

DIAGENETIC NEOFORMATIONS IN MIDDLE TRIASSIC EVAPORITIC AND CARBONATE ROCKS, MECSEK MTS. (S. HUNGARY)

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ABSTRACT

In the Middle Triassic (Anisian) evaporitic and carbonate rocks of the Mecsek Mts. diagenetic neoformation of corrensite, Mg-rich chlorite, magnesite, albite and quartz has been observed by microscopic and X-ray diffraction methods.

Fine-grained magnesite is found in clay mineral aggregates of siltstones of detrital origin. Magnesites of very similar lithologic character have been found in the Drauzug area of the Eastern Alps and in Hesse, Germany, in a comparable stratigraphic position.

Authigenic albites in limestones occur as euhedral crystals oriented according to (010).

The formation of euhedral quartz of prismatic shape is accompanied by the formation of corrensite in carbonate rocks.

The stability relations of these minerals are discussed in terms of thermodynamic considerations of LIPPMANN. The possible effects of Mg-enrichment in the solution and of transformation reactions of detrital expandable clay minerals are discussed.

INTRODUCTION

The Middle Triassic (Anisian) sequence of the Mecsek Mts. was studied by using the rock samples of the borehole Pécs-IX (see Fig. 1). Clay minerals were described in a previous publication (VICZIÁN 1993). In the present paper the results of microscopic investigations are presented and genetic problems of the mineral neoformations are discussed.

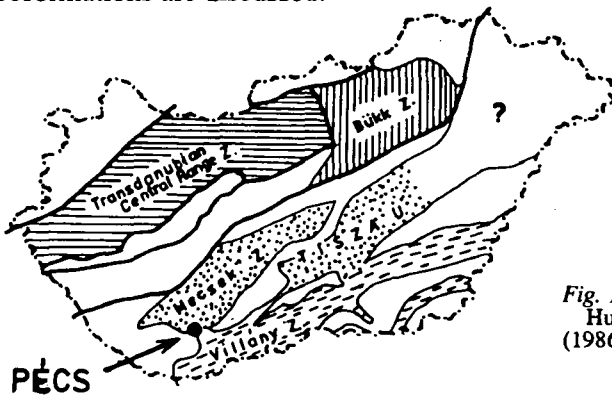


Fig. 1. Permian-Triassic facies units in Hungary according to HAAS *et al.* (1986). The location of the borehole at Pécs is shown.

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LELKES-FELVÁRI (1979, 1983), RÁLISCH (1981) and ORAVECZ-SCHEFFER and GÓCZÁN (1986). A detailed geological description of the region was given by NAGY (1968). Bulk mineralogical composition was determined by VICZIAN (1978, 1985, see Fig. 2) by X-ray diffraction analysis.

A brief macroscopic description of the rock samples investigated in the present study is given in Table 1.

Macroscopic description of the samples

TABLE 1.

1.	125.0—125.5 m	<i>Limestone, dark grey to grey, hard.</i>
2.	268.7—269.4 m	<i>Dolomitic marly limestone, dark grey and light grey angular grains.</i>
3.	272.5—274.0 m	<i>Calcitic dolomite, light grey, brecciated, with light red, loose matrix.</i>
4.	303.7—305.2 m	<i>Marly limestone, dark grey with thin white limestone layers and lenses.</i>
5.	312.4—312.6 m	<i>Marly limestone, dark grey, hard, homogeneous.</i>
6.	345.9—347.0 m	<i>Limestone, dark grey, hard, calcitic veins.</i>
7.	405.0—406.5 m	<i>Dolomite, grey, hard, with thin, dark grey clay layers, calcite veins.</i>
8.	408.0—410.4 m	<i>Limestone, grey, hard, with thin, dark grey clay layers, calcite veins.</i>
9.	503.0—505.0 m	<i>Anhydrite- and gypsum-bearing claystone. Alternation of grey, dark grey clay and white, glittering sulphate layers.</i>
10.	597.4—599.9 m	Alternation of thin a/ and b/ layers: a/ <i>Anhydrite-bearing dolomite marl, dark grey with desiccation cracks.</i> b/ <i>Gypsum- and dolomite-bearing siltstone, grey, with micas and sporadic plant debris.</i>
11.	704.8—707.3 m	<i>Dolomite marl, grey, dark grey, layered, hard.</i>
12.	799.4—800.4 m	<i>Siltstone, greenish and reddish grey, dark grey, layered, with micas and brown magnesite (?) grains.</i>
13.	864.2—864.8 m	<i>Anhydrite-bearing claystone, greenish grey with pink fibrous anhydrite veins and magnesite (?) grains.</i>
14.	878.0—878.6 m	<i>Siltstone. Alternation of thin dark grey and light grey layers with slightly undulating surface.</i>

METHODS

The investigations were carried out in the Institute of Mineralogy and Petrography of the Tübingen University, Germany, using laboratory methods of separation of grain fractions developed by LIPPMANN (see e.g. LIPPMANN and PANKAU, 1988).

Grains of the fraction 63 to 125 μm were scattered over a glass slide and mounted with canada balsam ($n=1.54$). Fractions containing high carbonate content (samples No. 2, 4, 9, 10/a and 10/b) were first examined in their original state and then treated with 0.5 n HCl. The insoluble residue of these fractions was mounted again on a glass slide with canada balsam.

Fractions were investigated under polarizing microscope and in special cases by X-ray diffraction (determination of albite in samples No. 6 and 8 and of magnesite in samples No. 12 and 13).

RESULTS

The results of the microscopic investigations are summarized in Tables 2 and 3.

The following types of mineral grains were found: aggregates of carbonate minerals including magnesite-bearing clay aggregates, carbonate-free clay aggregates, coarse-grained quartz + feldspar + mica + opaque aggregates as well as mineral grains of feldspars, quartz, micas (muscovite, biotite), pyrite, sparry carbonates, celestite, anhydrite and fluorite. In what follows, only aggregates of clay-carbonate composition, feldspar and quartz will be described in detail.

TABLE 2.
Type of carbonate grains, fraction 63 to 125 μm, untreated material, borehole Pécs-IX.

Stratigraphy	Sample No.	Fine-grained aggregates			sparry carbonates	carbonate minerals according to X-ray analysis
		carb.>clay (colour: light)	carb.>clay (colour: dark)	clay>carb.		
Báránytető M.	1.	+++				dol.?
Hetvehely + Viganvár	2.		+++		+	dol.>calc.
	3.	+++	+++		+	dol.
	4.		+++		+	calc.>dol.
Magyar-ürög M.	9.		+		+	magn., dol.?
	10/a. 10/b.		+++ +		(+)	dol. dol.
Patacs F.	12.			+++		magn.
	13.			+	+	magn.
	14.				(+)	—

+++ high
+ medium
+ low
(+) very low

calc.: calcite
dol.: dolomite
magn.: magnesite

Carbonate - clay aggregates including magnesite-bearing clay aggregates

Aggregates containing mainly carbonate minerals could be observed in samples where carbonate minerals were not dissolved by HCl (Table 2). The aggregates are sometimes build up of fine micritic carbonate grains sometimes resembling rhombohedrons. The carbonate aggregates contain clay minerals which are set free

when treated with HCl. The proportion of carbonate to non-carbonate material within the aggregates varies in the section. In the aggregates of samples No. 12 and 13, carbonate (magnesite) is less abundant than non-carbonate clay material. Aggregates of sample No. 14 are carbonate-free even in the untreated material. In other samples carbonate aggregates contain only little non-carbonate material.

TABLE 3.
Results of the microscopic investigation of the fraction 63 to 125 μ m, non carbonate minerals (carbonate-free samples or samples treated with 0.5n HCl), bore hole Pécs-IX

Stratigraphy	Sample No.	Fine-grained aggregates (dark grey, clay-pyrite)	Coarse-grained aggr. (q+fp+mica+opaque)	Quartz (detrital)	Quartz (euhedral)	Quartz (thin platelets)	Feldspar (detrital)	Feldspar (authigenic albite)	Mica (Muscovite)	Mica (biotite-Chlorite)	Pyrite	Celestite
Báránytető M.	1.		(+)			(+)			(+)			
Hetvehely+ Vigan-vár Members	2.	+	+		+		+			(+)	+	
	3.		+	+	(+)	+	(+)			(+)	+	(+)
	4.			+	+++		+				+	
	5.	+		+			(+)				+	
	6.	+					+	+			(+)	
	7.	+			(+)	(+)	+	(+)	(+)			+
	8.	+			(+)			+	(+)		+	
	9.	+	+	+	+		+	+				
Magyarürög M.	10/a.	+++	+	+			+				+	
	10/b.	+	+++	+		(+)	+		+			
	11.	+++			(+)		(+)		+	(+)		
Patacs F.	12.	+++					+		(+)			
	13.	+	+	+	+		+		(+)	(+)		(+)
	14.	+++		+					(+)	(+)		

+++ high
+ medium
+ low
(+) very low

q: quartz
fp: feldspar

As determined by comparison with X-ray analysis of the bulk samples most carbonate grains are dolomite. In samples No. 9, 12 and 13 magnesite was found by X-ray analysis. According to the shift of the reflection 104 toward higher d

values approximately one fourth to one third of Mg^{2+} is replaced by other ions, most probably Fe^{2+} :

	2 Θ	d
magnesite, JCPDS card No. 8-479:	32.65°	2.742 Å
samples No. 12 and 13:	32.5 °	2.76 Å
siderite, JCPDS card No. 29-696:	32.02°	2.795 Å.

A similar shift of the reflections 113 and 116 can be observed.

Magnesite has been found in the Eastern Alps and in Germany in rocks having similar stratigraphic and environmental relations. In particular, in the Drauzug area of the Eastern Alps magnesite is a normal constituent of terrigenous and evaporitic sedimentary rocks at the boundary of Upper Scythian - Lower Anisian in a zone extending 140 km (NIEDERMAYR *et al.* 1980, NIEDERMAYR 1989, NIEDERMAYR. *et al.* 1989). Magnesites found here may have Fe contents up to 20 weight per cent. Their genesis is related to deposition in "a broad coastal plain under transitional conditions between a fluvial setting and a coastal sabkha environment" (NIEDERMAYR *et al.* 1989, p. 55). In Germany, magnesite was determined by X-ray diffraction in gypsum-bearing silty sediments in a profile at Borken, Hesse (BÜHMANN and RAMBOW 1979). The age of these rocks is Upper Spathian (Upper Buntsandstein, Rötalinär) close to the boundary of Anisian (DOUBINGER and BÜHMANN, 1981).

Feldspars

Feldspars occur both as detrital grains and euhedral crystals. Feldspars were identified by their interference figure, optic sign, extinction angle and variation of index of refraction in relation to canada balsam.

Few grains of *detrital feldspar* of irregular shape and inhomogeneous extinction can be observed in nearly all samples. Both potassium feldspar and plagioclase occur, twinning is very rare. The grains are cloudy, altered and contain inclusions of carbonates, sometimes also pyrite and micas.

In limestone samples No. 5, 6 and 8 many *euhedral feldspar* crystals with well-developed crystal faces can be observed (Figs. 3 and 4). The rounded internal core of these crystals is altered and contains inclusions of carbonate grains, "sericite", pyrite and yellowish brown or black microlites. The core is surrounded by a clear overgrowth of the same optical orientation. No difference in the extinction angle of the core and the rim could be observed. Euhedral feldspars are untwinned. Roc Tourné type twins as described by FÜCHTBAUER (1948) cannot be observed because almost all crystals are oriented parallel to (010). The form of the crystals is usually an elongated hexagon formed by the traces of crystal faces (001), (110) and (111). Less frequently grains in other orientations, probably with the face (001) parallel to the plane of the preparation, occur.

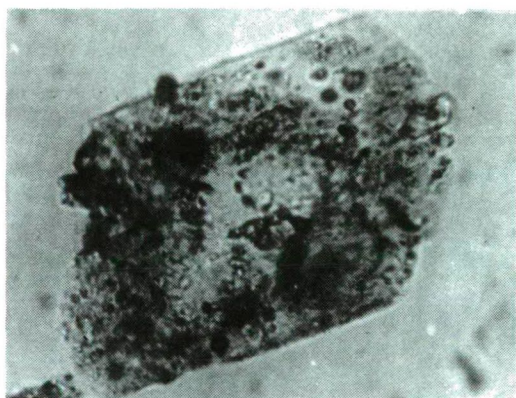
The index of refraction is lower than that of canada balsam. The relief is low but varies with the orientation of the grain if the crystal face (001) is nearly parallel to the plane of the preparation. Extinction is parallel to the elongation on (001) faces and oblique on (010) faces. Biaxial positive extinction figures with large axial angles can be observed on faces (010). Interference colours are most frequently yellow but vary from first order grey to blue. The optical properties can be interpreted as those of *low-albite* (ordered). As measured by extinction angles of the few albite twins observed the mineral may contain about 10 per cent anorthite molecule.

X-ray diffraction analysis was made of the samples No. 6 and 8 in order to prove the optical determination. The grains of the fraction 63 to 125 μm of the material previously treated with 0.5 n HCl were mounted on a glass slide with few drops of distilled water. The feldspar reflections are clearly those of low-albite (as compared with the JCPDS card No. 20-554). The intensities of the following reflection are clearly stronger than they should be in a powder free of preferred orientation:

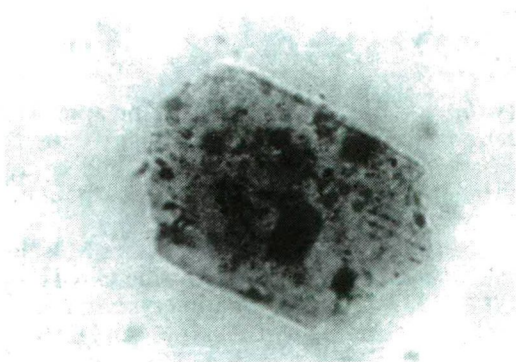
d (\AA)	hkl
6.38	001, 020
3.19	002, 040
2.125	060
1.594	080.

This is in agreement with the optical observation that most albite crystals have faces (010) or (001) parallel to the plane of preparation.

Authigenic albites were frequently reported from limestones and related rocks, e.g. from Liassic-Dogger of Zweisimmen, Switzerland (FÜCHTBAUER 1948), Muschelkalk of Göttingen (FÜCHTBAUER 1950), Zechstein near Braunschweig (FÜCHTBAUER 1956). Similar authigenic overgrowths of albite were observed in gypsum and gypsum-bearing claystone in the Muschelkalk of Nagold by LIPPMANN and PANKAU (1988).



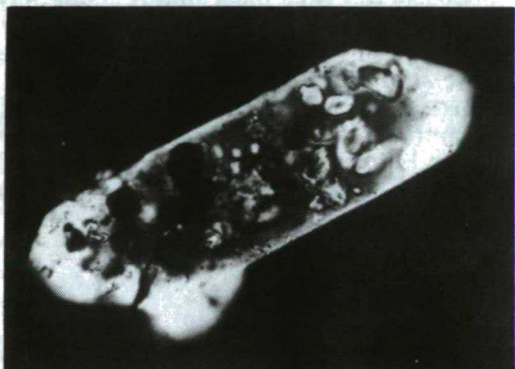
Figs. 3 and 4. Authigenic albites from sample No. 2, dolomitic marly limestone, borehole Pécs-IX, 268.7 to 269.4 m, Anisian, Viganvár Member. || N, 500X (left), 300X (right).



Quartz

Few *detrital* quartz grains of irregular shape were observed in almost all samples except pure carbonate rocks.

Euhedral quartz of regular prismatic shape terminating on one or both ends by rhombohedrons is very frequent in the insoluble residue of the limestone samples No. 2 and 4 but it occurs in many other samples too. The external rim of these crystals is clear but the core contains many inclusions such as carbonates and brownish-black opaque microlites (Figs. 5 and 6). This variety of quartz is considered to be authigenic. In some cases very small prismatic overgrowths can be observed on the surface of clay mineral aggregates (sample No. 13) or overgrowths of irregular shape on detrital quartz grains (samples No. 3, 10/a and 13) may occur.



Figs. 5 and 6. Authigenic quartzes from sample No. 6, limestone, borehole Pécs-IX, 345.9 to 347.0 m, Anisian, Hetvehely Member. +N, 300X.

Another variety of quartz are very *thin platelets* of irregular shape apparently perpendicular to the optic axis in direction *c* giving clear uniaxial positive interference figures. Also the plates may have inclusions of carbonate and opaque minerals. This variety is most abundant in sample No. 3 (so-called "boundary dolomite").

Euhedral prismatic quartz crystals were described from the Upper Muschelkalk at Haigerloch by LIPPMANN and SCHLENKER (1970) and from the Gipskeuper at Pfäffingen by LIPPMANN and STEINER (1983). Both prismatic crystals and thin plates were found in the Middle Muschelkalk at Nagold by LIPPMANN and PANKAU (1988).

Magnesite

The magnesites of the Drauzug area are considered by NIEDERMAYR (1989) to be converted from preexisting carbonates in an early stage of diagenesis. The genesis of magnesite will be considered here from the point of view of stability relations of rhombohedral carbonates in aqueous solutions (LIPPMANN 1980, 1982a, b).

Considering the stable phase diagram for the system $\text{CaCO}_3 - \text{MgCO}_3 - \text{H}_2\text{O}$ at 25 °C (LIPPMANN, 1982a) magnesite should crystallize from solutions having activity fractions $X_1 [\text{Mg}^{2+}]$ higher than 0.832. Magnesites should crystallize even from normal sea water because in sea water $X_1 [\text{Mg}^{2+}] = 0.865$ (LIPPMANN 1982b). The formation of magnesite, however, is inhibited at ordinary temperature by the hydration of Mg^{2+} ions, even if seeds of magnesite are present in the solution. Even at elevated activity fractions of Mg^{2+} either aragonite or hydrous magnesium carbonates such as nesquehonite, $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$, or hydromagnesite, $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$, precipitate firstly.

The transformation of these metastable phases into magnesite takes place first at elevated temperatures. Also huntite, $\text{CaMg}_3(\text{CO}_3)_4$, may be a metastable precursor of magnesite (LIPPMANN 1982a and LIPPMANN 1980).

As the activity fraction $X_1 [\text{Mg}^{2+}]$ of sea water, 0.865, is near to the eutectic point of the dolomite-magnesite pair, 0.832, a further enrichment of Mg^{2+} ions in the solution may favour the precipitation of magnesite. Simultaneous precipitation of gypsum may extract Ca^{2+} ions from the solution and produce high Mg^{2+} to Ca^{2+} ratios. This may be the case in the sequence studied where magnesite is accompanied with gypsum and anhydrite layers.

On the other hand, no concurrent formation of Mg-rich silicates such as chlorite, corrensite, talc, sepiolite or palygorskite could be observed in the portion of the sequence containing magnesite. All these minerals would be able to form as stable or metastable phases from high magnesian solutions according to the stability relations of these minerals in the system $\text{MgO} - \text{Al}_2\text{O}_3 - \text{SiO}_2 - \text{H}_2\text{O}$ (LIPPMANN 1979). The crystallization of clinocllore (chlorite, corensite) may not have taken place due to insufficient Al_2O_3 . No sepiolite or attapulgitite were formed instead, probably because of low activity of silica. Talc forms very slowly at low temperatures.

The high iron content presumed to be present in the magnesites studied can be explained by the solubility relations in the system $\text{MgCO}_3 - \text{FeCO}_3 - \text{H}_2\text{O}$ (LIPPMANN 1980 and LIPPMANN 1982b). By comparison of the solidus and solutus lines in the solubility diagrams of this system it is evident that high Fe concentrations in the solid are in equilibrium with rather low concentrations of Fe in the solution. In other words Fe readily precipitates from solution together with Mg to form carbonates. The high ferrous iron concentration is a consequence of the reducing conditions during diagenesis.

Albite

Albite was found in limestones of the Hetvehely Member deposited in sea water of normal salinity. Considering the stability relations in the system $\text{K}_2\text{O} - \text{Al}_2\text{O}_3 - \text{SiO}_2 - \text{H}_2\text{O}$ (LIPPMANN 1982c) and in the system $\text{Na}_2\text{O} - \text{Al}_2\text{O}_3 - \text{SiO}_2 - \text{H}_2\text{O}$ (unpublished diagram of LIPPMANN see Fig. 7) it is obvious that silica must be enriched in the solution as compared with normal sea water in order to reach

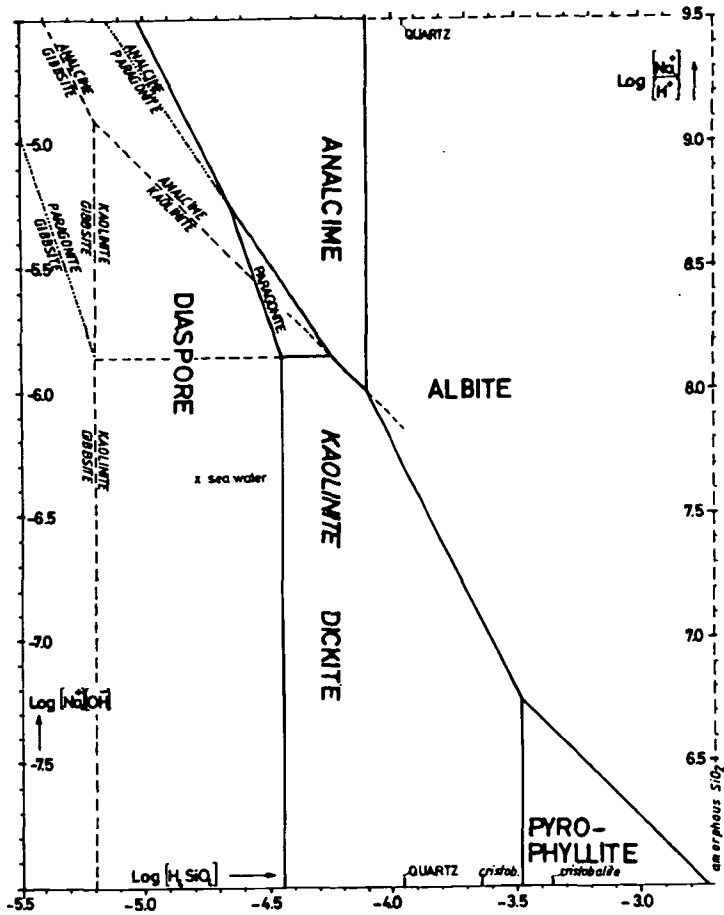


Fig. 7. The system $\text{Na}_2\text{O} - \text{Al}_2\text{O}_3 - \text{SiO}_2 - \text{H}_2\text{O}$ at 25°C with Al_2O_3 as immobile component calculated by LIPPMANN (unpublished diagram, improved version on Fig. 4 in LIPPMANN 1979). Explanations: CAPITALS: stable minerals. *ITALICS*: metastable minerals reacting in metastable equilibrium; *italics*: unstable minerals; continuous lines: stable phase boundaries; dashed and dotted lines: metastable phase boundaries.

the stability fields of feldspars. In the presence of sufficient silica, the type of the feldspar crystallized depends on the K^+ and Na^+ activity in the solution. The point for the composition of sea water is near the boundary of the stability fields of plagioclase and K-feldspar in the diagram $[\text{K}^+] \cdot [\text{OH}^-]$ versus $[\text{Na}^+] \cdot [\text{OH}^-]$ (unpublished diagram of LIPPMANN see Fig. 8). The composition of the solution may easily move into the stability field of albite if e.g., potassium is removed from solution by the reorganization of poorly crystalline micas or mixed-layer illite/smectites.

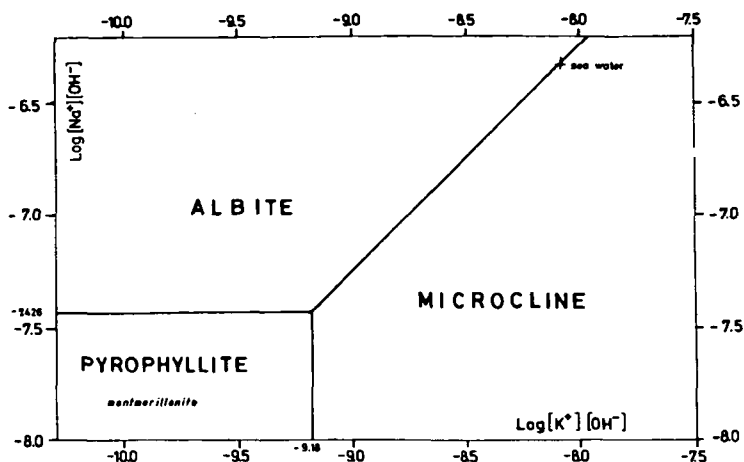
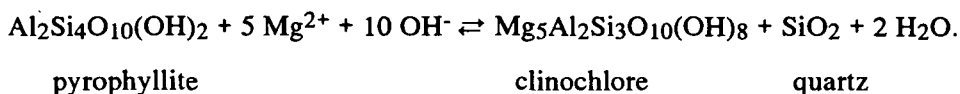


Fig. 8. The system $K_2O - Na_2O - Al_2O_3 - SiO_2 - H_2O$ at 25 °C with Al_2O_3 as immobile component and $\log [H_4SiO_4] = -3.3$ (30 mg/l SiO_2). Unpublished diagram constructed by LIPPMANN (1988) using the thermodynamic data of ROBIE *et al.* (1978).

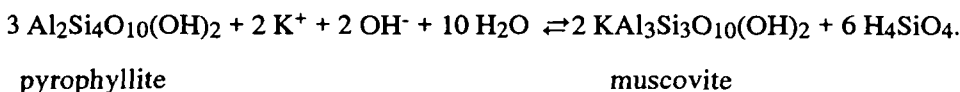
Explanations: CAPITALS: stable minerals, *italics*: unstable mineral. Note that the composition of sea water coincides with the boundary of the stability fields of albite and microcline within the accuracy limits of the data.

Quartz

Neof ormation of minor amounts of quartz has been observed in nearly all samples but authigenic quartz crystals are frequent only in marly limestones of the Hetvehely and Viganvár Members. The neof ormation of quartz coincides in these cases with the occurrence of corrensite in the clay fraction. It is probable that corrensite has formed by reaction of a smectite-like detrital clay material with solutions rich in Mg^{2+} and alkalinity (VICZIÁN 1993). The model of this transformation is the reaction of the end member pyrophyllite to clinochlore (Table 5 in LIPPMANN, 1979):



Quartz is a product of this reaction. Another source of silica may be the transformation smectite to illite which can be described by the reaction of the end member pyrophyllite to muscovite (Table 2 in LIPPMANN 1979):



Thus both the formation of corrensite and illite may produce quartz during diagenesis.

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