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Autor: Frey, Martin

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The reaction-isograd kaolinite + quartz = pyrophyllite + H₂O, Helvetic Alps, Switzerland*

by Martin Frey¹

Abstract

The reaction-isograd kaolinite + quartz = pyrophyllite + H₂O has been mapped in the Helvetic Alps, Switzerland, over a distance of about 200 km. Metamorphic conditions at the reaction-isograd were estimated from vitrinite reflectance and fluid inclusion data to be 1.3–2.1 kbar and 240–270 °C at a water activity of 0.6–0.8. Textural evidence obtained with a scanning electron microscope suggests that pyrophyllite grew through a dissolution-precipitation process.

Keywords: Isograd, kaolinite, pyrophyllite, metamorphic conditions, Helvetic Alps.

1. Introduction

Kaolinite, Al₂Si₂O₅(OH)₄, is a widespread clay mineral in clastic sedimentary rocks, where it occurs as a detrital constituent in near shore deposits as well as an authigenic phase in sandstones (see e.g. MILLOT, 1970). In addition, kaolinite is also known as a residual mineral from laterites. Pyrophyllite, Al₂Si₄O₁₀(OH)₂, on the other hand, has long been regarded as an essentially hydrothermal mineral (e.g. DEER et al., 1962, p. 119) but its occurrence during incipient regional metamorphism is now well established. More than sixty references referring to such pyrophyllite-bearing assemblages in shales, slates, phyllites, quartzites and conglomerates have been compiled by FREY (1987).

This paper documents the mapping of a low-temperature reaction-isograd pertaining to the reaction Al₂Si₂O₅(OH)₄ + 2 SiO₂ = Al₂Si₄O₁₀(OH)₂ + H₂O.

2. Mineral identification

Minerals were identified on a routine basis with aid of X-ray powder diffractometry complemented by Guinier camera technique. For kaolinite the first basal reflection coincided with the (002)-reflection of chlorite; but the second order basal reflection at 3.60 Å was clearly resolved from the (004)-reflection of chlorite at 3.56 Å (Fig. 1a, sample UB 171). Pyrophyllite was easily detected by its first three basal reflections at 9.2, 4.6 and 3.06 Å (Fig. 1b, c, samples MF 1243, MF 616) and was distinguished from talc by its (060)-reflection at 1.49 Å.

One sample containing both kaolinite and pyrophyllite was studied with a Phillips 515 scanning electron microscope (SEM) equipped with a Tracor Northern energy-dispersive spectrometer. Using a polished thin-section and working with backscattered mode provided textural and mineral chemical information (which will be discussed in section 5).

* Dedicated to Professor Ernst Niggli on the occasion of his 70th birthday.

¹ Mineralogisch-Petrographisches Institut der Universität, Bernoullistrasse 30, CH-4056 Basel.

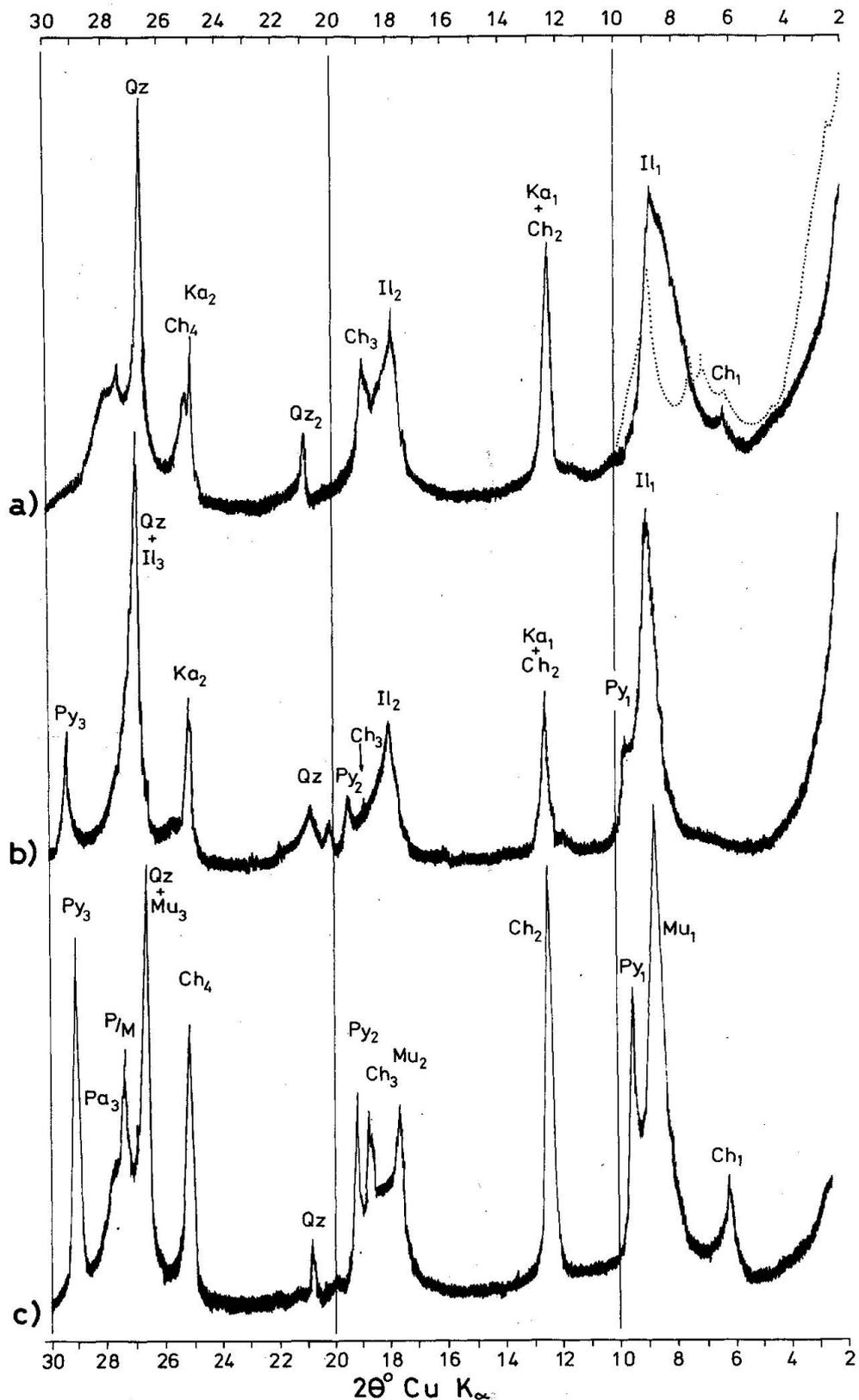


Fig. 1 X-ray diffractograms of three representative samples, < 2 μm fractions.

a) Kaolinite-bearing sample from locality 4 (cf. Fig. 2 and appendix). The stippled trace refers to the glycolated sample.

b) Kaolinite- and pyrophyllite-bearing sample from locality 51.

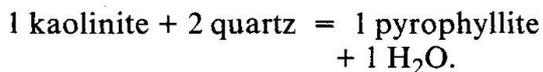
c) Pyrophyllite-bearing sample from locality 64.

Mineral abbreviations used: Ch = chlorite, Il = illite, Ka = kaolinite, Mu = muscovite, Pa = paragonite, P/M = paragonite/muscovite mixed-layer, Py = pyrophyllite, Qz = quartz.

Ch₁, Ch₂, ..., refer to the first and subsequent basal reflections of chlorite, etc.

3. Regional distribution of kaolinite + quartz vs pyrophyllite

The regional distribution of kaolinite + quartz and of pyrophyllite in the Helvetic nappes and the Prealps of Switzerland is shown in Fig. 2. For tectonic and palaeogeographic reasons these mineral assemblages have been found in various stratigraphic positions. Kaolinite + quartz occur mainly in Cretaceous marls and marly limestones while pyrophyllite has been discovered in Jurassic black shales as well as in Jurassic and Eocene laterites. Detailed information on sample localities (coordinates, tectonic and stratigraphic position), mineral assemblages and sources of data are given in the appendix. If the pyrophyllite occurrences of localities 85 and 88 are omitted for the moment, then a simple and regular distribution pattern emerges. Localities with kaolinite + quartz are found in a more external position in tectonically higher units, whereas pyrophyllite occurrences are found in a more internal zone in tectonically lower units. This mineral distribution pattern strongly suggests that kaolinite + quartz provides the starting material for the appearance of pyrophyllite by virtue of the reaction



In several areas the most internal occurrence of kaolinite + quartz and the most external occurrence of pyrophyllite are separated by a distance of some 10 km. In some cases this may be due to insufficient sampling, but in other cases this "gap" is lithologically controlled as e.g. in the Reuss valley SE of Lake Lucerne (BREITSCHMID, 1982). At four localities (Nos. 51–54), however, both kaolinite + quartz and pyrophyllite were found in the same specimen, while at locality 55 these sheet silicates occur in different specimens.

The presence of pyrophyllite at localities 85 and 88 will be discussed next. In the case of locality 85 this may be explained by an inverse metamorphic zonation, which is well documented by illite "crystallinity" (BAUD, 1984), coal rank (KÜBLER et al., 1979) and fluid inclusion data (MULLIS, 1979). The same explanation may be put forward also for locality 88 which belongs to the Ultrahelvetica "Zone des Cols", but metamorphic grade of this tectonic unit is not well constrained. A second possible

explanation takes the mineral assemblage into consideration, which yields some information about the composition of the fluid phase. At locality 88 pyrophyllite coexists with organic material, while at the nearby locality 29 kaolinite + quartz is found in a hematite-bearing specimen. Fluid inclusion studies by MULLIS (1979) have documented methane-rich fluids at such very low metamorphic grade, derived from the cracking of organic material. Therefore, in a rock containing organic material, a reduced water activity ($a_{\text{H}_2\text{O}} < 1$) is to be expected at these low temperatures while in a hematite-bearing rock the water-rich fluid phase will not be diluted by methane ($a_{\text{H}_2\text{O}} \approx 1$). For these reasons it seems plausible that, assuming constant temperature and fluid pressure, an assemblage containing kaolinite + quartz + organic material could have reacted to yield pyrophyllite while in another sample the assemblage kaolinite + quartz + hematite could still persist.

4. Metamorphic conditions at the reaction-isograd

The pressure (P), temperature (T) and fluid composition (X) conditions at the ka + qz = py + H₂O reaction-isograd can be estimated from vitrinite reflectance and fluid inclusion data. These results may then be compared with experimentally determined or thermodynamically calculated P–T data for the reaction ka + qz = py + H₂O.

4.1. COAL RANK DATA

The various coalification models have recently been reviewed by BUSTIN et al. (1983, 1985), ROBERT (1985) and KISCH (1987). While temperature is regarded uniformly as the main controlling variable, the role played by the time factor is controversial and ranges from "important" to "negligible". We have chosen here the model of BOSTICK et al. (1979) because it is widely accepted in the literature. This graphical method provides a relationship between vitrinite reflectance, maximum temperature and effective heating time (the time span during which the coal was within 15°C of its maximum temperature). Using an effective heating time of 5–10 Ma (STALDER, 1979, p. 466) and vitrinite reflectivity data given by

Tab. 1 P-T estimates for four localities along the reaction-isograd $ka + qtz = py + H_2O$

Locality*	coal rank data					fluid inclusion data			
	sample	coordinates	Rm (%)	Rmax (%)	T (°C)**	sample	coordinates	T (°C)	P (bar)
51	MF 1243	752.5/219.6	3.1	3.5	240	JM 77	750.2/217.7	236	2140
	MF 1244	752.5/219.6	3.2	3.5	240				
52	MF 582	738.7/217.3	4.7	5.2	260				
	MF 585	738.7/217.3	4.9	5.2	260				
54	BK 75/99	624.2/151.7	4.3	4.9	250	JM 9	623.7/151.8	262	1400
						JM 8	625.0/152.3	263	1340
						JM 10	625.0/151.4	267	1850
						JM 11	625.1/152.0	263	1710
						JM 12	625.4/151.6	257	—
55						JM 224	558.0/111.1	248	1750

* cf. Fig. 2 and appendix

** after BOSTICK et al. (1979)

FREY et al. (1980), maximum temperatures for three localities along the reaction-isograd range between 240 and 260°C (Table 1). Note that much higher temperatures of 300–370°C are obtained for the time-independent coalification models of BARKER (1983) and PRICE (1983).

4.2. FLUID INCLUSION DATA

MULLIS (1979, 1987) developed a method to determine approximative P and T values of fluid trapping based on fluid inclusion studies from fissure quartz. This microthermometric method takes advantage of immiscibility in H₂O-CH₄ fluids. As a result, the homogenization temperature (T_h) of water-rich inclusions saturated with CH₄ can be interpreted as temperature of formation. Methane-rich inclusions, on the other hand, allow the measurement of the density of CH₄ (ρ_{CH_4}). With these T_h and ρ_{CH_4} data the pressure of inclusion formation can be determined from experimental P-V-T-X properties.

P-T data for three localities along the reaction-isograd range between 1340–2140 bars and 236–267°C, respectively (Table 1). For localities 51 and 54 temperatures are also available from coal rank studies, and both methods yield remarkably similar results.

Because fissure quartz was generated possibly after the temperature climax of the meta-

morphic history, the above mentioned P-T data derived from fluid inclusions studies may be regarded as minimum values. However, these P-T values are believed to be near the conditions which prevailed during regional metamorphism (MULLIS, 1979, p. 535), and this

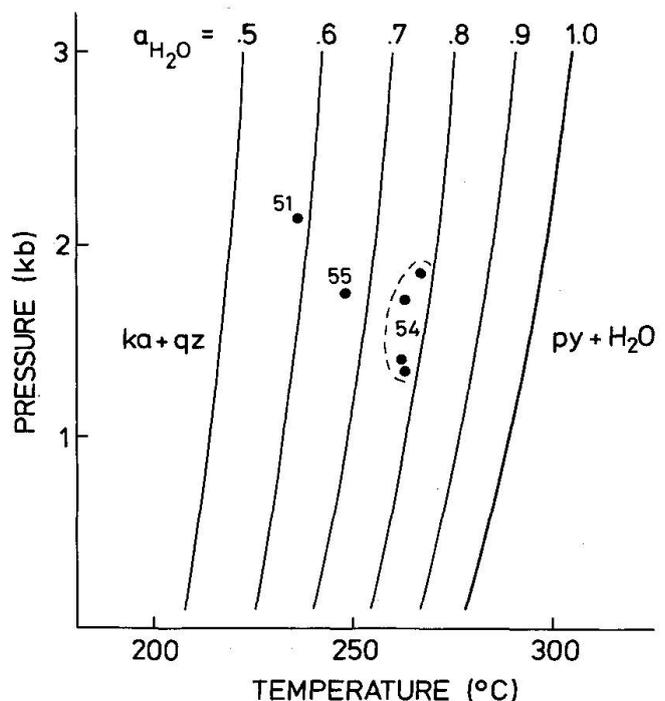


Fig. 3 Calculated P-T stability limits for the reaction kaolinite + quartz = pyrophyllite + H₂O at different water activities. The P-T conditions from Table 1 for localities 51, 54 and 55 at the reaction-isograd are indicated.

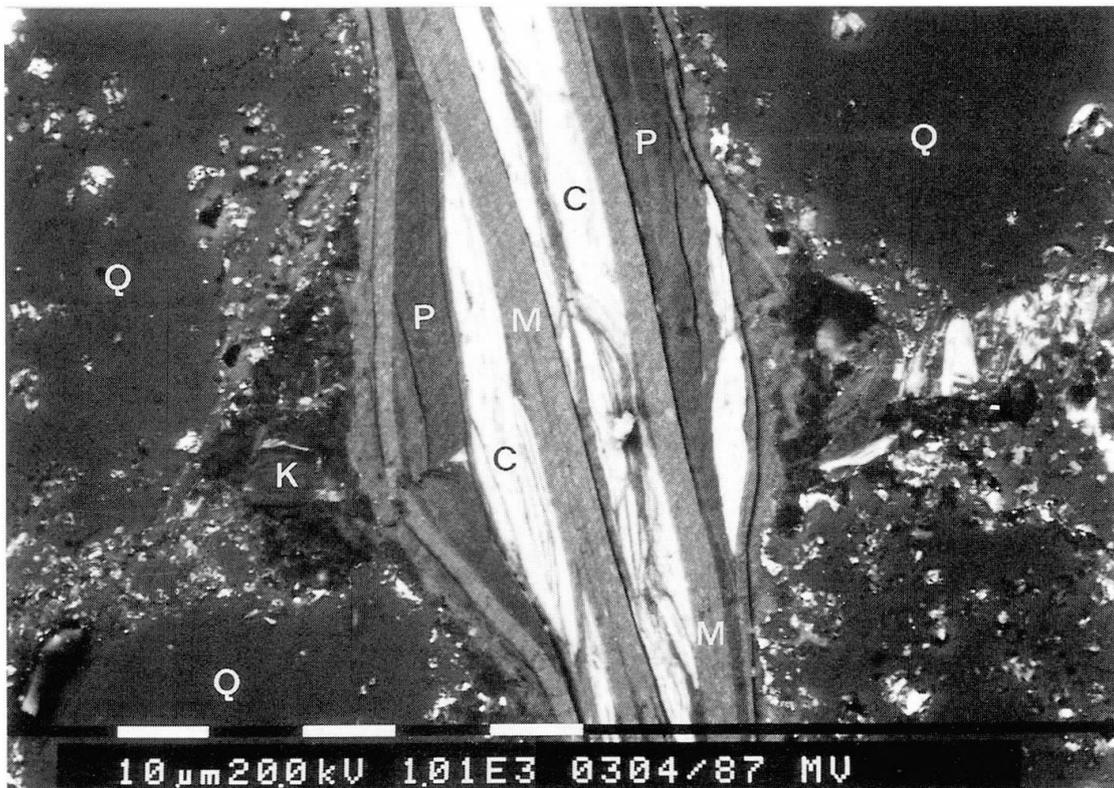
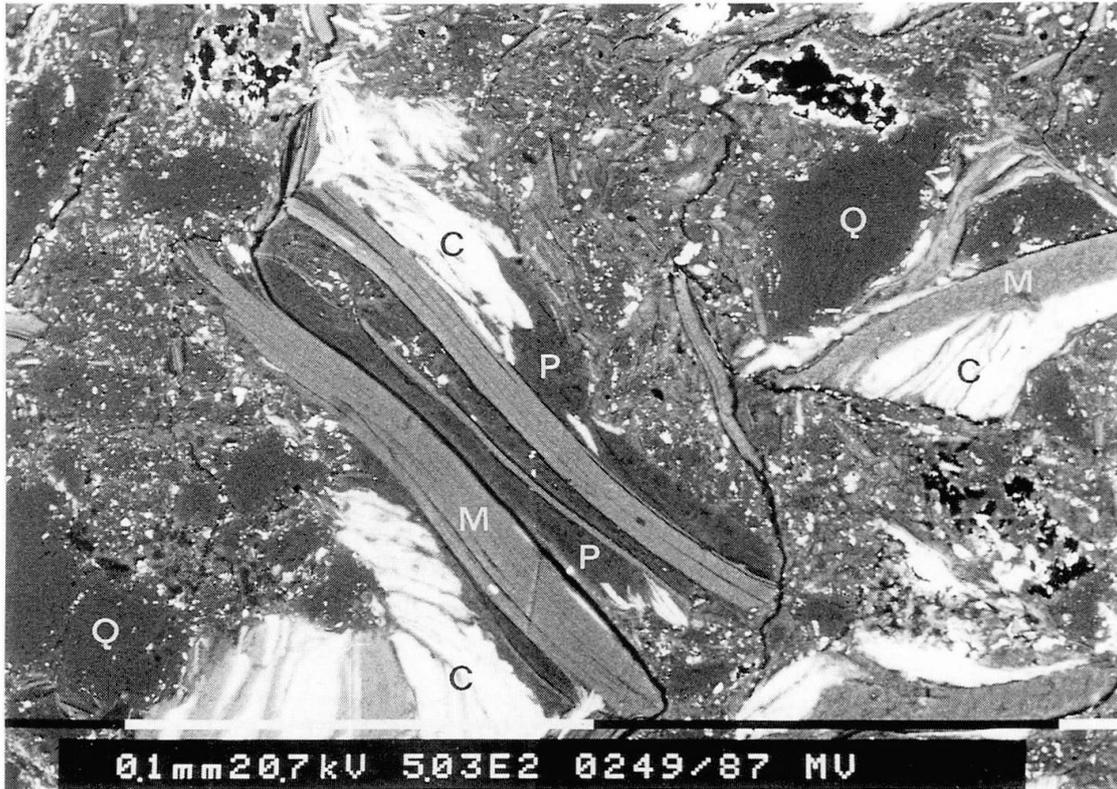


Fig. 4 SEM backscattered images of sample MF 2306 from locality 53.

A: Lamellar intergrowth of pyrophyllite and muscovite.

B: Lamellar intergrowth of pyrophyllite, muscovite and chlorite.

Mineral abbreviations: C = chlorite, K = kaolinite, M = Muscovite, P = pyrophyllite, Q = quartz.
See text for discussion.

conclusion is supported by the temperatures obtained from vitrinite reflectivity data.

4.3. WATER ACTIVITY AT THE REACTION-ISOGRAD

The equilibrium curve of the reaction $ka + qz = py + H_2O$ in P_{H_2O} - T space was calculated at desired values of a_{H_2O} using the thermodynamic database of BERMAN et al. (1985), (see Figure 3). If the P - T conditions along the reaction-isograd from Table 1 are plotted in this diagram, a_{H_2O} values in the range from 0.6 to 0.8 are obtained. These rather low values of a_{H_2O} are corroborated by fluid inclusion studies from the area of Figure 2 (MULLIS, 1979).

4.4. COMPARISON WITH OTHER AREAS

There is ample field evidence from other areas that kaolinite + quartz give way to pyrophyllite with increasing metamorphic grade (e.g. SCHRAMM, 1978; GOFFÉ, 1982; JUSTER and BROWN, 1984; FRANCESCHELLI et al., 1986). A corresponding reaction-isograd has been mapped by SCHRAMM (1978; see also FREY, 1986, Plate 1) over a distance of 15 km in Permian metaclastic sediments of the Eastern Alps. JUSTER and BROWN (1984) and FRANCESCHELLI et al. (1986), on the other hand, observed coexisting kaolinite + pyrophyllite + quartz over a range of conditions. This phenomenon might be explained by: the metastable persistence of kaolinite or the retrograde formation of the same phase; internal fluid buffering (this seems to be a reasonable explanation for the hematite-bearing mineral assemblages described by FRANCESCHELLI et al., 1986); or the effect of immiscibility in H_2O - CH_4 fluids as discussed by JUSTER and BROWN (1984) and CRAWFORD and HOLLISTER (1986, p. 17).

5. Textural evidence and reaction mechanism

A shale to slate from locality 53 was studied with a scanning electron microscope (SEM) in order to study textural relationships between

reactants and products at the reaction-isograd $ka + qz + H_2O$. It was found that pyrophyllite is preferentially localized within relatively large detrital muscovite (Fig. 4a) or sandwiched between muscovite and chlorite (Fig. 4b) sharing their (001) planes. Pyrophyllite was much less frequently observed in the matrix and only rarely found in direct contact with kaolinite. Note, for example, that xenomorphic kaolinite in Fig. 4b is not overgrown by pyrophyllite.

The intergrowth of pyrophyllite and muscovite has previously been described. CHENNAUX and DUNOYER (1967) and FREY (1978, p. 106) observed rims of pyrophyllite on detrital grains of muscovite in anchimetamorphic slates; ENGLAND (1972) reported lamellar intergrowth of pyrophyllite and muscovite in quartzites from the kyanite-pyrophyllite-quartz zone.

This textural evidence may be interpreted as a topotactic growth of pyrophyllite on muscovite, while an exsolution process seems less plausible. These two sheet silicates possess very similar crystal structures with two tetrahedral layers and one octahedral layer, abbreviated as *tot*, while kaolinite consists of *to* structural units. Obviously kaolinite was dissolved (as suggested by its irregular grain shape) and pyrophyllite was precipitated on the structurally similar muscovite for kinetic reasons.

Acknowledgements

Beat Künzi and Albert Matter provided information on mineral assemblages. Josef Mullis furnished new fluid inclusion data for locality 55. M. Düggelin und R. Guggenheim, SEM-Lab. University of Basel, supplied me with the SEM information. Steve Ayrton kindly improved the English. Thoughtful reviews were provided by N.D. Chatterjee, Christian Chopin, Bruno Goffé and Josef Mullis. To all these friends and colleagues my sincere thanks.

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Appendix:

Occurrences of kaolinite + quartz and pyrophyllite.

No.	#	Coordinates	Tectonic/Stratigraphic Position	Assemblage	Reference
1	5	745.55/234.3	Säntis nappe/Oehrli Fm + Vitznau Marl Fm	KQ IC CcDo 0	Burger (1982)
2	3	741.8/234.05	Säntis nappe/Oehrli Fm	KQ IC Cc 0	Burger (1982)
	2	742.8/235.0	Säntis nappe/Oehrli Fm	KQ IC Cc 0	Burger (1982)
3	11	736.0/228.2	Säntis nappe/Oehrli Fm + Vitznau Marl Fm	KQ IC CcDo 0	Burger (1982)
4	3	749.9/222.3	Säntis nappe/Drusberg Fm	KQ IC Cc 0	Briegel (1972)
5	1	748.9/219.0	Churfürsten-Drusberg nappe/Palfris Fm	KQ IC Cc 0	Burger (1982)
6	6	718.9/223.1	Subalpine Molasse/"Rupelian"	KQ IC CcDo 0	This study
7	6	714.6/203.9	Axen nappe/Oehrli Fm + Vitznau Marl Fm	KQ IC CcDo 0	Burger (1982)
8	9	708.8/206.8	Drusberg nappe/Palfris Fm + Vitznau Fm	KQ IC Cc 0	Burger (1982)
9	3	702.25/199.2	Axen nappe/Vitznau Marl Fm	KQ IC Cc 0	Burger (1982)
10	2	690.15/201.25	Drusberg nappe/Vitznau Marl Fm	KQ IC Cc 0	Breitschmid (1980, 1982)
11	1	689.7/199.15	Axen nappe/Orbitolina beds	KQ IC Cc 0	Breitschmid (1980, 1982)
12	4	685/207*	Border chain/Orbitolina beds + Vitznau Fm	KQ IC Cc 0	Breitschmid (1980, 1982)
	2	688.6/206.5	Ultrahelvetic Flysch	KQ IC Cc 0	Breitschmid (1980, 1982)
13	4	681.1/206.7	Border chain/Vitznau Marl Fm	KQ IC Cc 0	Burger (1982)
14	8	685/199*	Drusberg nappe/Vitznau Marl Fm	KQ IC Cc 0	Breitschmid (1980, 1982)
15	2	687.6/197.5	Axen nappe/Vitznau Marl Fm	KQ IC Cc 0	Breitschmid (1980, 1982)
16	2	681.3/199.3	Ultrahelvetic Flysch	KQ IC Cc 0	Breitschmid (1980, 1982)
	1	680.8/197.5	Drusberg nappe/Orbitolina beds	KQ IC Cc 0	A. Matter, pers. comm.
17	7	668.3/192.8	Drusberg nappe/Schrattenkalk	KQ IC Cc 0	Korner (1975)
18	5	661.9/203.2	Border chain/Orbitolina beds	KQ IC Cc 0	A. Matter, pers. comm.
19	15	653.85/201.15	Border chain/"Berriasian" Subalpine Flysch	KQ IC Cc 0	Ischi (1973)
20	13	659/198*	Schlieren Flysch	KQ IC Cc 0	Winkler (1983)
21	1	647.2/189.7	Schlieren Flysch	KQ IC Cc 0	Winkler (1983)
	2	648.0/194.1	Schlieren Flysch	KQ IC Cc 0	Winkler (1983)
22	9	649.7/181.65	Wildhorn nappe/Orbitolina beds	KQ IC Cc 0	A. Matter, pers. comm.
23	?	622/178*	Subalpine Flysch	KQ ISC CcDo 0	Dadelahi (1982)
24	1	623.2/164.4	Ultrahelvetic Flysch	KQ IC Cc 0	Frey et al. (1980)
25	?	621.2/151.7	Wildhorn nappe/"Berriasian"	KQ ... 0	B. Künzi, pers. comm.
26	40	596/177*	Gurnigel Flysch	KQ ISC Cc 0	van Stuijvenberg (1979)
27	9	595/173*	Gurnigel Flysch	KQ ISC Cc 0	van Stuijvenberg (1979)
28	1	560/145*	Gurnigel Flysch	KQ IC Cc 0	van Stuijvenberg et al. (1976)
29	4	580.85/131.75	Wildhorn nappe/Eocene palaeokarst	KQ IC H	Wieland (1979)
30	1	579.35/124.55	Morcles nappe/Eocene palaeokarst	KQ IC	Wieland (1979)
51	1	752.5/219.55	Säntis nappe/Drusberg Fm	PKQ IC Cc 0	This study
52	4	741/220*	Mürtschen nappe/Mols Fm	PKQ IRPaC CcDo 0	Frey et al. (1982)
53	1	725.8/210.6	Glarus nappe/Bommerstein Fm	PKQ I Cc 0	Frey et al. (1982)
54	1	623.8/151.7	Wildhorn nappe/"Aalenian"	PKQ ... 0	B. Künzi, pers. comm.
55	2	558.1/111.95	Morcles nappe/Eocene palaeokarst	KQ IC H PQ C	Wieland (1979)
61	6	750.8/214.3	Axen nappe/Mols Fm	PQ MPaC Cc 0	Frey & Wieland (1975)
62	2	744.2/209.65	Axen nappe/"Infralias"	PQ MPaC 0	Frey & Wieland (1975)
63	13	739.8/207.4	Axen nappe/"Infralias"	PQ MPaC Cc 0	Frey & Wieland (1975)
	1	740.3/208.6	Axen nappe/"Infralias"	PQ MPaC Cc 0	Frey & Wieland (1975)
	4	740.85/210.5	Axen nappe/Cardinia beds	PQ MPaC Cc 0	Frey & Wieland (1975)
64	5	737.8/206.4	Axen nappe/"Infralias"	PQ MPaC 0	Frey & Wieland (1975)
65	8	736.1/210.05	Axen nappe/Cardinia beds	PQ MPaC Cc 0	Frey & Wieland (1975)
66	6	737.65/214.8	Axen nappe/Prodkamm Fm	PQ MPaC 0	Frey & Wieland (1975)
	1	739.95/214.8	Axen nappe/Cardinia beds	PQ MPaC 0	Frey & Wieland (1975)

Appendix, continued.

No.	#	Coordinates	Tectonic/Stratigraphic Position	Assemblage	Reference
67	2	720.2/179.2	Autochthonous Aar Massif (AM)/Quartenschiefer	PQ MPaC H	Frey & Wieland (1975)
68	1	710.35/194.45	Axen nappe/"Aalenian"	PQ MPaC O	Frey & Wieland (1975)
69	7	703.3/192.7	Axen nappe/"Infralias"+ Cardinia beds	PQ MPaC CcDo O	Frey & Wieland (1975)
70	1	692.4/187.8	Autochthonous AM/"Aalenian"	PQ MPaC O	Frey & Wieland (1975)
71	2	688.4/186.1	Autochthonous AM/"Aalenian"	PQ MPaC Cc O	Breitschmid (1980, 1982)
72	3	681.8/185.35	Autochthonous AM/Eocene palaeokarst	PQ MPaC H	Wieland (1979)
	3	681.6/183.9	Autochthonous AM/"Aalenian"	PQ MPaC Cc O	Breitschmid (1980, 1982)
	2	683.0/183.2	Autochthonous AM/"Aalenian"	PQ MPaC O	Breitschmid (1980, 1982)
73	2	678.8/183.85	Autochthonous AM/Eocene palaeokarst	PQ MC H	Wieland (1979)
74	2	668.6/178.5	Autochthonous AM/Eocene palaeokarst	PQ MPaC	Wieland (1979)
75	6	646.75/162.5	Autochthonous AM/Eocene palaeokarst	PQ MPaC H	Wieland (1979)
76	1	640/160*	Wildhorn nappe/"Aalenian"	PQ MPaC Cc O	H.J. Drong, pers. comm.
77	?	635.3/154.3	Parautochthonous AM/"Aalenian"	PQ ... O	B. Künzi, pers. comm.
78	?	629.7/154.6	Wildhorn nappe/"Aalenian"	PQ ... O	B. Künzi, pers. comm.
79	1	626.3/154.4	Wildhorn nappe/"lower Liassic"	PQ MC CcDo O	Frey & Wieland (1975)
80	4	618.75/148.15	Doldenhorn nappe/Eocene palaeokarst	PQ MC Cc H	Frey & Wieland (1975), Wieland (1979)
81	1	619.85/138.3	Parautochthonous AM/"Hettangian"	PQ MPaC O	Frey & Wieland (1975)
82	1	618.35/133.3	Parautochthonous AM/"Liassic"	PQ MPaC Cc O	Morariu (1985)
83	1	613.8/135.7	Parautochthonous AM/"Aalenian"	PQ MPaC O	Frey & Wieland (1975)
84	1	611.5/137.85	Doldenhorn nappe/Eocene palaeokarst	PQ MPaC H	Frey & Wieland (1975), Wieland (1979)
85	3	600.3/153.5	Prealps/Jurassic palaeokarst	PD MC Cc H	Baud et al. (1979), Bertherin (1980)
86	3	595.4/124.9	Ultraschweizer nappes/"Aalenian"	PQ MPaC Do O	This study
87	3	587.0/127.5	Diablerets nappe/Eocene palaeokarst	PQ MC G	Wieland (1979)
88	3	579.15/133.2	Ultraschweizer nappes/"Aalenian"	PQ MPaC Do O	This study
89	3	598.8/125.0	Wildhorn nappe/"Aalenian"	PQ MPaC CcDo O	Burkhard (1986)
90	1	605.5/128.2	Wildhorn nappe/"Aalenian"	PQ MC O	Burkhard (1986)

No. = Localities refer to those of Fig. 2

= Number of samples per locality

* = Approximate coordinates only

Mineral abbreviations used:

C	chlorite	M	muscovite
Cc	calcite	P	pyrophyllite
D	diaspore	Pa	paragonite/muscovite mixed-layer
Do	dolomite		(including some paragonite at localities 62-63, 65-67, 69, 84)
G	goethite	O	organic material
H	hematite	Q	quartz
I	illite (including illite/smectite mixed-layers)	R	rectorite
K	kaolinite	S	smectite

Note: if more than one sample per locality is available, then the most complete assemblage is given only, neglecting existing sub-assemblages.