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MONOCLINIC AND TRICLINIC FORMS OF PYROPHYLLITE AND PYROPHYLLITE ANHYDRIDE

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ABSTRACT

X-ray powder patterns of pyrophyllites from twenty localities show differences indicating that both 1-layer triclinic and 2-layer monoclinic forms exist. Some samples are mixtures of the two forms; others are so disordered, either naturally or by mechanical grinding, that a differentiation is not possible. For the best crystallized material, the powder diagrams are indexed from the following unit cells:

triclinic: a 5.173, b 8.960, c 9.360 Å, α 91.2°, β 100.4°, γ 90°.
monoclinic: a 5.172, b 8.958, c 18.67 Å, β 100.0°.

The corresponding anhydride phases also are *triclinic:* a 5.140, b 9.116, c 9.504 Å, α 91.2°, β 100.2°, γ 90°; *monoclinic:* a 5.173, b 9.114, c 18.99 Å, β 100.0°. Expansion of b is interpreted as a relaxation of the twisted Si-O networks. After dehydroxylation, the Al ion coordination appears to change only slightly and this may impose a constraint on the structures in the a direction.

INTRODUCTION

The first crystal structure analysis of pyrophyllite by Gruner (1934) using X-ray powder diffraction data gave a monoclinic unit cell with a 5.14, b 8.90, c 18.55 Å, $\beta = 99.9°$, containing two structural layers and with probable space group $C2/c$. This structure was confirmed by Hendricks (1938), using single crystal diffraction data. He noted that disorder in the superposition of the component layers made it impossible to determine a unique space group. Rayner and Brown (1965) made a refined structure analysis using only the sharp diffractions and showed the ditrigonal nature of the silicon-oxygen tetrahedral sheets. They also remarked on the disorder present in all crystals they examined. Zvyagin, Mishchenko and Soboleva (1969), using electron diffraction data, again confirmed the two-layer monoclinic cell and additionally recorded a one-layer triclinic form for a synthetic pyrophyllite. They noted that the monoclinic structure may be a more or less regular alternation of two enantiomorphic triclinic structures analogous to the description given by Bailey (1963) for the relation between one-layer triclinic kaolinite and two-layer monoclinic dickite.

The present work was undertaken to study the relationship between the anhydride phase formed when pyrophyllite is dehydroxylated, and the original structure. It is well established that considerable structural order remains in the anhydride phase, and that the sequence of reactions from pyrophyllite, through the anhydride phase, eventually to mullite and cristobalite is a topotactic process (Thilo and Schünemann, 1937; Bradley and Grim, 1951; Nakahira and Kato, Personal Commun., 1961; 1964; Heller *et al.*, 1962; Heller, 1962).

Of particular interest is the question whether the loss of 'structural water' and the ensuing modification of the octahedral sheet of the 2:1 layer structure is accompanied by a relaxation of the silicon-oxygen tetrahedral sheets similar to that found in kaolinite (Brindley and Gibbon, 1968) and in muscovite (Eberhart, 1963; Nicol, 1964), both of which minerals show a significant increase of *b* with dehydroxylation. The results already published for pyrophyllite appear to be contradictory. The data of Thilo and Schünemann (1937) indicated an increase of *b* with dehydroxylation, and a similar observation was made by Lindqvist (1962); these authors describe the change by saying that dehydroxylated pyrophyllite is "talc-like," but this description seems to apply only to the lattice spacings. On the other hand, Heller *et al.* (1962) and Heller (1962) consider that a considerable decrease occurs in *b*, from 8.96 to 8.80 to 8.75 Å after heating to, respectively, 850° and 970°C, while *a* appears to increase.

EXPERIMENTAL PROCEDURES

The present experiments were made on fine powders of pyrophyllites from various sources with a view to obtaining the best crystalline material. Dehydroxylations were carried out in air at temperatures ranging from about 650–850°C and the extent of dehydroxylation was checked by cooling the samples in vacuo and then reweighing them. Dehydroxylation was taken to be complete when no further weight change occurred. The total weight loss was usually somewhat higher than the value 5.01 percent corresponding to the ideal composition and this was most probably due largely to incomplete removal of surface adsorbed water by preliminary drying at 105–110°C.

Pyrophyllite samples for X-ray powder analysis, when packed in rectangular cavities either from the front or the back face, commonly show considerable preferential orientation, particularly when the initial material has a fibrous or rosette form. This disadvantage was largely eliminated by filling the cavities through a side opening with a glass slide covering the front face while the holder was mounted on a vibrating table. The pyrophyllite powder, dried at 110°C and then sieved through a 200 mesh screen, packed sufficiently firmly so that the glass slide could be removed without disturbing the sample surface. This arrangement may tend to align the basal planes normal to the sample surface, but in fact no marked orientation of this kind was noticed. In any case, if the *hk*0 and *hkl* diffractions are slightly over-emphasized by this technique, it has advantages in bringing up weak reflections which might escape detection with normal sample preparation.

Powder diffraction patterns were recorded with nickel filtered CuKα radiation at $\frac{1}{4}°(2\theta)$/min and with $1°(2\theta)/2.5$ cm of chart recording paper. The instrument was calibrated with reference to spacings for silicon metal powder as given by the Philips Electronic Instruments Company based on $a=5.43062$ Å at 21.0°C and spacings were checked also by reference to KBr as an internal standard which in turn was checked against the silicon standard.

Almost all investigators of pyrophyllite using X-ray diffraction techniques, both powder and single crystal methods, have found considerable structural disorder. A survey of samples from many localities (see Appendix) was undertaken with a view to obtaining a few with a minimum of disorder, as judged by the number and sharpness of the diffraction peaks. Samples in macroscopically fibrous or rosette forms smeared readily with any mechanical treatment and could not be reduced satisfactorily to fine powders. Microcrystalline material was found to be more satisfactory, and, when supplied in a massive form, was powdered by crushing rather than by grinding, and with a continuous removal of the <200 mesh size powder.

RESULTS

The determination of the lattice parameter changes accompanying dehydroxylation was expected to be a simple problem because no questions had arisen previously concerning the validity of the 2-layer monoclinic unit cell for normal pyrophyllite. A search for the best crystalline material led to the selection of a pyrophyllite from New Zealand, supplied by Dr. P. K. Foster, Pottery and Ceramics Research Association, New Zealand and the Crown Lynn Potteries, Ltd., Auckland, N.Z. The geology of the area from which the material was obtained and a chemical analysis are given by Swindale and Hughes (1968). Unexpected indexing difficulties arose first with the anhydride phase, and then with the original mineral itself; no reasonable variations in the parameters of the 2-layer monoclinic cell gave agreement with the observed lattice spacings. Eventually a satisfactory indexing was obtained with 1-layer triclinic cells for both the initial mineral and the anhydride phase. When this work was essentially complete the results of Zvyagin *et al.* (1969) became available, showing the existence of a 1-layer triclinic form in synthetic pyrophyllite. A sample of the New Zealand material was sent to Dr. Zvyagin who confirmed its excellent crystallinity and its similarity to the Russian synthetic pyrophyllite (Zvyagin, personal communication).

Other pyrophyllites, however, do not show the characteristic diffraction features of the 1-layer triclinic form and these can be indexed on the basis of a 2-layer monoclinic cell similar to that given by Gruner and subsequent investigators. However, many pyrophyllites appear to be mixtures of the monoclinic and triclinic forms.

Figure 1 compares diffractometer traces recorded at 1° (2θ)/min of pyrophyllite from New Zealand (1-layer triclinic form) and from Honami, Japan (2-layer monoclinic form). Table 1 lists values of $d(hkl)$, observed and calculated for the two forms from recordings at $\frac{1}{4}$° (2θ)/min; the differences between d(obs) and d(calc) are generally less than 0.2 percent for all reflections which can be uniquely indexed.

The Appendix lists the results for twenty pyrophyllites which have been examined and gives references to descriptions of the minerals

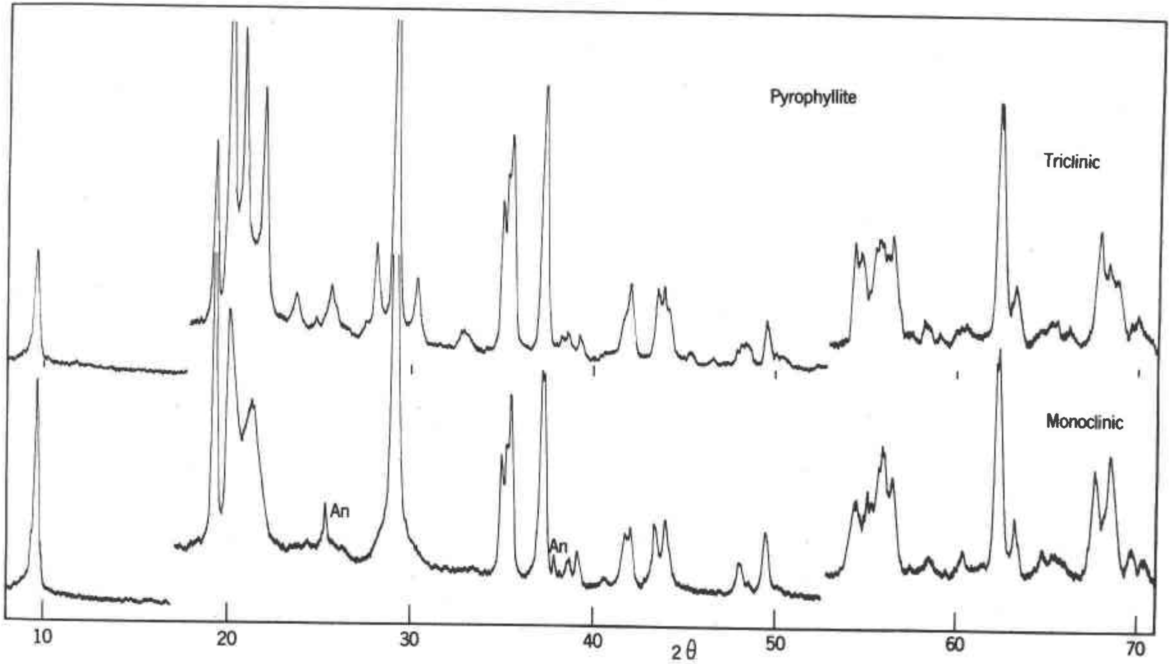

FIG. 1. Diffractometer patterns for triclinic pyrophyllite (New Zealand) and monoclinic pyrophyllite (Honami, Japan) taken with filtered CuKα radiation and recorded at 1°(2θ)/min. An=anatase impurity.

TABLE 1. X-RAY POWDER DATA FOR TWO FORMS OF PYROPHYLLITE.

Triclinic (New Zealand)				Monoclinic (Honami, Japan)			
hkl	*d*(calc)Å	*d*(obs)Å	*I*(obs)	*hkl*	*d*(calc)Å	*d*(obs)Å	*I*(obs)
001	9.204	9.20	80	002	9.204	9.21	100
002	4.602	4.60	30	004	4.602	4.61	40
020	4.479			020	4.479	4.42^{b}	60
110	4.417	4.42	100	110	4.428		
$11\bar{1}$	4.265	4.26	80	021	4.352		
$1\bar{1}\bar{1}$	4.239			$11\bar{2}$	4.248	4.18^{b}	90
$02\bar{1}$	4.061	4.06	60	111	4.164		
021	3.995			022	4.027		
$1\bar{1}1$	3.785	3.764	5				
111	3.749						
$11\bar{2}$	3.486	3.492*	5				
$1\bar{1}\bar{2}$	3.451	3.454*	5				
022	3.177	3.178	20				
003	3.068	3.068	100	006	3.068	3.069	100
112	2.953	2.953	20				
$11\bar{3}$	2.740	2.741*	3				
$1\bar{1}\bar{3}$	2.712	2.710*	4				
$1\bar{3}0$	2.580			130	2.576	2.571	15
$20\bar{1}$	2.572	2.569	30	$20\bar{2}$	2.572		
130	2.571						
$13\bar{1}$	2.548	2.547	30				
200	2.544						
$1\bar{3}\bar{1}$	2.532	2.532	40	200	2.547	2.550*	15
023	2.507			$13\bar{2}$	2.539	2.534	20
$20\bar{2}$	2.416	2.416	80	026	2.531		
131	2.410			132	2.426	2.419^{b}	45
$13\bar{2}$	2.355			$20\bar{4}$	2.413		
201	2.347	2.341^{b}	10	202	2.352	2.352*	1
113	2.346			$13\bar{4}$	2.337	2.334	3
004	2.301	2.300	5				
$2\bar{2}0$	2.216	2.215^{b}	4	008	2.301	2.303	4
220	2.209						
$1\bar{1}\bar{4}$	2.174	2.170*	15	220	2.214	2.216^{b}	1
041	2.166						
$20\bar{3}$	2.155	2.152	15				
132	2.148			134	2.168	2.166*	9
$22\bar{2}$	2.133	2.135*	2	$20\bar{6}$	2.152	2.151*	9
$2\bar{2}\bar{2}$	2.120	2.116*	2				
$2\bar{2}1$	2.087	2.083	20				
$13\bar{3}$	2.084						
221	2.072						
$02\bar{4}$	2.064	2.059*	25	222	2.082	2.086	10
$1\bar{3}\bar{3}$	2.048						

(Continued on following page)

TABLE 1 (*continued*)

Triclinic (New Zealand)				Monoclinic (Honami, Japan)			
hkl	*d*(calc)Å	*d*(obs)Å	*I*(obs)	*hkl*	*d*(calc)Å	*d*(obs)Å	*I*(obs)
$04\bar{2}$	2.031	2.026^b	3				
024	2.030			$13\bar{6}$	2.064	2.059	15
042	1.997	1.998	2	028	2.047		
$22\bar{3}$	1.951	1.952^b	<1				
$2\bar{2}2$	1.893	1.887^{vb}	12				
$20\bar{4}$	1.881			136	1.893	1.892	6
133	1.874			$20\bar{8}$	1.877	1.876*	1
005	1.841	1.841	10	0010	1.841	1.842	10
$04\bar{3}$	1.827	1.823*	2				
$13\bar{4}$	1.815	1.812*	3				
$11\bar{5}$	1.808						
203	1.807			206	1.811	1.814^b	1
$22\bar{4}$	1.7430	1.7439	1				
$2\bar{2}\bar{4}$	1.7252	1.7223	1				
$24\bar{1}$	1.6907	1.6894	12	$31\bar{2}$	1.6928	1.6879*	14
025	1.6902			150	1.6901		
150	1.6879			$24\bar{2}$	1.6889		
$2\bar{4}\bar{1}$	1.6873			240	1.6817		
$2\bar{2}3$	1.6846						
$2\bar{4}0$	1.6841						
$15\bar{1}$	1.6839						
240	1.6778		12				
$1\bar{5}\bar{1}$	1.6758	1.6674*					
310	1.6653			310	1.6681	1.6662*	15
$31\bar{2}$	1.6636			$31\bar{4}$	1.6617		
$1\bar{3}4$	1.6589	1.6529*	30				
$1\bar{5}1$	1.6525						
$24\bar{2}$	1.6483			134	1.6458	1.6469*	20
151	1.6375	1.6327*	20	152	1.6455		
$2\bar{4}\bar{2}$	1.6362						
$20\bar{5}$	1.6356			$20\overline{10}$	1.6325	1.6313*	15
134	1.6296						
$2\bar{4}1$	1.6276						
$15\bar{2}$	1.6266						
$04\bar{4}$	1.6219	1.6213*	3				
$1\bar{1}5$	1.6192						
$1\bar{5}\bar{2}$	1.6086	1.6068	<1				
115	1.6076			1110	1.6155	1.6104^b	2
311	1.5898	1.5851	4	048	1.6049		
044	1.5883						
$31\bar{3}$	1.5869			$31\bar{6}$	1.5832	1.5789^b	4
$3\bar{1}\bar{3}$	1.5829			208	1.5766		

TABLE 1 (*continued*)

Triclinic (New Zealand)				Monoclinic (Honami, Japan)			
hkl	*d*(calc) Å	*d*(obs) Å	*I*(obs)	*hkl*	*d*(calc) Å	*d*(obs) Å	*I*(obs)
$1\bar{5}2$	1.5682	1.5653	3				
$24\bar{3}$	1.5620						
152	1.5459	1.5443*	2				
$22\bar{5}$	1.5445						
$2\bar{4}\bar{3}$	1.5441						
006	1.5340	1.5318*	3	0012	1.5340	1.5348	4
$2\bar{4}2$	1.5320			$22\bar{5}$	1.5338		
$15\bar{3}$	1.5306						
$1\bar{1}\bar{6}$	1.5189	1.5218*	<1				
242	1.5130	1.5142*	<1				
$1\bar{5}\bar{3}$	1.5068	1.5053*	1				
060	1.4930	1.4929	30	060	1.4930	1.4935	20
$33\bar{1}$	1.4927			$33\bar{2}$	1.4929		
$3\bar{3}\bar{1}$	1.4926						
$2\bar{2}4$	1.4921						
$3\bar{1}2$	1.4885	1.4894*	10	314	1.4890	1.4898*	5
330	1.4723	1.4716	8				
$3\bar{3}\bar{2}$	1.4695			$33\bar{4}$	1.4715	1.4708	7
061	1.4689						
$24\bar{4}$	1.4505	1.4513*	<1				
$1\bar{3}\bar{5}$	1.4499						
026	1.4420	1.4433*	2				
153	1.4321	1.4291*	6	156	1.4456	1.4407*^{b}	3
$20\bar{6}$	1.4308			1310	1.4391		
$2\bar{4}\bar{4}$	1.4302			$24\bar{8}$	1.4386		
$06\bar{2}$	1.4289			$20\overline{12}$	1.4282	1.4260*^{b}	4
$3\bar{3}1$	1.4266			332	1.4241		
135	1.4258			0410	1.4221		
$3\bar{3}\bar{6}$	1.4130	1.4118	4	$33\bar{6}$	1.4161	1.4154*^{b}	2
062	1.4115						
$13\bar{6}$	1.3846	1.3831	18				
116	1.3811			1112	1.3862	1.3858	15
$3\bar{1}3$	1.3703	1.3703*^{b}	30	2010	1.3823		
$22\bar{6}$	1.3699			$13\overline{12}$	1.3720	1.3697	20
$3\bar{3}2$	1.3509	1.3462*^{b}	10	316	1.3706		
$33\bar{4}$	1.3446			334	1.3475	1.3491*	4
332	1.3398						
$1\bar{5}4$	1.3382	1.3347*	2				
$3\bar{3}\bar{4}$	1.3336						
063	1.3316			066	1.3425	1.3432*	1
$24\bar{5}$	1.3311			$33\bar{8}$	1.3374	1.3365*	2
007	1.3149	1.3137*	3	0014	1.3146	1.3159	1.5
154	1.3127						
225	1.3119						

vb = very broad; b = broad; * not completely resolved where these are known. Samples from Japanaese sources are described in the recent publication "The Clays of Japan," prepared for the 1969 International Clay Conference, Tokyo, and edited by Iwao (1969). Kodama (1958) also has given data on some of the materials used.

Only one other pure, or nearly pure, triclinic form has been identified, namely from Bontenyama, Japan. Several nearly pure monoclinic varieties have been found. Possibly many samples occurring as fibrous and rosette forms may be monoclinic, but the disorder introduced in powdering these materials makes any identification by X-ray powder diffraction rather uncertain. Some samples are clearly mixtures of the two forms.

Figure 2 illustrates in more detail the diffraction range 18°–32° (2θ) (Cu$K\alpha$) and shows the modification of the monoclinic pattern by 10 percent and 20 percent additions of the triclinic form. A naturally occurring sample from Shokozan, Japan, gave the pattern shown and appears to have about 20 percent of the triclinic form.

While many pyrophyllites appear to be mechanical mixtures or phase mixtures of the two forms, other samples give much poorer diffraction patterns and may represent mixtures on a unit-cell scale, *i.e.*, interstratifications or simply 'mistakes' in the layer stacking sequence. So far these patterns have not been evaluated in any detail but they appear to be rather unpromising material for detailed analysis.

Figure 3 illustrates the diffraction patterns of the anhydride phases developed from the New Zealand (triclinic) and the Honami (monoclinic) pyrophyllites by heating at 800°C for 96 hr. The patterns are of comparable sharpness to those given in Figure 1, and the indexing gives a similar close agreement between observed and calculated spacings. However, the very strong reflection at $2\theta = 20.10°$ from the triclinic anhydride, when recorded at $\frac{1}{4}°$ (2θ)/min, shows an additional reflection at $2\theta = 20.48°$ for which no indexing has yet been obtained.

The unit cell parameters of the initial minerals and of the anhydride forms are compared in Table 2, where it is seen that the b parameter *expands* by 1.74 percent in going from the normal to the anhydride form for both structural types, while the a parameter appears to *decrease* by 0.64 percent for the triclinic form but shows no change for the monoclinic form. Both forms show an expansion of $d(001)$ by about 1.62 percent.

DISCUSSION

Since detailed structure analyses of the initial and anhydride phases will be undertaken to elucidate the changes accompanying dehydroxylation, it is unnecessary here to develop elaborate hypotheses. However, the following simple considerations can be given.

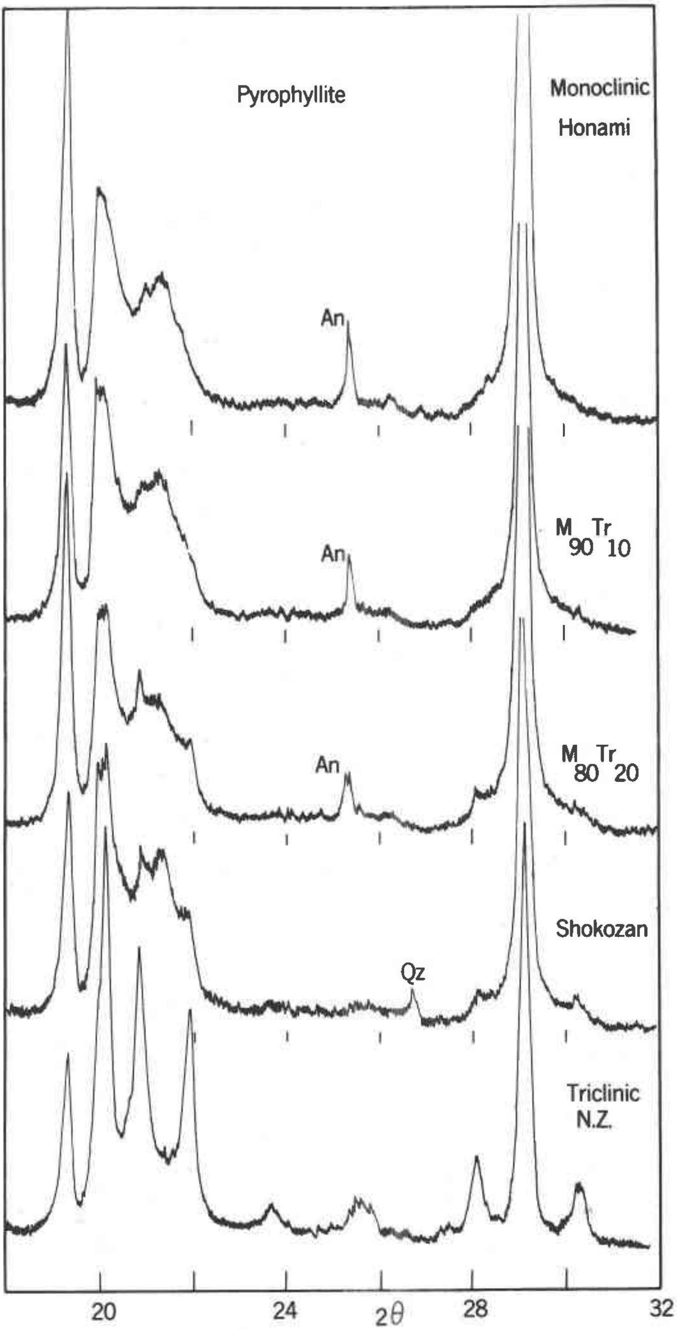

FIG. 2. Expanded diffraction patterns of monoclinic and triclinic pyrophyllites, and of synthetic and natural mixtures, taken with CuKα radiation at $\frac{1}{4}°$ (2θ)/min.

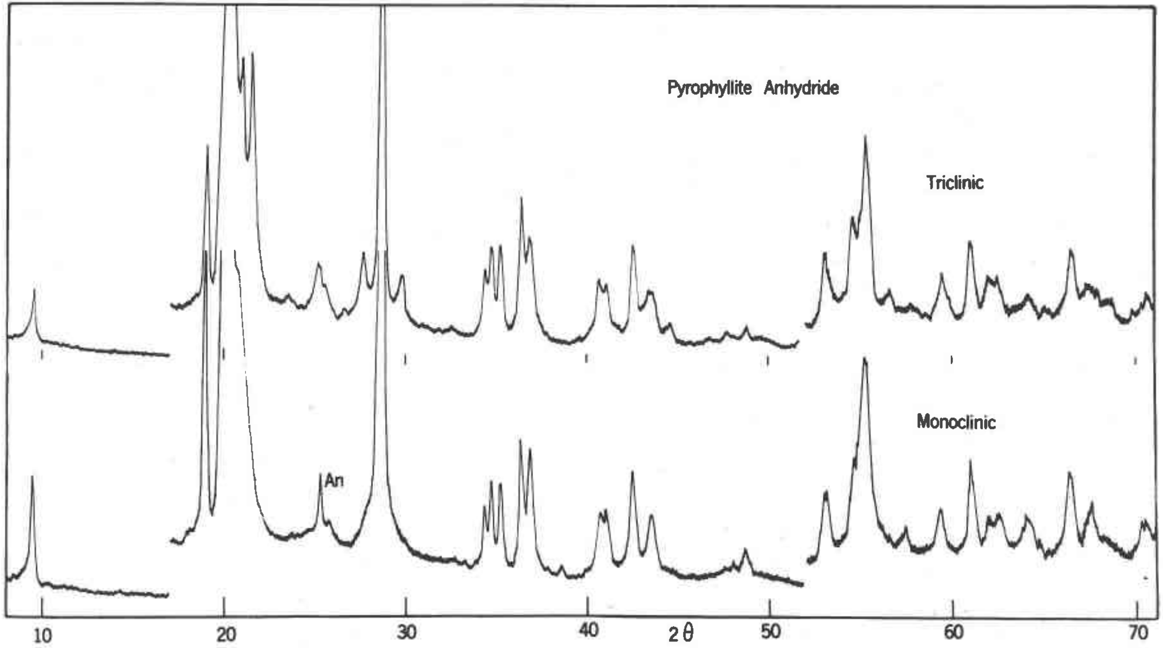

FIG. 3. Diffractometer patterns of the anhydride phases from triclinic pyrophyllite (New Zealand) and monoclinic pyrophyllite (Honami Japan) taken with filtered CuKα radiation and recorded at 1°(2θ)/min. An=anatase impurity.

TABLE 2. LATTICE PARAMETERS OF TRICLINIC AND MONOCLINIC PYROPHYLLITES AND THEIR ANHYDRIDE FORMS.

	Triclinic			Monoclinic		
	Normal	Anhyd.	% Exp.	Normal	Anhyd.	% Exp.
a	5.173 Å	5.140 Å	−0.64	5.172 Å	5.173 Å	0
b	8.960	9.116	+1.74	8.958	9.114	+1.74
c	9.360	9.504	—	18.676	18.995	—
α	91.2°	91.2°	—	90.0°	90.0°	—
β	100.4	100.2	—	100.0	100.0	—
γ	90.0	90.0	—	90.0	90.0	—
$d(001)$	9.204 Å	9.352 Å	+1.61	18.408 Å	18.707 Å	+1.625
$b/a\sqrt{3}$	1.000	1.024	—	1.000	1.017	—

The mean b parameter of pyrophyllite, 8.959 Å, is significantly less than the ideal value calculated for a hexagonal silicon-oxygen sheet with Si–O $=1.62$ Å, namely $4\sqrt{2}\times 1.62=9.15$ Å. The observed value of b corresponds to a di-trigonal silicon-oxygen sheet with an angle of twist of $\cos^{-1}(8.959/9.15)=11°42'$ which can be compared with the angle 10.0–10.5° given by Rayner and Brown (1965).

The expanded value of b for the anhydride phases averages 9.115 Å which is only slightly less than 9.15 Å. This small difference cannot be uniquely interpreted. It could be attributed to a small residual twist of the tetrahedra, but a small change in the three basal Si–O bond lengths to a value slightly less than 1.62 Å, or a small deviation from ideal tetrahedral angles could explain the observed value. Therefore, at this time, it can be concluded only that the twisted tetrahedral networks of the initial structures appear to be largely untwisted in the anhydride phases.

There seems no doubt that the parameter a does not show the same expansion as b does and tentatively it appears that after dehydroxylation the octahedral sheet still exerts a constraint in the a direction.

Whereas in the case of kaolinite (Brindley and McKinstry, 1961) X-ray fluoresence measurements of the AlKα wavelength indicated that Al changed from 6-fold to 4-fold coordination following dehydroxylation, preliminary measurements with the pyrophyllite samples heated at 800°C for 96 hr have indicated a smaller change in emission wavelength[1] and presumably the Al does not change to 4-fold coordination. While it is necessary to confirm this conclusion by X-ray structure analysis, it seems likely that the Al ions still impose a constraint on the relaxation of

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[1] More extended measurements by Dr. S. Udagawa (personal communication) appear to confirm the magnitude of the AlKα wavelength change which we have found.

the silicon-oxygen network. The one reflection in the anhydride pattern of the triclinic material not yet satisfactorily indexed may represent a polymorphic variety but an experimental verification has not yet been obtained.

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APPENDIX. PYROPHYLLITE SAMPLES EXAMINED

Source (reference)	Physical characteristics	X-ray diffraction characteristics
1. New Zealand (Swindale & Hughes, 1968) Dr. P. K. Foster, and Crown Lynn Potteries[a]	Massive, white, soft, easily crushed	Triclinic
2. Bontenyama, Koshyoku City, Nagano pref., Japan. Dr. H. Minato[a]	Massive, white, readily crushed and ground	Essentially triclinic. Impurity? Reflections at 3.98, 3.24 Å. Almost identical with #1
3. Honami, Shimotakai-gun, Nagano pref., Japan. (Iwao, 1969, pp. 81–2) (Kodama, 1958) Dr. T. Sudo[a]	Massive, pale blue-green, soft, easily crushed and ground	Essentially monoclinic. Anatase present
4. Yoji, Kanra-gun, Gumma pref., Japan. (Iwao, 1969, p. 82) (Kodama, 1958) Dr. T. Sudo[a]	Massive, off-white, easily crushed and ground	Essentially monoclinic. Small amount of quartz. Similar to #3
5. Robbins, N. Carolina. Wards Natural Science Est. Inc.	Massive, white, lustrous, silky touch, easily ground	Essentially monoclinic. Mica ~5%; trace of quartz
6. Newfoundland American Olean Tile Co., Pa.[a]	Massive, off-white, readily crushed and ground	Essentially monoclinic. Traces of quartz and mica. Similar to #3
7. Shokozan, Hiroshima pref., Japan (Iwao, 1969, p. 74) Dr. T. Sudo[a]	Massive, light-green, friable, easily ground	Monoclinic ~80%; triclinic ~20%. Trace of quartz
8. Goto Mine, Fukue Island, Nagasaki pref., Japan. (Iwao, 1969, pp. 72–73)	Massive, white, very soft, powdery	Monoclinic ~80%, triclinic ~20%. Quartz impurity; a second impurity giving 3.98 Å line. Similar to #7
9. Wondong, Korea, Dr. Sang, Geol. Survey, Korea[a]	Massive, slight buff color, white when crushed, easily ground	Monoclinic + some triclinic. Quartz impurity. Similar to #7

Table 1 (*continued*)

10. Wolsung, Korea. Dr. Sang, Geol. Survey, Korea[a]	Massive, very pale green, waxy appearance, fractures with difficulty, easily crushed	Monoclinic ~70%; triclinic ~30%
11. Berosovska, Urals, USSR Smithsonian Inst.[a] (USNM 96247)	Rosettes, blue-green, difficult to grind	Essentially monoclinic. Similar to #7. Mica, quartz impurities
12. Zacvalpan, Mexico Dr. L. de Pablo[a]	Massive, off-white, waxy appearance, not readily ground	Probably monoclinic Quartz impurity
13. Municipio de Diamantina, Minas Gerais, Brazil. Dr. P. de Souza Santos[a]	White powder	Probably monoclinic Trace of quartz; ~5% mica
14. St. Niklas, Zermatt, Switzerland. Smithsonian Inst.[a] (R11384)	Rosettes, pale blue, shiny	Probably monoclinic Trace of quartz
15. Gueser Gulch, Calif. Smithsonian Inst. (14814)	Rosettes, brown	Probably monoclinic
16. Orange Co., Pa. Smithsonian Inst.[a] (C3838)	Rosettes, light buff color black impurity	Possibly monoclinic

Obscure patterns were obtained from the following samples: Graves Mountain, Lincoln County, Ga. (USNM 86292); Staley, N. Carolina (USNM 97369); Gargol, Adams Co., Penna.; Guildford County, N. Carolina.

[a] Donor.

References

Bailey, S. W. (1963) Polymorphism of the kaolin minerals. *Amer. Mineral.* **48,** 1196–1209.

Bradley, W. F. and R. E. Grim (1951) High temperature thermal effects of clay and related materials. *Amer. Mineral.* **36,** 182–201.

Brindley, G. W. and D. L. Gibbon (1968) Kaolinite layer structure: Relaxation by dehydroxylation. *Science* **162,** 1390–1391.

———, and H. A. McKinstry (1961) The kaolinite-mullite reaction series: IV, The coordination of aluminum. *J. Amer. Ceram. Soc.* **44,** 506–507.

Eberhart, J. P. (1963) Étude des transformations du mica muscovite par chauffage entre 700 et 1200°C. *Bull. Soc. Fr. Mineral. Cristallogr.* **86,** 213–251.

Gruner, J. W. (1934) The crystal structure of talc and pyrophyllite. *Z. Kristallogr.* **88,** 412–419.

Heller, L. (1962) Thermal transformation of pyrophyllite to mullite. *Amer. Mineral.* **47,** 156–157.

——— V. C. Farmer, R. C. MacKenzie, B. D. Mitchell and H. F. W. Taylor (1962) Dehydroxylation and rehydroxylation of triphormic dioctahedral clay minerals. *Clay Miner. Bull.* **5,** 56–72.

Hendricks, S. B. (1938) The crystal structure of talc and pyrophyllite. *Z. Kristallogr.* **99,** 264–274.

Iwao, S. (1969) (Ed) *The Clays of Japan.* Geological Survey of Japan, 209 pp.

Kodama, H. (1958) Mineralogical study on some pyrophyllites in Japan. *Mineral. J.* [*Tokyo*] **2,** 236–244.

Lindqvist, B. (1962) Polymorphic phase changes during heating of dioctahedral layer silicates. *Geol. Foren. Förh.* **84,** 224–229.

NAKAHIRA, M. AND T. KATO (1964) Thermal transformations of pyrophyllite and talc as revealed by X-ray and electron diffraction studies. *Clays and Clay Minerals* **12,** 21–27.

NICOL, A. W. (1964) Topotactic transformation of muscovite under mild hydrothermal conditions. *Clays Clay Miner.* **12,** 11–19.

RAYNER, J. H. AND G. BROWN (1966) Structure of pyrophyllite. *Clays Clay Miner.*, **13,** 73–84.

THILO, E. AND H. SCHÜNEMANN (1937) Über das Verhalten des Pyrophyllits beim Erhitzen und die Existenz eines "wasserfreien Pyrophyllits" *Z. Anorg. Allg. Chem.* **230,** 321–335.

SWINDALE, L. D. AND I. R. HUGHES (1968) Hydrothermal association of pyrophyllite, kaolinite, diaspore, dickite, and quartz in the Coromandel area, New Zealand. *N. Z. J. Geol. Geophys.*, 11 (5), 1163–1183.

ZVYAGIN, B. B., K. S. MISHCHENKO, AND S. V. SOBOLEVA (1969) Structures of pyrophyllite and talc in relation to the polytypes of mica-like minerals. *Kristallografiya* **13,** 599–604 (1968) [in Russian]. Transl. *Sov. Phys. Crystallogr.* **13,** 511–515.

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