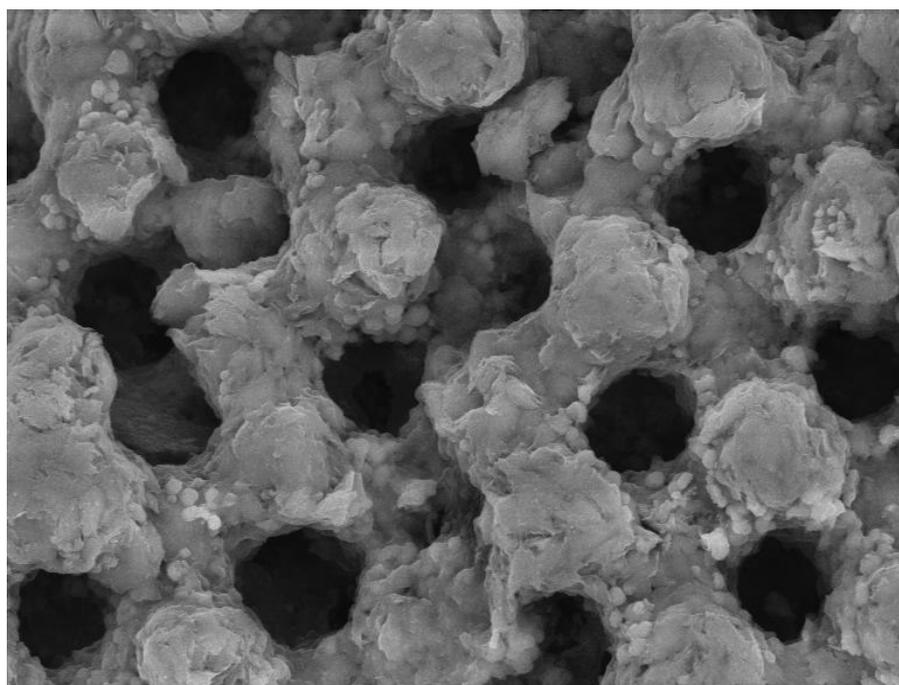


Theses of PhD dissertation

ERZSÉBET TÓTH

ANALYTICAL PRACTICE AND CRYSTAL CHEMISTRY OF THE CELADONITE–GLAUCONITE
GROUP. THE CRYSTAL CHEMISTRY OF GLAUCONITISATION,
BASED ON SOME HUNGARIAN EXAMPLES



HV: 25.0 kV DET: BS Det - SE Dete 
Satellite ©Tescan DATE: 11/22/03 20 µm

Supervisor: Tamás G. Weiszbürg, habil associate professor

*PhD School in Earth Sciences
Head: Prof. Miklós Monostori
Geology-Geophysics PhD Programme
Head: Prof. Miklós Monostori
Field: Geology*

DEPARTMENT OF MINERALOGY
EÖTVÖS LORÁND UNIVERSITY
BUDAPEST

2007

Introduction

Green earth varieties have been widely used since the Roman times. From the 19th century on, most green earth types have been called *glauconite* or *celadonite*, and the two terms are present in mineralogy and geology also today. Scientific content and relationship of these two terms have been changing continuously and they are mixed up in scientific papers even today.

In the first eight decades of the 20th century they have been distinguished upon *genetic environment*: green grains and earthy masses found in sedimentary environment were called glauconite, whereas those originating from volcanic environment were called celadonite. However, modern nomenclatures (AIPEA, 1980; AIPEA, 1986; IMA, 1998) are clearly based on *crystal chemistry*, although ambiguity is still preserved and reflected in the fact the three nomenclatures apply all the three possible crystal chemical cation position (tetrahedral, octahedral and interlayer, resp.) to differentiate between the two minerals.

The latest nomenclatures (IMA 1998≡AIPEA 2006) agree that celadonite is an iron-rich dioctahedral mica with TOT charge originating predominantly from the octahedral layer, while glauconite is its interlayer-cation-deficient counterpart. Differentiation is thus based on crystal chemistry, but in practice – due to analytical problems arising from small grain size and inhomogeneities, the two minerals are mainly differentiated upon structural characteristics (XPD and FTIR behaviour – according to the 1980 definitions of AIPEA). Up to present, references have not revealed that there are serious discrepancies between the two approaches.

To further complicate the situation, minerals of this group form in certain environments by direct crystallisation, whereas under different conditions (on the sea-bottom, near the sediment-water interface) they appear as a result of a complex crystal chemical evolution. This latter process is known as *glauconitisation*, and the understanding of this process is vital for both oceanology and in palaeoenvironmental reconstructions.

The *crystal chemical relationship* between celadonite and glauconite is currently thought to be similar to the relationship between illite – studied in details due to its significance in hydrocarbon prospecting – and muscovite. *Formation conditions* are significantly different, however: during illitisation, disperse expandable dioctahedral layer silicates of the sediment turn gradually into (interlayer-deficient) mica, as a result of progressing burial: increasing pressure and temperature. Glauconitisation is on the other hand a predominantly chemistry-driven process occurring near the sediment-water interface. In the lack of detailed studies on glauconitisation, the two processes are often made parallel, although we have no proven thermodynamic reason to do that.

Main goals

My work had two basic goals.

The first main goal was to find out the potential and limits of the *chemical investigation* of the celadonite-glaucanite group by collecting statistical amount of analytical data on a number of well-characterised samples, applying and comparing widely used analytical methods. The *quality of chemical data* was measured by *their crystal chemical applicability*, as it has been shown that the modern characterisation of this mineral group is based on crystal chemistry. The basic question was therefore the following: Is it possible to obtain chemical data of high enough quality for crystal chemical calculations on a large number of samples with modern analytical methods? If so, what are the limitations? It was necessary to handle the frequent lack of Fe²⁺-determination and the propagation of the analytical error during formula calculation. Analytical comparability required the highest possible chemical homogeneity of the samples.

The second main goal was the crystal chemical characterisation of the *glaucanisation process*. A number of models have been proposed in the last 50 years to explain / describe this process. Model proposing researchers tried to reconstruct the stages of glaucanisation through the comparison of recent or fossile samples, which led to camouflaging or rendering uncertain the gist of the process. To avoid this, we were the first to study *evolution series* represented by fractions obtained through the sequential separation of single samples. This approach eliminates all factors independent of glaucanisation (different starting material, formation conditions and diagenetic history), and the remaining differences between the fractions will directly unveil /reveal the glaucanisation process itself. The studied evolution series originate from Hungarian Cretaceous and Oligocene formations, facilitating the later application of mineralogical results in the Hungarian geological research.

During my work it turned out that the general confusions surrounding the nomenclature of this mineral group even today make it impossible express ideas and results accurately. Therefore it was unavoidable to trace the birth and evolution of the terms glaucanite and celadonite, just to be able to handle them on a uniform and unequivocal platform.

The need to show lucidly/ expressively the multivariate process of glaucanisation arose during my work, too. Published graphical representation systems of crystal chemistry were therefore listed and evaluated upon their applicability to iron-rich dioctahedral micas, highlighting their advantages and disadvantages, too. Recognising their disadvantages, we set the goal of designing a new graphical representation system that elucidates the process of glaucanisation.

Methods

The applied methods can be divided into several groups.

Beside the routine processing of related current references, *outlining the theoretical background* required the setting up of a coherent crystal chemical database, uniform in its approach. This database includes earlier published analyses (approximately 600 entries) complemented by crystal chemical calculations. Confusions in the terms rooted in the distant past required the systematic processing of historical original works back to the middle of the 18th century.

Sample selection was the result of field work supported by geologists familiar with the studied formations. To better understand the crystal chemical changes during glauconitisation and the crystal chemistry of the celadonite-glauconite group, properly separated green layer-silicates of six sedimentary formations were studied in detail. These formations are: Upper Oligocene Eger Formation (sandstone, sample codes: NY1, NY3, EWT), Lower–Middle Cretaceous Sümeg Marl Formation (limestone, sample code: SK), Middle Cretaceous Pénteskút Marl Formation (limestone, sample code: PM) and the Lower Jurassic Úrkút Manganese Ore Formation (banded manganese carbonate ore).

Each sample was submitted to *special separation processes* designed by our working group in order to obtain chemical homogeneity required by our goals and not yet achieved by earlier researchers of this field.

Following traditional *optical microscopy* (stereomicroscope, transmitted light polarizing microscope), appr. 900 grains were selected for detailed morphological study by *scanning electron microscopy* (SEM). In the next stage, grains were embedded in epoxy resin, and their chemical composition was studied by *quantitative local chemical methods* (WDX, EDX, LA-ICP-MS).

Main and trace element composition of separated fractions of evolution series were also determined by *bulk methods* (ICP-AES, ICP-MS). Wherever possible, Fe²⁺-content was determined by titration, too.

Comparison of laboratories (Geological Institute of Hungary Budapest and the Natural History Museum, London) *and methods* (local vs. bulk; WDX vs. EDX) played an important role during designing the measurements. At the local chemical methods (WDX, EDX), the effect of *analytical conditions* (size of the analysed area, analysis order etc.) on the measured compositions was also tested.

The *structural methods* X-ray powder diffraction (XPD) and Fourier-transform infrared spectroscopy (FTIR) were mainly applied for phase identification, at some evolution series for the documentation of the structural changes and to check sample purity.

Theses

1. With the help of appr. 1300 new chemical analyses it has been confirmed that all of the applied main element analytical methods – designed in accordance with our approach – are capable of yielding chemical data suitable for crystal chemical calculations in the celadonite-glaucanite group. Systematic differences were recorded, however, between WDX and EDX analyses, local (WDX, EDX) and bulk (ICP-AES) methods and the single ICP-AES laboratories. These results allow the crystal chemical evaluation of our own data, as well as the crystal chemistry related quality assessment of analyses available in the literature (appr. 600 analyses).

2. It has been shown that concerning the Fe^{3+} -Al substitution in the octahedral sheet of dioctahedral layer-silicates the crystal chemical space is continuous at both smectites and interlayer-deficient micas. For smectites, proof is provided by traditional green grain series undergoing glaucanitisation, while for interlayer-deficient micas by the atypical green grain series of the Sümeg Marl Formation. These results suggest that the iron-rich (${}^{\text{VI}}\text{Al} / ({}^{\text{VI}}\text{Al} + {}^{\text{VI}}\text{Fe}^{3+}) < 0,5$) counterpart of montmorillonite should be defined as a new mineral species and that the illite definition of IMA should be extended.

3. The starting material of glaucanitisation was found to be montmorillonite, which is different from “traditional” montmorillonites in having K as the dominant cation in the interlayer (and not Na or Ca). The crystal chemical evolution goes through the steps montmorillonite => „ Fe^{3+} -montmorillonite” / nontronite => glaucanite-GG => glaucanite-CG.

4. It has been shown that the octahedral occupancy ($B_{\text{O}} > 2.00$) is a real feature of glaucanitising grains (*i.e.* green grains evolving in the sediment) and not a consequence of analytical error. Fe^{2+} content estimation assuming $B_{\text{O}} = 2.00$ is therefore not a proper formula calculation method at these materials. Giving a rough estimate of the Fe^{2+} content upon the grain colour seems to be more adequate.

5. From a crystal chemical viewpoint, glaucanitisation has been divided into two stages. In the first stage, the amount of ${}^{\text{IV}}\text{Al}$ increases, and the accompanying decrease of the tetrahedral cation charge facilitates the incorporation of K into the interlayer space (a feature similar to illitisation – relevant samples: Pénteskút Marl Formation, **Fig. 1**, whitish grains of the Sümeg Marl Formation). In the second stage of glaucanitisation, the decrease of the octahedral cation charge facilitates the incorporation of K into the

interlayer space, and the composition of the tetrahedral layer is not changing significantly (a feature different from illitisation – relevant samples: Eger Formation – NY1 – **Fig. 2**, NY3, EWT).

6. It was demonstrated that at the beginning of glauconitisation, octahedral occupancy is significantly higher (B_O) than the ideal dioctahedral value, and it is not changing much in the first stage of glauconitisation. In the second stage, however, the release of ^{VI}Al exceeds the incorporation of $^{VI}Fe^{3+}$ and $^{VI}Fe^{2+}$, thus B_O decreases, approaching ideal dioctahedrality. The decrease of the octahedral cation charge (X_O) originates predominantly from the decrease of B_O („increase of vacancies”, **Fig. 3**). The composition of the octahedral sheet changes the same way during both stages of glauconitisation: Al content decreases, Fe content increases (in both valence states), while the amount of Mg is relatively stable.

7. A new, combined graphical representation system has been designed that represents the crystal chemistry of the celadonite-glauconite group better than the previous ones. This system consists of the combined application of the *3D charge diagram* (axes x , y , z are the octahedral cation charge, X_O , the ratio $^{VI}Al / (^{VI}Al + ^{VI}Fe^{3+})$ and the interlayer cation charge, X_{IL} , resp.; **Figs. 1–2**) and the *octahedral occupancy–octahedral charge (B_O – X_O) diagram* (**Fig. 3**). The 3D charge diagram is able to show the main discrimination borders of all three nomenclatures and allows the definition of smaller crystal chemical space sections by the combination of nomenclatures. Some nomenclature borders (IMA: $^{VI}R^{2+} / (^{VI}R^{2+} + ^{VI}R^{3+})$) can migrate as a function of octahedral occupancy, but the *diagram retains its crystal chemical character in all cases*. The crystal chemical space occupied by the celadonite-glauconite group is best dissected by the combination of the AIPEA 1980 and IMA 1998 nomenclature. The introduction of the two-letter suffices allows the unequivocal expression of chemical composition, without the introduction of new terms.

8. LA-ICP-MS has been applied for the first time for the *systematic* study of the rare earth element (REE) content of the celadonite-glauconite group. It has been shown that the true REE content of these green layer-silicates is significantly lower (by 1–2 orders of magnitude) than previously published values (ICP-MS, INAA). Comparing local (LA-ICP-MS) and bulk (ICP-MS, INAA) analytical data, we managed to explain this phenomenon: higher REE concentrations are caused by the subordinate, intragrain presence of REE-concentrating phases that cannot be removed by separation.

9. The true REE pattern of glauconites is similar in shape to the REE patterns of sedimentary rocks and the upper crust, though the REE concentrations are lower (0.1–0.5 PAAS) already at the beginning of glauconitisation and are further decreasing during the process.

10. Pores and cracks of the high-density ($\rho > 2.63 \text{ g/cm}^3$) grains of sample EWT are filled by a yet unknown Ca-REE-phosphate phase, the composition of which can be expressed as (normalised to 4 O) $(\text{Ce}_{0.42}\text{La}_{0.16}\text{Nd}_{0.12})\text{Ca}_{0.36}\text{P}_{1.05}\text{O}_4$. The REE : Ca ratio is approximately 2 : 1.

11. The green grain series of the Sümeg Marl Formation differs in both appearance and composition from the traditional sedimentary green interlayer-deficient micas: the high-density grains may be regarded as the first description of allochthonous granular celadonite-CC. The grains are sometimes pseudomorphs after elongated minerals (with traces of prismatic cleavage), and often preserve signs of alteration.

12. The identification of the illitising(?) gray grain series (up to present without chemical data) in the non-magnetic fractions of the Sümeg Marl Formation revealed that not only iron-rich dioctahedral layer silicates (glauconites evolving in sediments) may form aggregates by orders of magnitude larger than the size of clay minerals. It is an important methodological conclusion that during the clay mineralogical study of sedimentary rocks it is necessary to conduct μm range textural studies beside the conventional preparation (crushing, carbonate dissolution *etc.*) of the clay fraction.

Conclusions

With the acquisition and evaluation of appr. 1300 new main element analyses, obtained under controlled and comparable conditions, complementing the appr. 600 analyses published so far on the celadonite-glauconite group, we managed to set up the requirements of obtaining high-quality analytical data (good for crystal chemical calculations) on the celadonite-glauconite group minerals. This gives us the chance to identify the minerals of this group – in agreement with both IMA and AIPEA recommendations – upon chemical data, obtained even by local chemical methods (EPMA, SEM+EDX), allowing the study of a high number of samples within reasonable time and cost limits.

In agreement with our preliminary assumptions, the multi-step separation technique (involving separation upon grain size, magnetic susceptibility and density) designed by our working group separates glauconitic grains of a given population according to maturity (interlayer cation content

etc.). Fractions obtained by the labour-intensive separation represent evolution series, offering the only opportunity to the direct study of glauconitisation.

With a statistical number of analyses, obtained on the evolution series of five Hungarian glauconite-bearing samples, we managed to reconstruct convincingly the crystal chemical changes (reflected in changing chemical composition) occurring during glauconitisation.

Having documented the true REE content of the celadonite-glauconite group and revealed the REE pattern distorting effect of syn- or postdiagenetic REE-concentrating accessory minerals we can avoid the inadequate geochemical application of this mineral group in the future.

Publications related to the subject of the PhD dissertation

Papers

1. Weiszburg T., **Tóth E.**, Kuzmann E., Lovas Gy. (1999): Iron-rich trioctahedral smectite from Csódi Hill, Dunabogdány, Hungary (*in Hung.*). *Topographia Mineralogica Hungariae*, 6, pp. 179–190.
2. Weiszburg, T. G., **Tóth, E.**, Beran, A. (2004): Celadonite, the 10-Å green clay mineral of the manganese carbonate ore, Úrkút, Hungary. *Acta Mineralogica-Petrographica Szeged*, 45/1, pp. 65–80.
3. Weiszburg, T. G., Nagy, T., **Tóth, E.**, Mizák, J., Varga, Zs., Lovas, Gy. A., Váczi, T. (2004): A laboratory procedure for separating micas from quartz in clay-sized materials. *Acta Mineralogica-Petrographica Szeged*, 45/1, pp. 133–139.

International conference abstracts

1. **Tóth, E.**, Weiszburg, T. G. (2000): Chemical composition of celadonite in a mixed clay-sized material from Úrkút, Hungary. M&M4 Conference (Melbourne), Program and Abstract Volume, p. 97.
2. **Tóth, E.**, Mizák, J., Nagy, T., Varga, Zs. (2001): A new procedure for the separation of clay minerals from mixed clay-sized materials. *Solid Solutions in Silicate and Oxide Systems of Geological Importance (Lübeck)*, Programme and abstract volume, p. 35.
3. Weiszburg, T. G., Pop, D., **Tóth, E.** (2001): Glauconites and celadonites in Central Europe: a nomenclatural review. *Mid-European Clay Conference '01 (Stará Lesná)*, Book of abstracts, p. 116.
4. **Tóth, E.**, Weiszburg, T. G., Pop, D. (2001): Celadonite: the colour-giving green clay mineral of the carbonate manganese ore, Úrkút, Transdanubian Central Range, Hungary. *MinPet 2001*

Conference (Wien), Mitteilungen der Österreichischen Mineralogischen Gesellschaft, 146, 288–289.

5. **Tóth, E.**, Weiszbürg, T. G. (2002): New genetical environment for celadonite: the Úrkút carbonate manganese ore, Transdanubian Range, Hungary. 18th General Meeting of the International Mineralogical Association (Edinburgh), Programme with abstracts, p. 169.
6. Weiszbürg, T. G., **Tóth, E.**, Pop, D. (2002): Glauconite and celadonite: a nomenclature review. 18th General Meeting of the International Mineralogical Association (Edinburgh), Programme with abstracts, p. 135.
7. Fekete, J., Weiszbürg, T. G., **Tóth, E.** (2003): Tracing glauconite formation in Oligocene-Miocene sandstones in Hungary. Mineral Sciences in the Carpathians Conference (Miskolc), Acta Mineralogica-Petrographica Abstract Series, 1, p. 31.
8. Weiszbürg, T. G., Pop, D., **Tóth, E.** (2003): Chemistry-based nomenclatures versus discriminating analytical methods (FTIR, XPD) in the celadonite-glauconite family. Mineral Sciences in the Carpathians Conference (Miskolc), Acta Mineralogica-Petrographica Abstract Series, 1, p. 111.
9. Weiszbürg, T. G., **Tóth, E.**, Pop, D. (2003): Misfits between the chemistry-based classification schemes and the routinely used discriminating analytical methods in the celadonite-glauconite family. Euroclay 2003, 10th Conference of the European Clay Groups Association (Modena), Abstracts volume, p. 287.
10. **Tóth, E.**, Weiszbürg, T.G., Pop, D. (2003): Bottomlines in calculating crystal chemical formulae in the celadonite-glauconite family. Euroclay 2003, 10th Conference of the European Clay Groups Association (Modena), Abstracts volume, p. 272.
11. **Tóth, E.**, Weiszbürg, T.G. (2004): The evolution of REE and other trace element patterns in the course of glauconitization. 13th Annual V.M. Goldschmidt Conference (Copenhagen), Geochimica et Cosmochimica Acta 68/11S (Special Supplement – Abstracts of the 13th Annual V.M. Goldschmidt Conference Copenhagen, Denmark, June 5–11, 2004), p. A378.
12. **Tóth, E.**, Weiszbürg, T.G. (2004): Causes of misfit between local and bulk chemical data: the effect of internal inhomogeneity of a glauconite population. 32nd International Geological Congress, 20–28 August, 2004 (Florence). Abstract volume, p. 218 (42-20).
13. Weiszbürg, T.G., Pop, D., **Tóth, E.** (2004): The process of glauconitization – traced by the study of glauconite populations from the Upper Oligocene Eger Formation (North Hungary). 32nd International Geological Congress, 20–28 August, 2004 (Florence). Abstract volume, p. 216 (42-8).

14. **Tóth, E.**, Weiszburg, T.G. (2004): Layer charge evolution during glauconitisation. LCCM '04, International Workshop on Current Knowledge on the Layer Charge of Clay Minerals, September 18–19, 2004, (Smolenice). Abstract volume, p. A-18.
15. **Tóth, E.**, Weiszburg, T.G. (2004): Substitution processes during glauconitisation. MECC '04, 2nd Mid-European Clay Conference, 20–24 September, 2004 (Miskolc). Acta Mineralogica-Petrographica Abstract Series, 4, p.107.
16. Weiszburg, T.G., **Tóth, E.** (2004): The crystal chemical evolution of glauconites. MECC '04, 2nd Mid-European Clay Conference, 20–24 September, 2004 (Miskolc). Acta Mineralogica-Petrographica Abstract Series, 4, p.115.
17. **Tóth, E.**, Weiszburg, T.G., Williams, C.T., Jeffries, T.E., James, S., Bartha, A., Rohonczy, J. (2004): Reliability of chemical data on glauconites. New Developments in the Study of Clay Minerals and Fine-Grained Materials, One-Day Meeting of the Clay Minerals Group of the Mineralogical Society, 19th October, 2004 (London). Programme and abstracts volume, p. 18.
18. Weiszburg, T.G., **Tóth, E.** (2004): The crystal chemistry of glauconitisation. New Developments in the Study of Clay Minerals and Fine-Grained Materials, One-Day Meeting of the Clay Minerals Group of the Mineralogical Society, 19th October, 2004 (London). Programme and abstracts volume, p. 19.
19. **Tóth, E.**, Weiszburg, T.G., Pop, D. (2006): Critical evaluation of the analytical data on dioctahedral iron-rich micas and related mineral phases published in the literature. 3rd Mineral Sciences in the Carpathians International Conference, 9–10 March, 2006 (Miskolc). Acta Mineralogica-Petrographica Abstract Series, 5, p. 120.
20. Cora, I., **Tóth, E.**, Weiszburg, T.G., Zajzon, N. (2006): Mineralogical study of Upper Cambrian glauconites from Texas, USA. 3rd Mineral Sciences in the Carpathians International Conference, 9–10 March, 2006 (Miskolc). Acta Mineralogica-Petrographica Abstract Series, 5, p. 21.
21. Váczi, T., **Tóth, E.**, Weiszburg, T.G. (2006): Modelling IR absorption for Si-O stretching vibrations of 2:1 layer silicates based on bond valence calculations. 3rd Mineral Sciences in the Carpathians International Conference, 9–10 March, 2006 (Miskolc). Acta Mineralogica-Petrographica Abstract Series, 5, p. 128.
22. **Tóth, E.**, Pop, D., Weiszburg, T.G. (2006): Graphical representation systems for TOT layer silicates: a case study for iron-rich dioctahedral phases (celadonite, glauconite). 19th General Meeting of the International Mineralogical Association, Expansion to Nano, Bio and Planetary Worlds, 23–28 July, 2006 (Kobe). Program and Abstracts, p. 152.
23. Weiszburg, T.G., **Tóth, E.**, Pop, D. (2006): Continuous crystal chemical space for the dioctahedral iron-rich micas and related phases (celadonite, glauconite, Fe-illite). 19th General

Meeting of the International Mineralogical Association, Expansion to Nano, Bio and Planetary Worlds, 23–28 July, 2006 (Kobe). Program and Abstracts, p. 152.

24. Cora, I., **Tóth, E.**, Weiszburg, T.G. (2006): Mineralogical study of Upper Cambrian glauconites from Texas, USA. 19th General Meeting of the International Mineralogical Association, Expansion to Nano, Bio and Planetary Worlds, 23–28 July, 2006 (Kobe). Program and Abstracts, p. 278.
25. **Tóth, E.**, Weiszburg, T.G. (2006): Two in one: two different glauconite series from the same rock, Sümeg Marl Formation, Lower Cretaceous, Bakony Mountains, Hungary. 3rd Mid-European Clay Conference – MECC 2006, 18–22 September, 2006 (Opatija). Abstracts Book, p. 116.
26. Pop, D., **Tóth, E.**, Weiszburg, T.G., Sásáran, E., Stichleutner, S. (2006): The TOT layer silicate of the lacustrine green clay, Rona limestone, Romania. 3rd Mid-European Clay Conference – MECC 2006, 18–22 September, 2006 (Opatija). Abstracts Book, p. 95.
27. Weiszburg, T.G., **Tóth, E.**, Pop, D. (2006): Continuous crystal chemical space for the dioctahedral iron-rich micas and related phases (celadonite, glauconite, Fe-illite). 3rd Mid-European Clay Conference – MECC 2006, 18–22 September, 2006 (Opatija). Abstracts Book, p. 122.

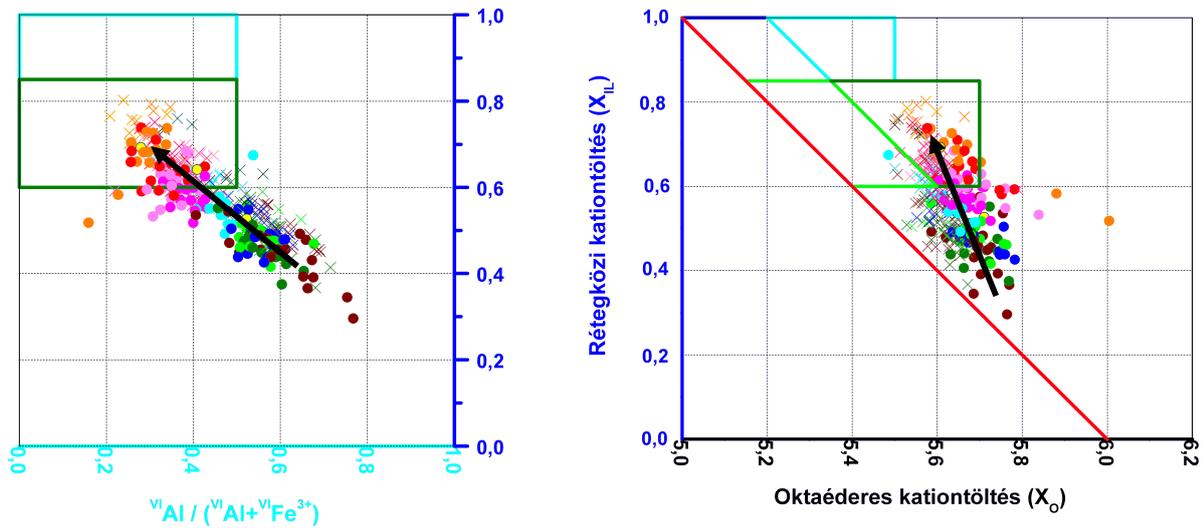


Fig. 1. The first stage of glauconitisation, demonstrated by the green grain series of the Pénzeskút Marl Formation, on two sections of the 3D charge diagram. The direction of evolution (increasing aggregate density) is shown by an arrow. Analyses clearly get diverge from the red diagonal (Si = 4), reflecting increase of ^{IV}Al . Chemical evolution of the grains: montmorillonite => „ Fe^{3+} -montmorillonite” => glauconite-GG.

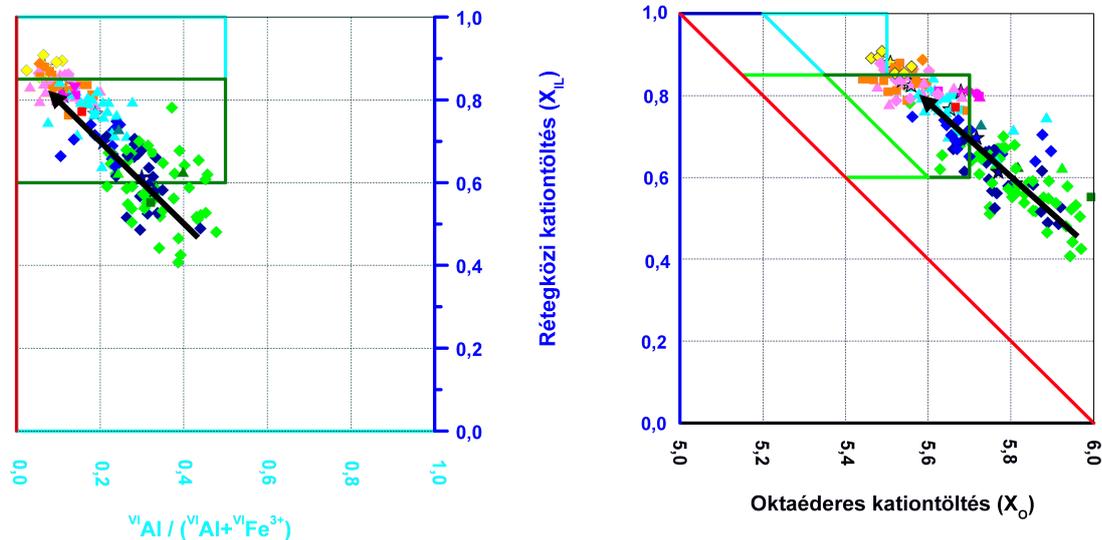


Fig. 2. The second stage of glauconitisation, demonstrated by the green grain series of sample NY1, Eger Formation, on two sections of the 3D charge diagram. The direction of evolution (increasing aggregate density) is shown by an arrow. Analyses plot parallel to the red diagonal (Si = 4), indicating that the evolution is „isotrahedral”. Chemical evolution of the grains: nontronite => glauconite-GG => celadonite-CG.

Fig. 3. The second stage of glauconitisation, demonstrated by the green grain series of sample NY1, Eger Formation, on the B_O - X_O diagram. The direction of evolution (increasing aggregate density) is shown by an arrow. For the decrease of the octahedral charge (X_O) the release of ^{VI}Al , and later the $^{VI}Al \Rightarrow ^{VI}Fe^{2+}$ substitution is responsible.

