996). JUHÁSZ (1989) identified the principal component of the basaltic bentonites as Fe-bearing beidellite. It was mainly rhyolite-related bentonite deposits of the Carpathian Basin that were systematically reviewed by KOVÁCS-PÁLFFY (1998). Fine details of the sedimentation conditions of a bentonite deposit were revealed using sequence stratigraphic and geochemical analysis (PÜSPÖKI et al. 2005, 2008).

Lithostratigraphic applications, diagenesis

The clay minerals of Hungarian sedimentary formations were systematically characterised by VICZIÁN (1995). This type of clay study was combined with geochemical research carried out by VARGA et al. (2007), RAUCSIK & VARGA (2008). The diagenetic transformation of smectite into illite was applied for hydrocarbon exploration by VICZIÁN (HÁMOR-VIDÓ & VICZIÁN 1993, TANÁCS & VICZIÁN 1995), J. MÁTYÁS (HILLIER et al. 1995) and others.

Phyllosilicates in very low grade metamorphic rocks

The very low grade metamorphic stage of transformation has been studied by ÁRKAI et al. (2002). Together with M. TÓTH he improved the "illite crystallinity" (Kübler index) method and introduced the characteristic parameter "chlorite crystallinity" (Árkai index), based on X-ray diffraction (ÁRKAI 1991, ÁRKAI et al. 1996). In addition to potassic white mica paragonite and margarite were also studied (LIVI et al. 2008). The X-ray results were compared with textural analysis using the TEM method.

In cooperation with P. Horváth, Judik etc., his studies extended to the anchimetamorphic formations of the Swiss Alps, also south-eastern territories of Europe (ÁRKAI et al. 1995, JUDIK et al. 2003).

Hydrothermal alteration of volcanic rocks

The classical studies of PÁLFY (1911) and INKEY (1906) found a correlation between propylitic alteration and gold mineralization in andesitic rocks. Propylitization is a large-scale alteration process in which chlorite forms in place of mafic silicates. SZÁDECZKY-KARDOSS (1958, 1960) drew attention to the uptake of volatiles by magma (transvaporisation), the crystallisation of volatile-rich magmas (formation of hypo-magmatic rocks) and alteration due to secondary processes (meta-magmatic rocks) which produce various clay minerals. In the Tokaj Mts, zones of *hydrothermal alteration* were first recognised by SZÉKY-FUX (1970) and later intensively studied by NEMECZ et al. (1963), NEMECZ & VARJÚ (1970). In the Velence Hills a deeper level of hydrothermal alteration was explored by the team of DARIDA-TICHY, FÖLDVÁRI, FARKAS, led by I. HORVÁTH (DARIDÁNÉ TICHY et al. 1984). Recently in both regions this line of research has been continued mainly by MOLNÁR (MOLNÁR et al. 1999, PÉCSKAY et al. 2005).

Economic and environmental geology

A comprehensive inventory of the economic clay deposits of the historical Hungary (i.e. the pre-1920 borders of Hungary) was prepared by KALECSINSZKY (1905). The kaolin, bentonite, illite, zeolite and siliceous earth deposits of the Tokaj Mts and other areas of the country were studied for several decades by E. MÁTYÁS (1966, 1974) and ZELENKA (1994, PÉCSKAY et al. 2005). A comprehensive review was compiled by VÉGHNÉ (1967).

Model experiments of *environmental pollution* by heavy metals were carried out by NÉMETH et al. (2005) and applied to soils by SIPOS (2006–2007). The interaction of high arsenic groundwater with the rock-forming minerals was studied by VARSÁNYI & Ó. KOVÁCS (2005).

Clay liners for communal *waste depositories* were developed by SZABÓ (CZURDA & SZABÓ ed. 1996). Pelitic host rocks and the fault gouge of the granitic host rock for radioactive waste depositories were studied in Mecsek Mts (FÖLDVÁRI 2006, R. VARGA et al. 2005).

Extraterrestrial applications

GUCSIK (GAVIN et al. 2010) has started to develop models for the formation of the recently discovered clay minerals on the planet Mars.

Conclusions

Three main periods can be distinguished in the history of the Hungarian clay mineral research.

In the first period which comprises roughly the 1960–70's, the main interest was focused to the study of hydrothermal alteration in volcanic regions, including the study of specific minerals and economic deposits.

In the second period, approximately in the 1980–90's, investigations concentrated to the study of sedimentary formations, including shales, pelitic rocks and bauxites. Special attention was given to diagenetic processes related to hydrocarbon genesis and very low grade metamorphism.

In the most recent period, on the turn of the 20th and 21st centuries the importance of young sediments such as soils and Quaternary deposits is growing. There is an increasing interest to environmental applications such as lining of radioactive waste depositories and heavy metal pollution. Thermal analysis was applied on high level in the former and in this period. There are instrumental analyses of specific mineral groups like serpentines and iron clay minerals of high international standard.

In general, the research was able to attain a reliable, medium to good level which matched the international standards. In a few fields, however, such as in the study of bauxites, very low grade metamorphism or in some mineralogical studies Hungarian researchers played a pioneering role in the general clay science.

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