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# Thermal Studies of Some Minerals. No. 1.

By

Yoshiharu UMEGAKI and Suguru NAGATOMI

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*With 6 Tables and 10 Text-figures*

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**ABSTRACT:** Thermal effects on the inner structures of some minerals were researched with use of special apparatus, a sort of thermal increment diffractometer, through which variations of the interplaner spacings in question were pursued not only at ordinary states, as were so far often carried out, recovered from high-temperature condition but also during continuously heated state at elevated temperature up to 1000°C. The results thus obtained have been found considerably different from those referred to in the previous works.

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## I. INTRODUCTION

Most of the previous researches under the heading seem to have been accustomed to comparing the modifications revealed by the inner structures of certain minerals at ordinary state with those restored through cooling after thermal treatments. It however is difficult to trace the actual variations in the inner structures, even though remarkable differences in modifications be discernible at the final state recovered from heating at given temperatures, by ordinary means of X-ray (*cf.* UMEGAKI *et al.*, 1957; LIMA-DE-FARIA, 1958), infra-red absorption (*cf.* STUBIČAN, 1959; DACHILLE *et al.*, 1959; STUBIČAN *et al.*, 1961; TETTENHORST, 1962) and so forth, inasmuch as either reversible or irreversible variations, if any or all, are to be anticipated in the course of treatments. On the other hand, *DTA* methods (*cf.* KULBICHI *et al.*, 1959; SCHMIDT *et al.*, 1959) widely adopted may also be useful for pursuing conversions appeared in the modifications of materials bearing small heat contents and as a scheme for rapid, if not complete, identification of the specimens. At the same time, it is to be remembered that in such case the heating or cooling, for instance, in the rate of some degrees per minute, was too speedy, and duration of time at given state was too short, to cause the real and perfect conver-

sion for the comparatively stable structures, resulting in that seeming variations have been liable to occurring almost always on the sides of temperatures higher than are really necessitated. In view of this, some experiments have been carefully carried into effect concerning some of specific minerals with layer- and metamict structures.

## II. EQUIPMENTS AND CONDITIONS FOR EXPERIMENTS

In pursuit of variations emerging in the inner structures, thermal effects on the pulverized specimens under consideration have been continuously traced during heated state at respective temperatures. The equipments used and the conditions settled in series of the experiments are outlined in the following:

### (1) *X-ray apparatus.*

X-ray diffractometer provided for the research is that of GX-2B type, manufactured by Shimazu Man. Co. Ltd., to which the auto-scaler, recorder, powerful transformer putting out 60 KV in maximum and so on are attached.

### (2) *Heating apparatus.*

The goniometer, on which the supporter for the sample-holder is placed, is enclosed in a cylindrical metallic case with the windows covered with double foils of nickel and special metal, through which both incident and diffractive X-ray can be penetrated. The holder is made of nickel, and platy in shape (80 mm in length, 9 mm in width, and 3 mm in thickness), in the central part of which a rectangular space (20 mm in length and 6.2 mm in width) is bored and bridged across by six Pt wires. The metallic case mentioned above is devised to be kept in air-tight condition and aspiratable strongly through a hole combined with a vacuum pump.

The paste of the pulverized specimen (finer than 250 mesh in grain-size) kneaded with nitrocellulose diluted in amyl acetate as well as, if necessary, a little amount of quartz powder as a sort of indicator standardizing the spacings are filled closely in the space of the holder, flattened on the glass plate and allowed to be dried up in the air.

The holder containing the sample thus prepared is inserted in a cylindrical electric furnace (60 mm in length and 15 mm in inner diameter) connected with a current regulator and heated at any given temperatures, the variations of which are continuously measurable on the reading of millivoltmeter combined with Pt-PtRh thermo-couple held in the sample. Both the supporter of the holder and the metallic case are cooled with running water so as to be insulated from unnecessary thermal effects.

### (3) *X-ray conditions.*

In accordance with necessity, 25-35 KV and 15-25 mA were yielded on the X-ray tube with Cu target and increase of some KV as well as of some mA was re-

quisite for putting out the sufficient intensity of diffraction from the specimens protected within the metallic case during thermal treatment, compared with in case of ordinary experiments in the air. Taking into account of counting miss ascribed to the electronic circuit of *G. M.* counter, probable errors in numbers of pulses, opening angle of the slit, effect of duration of diffractive rays into the counter and the characteristics of the apparatus used, conditions suitable for the experiments were kept constant as follows:

X-ray:  $\text{CuK}\alpha$ , time constant: 1.25 sec for 1000 cps (excepting the special cases), scanning speed:  $1^\circ (2\theta)/\text{min}$ , rotating speed of chart: 1 cm/min, divergent slit: 1.5 mm in width, and receiving slit: 0.2 mm in width.

### III. PRELIMINARY TESTS

Referring to the data given by BASSETT and LAPHAM (1957), the specimens of low quartz ( $a_0$ : 4.91 Å and  $c_0$ : 5.39 Å at room temperature) collected from a small pocket of pegmatite in biotite granite occurring at Kuritani, Otake City, Hiroshima Prefecture were preliminarily provided for thermal treatments, as are briefly presented in the following:

#### (1) Heating for an hour at several temperatures.

X-ray diffraction figures revealed between  $17^\circ$  and  $52^\circ$  in  $2\theta$  were respectively recorded on charts in such way that the specimens were held at a given temperature and then successively at the temperature higher than the precedings for an hour plus time necessary for recording. A part of the data obtained are listed in Table 1, showing simply the expansion in each spacing, diminution of (110) and (102), and vanishing of (111), compared with those recognized at  $25^\circ\text{C}$ .

TABLE 1. RÖNTGENOMETRIC DATA FOR QUARTZ

Condition	Specimen from Kuritani, Otake City, Hiroshima Prefecture						Beta-quartz from Arkansas (BASSETT & LAPHAM)	
	at $25^\circ\text{C}$		heated for 1 hr. at $600^\circ\text{C}$ .		heated for 1 hr. at $900^\circ\text{C}$ .		heated at $625^\circ\text{C}$ .	
hkl	d (Å)	I	d (Å)	I	d (Å)	I	d (Å)	I
100	4.255	26	4.332	28	4.332	30	4.340	22.8
101	3.342	>100	3.390	>100	3.390	>100	3.399	109.4
110	2.456	14	2.498	7	2.500	9	2.500	2.3
102	2.281	12	2.307	3	2.308		2.310	1.9
111	2.236	11		1		0		
200	2.127	9	2.165	7	2.165	7	2.166	3.2
201	1.980	7	2.010	8	2.012	8	2.013	3.9
112	1.818	26	1.842	29	1.842	28	1.837	10.3

(2) *Continuous heating*

To find out whether variations of the inner structures may or not depend on the conditions in heat treatment, the pulverized specimens of quartz were heated continuously in the rate of  $5^{\circ}\text{C}$  per minute from room temperature to  $1000^{\circ}\text{C}$ . The results obtained together with those remained not included in Table 1 are illustrat-

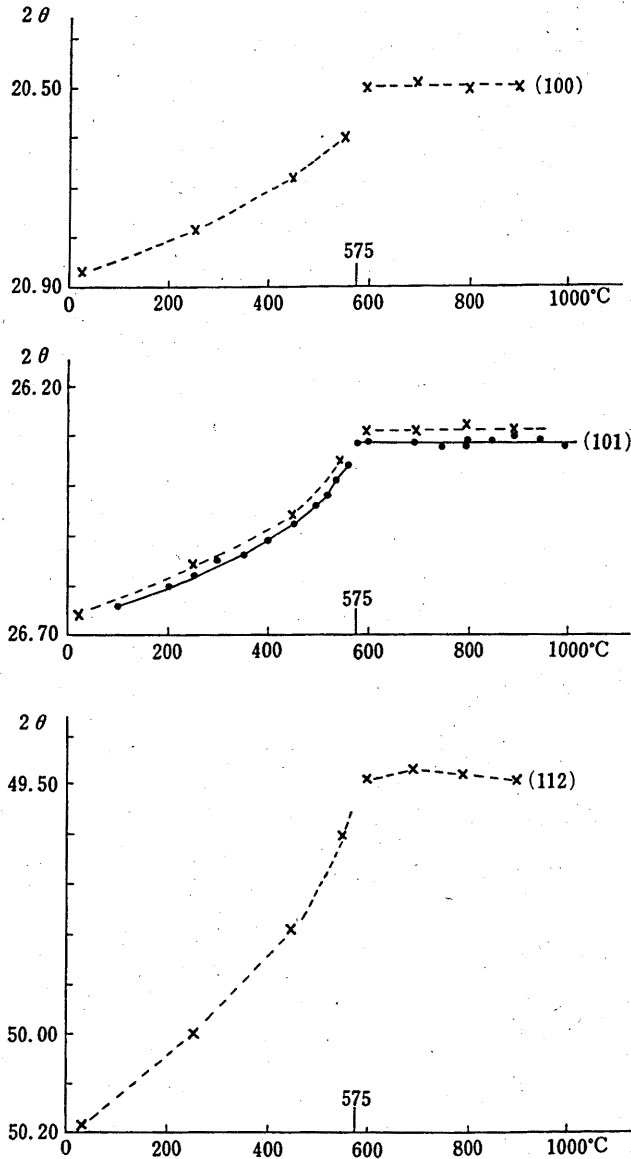


FIG. 1. Variation of spacings quartz

- × kept constant for 1 hr. at each temperature checked
- heated continuously

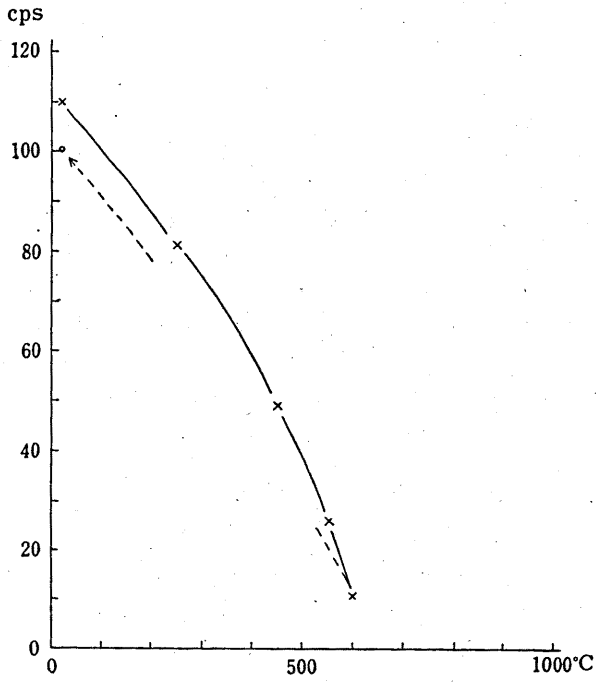


FIG. 2. Intensity variation revealed by (111) of quartz.  
 x kept constant for 1 hr. at each temperature checked.  
 o allowed to cooling.

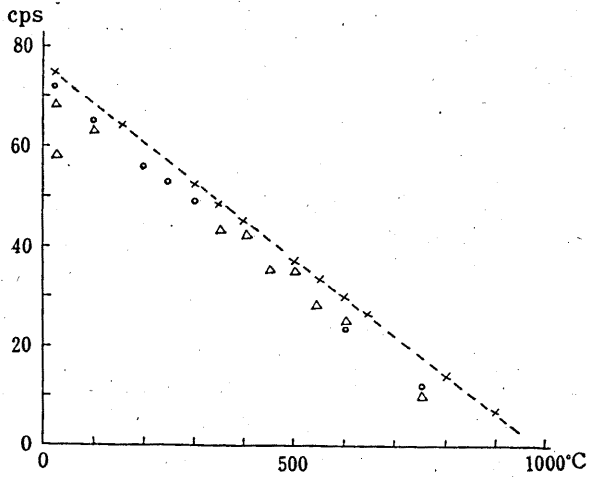


FIG. 3. Intensity variation revealed by (111) of quartz.  
 x heated continuously (5°C/min).  
 o cooled continuously.  
 Δ allowed to cooling.

ed in Figs. 1, 2, and 3.

Inspection of these figures illustrates that certain spacings shown by pure quartz evidently reveal thermal expansion at the temperatures up to  $575^{\circ}\text{C}\pm$  and thenceforth invariable values or no trace of conversion into tridymite or others up to  $1000^{\circ}\text{C}$  indifferent to heat treatment. To be noticed is however that this is, as will later be alluded to, not the case with other kinds of minerals and, furthermore, that the intensity of diffraction relating to certain spacings is liable to be intensely controlled by the heating speed. As is deducible easily from comparison of Fig. 2(a) with 2(b), rapid heating, for example, in the rate of  $5^{\circ}\text{C}/\text{min}$  seems not suitable for complete transition from low-temperature modification to higher one, and duration of heating for an hour at respective temperature is likely to making it easier even at the temperature fairly lower than that so far determined.

### III. EXPERIMENTS

In the light of the results mentioned above, similar experiments were put into operation in order to clarify the essence of variations in the inner structures of some layer-structured and metamict minerals in comparison with the previous works (*cf.* TAKUBO *et al.*, 1953; WHITE, 1958; CARROLL, 1959; ERNST *et al.*, 1961; WARSHAW *et al.*, 1961).

#### (1) Tests for pyrophyllite

The specimens collected from Shôkôzan, Shôbara City, Hiroshima Prefecture were provided for thermal treatments. The main spacings obtained both at room temperature and at  $1000^{\circ}\text{C}$  are listed in Table 2.

TABLE 2 RÖNTGENOMETRIC DATA FOR PYROPHYLLITE FROM SHÔKÔZAN

hkl	Room temp.		Kept const. at $1000^{\circ}\text{C}$ for 1 hr.		ASTM	
	d	I	d	I	d	I/I <sub>1</sub>
002	9.18	56	9.41	31	9.14	40
004	4.596	40	4.713	35	4.57	50
111, 112	4.186	10	4.418	19	4.15	20
					3.87	5
113	3.389	3	3.486	4	3.34	20-40
006	3.064	79	3.145	78	3.04	100
	2.567	6	2.607	3		
	2.548	7	2.574	3		
$\bar{1}32$ , 200	2.529	9	2.539	5	2.52	20
$\bar{2}04$ , 132	2.414	12	2.430	4	2.40	40
008	2.300	4	⋮	⋮	2.29	20
	⋮	⋮	⋮	⋮	⋮	⋮





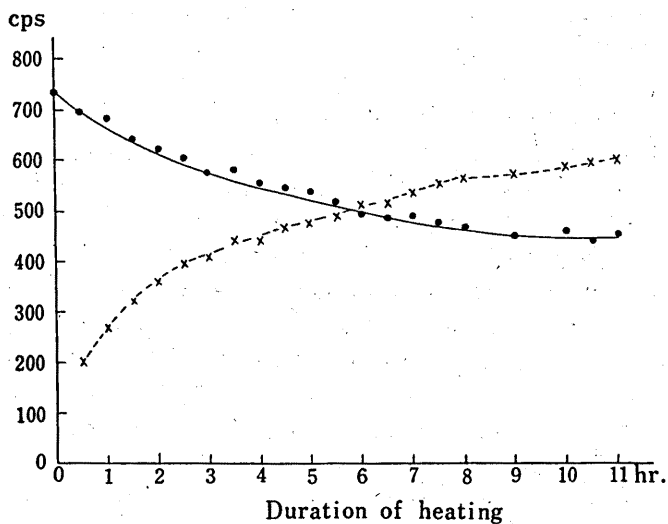


Fig. 5(a) Intensity variations resulted from Fig. 4(a) indicating splitting of (006) of pyrophyllite with duration of heating at 580°C.

- (006)
- × Spacing splitted from the former.

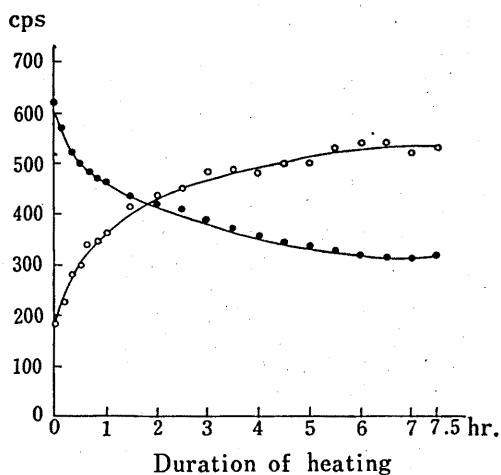


Fig. 5(b) Intensity variations indicating splitting of (006) of pyrophyllite with duration of heating at 600°C.

- (006)
- × Spacing splitted from the former.

the structural variation of mineral.

On the other hand, a sort of endothermal peak of pyrophyllite, ascribed to setting-free of  $[\text{OH}]$ , observed on *DTA* curve has commonly been believed to be

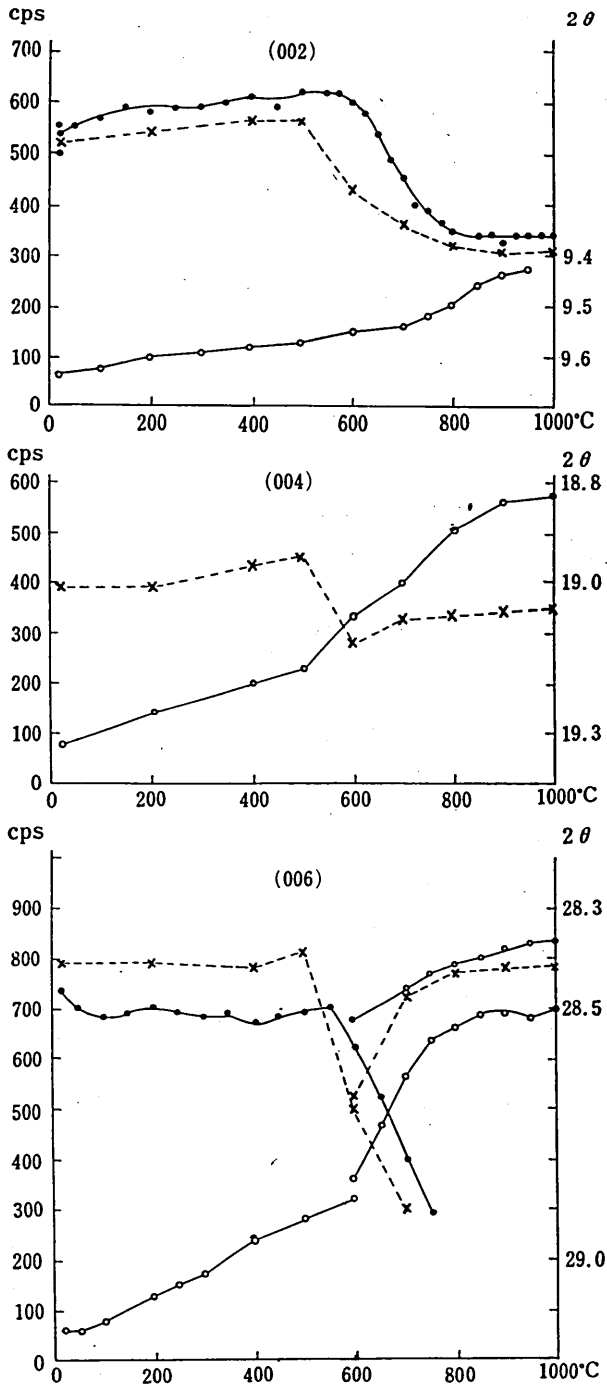


FIG. 6 Variation of spacings of pyrophyllite

× (cps) kept constant for 1 hr. at each temperature. • (cps) heated continuously.  
 o (2θ) kept constant for 1 hr. at each temperature.

completed at a temperature between 650° and 680°C, whereas the real variation is to be taken place in the representative layer spacings with duration of heating at the temperature lower than that generally expected through *DTA* and thenceforth becomes more complicated not simply because of disappearance of [OH] but rather on account of evolution of new spacing. This may also be the case with (004) to a certain extent.

(2) *Tests for muscovite*

Pure specimens sampled from Nako, Abu-gun, Yamaguchi Prefecture were subjected to the similar procedures excepting that *EMF* and current intensity of the X-ray tube were raised respectively to 35 *KV* and 20 *mA*. Most of the main spacings recognized both at room temperature and at 1000°C are listed in Table 3.

TABLE 3 RÖNTGENOMETRIC DATA FOR MUSCOVITE FROM NAKO

hkl	Room temp.		Kept const. at 1000°C for 1 hr.		A S T M	
	d	I	d	I	d	I/I <sub>1</sub>
002	10.02	49	10.19	26	9.95	95
004	5.01	30	5.11	36	4.97	31
110					4.47	21
111	4.47	6	4.511	10	4.30	4
022					4.14	4
112					3.95	6
113					3.882	14
023					3.731	17
114					3.489	22
024					3.342	23
006	3.340	65	3.413	68	3.220	100
114	3.198	7	3.262	6	3.199	28
115					3.122	2
025	2.991	8	3.059	5	2.987	34
115	2.858	7	2.921	5	2.859	24
116	2.793	6	2.840	4	2.789	21
131					2.596	16
202	2.560	7			2.566	54
008	2.503	8	2.560	20	2.505	7
132					2.491	14
133					2.465	8
202					2.450	7
204					2.398	10
133					2.384	27
134					2.254	9
135					2.236	4
221, 204					2.208	7
223					2.189	4
206					2.149	15
135					2.132	21
223	2.127	4			2.070	4
044					2.053	6
00, 10	2.002	28	2.049	15	1.993	46

With specific regard to (002), (004) and (006), variations were continuously pursued during being held for an hour at each temperature. The data concerned are plotted in Fig. 7.

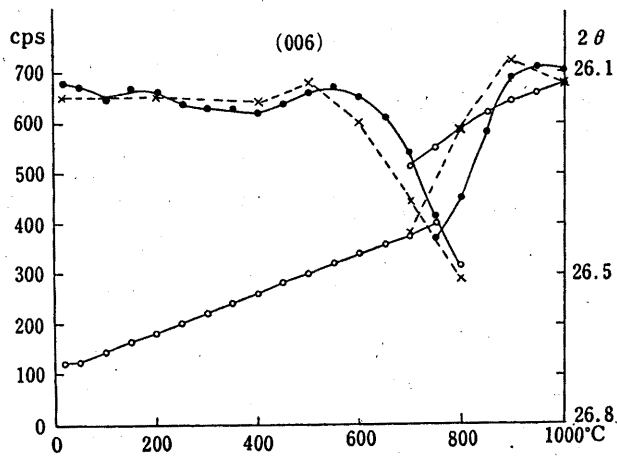
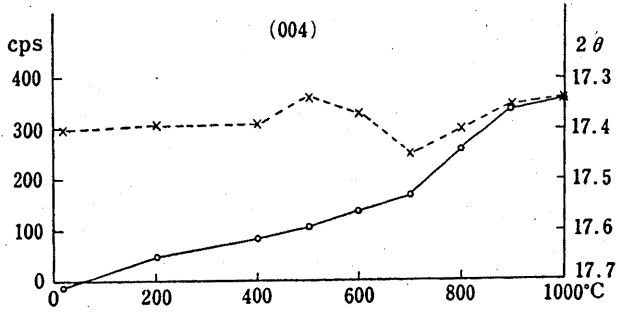
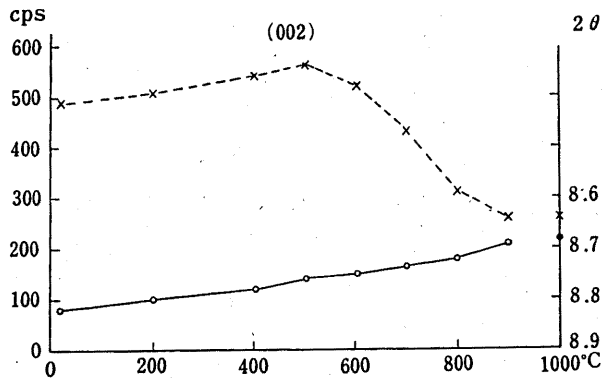


Fig. 7. Variation of spacings of muscovite  
 × (cps) kept constant for 1 hr. at each temperature  
 • (cps) heated continuously  
 ○ (2θ) kept constant for 1 hr. at each temperature or heated continuously

The figure shows that the spacings of (002) and (004) reveal gradual variations without conspicuous discontinuity and that of (006) as well as its intensity illustrate a striking gap at 700°C, although the continuous heating in the rate of 8°C/min without staying at each temperature seems to cause deviation of more than 50°C toward higher-temperature side.

### (3) Tests for dickite

The specimens taken from Shōkōzan were thermally treated in the conditions quite similar to in the preceding cases. Most parts of the principal spacings are tabulated in Table 4.

TABLE 4 RÖNTGENOMETRIC DATA FOR DICKITE FROM SHŌKŌZAN

hkl	Room temp.		Kept const. at 1100°C for 1 hr.		ASTM	
	d	I	d	I	d	I/I <sub>1</sub>
002	7.16	69	8.12	4	7.153	90
020, 110	4.447	5	6.17	3	4.451	70
11 $\bar{1}$	4.380	6	5.42	3	4.366	70
021	4.270	4	5.19	3	4.254	40
111	4.137	6	5.13	3	4.118	80
11 $\bar{2}$	3.960	3	4.191	4	3.953	40
022	3.802	4	3.460	3	3.790	70
004, 112	3.579	72	3.391	3	3.578	100
11 $\bar{3}$	3.430	4	2.543	5	3.428	50
023					3.262	30
113					3.094	40
11 $\bar{4}$					2.936	40
024					2.794	40
114					2.650	10
13 $\bar{1}$ , 200	2.560	5			2.558	70
11 $\bar{5}$					2.524	5
131, 20 $\bar{2}$	2.507	6			2.503	80
006, 132	2.386	10			2.383	50
13 $\bar{3}$ , 202	2.326	11			2.322	90
	⋮	⋮			⋮	⋮

Date for the layer spacings such as (002) and (004) are plotted in Fig. 8, indicating that both are simply expanded up to ca. 800°C, while an abrupt diminution in their intensity seems to take place at ca. 500°C in the case of being kept at respective temperatures for an hour and at ca. 550°C in the case of being continuously heated with higher speed.

Remarkable is that both spacings corresponding to (002) and (004) are almost perfectly disappeared at the temperature above 700°C with very slow heating and

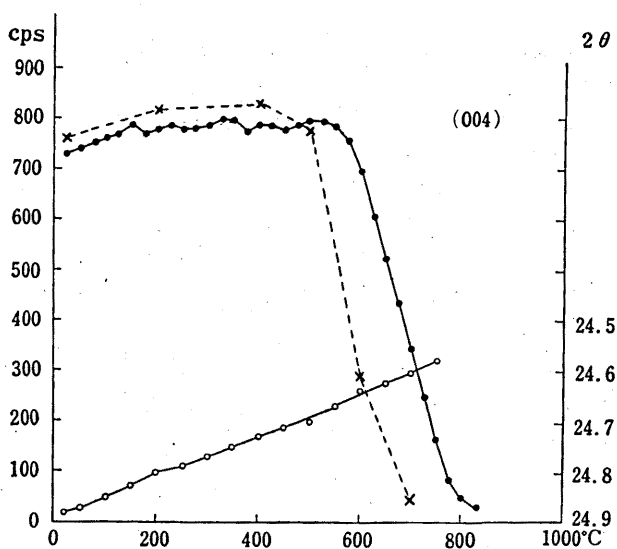
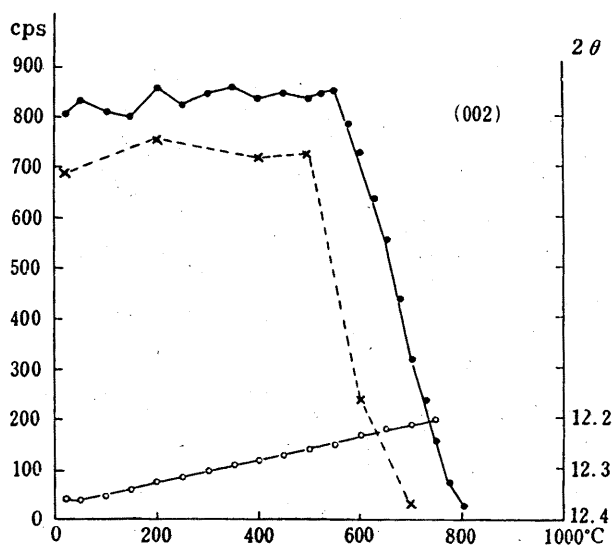


FIG. 8. Variation of spacings of dickite  
 $\times$  (cps) kept constant for 1 hr. at each temperature.  
 $\bullet$  (cps) heated continuously.  
 $\circ$  ( $2\theta$ ) heated continuously.

above 800°C with rapid treatment, and then quite different spacings assumed to represent the growth of a sort of meta-kaolin, though be yet remained to be researched in more details, are appeared.

(4) Tests for fergusonite

Blackish grey-colored specimens selected out from the coastal placer at Hagata-



Inspection of the table surely reveals that simple expansion of certain spacings is, in general, observable with heating of the specimens, while splitting of certain ones into some parts becomes more conspicuous with elevation of temperature and is almost completely remained even after cooling down to room temperature. This surely represents irreversibility of the process at least at the temperature lower than 1000°C. Similar relation is also deducible from the data presented in ASTM card, although the latter be of course connected with those obtained at the temperature 200°C higher than in the case of this experiment and, in consequence, splitting of the spacings be therein more obvious.

(5) *Tests for thorogummite*

Earthy matters picked up from pegmatite deposit occurring in the Tateiwa mine, Hôjô-chô, Onsen-gun, Ehime Prefecture were provided for the experiments. The specimens, faint brownish in color, are sporadically enclosed mainly in albanite and characterized with considerably intense radioactivity (ca. 50 cps/g). On the basis of the same reason as in the case of fergusonite, thermal effects on the mineral in question were pursued at each temperature above 600°C during being kept constant respectively for 10 hours, resulting in overlapping of the heating at higher temperatures. The results are listed in Table 6, wherein the spacings with asterices in the column of 1000°C are considered to be corresponding to those for thorite produced through thermal effects.

In this case, the fact is that the mineral under consideration is originally well-crystallized at ordinary state in spite of its earthy appearance, and nevertheless, it was accountably difficult to determine the positions of diffractive peaks drawn on the recorder because of their severe fluctuation and higher background ascribed to radioactivity of the elements contained. At any rate, scattering of almost all spacings seems to taken place already at ca. 600°C and to reach its culmination at ca. 900°C, whereas crystallinity recognized at 1000°C is apparently of the highest grade if merely based on numerous spacings but this is to be ascribed to a quite different condition due to production of thorite. On cooling down to room temperature, all the spacings appeared during thermal manipulation indicate a nearly complete recovery to the initial state without remainders of variation either in  $2\theta$  or in intensity, suggesting evidently a sort of reversible process. As has already been anticipated, the data obtained in these experiments are believed more accurate and useful for confirming the real variation appeared in the inner structures of certain minerals through thermal effects than the previous ones given at ordinary state simply after heating and, at the same time, more essential for enlightening the relation of secondarily produced minerals to the original one, metamictization of certain minerals and so on than those so far presented only on the basis of chemical connection with any other resembling minerals.



TABLE 6 RÖNTGENOMETRIC DATA FOR THOROGUMMITE

hkl	Room temp.		at 600°C		at 700°C		at 800°C		at 900°C		at 1000°C		allowed to cooling down to room temp.		A S T M	
	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>	d	I/I <sub>1</sub>
011	4.690	100	4.690	100	4.70	100	4.70 <sub>B</sub>	100	(4.76) (4.70)	90	5.28 (4.77) (4.68)	40 100	4.720	100	4.695	90
200	3.539	90	4.150 (3.531) (3.500)	50 70	4.14 (3.537) (3.507)	60 80	4.15 <sub>B</sub> (3.537) (3.490)	60 80 70	4.15 <sub>B</sub> (3.555) (3.520)	60 100	4.15 (3.572) (3.541*)	70 100	3.548	90	3.537	100
121	3.267	60	3.267	60	3.273	70	3.270	60	3.270	60	3.275 3.226* 3.172*	50 60 60	3.175	60		
112	2.821	50	2.823 <sub>B</sub>	50	2.820	60					(2.858) (2.831)	40	2.840	50	2.821	40
220	2.671	60	2.671	60	2.675	50	(2.720) (2.680 <sub>B</sub> )	50 50	2.675 <sub>B</sub>	60	2.679* 2.546*	60 40	2.694	60	2.653	60
022	2.644	50									2.227 (2.219*)	30	2.663	70	2.499	30
031	2.205	30	(2.195) (2.186)	40	(2.201 <sub>B</sub> ) (2.187 <sub>B</sub> )	40	(2.212) (2.191)	40	2.212 <sub>B</sub>	40	2.006 1.979	30 40	2.206	40	2.203	40
013	2.002	30	2.002	30			1.947	40	1.945 <sub>B</sub>	50	(1.955*) (1.938)	30 40			2.000	40
321	1.868	40	1.866	40	1.871	50			1.859	50	(1.879) (1.866)	40	1.878	40	1.869	30
312	1.819	50	1.821	50	1.817	50	1.824	50	(1.829) (1.817)	70 60	(1.843*) (1.833 <sub>B</sub> *)	50			1.818	60
400	1.768	40	1.794	40			1.801	50	1.794	50	1.812	50	1.826	70	1.767	10
123							1.786	50	1.768	40	1.765 <sub>B</sub>	30	1.763	30	1.740	10
411							1.697	40	1.656	50	1.689 1.661*	30 40	1.650	40	1.653	10

\* Probably thorite

## V. CONCLUSION

- (1) Thermal variations of certain minerals bearing the layer-structures are considered to be taken place more easily in their sub- or inter-layers than in the main ones and in the case of other manipulations such as *DTA* used to resulting in the apparent transition appeared generally at the temperature higher than be really so. In other words, conversion of one modification into the other is apt to being observed during being kept constant for long hours at the temperature considerably lower than in the case of continuous rapid heating, exclusive of the transition from low- to high-quartz.
- (2) In spite of simple expansion revealed by the spacings with elevation of temperature, it often happens that the intensity corresponding to the same ones independently indicates the remarkable discontinuity at the related temperature. This may also be reflected on the data obtained through rapid heating as a sort of discontinuity even though at the temperature considerably higher than that necessary for real transition of the inner structure.
- (3) In the layer-structures, thermal variations of the spacings other than those of basal planes are not so much distinct as be expected in other manipulations and accordingly useless for inspection.
- (4) Special cares must be taken on researching the metamict structures, inasmuch as their variations are more complicated than were presented in the previous views that the metamict state should be 'irreversibly' converted into the crystalline one through simple heating regardless of difference of the process in that both reversible and irreversible variations are possible in accordance with respective characteristics attached to the structures concerned.
- (6) It seems necessary to allude to that conspicuous fluctuation of the positions of the spacings recorded in the structures containing radioactive elements in more or less amount and the increment of the spacings in number through thermal effects are attributed either to raising of crystallinity of the related structure or to the formation of new modification, necessitating the re-inspection of the previous works on one hand and furthermore scrutiny concerning the inner structures of various minerals on the other.
- (7) It has become clear that duration of heating at each temperature and detailed pursuit of the crystal structures both at the state during heated or cooled at respective temperatures and in ordinary condition recovered from the preceding state are considered most indispensable for this kind of study, and it is to be added to that deduction derived merely from chemical analyses is liable to falling into false understanding in relation to mineralogenesis.

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