A NEW OCCURRENCE OF BARIUM-FELDSPAR AT OTJOSONDU, SOUTH-WEST AFRICA, AND AN X-RAY METHOD FOR DETERMINING THE COMPOSITION OF HYALOPHANE

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Abstract

Four new barium-feldspars Cn_{94} , Cn_{55} , Cn_{50} and Cn_{10} are described. The celsian (Cn_{94}) contains 2.24% Fe₂O₃ (Fe³⁺ probably replacing Al³⁺) and has refractive indices much higher than material of about the same composition but for iron viz. $\gamma = 1.608$, $\beta = 1.599$ and $\alpha = 1.593$. Similar divergencies occurred in its x-ray diffraction pattern.

Adularia and not sanidine is favoured as the potassium end-member of the series $KAlSi_3O_8-BaAl_2Si_2O_8$. A sudden change in the intensities for similar reflections between the compositions Cn_{37} and Cn_{50} indicates two isomorphous series ($Cn_0-Cn_{40\pm}$ and $Cn_{45\pm}-Cn_{100}$) rather than one (Cn_0-Cn_{100}).

Four variation diagrams based on x-ray diffraction data are offered by means of which the BaO-content of hyalophane ($Cn_0-Cn_{40\pm}$) can be determined to an accuracy of $\pm 5\%$.

INTRODUCTION

In a study of the manganese ores of Otjosondu, South-West Africa, de Villiers (1, p. 95) described two barium-feldspars, hyalophane (Cn_{10}) and celsian (Cn_{94}) . Subsequently two feldspars of intermediate composition, Cn_{55} and Cn_{50} , were also found at the same locality.

All these feldspars do not appear to be confined to any particular type of ore. The celsian is associated with a fine-grained vredenburgite-garnet rock, whereas the hyalophane is present as small bodies of irregular shape in a rock composed chiefly of calderite garnet, or as small veinlets traversing the garnet. This celsian and hyalophane were present in amounts large enough to allow their separation from the country rock by mechanical means, and in quantities large enough for detailed chemical analyses. The two intermediate ones (Cn_{55} and Cn_{50}) occur in rocks composed of braunite, garnet and hollandite. They were present in very small quantities and the material eventually submitted for chemical analyses was insufficient for detailed analytical determinations.

The chemical analyses listed are corrected for impurities originally contaminated with the material analyzed. The spectrographic analyses were carried out on material handpicked under the microscope.

CHEMICAL COMPOSITION

(a) Celsian (Cn_{94})

Spectrographic analysis Present: Ba, Al, Si, K, Na, Fe, Ca. Trace: Mg, Mn. Absent: Rb, Cs, Li, Sr, B, Be, Ti.

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Chemical analysis		Mol. prop.	Molecular composition	
SiO_2	33.20	0.5528	Celsian (Cn)	94.49
Al_2O_3	25.20	0.2472	Orthoclase (Or)	3.06
Fe_2O_3	2.24	0.0140	Albite (Ab)	1.05
CaO	0.12	0.0030	Anorthite (An)	0.61
Na ₂ O	0.12	0.0019		
K_2O	0.52	0.0055	Total	99.16
BaO	38.62	0.2518		
Total	100.04			

TABLE 1

Chemical analysis by C. F. J. van der Walt.

A very unusual aspect of the chemical composition of this celsian is the relatively large proportion of Fe³⁺ which probably replaces Al³⁺. Proof that the Fe³⁺ is contained in the celsian molecule was obtained by grinding handpicked grains of the mineral to a fine powder and boiling the powder in concentrated hydrochloric acid for long intervals over a period of 72 hours. After thorough washing, a spectrographic analysis showed no weakening of the iron lines 3719.9 Å, 3099.9 Å, 3099.97 Å and 3100.3 Å.

The refractive indices of this celsian are also much higher than any recorded in the literature for material of about the same composition, except for the iron. Its x-ray diffraction pattern also shows deviations from that of iron-free celsian (Cn_{89}) from Jakobsberg, Sweden. These deviations support the view that at least a large proportion, if not all the Fe₂O₃ indicated by the chemical analysis, is contained in the celsian molecule. This Fe³⁺ in the molecule evidently causes the abnormalities in its physical properties.

The remaining minor constituents, viz. Na and Ca, are probably not responsible for either the relatively high refractive indices or the divergencies in the angle 2θ for similar reflections, because in most alkali feld-spars (which contain these elements in small quantities) their influence is negligible. The spectrographic analysis also indicates that no other elements are present in quantities large enough to have caused these differences.

(b) Hyalophane (Cn_{10})

Spectrographic analysis Present: Si, Al, K, Na, Ba. Trace: Sr, Mg, Fe, Ca. Absent: Rb, Cs, Li, B, Be, Mn, Ti.

Chemical analysis		Mol. prop.	Molecular composition		
SiO ₂	61.17	1.0185	Celsian (Cn)	9.51	
Al_2O_3	20.35	0.1996	Orthoclase (Or)	68.15	
Na_2O	2.92	0.0471	Albite (Ab)	18.28	
$K_{2}O$	11.52	0.1223	Nepheline (Ne)	3.44	
BaO	3.89	0.0254			
			Total	99.38	
Total	99.85				

TABLE 2

Chemical analysis by C. F. J. van der Walt.

In the calculation of the norm of hyalophane there is a deficiency of silica which results in the formation of a little nepheline. At the same time there is a surplus of 0.47 per cent Al_2O_3 . It is, therefore, likely that some Al^{3+} and Ba^{2+} have replaced Si⁴⁺ and Na⁺ and/or K⁺.

The presence of iron (Fe³⁺ or Fe²⁺) in this hyalophane (Cn₁₀) is doubtful because after treatment with acid, in the same way as the celsian, only faint traces of iron could be detected spectrographically.

(c) The two intermediate felds pars $(Cn_{50} \text{ and } Cn_{55})$

Due to the insufficient amounts of available material only the BaO content of these two samples could be determined chemically. They contained 22.5 per cent BaO (Cn_{55}) and 20.4 per cent BaO (Cn_{50}). Spectrographically it was proved that the former (Cn_{55}) contains iron, but the latter was iron-free.

PHYSICAL PROPERTIES

(a) Optical

As seen in thin section these barium-feldspars are rarely twinned. They

Sample	F	Refractive indice	Axial angle		
	α	β	γ	2V (meas.)	2V (cal.)
Cn94	1.593	1.599	1.608	+85°	+78°30'
Cn ₅₅	1.565	1.572	1.574	-40°	$-41^{\circ}22'$
Cn ₅₀	1.555	1.562	1.564	-43°	$-41^{\circ}22'$
Cn ₁₀	1.525	1.530	1.532	-62°	$-64^{\circ}36'$

TABLE 3. OPTICAL PROPERTIES OF S.W.A. BARIUM-FELDSPARS

NT.	Hyalophane (Cn ₁₀)		(Cn10)	Celsian, S.W.A. (Cn ₉₄)			Celsian, Sweden (Cn ₈₉)		
No.	Int.	2θ(Cu)	d Å	Int.	20(Cu)	d Å	Int.	2θ(Cu)	d Å
1	3	13.55	6.53	4	13.83	6.39	3	13.98	6.33
2	1	15.09	5.87						
3				2	19.42	4.57	3	19.59	4.53
4	6	21.11	4.01						
5	2	22.61	3.93						
6	8	23.60	3.77	3	23.56	3.77	3	23.75	3.74
7	1	24.69	3.60						
8	5	25.71	3.46	6	25.88	3.44	6	26.06	3.42
9	9	26.98	3.30	7	26.76	3.33	7	26.97	3.30
10	10	27.65	3.22	7	27.52	3.24	7	27.73	3.21
11	8	29.92	2.98	7	29.72	3.00	8	29.94	2.98
12	7	30.80	2.901	4	30.80	2.901	4	31.00	2.882
13	5	32.42	2.759	5	32.43	2.758	5	32.64	2.741
14	8	34.85	2.572	10	34.82	2.574	10	35.06	2.557
15	1	37.01	2.427	3	37.35	2.406	3	37.42	2.401
16	2	38.79	2.319	3	38.85	2.316	3	38.96	2.310
17				2	39.80	2.263	1	39.93	2.256
18	6	41.75	2.162	10	41.65	2.167	10	41.85	2.157
19	1	42.76	2.113	1	42.94	2.104	1	43.02	2.101
20	1	43.98	2.057	1	44.00	2.056	1	44.31	2.042
21	1	45.20	2.004	2	45.46	1.994	2	45.70	1.984
22	1	46.05	1.969	2	46.62	1.946	2	46.68	1.944
23	2	47.30	1.920					n (
24	1	49.15	1.852	1	49.00	1.857	1	49.41	1.843
25	8	50.78	1.796	5	50.63	1.801	4	51.00	1.789
26	1	54.81	1.673	1	55.71	1.676	1	55.83	1.673
27	2	56.54	1.626	2	56.58	1.625	3	56.72	1.622
28	2	58.78	1.570	2	58.53	1.576	1	58.71	1.571
29	1	60.50	1.529	1	60.87	1.521	1	60.93	1.519
30	7	62.05	1.494	5	63.93	1.455	5	62.02	1.495
31				6	65.30	1.428	5	65.42	1.425
32				2	70.00	1.343	2	70.09	1.342
33				2	71.58	1.317	2	71.72	1.315
34				1	73.50	1.287			

TABLE 4. X-RAY DIFFRACTION DATA OF BARIUM-FELDSPARS

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usually exhibit only one good cleavage (001); but in some cases a cleavage parallel to (010) is also developed. Refractive indices were determined according to a single variation method on a Universal stage. The optical properties are listed in Table 3 below. Refractive indices are correct to ± 0.003 at 22° C.

X-ray examination

X-ray diffraction patterns were obtained by using 114.59 mm. diameter cameras and exposing for 2.5 hours to unfiltered Cu-radiation at 35 kV. and 20 mA. The mounting of the films in the cameras, the shrinkage correction factors and the measuring of the films were carried out in the same way as described by the author (2, pp. 946–947). The values of 2θ as listed in Table 4 are considered to be correct to ± 0.03 degrees.

A sample of celsian from Jakobsberg, Sweden (ex. Harvard Univ. Museum No. 10062) containing 36.4 per cent BaO (Cn₈₉) (analysis by C. F. J. van der Walt) was also examined for comparative purposes (see Table 4). This sample is free from iron, its chemical composition is otherwise comparable with that of the S.W.A. celsian, but the *x*-ray diffraction data on these two minerals vary greatly, no doubt because of the Fe³⁺ which is contained in the unit cell of the South-West African sample.

Table 4 clearly reveals the effect of the Fe³⁺ in the unit cell of the S.W.A. celsian. Similar differences are also encountered when the x-ray diffraction data for the two intermediate samples Cn_{50} (containing no iron) and Cn_{55} (containing 2.9% Fe₂O₃) are compared.

Another striking feature revealed by Table 4 and Plate 1 is that the intensities of reflections from similar crystal planes for hyalophane and celsian are not the same. Table 4 only allows a vertical comparison of intensities visually estimated using an arbitrary scale from zero to 10 for each spectrum. It is clear that the strongest reflections of hyalophane have lost their prominence in the celsian pattern. Fairly weak reflections of hyalophane have become the more prominent ones in the celsian pattern. This alteration of intensities is indicative of some change that has taken place in the crystal structure of barium-feldspars on proceeding from hyalophane (Cn₁₀) in the series KAlSi₃O₈—BaAl₂Si₂O₈ to celsian (Cn₉₄).

Taylor, Darbyshire and Strunz (3, p. 476) suggested that the potassium-rich members of the series are monoclinic, but that celsian is triclinic and only pseudo-monoclinic dimensionally. From this suggestion it would follow that there should be a transition point from KAlSi₃O₈ to $BaAl_2Si_2O_8$, indicating the change in the nature of the dominant atom and separating two isomorphous series from one another. The position of

such a transition point will be determined by the amount of Ba^{2+} that can replace K^+ in hyalophane and at the same time still be bonded into a monoclinic unit. When this critical point is exceeded the structure breaks down to adopt triclinic symmetry which is maintained until all K^+ has been replaced by Ba^{2+} .

NOTES ON THE SERIES KAISi₃O₈-BaAl₂Si₂O₈

Winchell (4, p. 354) suggested a single isomorphous series between adularia and celsian. He regards hyalophane as a barium-bearing adularia, but is uncertain whether celsian is related to adularia or sanidine, the high temperature form of potash-feldspar. According to Winchell's variation diagram the axial angle of the feldspar Cn_{70} is nearly zero. The low axial angles determined on the two South-West African samples supports the decrease in 2V observed for members between Cn_{60} and Cn_{80} .

The variation diagram of Yosimura (6, p. 372) shows notable differences when compared with that of Winchell. Yosimura indicates a break in properties at a point representing hyalophane of about Cn_{40} (BaO 16%).

A persusal of the literature on the Ba-feldspars (some 45 papers were consulted) reveals such irreconcilable data, that even after verification with new data of South-West African samples, no reliable variation diagram of the optical properties of this series could be compiled.

In order to construct variation diagrams based upon x-ray diffraction it was decided also to investigate a few samples from known localities, bringing the total number of samples examined to fourteen.

THE MATERIAL USED FOR CONSTRUCTING VARIATION DIAGRAMS

(a) Adularia

(i) Binnenthal, Valais, Switzerland. (ex. Pretoria Univ. Museum). $2V_{\alpha}=62^{\circ}$. Spectrographic analysis indicates traces of Ca, Ba, Mg and Sr apart from the major constituents Si, Al, K, and Na.

(ii) Gotthard, Switzerland. (ex. Transvaal Museum). $2V_{\alpha}=64^{\circ}$. Apart from the major constituents Si, Al, K, and Na; traces of Mn, Ca, Ba, Sr, Mg and Rb were encountered.

(iii) Mayrhofen, Zillertal, Tyrol. (ex. Transvaal Museum). $2V_{\alpha}=63^{\circ}$. Traces of Ba and Sr were found on the spectrograph in addition to the major components.

The x-ray diffraction patterns of these three adularia samples are identical. The angle 2θ for similar reflections was found to vary only within the limits of experimental error. The average result for these three samples was used in the variation diagrams where it is represented by the letter a.

(b) Sanidine

(i) Mt. Cumini, Italy. (ex. Transvaal Museum). $2V_{\alpha} = 14^{\circ}$. The major constituents are Si, Al, Na, and K with traces of Ba, Sr, Rb and Ca.

(ii) Bolanitos Quarry, Mt. Guagujuata, Mexico. (ex. Transvaal Museum). $2V_{\alpha} = 40^{\circ}$. Sodium is not a major constituent but only present as a trace, so also with Ca, Ba and Rb.

(iii) Laacher See, Eifel. (ex. Transvaal Museum). $2V_{\alpha}=8^{\circ}$. Apart from the major constituents Si, Al, K and Na, it also contains traces of Fe, Sr, Ba, Rb and Ca.

The angle 2θ for similar reflections for these three samples of sanidine was also identical within the limits of experimental error. In the construction of the variation diagrams the average value for these three samples was used and is indicated by the letter b.

The soda content of five of the above-mentioned samples of adularia and sanidine lies between 0.12 per cent and 2 per cent, and that of the specimen from Mexico was determined to be about 0.12 per cent. This conclusion was reached by taking the three barium-feldspars of known Na-content as standards and deducing by comparison of the intensities of the following sodium lines 3302.3 Å, 3303.0 Å, 4494.3 Å, 4497.7 Å and 4748.0 Å, which represent the sodium content.

(c) Barium-feldspars

(i) Hyalophane, S.W.A. (Cn₁₀). Represented by the letter c in the diagrams.

(ii) Hyalophane, Franklin, New Jersey, U.S.A. (ex. Harvard Univ. Museum). Material described by Bauer and Palache (5, p. 174). The chemical analysis of this material is: $SiO_2=49.51\%$, $Al_2O_3=25.48\%$, $K_2O=9.98\%$, $Na_2O=3.56\%$ and BaO=11.47% (Cn₂₈). A spectrographic analysis of the acid purified material reveals a certain amount of Ca, thus indicating that the BaO-content of the chemical analysis may be slightly too high. Its refractive index also indicates a chemical composition of Cn₂₄ rather than Cn₂₈. The specimen is represented by the letter *d* on the graphs.

(iii) Hyalophane, Binnenthal, Valais. (ex. Transvaal Museum). The following is the analysis by C. F. J. v.d. Walt.

Chemical analysis		Mol. prop.	Molecular composition	
SiO_2	51.10	0.8508	Celsian (Cn)	37.28
Al_2O_3	22.10	0.2168	Orthoclase (Or)	47.23
Fe_2O_3	0.40	0.0025	Albite (Ab)	8.45
MgO	0.54	0.0134	Anorthite (An)	3.07
CaO	0.63	0.0112	Nepheline (Ne)	4.28
BaO	15.24	0.0994		
Na ₂ O	2.00	0.0323	Total	100.31
K_2O	8.00	0.0849		
Total	100.01			

TABLE 5. CHEMICAL ANALYSIS OF HYALOPHANE, BINNENTHAL, VALAIS

Represented by the letter e on the diagrams.

(iv) Hyalophane, Binnenthal, Valais. (ex. Harvard Univ. Museum No. 10065). The refractive indices of this mineral indicate that its chemical composition is about Cn_{30} . It is represented by the letter f on the diagrams.

(v) Hyalophane, South-West Africa. Cn50, represented by the letter g on the diagrams.

(vi) Hyalophane, South-West Africa, Cn_{55} , represented by the letter h on the diagrams.

(vii) Celsian, Jakobsberg, Sweden. (ex. Harvard Univ. Mus. No. 10062). The BaOcontent, determined by C. F. J. v.d. Walt, is 36.4 per cent. The spectrographic analysis indicates less than 2 per cent Na₂O. It appears, therefore, that this material is identical to that analyzed by Strandmark (7, p. 298). According to his analysis the material contains 0.22% K₂O and 0.77% Na₂O apart from SiO₂, Al₂O₃ and BaO. The molecular composition of this feldspar is, therefore, Cn₈₉. It is represented by the letter *i* on the graphs.

(viii) Celsian, South-West Africa, represented by the letter j on the graphs.

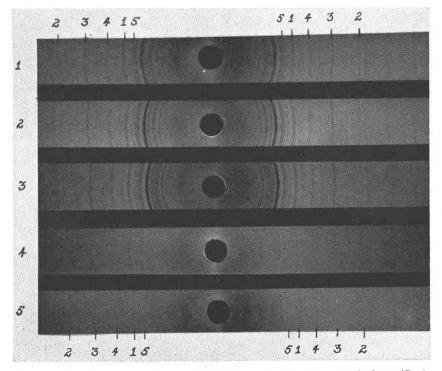


PLATE 1. X-RAY DIFFRACTION PATTERNS (CuK) OF: 1. Adularia. 2. Hyalophane (Cn₁₀). 3. Hyalophane (Cn₃₇). 4. Hyalophane (Cn₅₀). 5. Celsian (Cn₅₀).

It seems that the potassium end-members of the series is represented by adularia, the low temperature form of orthoclase rather than by sanidine. This is suggested, in the first place, by the axial angles: $63^{\circ} \pm 1^{\circ}$ for almost Ba-free adularia and 62° for hyalophane of composition Cn₁₀, whereas the values for sanidine lie between 8° and 42°.

Moreover, adularia fits much better than does sanidine on the variation curves in Figs. 1 to 4, which are based on x-ray diffraction data.

The intensities of the reflections in the x-ray patterns are the same for adularia and sanidine, as well as for hyalophane of composition Cn_{10} ,

Cn₂₄, Cn₃₀ and Cn₃₇. These patterns are characterized by the prominent doublet (d=3.29 and d=3.22 Å) for Cn₃₇. (Lines nos. 9 and 10 in Table 4). This doublet loses its prominence in the patterns of the members of composition Cn₅₀, Cn₅₅, Cn₈₉ and Cn₉₄. An examination of Plate 1 discloses numerous other differences in the relative intensities. This sudden change in the intensities for similar reflections indicates that the "transition point" from a monoclinic to a triclinic unit cell is located between the compositions Cn₃₇ and Cn₅₀, thus giving rise to two series Cn₀ - Cn_{40±} and Cn_{45±} - Cn₁₀₀.

Theoretically one would have expected the transition point to be at the composition Cn_{50} , but it clearly occurs displaced slightly towards the potassium end.

Claisse (8, p. 419) has pointed out that in the case of the plagioclase series (having trinclinic symmetry from Ab₀ to Ab₁₀₀) two isomorphous series exist, with a transition point at about Ab₆₆ instead of Ab₅₀. In albite the Na⁺ ion is surrounded by 6 oxygen atoms, whereas the dominant Ca^{2+} ion of anorthite is probably surrounded by 7 or 8 oxygen atoms, indicating that albite would more easily replace anorthite than vice versa, since the Na⁺ ion would find its 6 oxygen atoms in the anorthite

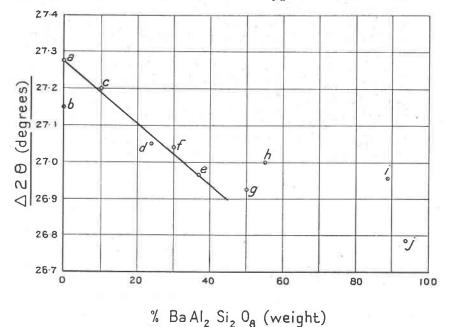


FIG. 1. Graphical representation of the variation of $\Delta 2\theta$ for lines 2-1 with the variation in the chemical composition of hyalophane.

June i =
$$\#(4) = 34.85^{\circ} = 2.57^{2}$$

June 2 = $\# 30 = 62^{\circ}.05^{\circ} = 1.494$

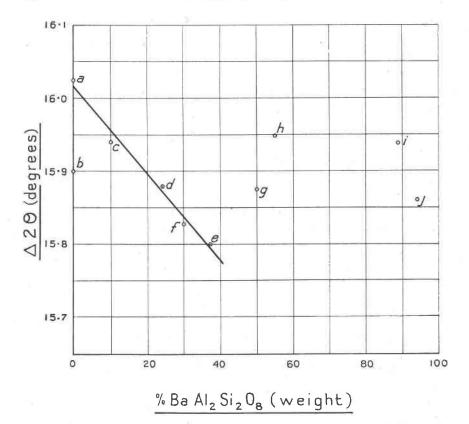


FIG. 2. Graphical representation of the variation of $\Delta 2\theta$ for lines 3-1 with the variation in the chemical composition of hyalophane.

structure more easily than the Ca^{2+} ion could find 7 or 8 in the albite structure.

Taylor, Darbyshire and Strunz (3, p. 477) say that the oxygen atom arrangement is probably substantially the same in all potassium-barium feldspars, although small differences may cause the transition point to be about Cn_{40-45} instead of Cn_{50} .

When the two series NaAlSi₃O₈-CaAl₂SiO₈ and KAlSi₃O₈-BaAl₂Si₂O₈ are compared in general, the first and most obvious resemblance is the similarity of the replacement. In both cases the smaller univalent ion $(K^+=1.33 \text{ Å}, Na^+=0.98 \text{ Å})$ is replaced by the larger divalent ion $(Ba^{2+}=1.43, Ca^{2+}=1.06 \text{ Å})$ accompanied by an equal replacement of Si⁴⁺ by Al³⁺. From a geometrical viewpoint it can therefore be expected that similar changes in the environment of the dominant cation will take place in both series when the nature of the dominant cation changes. Direct comparison of the change in the environmental condition of the two series at the transition point is however impossible, as in the case of the Ba-feldspars the transition point also denotes a change in crystal symmetry from monoclinic to triclinic whereas in the plagioclases the triclinic symmetry is maintained throughout the entire series.

An X-ray Method for Determining the Composition of Hyalophane

According to Taylor, Darbyshire and Strunz (3, p. 472) the unit cells of barium-feldspars ranging in composition from Cn_{10} to Cn_{37} have approximately the same dimensions. As the physical properties of solid solutions vary with their composition, a study of this series was undertaken by *x*-ray diffraction as a basis of future identification in view of the difficulties experienced by using optical and other methods for this purpose.

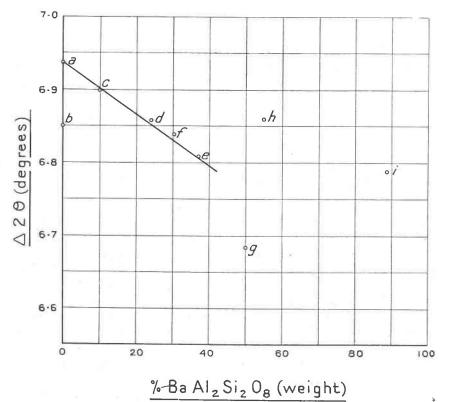
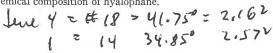


FIG. 3. Graphical representation of the variation of $\Delta 2\theta$ for lines 4-1 with the variation in the chemical composition of hyalophane.



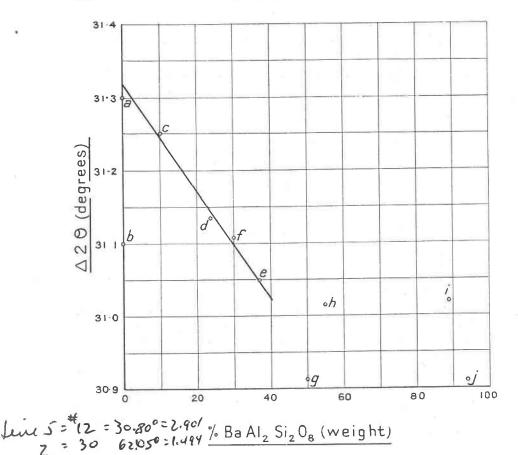


FIGURE 4. Graphical representation of the variation of $\Delta 2\theta$ for lines 5-2 with the variation in the chemical composition of hyalophane.

In any case, there are insufficient representatives known in the range Cn_{50} to Cn_{100} to construct a variation diagram for this second series. Nevertheless, the relevant properties of the few known examples of this composition have also been plotted on the diagrams (Figs. 1 to 4), in order to illustrate clearly the transition point as well as the effect of the replacement by Fe^{3+} of Al^{3+} and possibly of other cations.

In the diffraction patterns of hyalophane of composition ranging from Cn_0 to Cn_{37} it was found that for a given pair of lines the shift was in opposite directions. Because of this complementary shifting of lines due to a change in composition it was found that the differences between the distances of a given pair of lines is a more sensitive method than the differences in the actual distances of each line for different samples.

The data for adularia (a) and sanidine (b) as plotted on all the diagrams

indicate the differences between them and also show that adularia and not sanidine is to be regarded as the potassium end-member.

For the construction of the variation diagrams the lines marked 1, 2, 3, 4 and 5 on Plate 1 were selected as they are relatively prominent, sharply defined and were found to have shifted more than the others. The positions of these five lines are as follows for hyalophane (Cn_{10}) in Table 4.

Line no.	Line no. in Table 4	$d A.$ (for Cn_{10})
1	14	2.574
2	30	1.496
3	25	1.798
4	18	2.163
5	12	2.903

The following four variation diagrams will suffice for purposes of identification.

Precision

The values of 2θ are correct to 0.03 degrees. Therefore, the maximum error of $\Delta 2\theta$ could be 0.06 degrees. Hence, each diagram represents the BaAl₂Si₂O₈-content accurately to within 8 per cent. By using three or more of the diagrams accuracy should be ± 5 per cent BaAl₂Si₂O₈, corresponding to a BaO-content of ± 2 per cent.

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