



GEOLOGY AND K-AR GEOCHRONOLOGY OF ILLITE FROM THE CLAY DEPOSIT AT FÜZÉRRADVÁNY, TOKAJ MTS., HUNGARY

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ABSTRACT

Illite deposit of Füzérradvány is a part of a low sulphidation type epithermal system hosted by an Upper Miocene (Sarmatian) volcanoclastic and sedimentary sequence. Mineable high quality illite bodies were formed along siliceous veins due to interaction of hydrothermal fluids with permeable rock units. In order to get highly reliable K-Ar data for timing of the hydrothermal activity, careful sample preparation and duplicate analyses on illite samples in two independent laboratories (Institute of Nuclear Research, Hungarian Academy of Sciences and Okayama University of Science, Japan) were carried out. On the basis of the results, the most probable age of mineralization is 11.89 ± 0.3 Ma. This result is in agreement with previous data suggesting that hydrothermal mineralization exposed in the Tokaj Mts. took place during the Middle-Upper Miocene (Sarmatian) paroxysms of volcanism.

Key words: K-Ar dating, illite, Carpathians, Tokaj Mts., epithermal system

INTRODUCTION

The recognition and exploitation of the ceramic clay locality of Korom Hill at Füzérradvány (Tokaj Mts.) dates back to the first half of the 19th century. In the 30's of the 20th century the material became famous as one of the first known occurrences of micaceous clay minerals. The Füzérradvány illite was first described by Maegdefrau and Hofmann (1937) who compared their results on the "mica of Sárospatak" with those of a "sericite-like mineral" from Illinois, U. S. A., the material which the name "illite" was given by Grim et al. (1937) and found considerable similarities. Thus the "mica of Sárospatak" (i. e. Füzérradvány) has a very close relation to the birth of the widely used mineralogical term "illite". Since this time the material was subject of numerous investigations. By these studies the Füzérradvány "illite" became probably the most famous Hungarian clay mineral (Viczián 1997). Recently it has been used as standard material called "Zempleni illite". The identity of "Zempleni illite" with the Füzérradvány "illite" and the main results of its recent investigations were discussed by Viczián (1996).

The mineralogical properties of illite from the clay deposit at Füzérradvány (e.g. "Zempleni illite") have already been studied in detail (Nemecz and Varjú 1970, Patzkó and Szántó 1983, Środoń 1984, Dódony 1985, Ahn and Buseck 1990, Veblen et al. 1990, Reynolds 1992, Środoń et al. 1992), however, genesis of the deposit and especially age of mineralization has not been clarified in more detail. Mátyás (1972) established a generalized model for the processes leading to the formation of the deposit and a further impact on the studies of the area was the discovery of precious metal anomalies in relation to the deposit (Hartikainen et al. 1992). Detailed field surveying (Csongrádi and Zelenka 1995) has

clarified the relationships between precious metal enrichment and argillic alteration and new results of ore deposit modelling in the Tokaj Mts. (Molnár 1993, Molnár et al. 1999) provided background for re-evaluation of earlier data.

An important aspect of understanding of hydrothermal systems is the knowledge of age of mineralization and its relationship to volcanic processes on the local, as well as, regional scale. Because illite and other K-bearing minerals also occur in other hydrothermal systems of the Tokaj Mts., K-Ar dating appears to be useful tool for such studies. Previous studies by Środoń et al. (1992) suggested a Badenian age for the mineralization on the Korom Hill, however, this was not reasonable because of the geology of the area. This paper aims to summarize our knowledge about the clay deposit at Füzérradvány and provides an upgraded model for the mineralizing processes taking into account results of new geochronological data.

GEOLOGY AND CHARACTERISTICS OF THE CLAY DEPOSIT

The Korom Hill in the vicinity of Füzérradvány village is situated in the northeastern part of the Tokaj Mts. and 18 km northwest of the town of Sátoraljaújhely (Fig. 1). The Tokaj Mts. is a part of the Carpathian intermediate-acidic calc-alkaline volcanic belt of Neogene-Quaternary age (Pécskay et al. 1995) and locates in the turning area of the Western and Eastern Carpathians. The syn- to post-collisional intermediate-acidic volcanism of the Western Carpathians and the andesitic volcanic arc of the Eastern Carpathians differ in petrochemical and petrogenetical aspects, however, both areas are intensely mineralized and subjects of exploration for epithermal gold. The Tokaj Mts. is located on a NNE-trending graben structure formed during the Miocene (Badenian-Sarmatian-Pannonian; 15.0 – 9.4 Ma according to

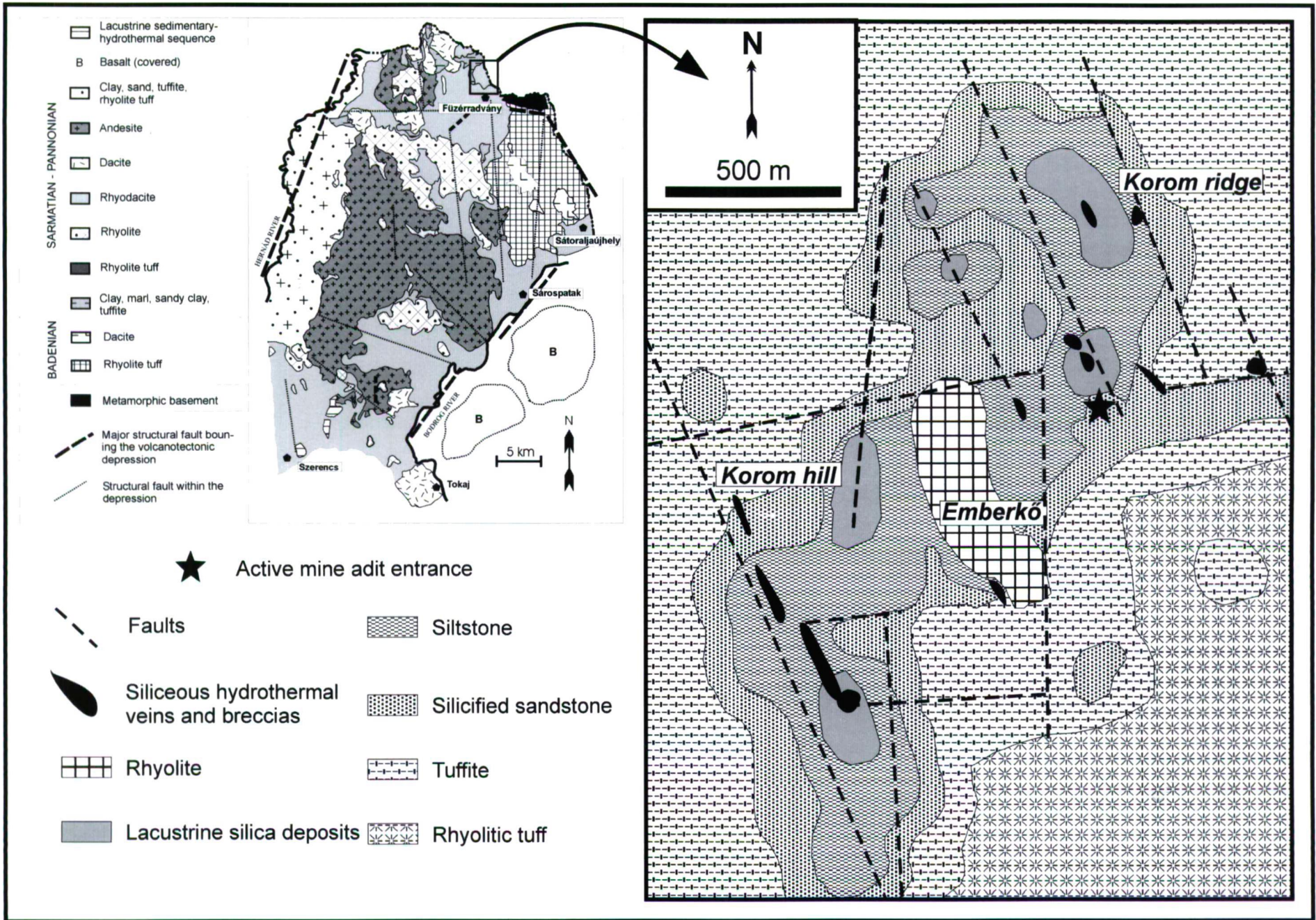


Fig. 1. Geology of the Korom Hill area (after Csongrádi and Zelenka 1995). Inset shows the geological sketch of the Tokaj Mts. with indication of study area.

Pécskay et al., 1986) volcanic activity. Most of the volcanic rocks exposed belong to the Sarmatian-Pannonian volcanic cycle; volcanic rocks of Badenien age crop out in the northeastern part of the mountains and are known under the up to 800-1000 m thick cover of younger volcanic products in drillholes. K-Ar dating on many mineralized areas showed that most of hydrothermal systems were active between 13 and 10 Ma ago (Pécskay and Molnár 2002).

In the area of Füzérradvány, the basement of the Tokaj Mts. is represented by a sequence of Paleozoic age consisting of gneiss, amphibolite and mica schist. These metamorphic rocks are exposed east of the Korom Hill, both in Hungary and in Slovakia (Fig. 1). Based on drillhole data, the basement rocks are overlain by Miocene (Badenian) sediments.

The area of Korom Hill represents a tectonically uplifted block in which the oldest exposed rocks are marine clays of Upper Miocene (Sarmatian) age overlain by ignimbrite (pumiceous rhyolitic tuff) and tuffite (Fig. 1). Considering the intense hydrothermal alteration of these rocks, their radiometric age could not be determined. A lacustrine sequence of about 100 m thickness forms the cover of the acidic volcano-sedimentary units (Mátyás 1974, Gyarmati 1977). Most of the lacustrine sediments consist of siliceous rocks partly due to the sedimentation environment of high silica content and partly due to the later hydrothermal alteration. The main rock types of this lacustrine sequence are silicified fine grained sandstone; silicified siltstone with prints of fossil plants and Mollusca and lacustrine silica of white, black and red colour observed usually at the top of the sequence. Those deposited in small lakes which were fed by hot springs, whose major outflow zones are characterized by

opaline-chalcedony bodies of a few tens of meters diameter. The above described sequence is penetrated by a rhyolite extrusion of northwesterly strike and with 200 m by 700 m aerial extension (the Emberkő Hill, Fig. 1). All of the above mentioned rock types were penetrated by siliceous hydrothermal breccia containing fragments of the country rocks and quartz veins. Quartz veins have microcrystalline massive, or occasionally banded appearance. Thickness of subvertical veins is from 0.1 m (quartz veins) to up to 20 m (breccia veins) with predominantly northwest-southeast orientation (Fig. 1). The orientation of veins corresponds to the most common orientation of faults in the mineralized area, however, east-west and north-south trending unmineralized faults also occur.

In the area of the Korom Hill, lithochemical and soil sampling revealed geochemical anomalies of Au, As, Sb and Hg (Hartikainen et al. 1992). Hydrothermal activity and mineralization is clearly confined to the northwesterly striking tectonic elements with strongly silicified rock units of 10 to 50 m diameter sitting along these fault zones. Silicification and alteration is known in an area of about 10 km² and extend to the Early Paleozoic crystalline basement units. Mineralization in the basement represents a deeper level, possibly a feeder zone of the same system.

The formation of the illite deposit at Füzérradvány can be confined to the shallow zones of hydrothermal activity affecting the ignimbrite and tuff units and lacustrine sedimentary sequence of the Korom Hill. Illite occurs in the hydrothermal alteration zone along the major siliceous veins forming the feeder zones of the shallow hot spring environment (Fig. 2). In the vicinity of the hydrothermal

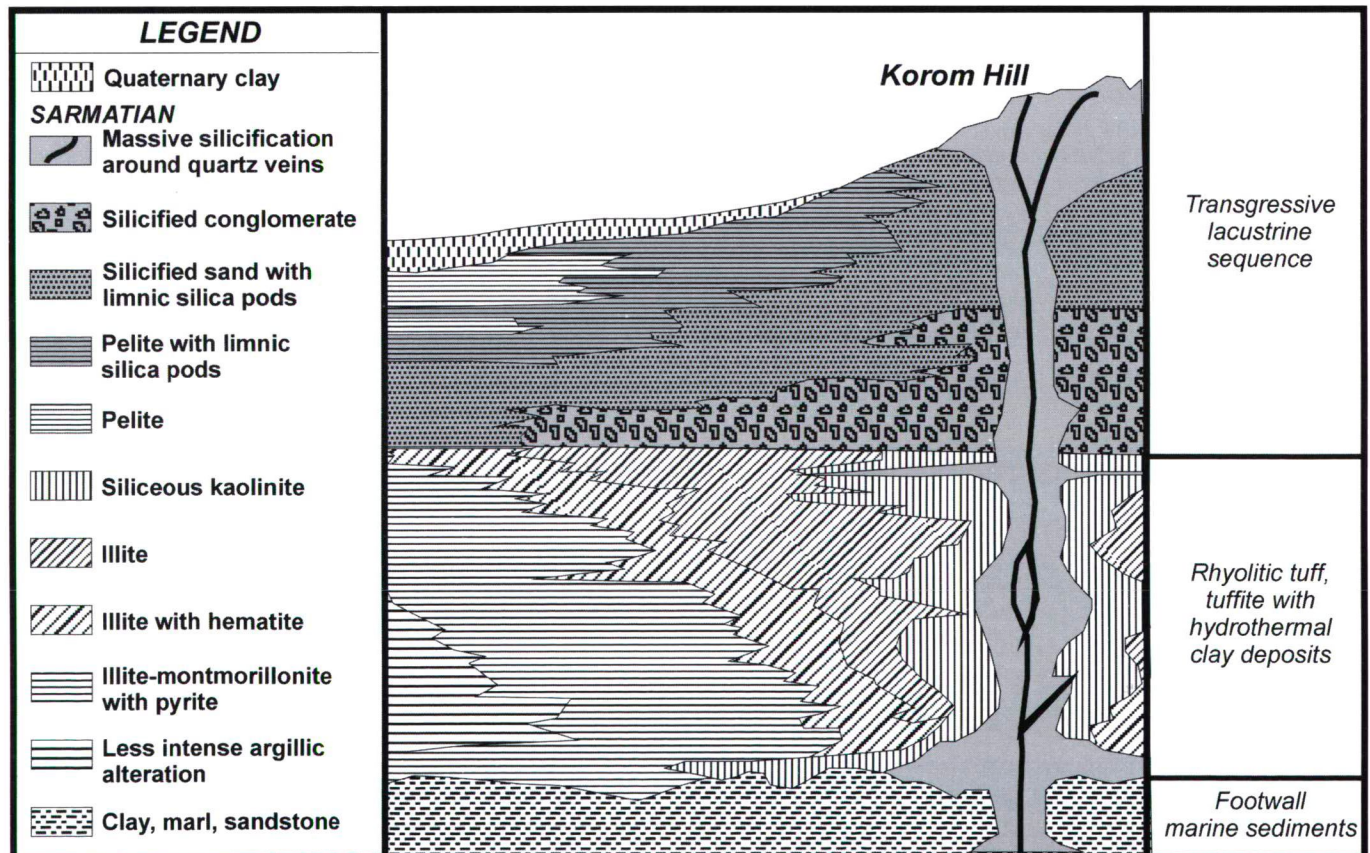


Fig. 2. Hydrothermal alteration and distribution of clay deposits in conceptual cross section of the Korom Hill (after Mátyás, 1972).

vents, silicification and kaolinite alteration occur proving the acidic, silica rich nature of hydrothermal fluids. Silica content of these fluids caused intense silicification of clastic sediments and deposition of siliceous layers in the covering lacustrine-sedimentary sequence. The lacustrine environment also supported syngenetic precipitation of clays in periods of restricted transport of terrigenous materials. Neutralization of hydrothermal fluids during interaction with rhyolitic tuff units at depth resulted in illitization further away from the hydrothermal vents. These illite rich zones form the area of recent mining. Towards the marginal zones of the hydrothermal alteration smectite and less altered rock units can be found.

The mineable illite pods are up to 10 m thick and are outlined by the K_2O content of the altered ignimbrite and rhyolitic tuff and tuffite and high values reflect almost pure illite composition (Fig. 3). Excess potassium content probably reflects association of hydrothermal K-feldspar (adularia) with illite. The high quality pure industrial illite has the following composition (Mátyás 1972):

	wt. %
SiO_2	48.51
Al_2O_3	31.26
$Fe_2O_3 + FeO$	0.50
TiO_2	0.04
CaO	1.75
MgO	1.86
K_2O	7.85
Na_2O	0.10

SO_3	0.06
L.O.I.	6.86

It is remarkable that chemical composition of clay is almost the same as the composition of "Zempleni illite" determined by Veblen et al. (1990) using electron microprobe analyses.

K-AR DATING

Sampling and analytical methods

Four illite samples were collected for detailed studies. Among those ND-1 and ND-2 represent samples from massive illite pods exploited in the mine (Fig. 3). These samples are totally altered ignimbrite/rhyolite tuff in which all of the original texture and minerals are absent with exception of less altered lithoclasts and quartz grains. ND-3 is a pumice fragment from not so intensely altered ignimbrite. Texture of the fragment is still preserved, however, the whole mass is intensely argillic. ND-4 is from a small clay pocket in the ignimbrite in the vicinity of the sampling point of ND-3. Again, the original characteristics of host rock are totally eliminated by the intense alteration and only lithoclasts remained relatively fresh.

Suspensions of clay minerals were prepared by wet sieving of samples in order to eliminate presence of lithoclasts and mineral fragments then the suspension was separated by decantation. After 5 minutes of settling time the suspension from the upper 10 cm of the decantation tube was sampled then centrifuged. For ND-3 longer settling time (10 minutes) was used to eliminate glass and mineral particle fragments.

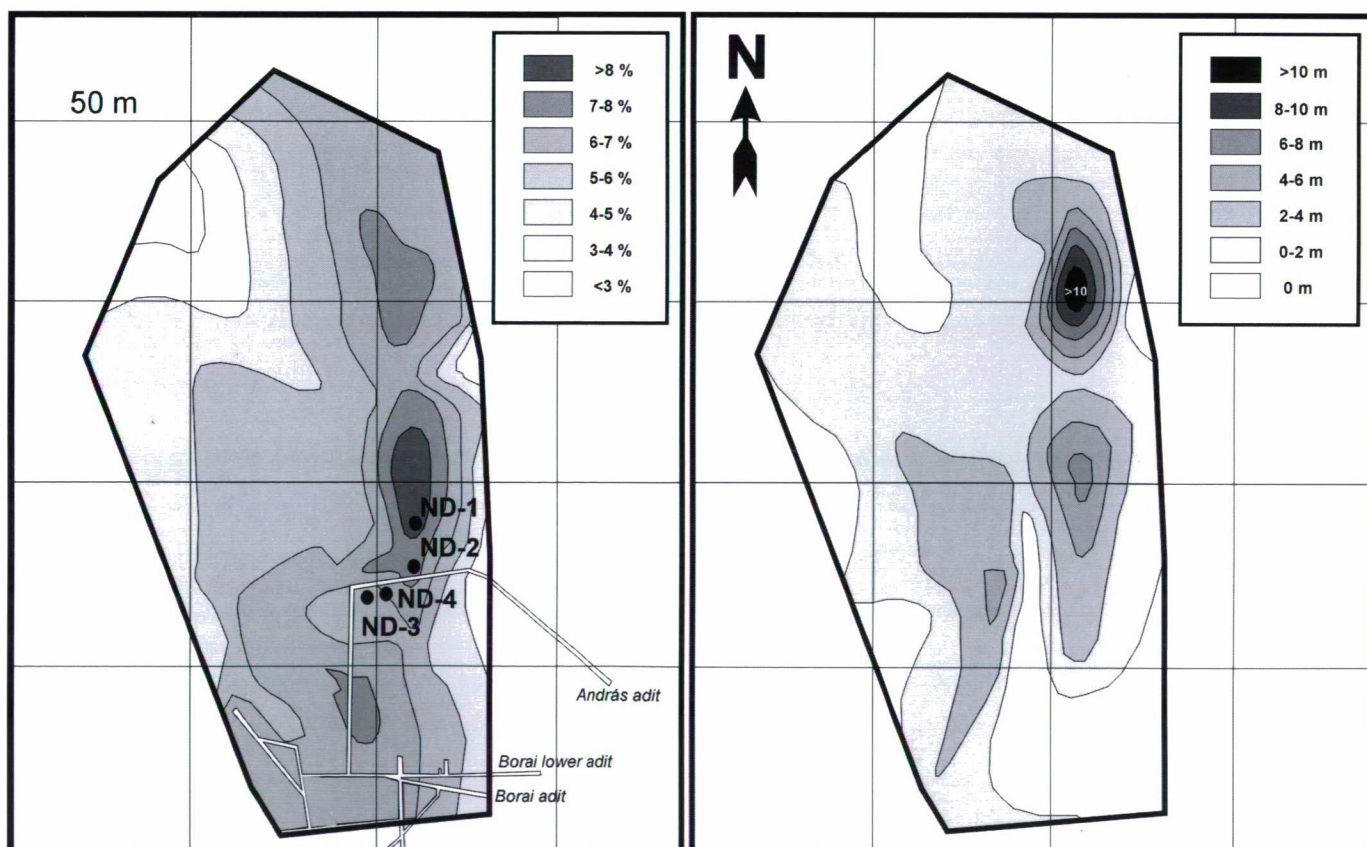


Fig. 3. Distribution of the K_2O content (A) and thickness of mineable clay deposits (B) in the Korom Hill (after Mátyás, 1972) with locations of samples.

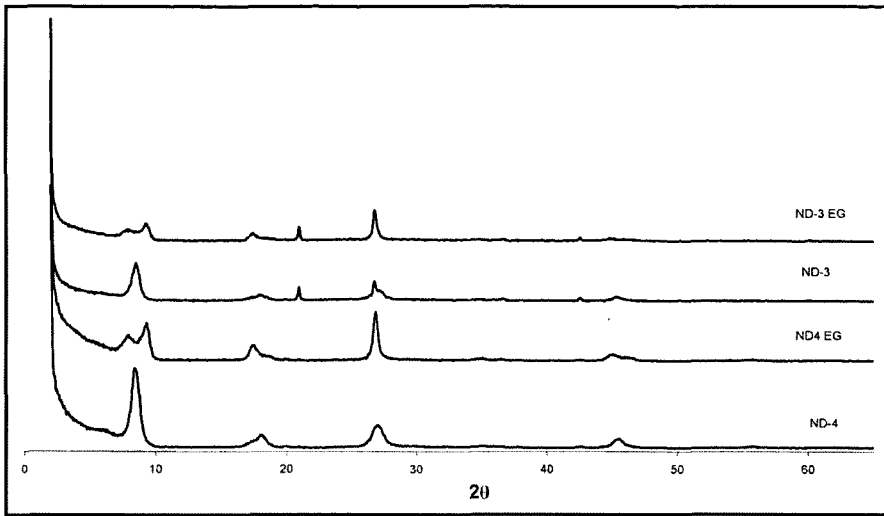


Fig. 4. X-ray powder diffraction patterns for oriented and glycolated (EG) samples ND-3 and ND-4.

Dry and glycolated oriented clay samples were analysed by X-ray powder diffraction method in order to check purity of separates and determination of structural properties. XRD analyses were performed by using a Siemens D 5000 type equipment (Bragg-Bentano geometry, theta-theta running mode, Cu K α ray, secondary graphite monochromator, 0.05° step interval, 2 seconds detection time at each steps).

The K-Ar datings were performed on duplicates of illite samples at the Institute of Nuclear Research, Hungarian Academy of Sciences (Debrecen) and at the Okayama University of Science, Japan. In Debrecen, conventional experimental techniques were used for the argon and potassium analyses. Details of procedures are those described in Pécskay and Molnár (2002). The results of calibration of instruments and applied methods have been described by Balogh (1985).

In Japan, the potassium contents of samples were analysed by flame photometry using a 2000 ppm Cs buffer. The analytical error of this procedure was 2% at the 2 σ confidence level. Ar was dated by a 15 cm radius sector type mass spectrometer with a single collector system using an isotopic dilution method with ³⁸Ar spike. The analytical error of Ar analyses is about 1% at the 2 σ confidence level. Details of methods were described by Nagao et al. (1984) and Itaya et al. (1991). Age calculations of K/Ar analyses were made using the decay constants given by Steiger and Jäger (1977).

Results

XRD analyses on oriented and glycolated samples provided identical patterns for ND-1, ND-2 and ND-4 samples and this is exemplified by the XRD pattern for the sample ND-4 on Fig. 4. XRD patterns after glycolation

proved that all of the samples have small amount of smectite interlayering in agreement with the results of previous studies on the illite from this deposit (see Introduction). Slight difference exists for sample ND-3 which can be related to the presence of volcanic glass and quartz particles from the altered pumice in spite of the longer settling time during sample preparation (Fig. 4).

Results of K-Ar age determination are summarized in Table 1. Range of age data is from 12.2 Ma to 11.5 Ma. Comparing results of laboratories it can be concluded that slightly older K-Ar ages were measured in Debrecen, except of sample ND-4 when an older age was obtained in Okayama. This latter age difference is the obvious consequence of the differences in potassium contents measured at both laboratories, since the argon analyses show excellent agreement. For this sample the best estimation can be done by taking the average value of age calculations, which is 11.81±0.4 Ma.

Sample ND-3 contains the least amount of potassium. This is due to the origin of this sample (i.e. altered pumice fragment). On the other hand, this sample provided the youngest average K-Ar age (11.53±0.4 Ma). Hurley et al. (1963) showed that very fine crystal size allows loss of radiogenic argon to occur at a temperature little above ambient causing the measured analytical age to be young. This is applicable for ND-3 because its settling time was longer in comparison to the other samples. Therefore the K-Ar age of sample ND-3 should be considered as minimum age.

Table 1. Results of K-Ar dating on illite samples from the Korom Hill, Füzéradvány, Tokaj Mts.

Sample No.	Laboratory	Analytical No.	Type of sample	K (wt.%)	Average K (wt.%)	⁴⁰ Ar rad (ccSTP/g)	Average ⁴⁰ Ar rad (ccSTP/g)	⁴⁰ Ar _{rad} (%)	K-Ar age (Ma)	Average K-Ar age (Ma)
ND-1	ATOMKI	6366	massive clay	7.05	7.09	3,316*10 ⁶	3,288*10 ⁻⁶	31,4	12,1 +/- 0,5	11,89 +/- 0,30
	OUS	S39-154		7.12		3,259*10 ⁶		93,4		
ND-2	ATOMKI	6367	massive clay	6.80	6,81	3,228*10 ⁶	3,156*10 ⁻⁶	32,1	12,2 +/- 0,6	11,88 +/- 0,3
	OUS	S39-155		6.81		3,084*10 ⁶		94,6		
ND-3	ATOMKI	6368	altered pumice	4.93	4,95	2,227*10 ⁶	2,226*10 ⁻⁶	43,7	11,6 +/- 0,4	11,53 +/- 0,4
	OUS	S39-156		4.97		2,225*10 ⁶		89,9		
ND-4	ATOMKI	6369	clay pocket, alt.tuff	6.44	6,27	2,898*10 ⁶	2,888*10 ⁻⁶	26,8	11,5 +/- 0,7	11,81 +/- 0,4
	OUS	S39-157		6.09		2,879*10 ⁶		85,2		

Highly consistent average radiometric ages (11.89 ± 0.3 Ma and 11.88 ± 0.3 Ma) were measured on samples with high K-content (ND-1 and ND-2 samples). These data can be considered as the most reasonable results for the age of formation of illite.

DISCUSSION

Geology of the clay deposit at Füzérradvány indicates that a volcanic and sedimentary sequence was cut by hydrothermal vents and argillic alteration zones are centred on those vents (Fig. 2). The relatively acidic nature of fluids in the main channels is expressed by the occurrence of kaolinite along the veins. Illite forms in less acidic conditions in epithermal systems (Hedenquist et al. 1996). Thus illite occurrence in certain zones moving apart from the main hydrothermal channels can be explained by neutralization of acidic hydrothermal fluids during their interaction with permeable rock units. The overall zoning of hydrothermal alteration on the Korom Hill is very similar to other mineral deposits of the Tokaj Mts. (Mátyás 1972) and corresponds to shallow levels of low sulphidation type epithermal systems (Molnár 1993, Molnár et al. 1999). The only important difference in comparison to other shallow hydrothermal systems of the Tokaj Mts. is the lack of alunite-bearing steam-heated alteration zone in the uppermost part of the hydrothermal system (Fig. 2). Steam-heated alteration in an epithermal systems forms above the palaeogroundwater table (Hedenquist et al. 1996). The lack of this alteration zone on the Korom Hill can be explained by two ways: One possibility is that the hydrothermal alteration overprinted the lacustrine-sedimentary environment and the steam heated alteration zone has already eroded away. Because the deposition of silica in the lacustrine environment was due to presence of hot springs in the sedimentary basins, this invokes that hydrothermal activity had two stages. On the other hand, the feeder zones of discharges might be the zones with quartz and breccia veins and thus hydrothermal fluids reached the bottom of the basin under the paleogroundwater table and therefore formation of steam-heated alteration zone was limited.

Our K-Ar studies suggest that there is no significant argon loss from the dated mineral separates. Also, we can exclude the presence of excess/inherited argon caused by some detrital mica in the concentrates. Therefore assuming that the most reliable ages can be related to samples with high potassium content, the age of the hydrothermal activity on the Korom Hill is 11.89 ± 0.3 Ma. and if there existed two overprinting hydrothermal events they cannot be distinguished behind the resolution of K-Ar dating.

CONCLUSIONS

Mineralization of the Korom Hill at Füzérradvány represents shallow levels of low sulphidation type epithermal system in which illite formed in certain zones along hydrothermal channels. Using appropriate sample preparation of illite for K-Ar studies, geologically reliable age data were achieved for the hydrothermal activity and formation of illite deposit. Results of samples with the highest potassium content suggest that illite formation can be related to a single hydrothermal event which took place at 11.89 ± 0.3 Ma. This is in agreement with the Sarmatian age

of the host rock and rhyolite domes located nearby the deposit. Recently obtained analytical data confirm that the mineralization at Füzérradvány was formed synchronously with other hydrothermal systems in the region, and therefore all hydrothermal systems exposed in the area of the Tokaj Mts. were formed in relation to the Sarmatian intermediate-acidic volcanic activity.

ACKNOWLEDGMENTS

This work was supported by the bilateral research agreement between the Hungarian Academy of Sciences and the Japan Society for Promotion of Sciences. The HNSF (OTKA) found No. T 046886 and the HNSF (OTKA) found No. M 041434 provided additional support. Special thanks to István Viczián and Kadosa Balogh for helpful comments during the project and preparation of the manuscript.

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Received: August 27, 2005; accepted: January 10, 2006