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BULLETIN No. 75

CONTRIBUTIONS TO
THE MINERALOGY OF ILÍMAUSSAQ
Nos 9–11

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KØBENHAVN
BIANCO LUNOS BOGTRYKKERI A/S
1968

GRØNLANDS GEOLOGISKE UNDERSØGELSE

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Contributions to the mineralogy of Ilimaussaq nos 9-11

- 9 The mineralogy of naujakasite O. V. PETERSEN
- 10 Ilimaussite, a new rare earth-niobium-barium silicate from Ilimaussaq, South
Greenland E. I. SEMENOV, M. E. KAZAKOVA and V. J. BUKIN
- 11 On the mineralogy of pyrochlore from the Ilimaussaq alkaline intrusion, South
Greenland E. I. SEMENOV, H. SØRENSEN and Z. T. KATAJEVA

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GRØNLANDS GEOLOGISKE UNDERSØGELSE

THE MINERALOGY OF NAUJAKASITE

CONTRIBUTION TO
THE MINERALOGY OF ILÍMAUSSAQ, No. 9

BY

OLE V. PETERSEN

WITH CHEMICAL ANALYSIS

by I. Sørensen

WITH 3 FIGURES AND 3 TABLES IN THE TEXT
AND 2 PLATES

KØBENHAVN

C. A. REITZELS FORLAG

BIANCO LUNOS BOGTRYKKERI A/S

1967

Abstract

Naujakasite occurs as platy rhomb-shaped, silvery white crystals in the lujavrite at two localities Taperssuatsiait and Kvanefjeld, in the Ilimaussaq alkaline intrusion, South Greenland. Occasionally naujakasite constitutes about three quarters of the rock.

The crystals are monoclinic with three distinct cleavages; $\{001\}$ mica-like, $\{010\}$ and $\{\bar{4}01\}$ distinct but somewhat irregular. $(001):(\bar{4}01) = 93^\circ$.

The density is 2.262 g/cm³, the hardness 2¹/₂-3.

Naujakasite is biaxial negative, with $2V_\alpha$ between 52° and 71° , $n_\alpha = 1.537$, $n_\beta = 1.551 - 1.549$, $n_\gamma = 1.556$, $2V_\alpha$ calculated = $62^\circ - 75^\circ$. $\perp(001): \gamma = +45^\circ$.

X-ray diffraction yields: $a_0 = 15.039 \text{ \AA}$, $b_0 = 7.991 \text{ \AA}$, $c_0 = 10.487 \text{ \AA}$ and $\beta = 113.67^\circ$. Vol. 1154 \AA^3 . Space groups C 2/m, Cm or C2.

The formula $(\text{Na}, \text{K})_6 (\text{Fe}^{\text{II}}, \text{Mn}, \text{Ca}) (\text{Al}, \text{Fe}^{\text{III}})_4 \text{Si}_6\text{O}_{26} \cdot \text{H}_2\text{O}$, $Z = 2$ is proposed for the naujakasite although there are some differences between observed and theoretical values.

CONTENTS

	Page
Abstract	2
Introduction	5
Occurrence	6
Mineralogy	7
General description	7
Optical properties	7
X-ray diffraction	9
Chemistry	11
Alteration	14
Acknowledgements	16
References	17
Plates	18

INTRODUCTION

Naujakasite was first described by BØGGILD (1933) from a loose boulder, collected by FLINK in 1897 at Naujakasik on the south coast of the fjord Tunugdliarfik.

BØGGILD (1933) described the mineral as monoclinic, with pseudo-hexagonal habit. It was found to be biaxial negative with the axial angle near 90° , one optic axis being nearly normal to the mica-like basal cleavage. Only the refractive index $n_\beta = 1.537$ was given. The Formula H_2O , 3 $(\text{Na}_2\text{Fe})\text{O}$, 2 Al_2O_3 , 8 SiO_2 was proposed. BØGGILD (1933) himself stated that there were rather large differences between the chemical analysis and the theoretical values. In order to overcome these discrepancies he proposed to disregard the iron, which he considered to be due to the inseparable inclusions of arfvedsonite, and presented the formula $\text{HNa}_3\text{Al}_2\text{Si}_4\text{O}_{13}$. The X-ray data were give as follows: $a_0 = 15.06 \text{ \AA}$, $b_0 = 7.98 \text{ \AA}$, $c_0 \simeq 19.5 \text{ \AA}$, with β near 90° .

Naujakasite was first found in outcrop in the summer of 1955 in the bay Tuperssuatsiait on the south coast of Tunugdliarfik, and in the summer of 1956 it was also found on Kvanefjeld (DANØ and SØRENSEN, 1959). In the summer of 1964 the author had the opportunity of visiting both localities, and collected many fresh specimens.

OCCURRENCE

Naujakasite is found in naujakasite lujavrite, a name given to an arfvedsonite lujavrite containing naujakasite.

Naujakasite is sometimes so abundant that three quarters of the rock consists of this mineral. The easily recognizable eu- and subhedral, silvery white plates of naujakasite are larger than the average grain-size of the rock. Most of the remaining part of the rock is arfvedsonite. The arfvedsonite occurs for the most part as masses of subhedral grains between the crystals of naujakasite, but it also occurs as small euhedral prisms enclosed in the naujakasite. Brownish grains of steenstrupine are enclosed in the naujakasite and also occur as subhedral grains in the arfvedsonite groundmass, together with subhedral grains of analcite.

MINERALOGY

General description

The naujakasite occurs in platy crystals, the diameter of which ranges from 1 to 5 mm, the thickness being less than 1 mm. The plates are rhomb-shaped.

There is one direction of mica-like cleavage, parallel to the platy development. In the plates traces of numerous irregular cracks, and two mutually perpendicular directions of cleavage are seen. The two mutually perpendicular cleavage directions are diagonal in the rhombs. The cleavage plates are brittle and cannot be bent without being broken.

The cleavage faces are silvery white, with a strongly pearly, to a certain degree metallic, lustre. When seen from other directions the crystals appear grey.

The hardness, measured on the cleavage plane, is: $2\frac{1}{2}$ -3.

The density as determined by suspending the mineral in heavy liquids is 2.622 ± 0.005 g/cm³.

No crystals were found, which could be measured with the goniometer.

Optical properties

In powder preparations (150 to 200 mesh) the mineral is seen to be colourless.

The fact that the interference figure is always the same, an uncentred biaxial optic-axis figure in an optically negative crystal, confirms that one of the three cleavages is mica-like. One of the remaining two mutually perpendicular directions of cleavage is seen to be perpendicular to the direction of the mica-like cleavage, the other being only nearly so. The extinction directions are parallel to the two mutually perpendicular directions of cleavage.

Thin sections of the crystals show that the naujakasite contains numerous unorientated inclusions of well developed prismatic crystals of arfvedsonite. Brownish, subhedral grains of steenstrupine are also found enclosed. Sections cut obliquely to the {001} cleavage show very thin layers of analcite and/or air, along the {001} cleavage planes. The refractive indices of the material which constitutes the layers were determined

by measuring the angle of total reflection by means of the universal stage, as proposed by Dr. H. MICHEELSEN. The analcite is considered to be an alteration product. It is believed that total reflection at these layers of analcite and/or air are the main reason for the pearly to metallic lustre.

Further alteration causes the appearance of the "fields of faintly translucent, brownish substance", mentioned by BØGGILD (1933). The brown colour is due to pigmentation, probably by iron oxides. Because of its low specific gravity this brownish substance could be separated by means of heavy liquids. X-ray diffraction showed it to be analcite.

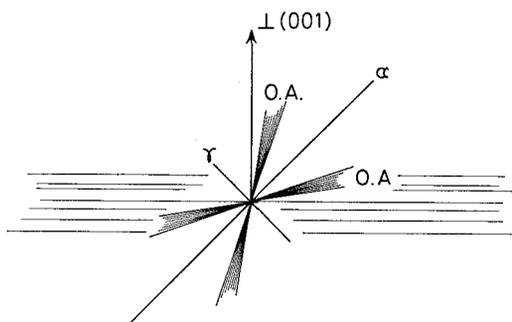


Fig. 1. The orientation of the optical indicatrix in naujakasite, as seen along the b axis. The axial plane is $\perp b$. The horizontal lines show the mica-like $\{001\}$ cleavage.

When examined by means of the universal stage the mineral is seen to be monoclinic, with the axial plane parallel to (010) . The axial angle $2V_\alpha$ varies from $52^\circ \pm 1^\circ$ to $71^\circ \pm 1^\circ$. The cleavage directions are: $\{001\}$ mica-like, $\{\bar{4}01\}$ (in the setting of GOSSNER and KRAUSS (1933) given as $\{100\}$) and $\{010\}$ distinct but somewhat irregular.

Naujakasite shows a distinct axial dispersion.

The orientation of the indicatrix is shown in fig. 1. $\perp(001)$: $\gamma = +45^\circ \pm 1^\circ$.

Occasionally naujakasite may be twinned. The twin plane which is also the composition plane is (001) .

The principal indices of refraction were determined by means of the universal stage, using the λ - T variation method with optical glass as internal standard, MICHEELSEN (1957). The principal indices of refraction for $\lambda = 589$ nm are:

$$\begin{aligned} n_\alpha &= 1.537 \pm 0.001 - 1.537 \pm 0.001 \\ n_\beta &= 1.551 \pm 0.001 - 1.549 \pm 0.001 \\ n_\gamma &= 1.556 \pm 0.001 - 1.556 \pm 0.001 \\ n_\gamma - n_\alpha &= 0.019 \pm 0.001 - 0.019 \pm 0.001 \end{aligned}$$

$$2V_\alpha \text{ calculated} = 62^\circ \pm 5^\circ - 75^\circ \pm 5^\circ.$$

X-ray diffraction

X-ray diffraction data given in the literature are:

Cell parameters.

GOSSNER and KRAUSS (1933)	NEUMANN and BERGSTØL (ASTM Powder Diffraction File, no. 14-461).
a_0 = 15.06 Å	15.09 Å
b_0 = 7.98 Å	8.00 Å
c_0 (for $\beta = 90^\circ$)... = 19.16 Å	
c_0 (for $\beta \simeq 90^\circ$)... =	19.15 Å
c_0 (for $\beta = 100^\circ$).. = 19.8 Å	

The orientation of the unit cell in relation to the axial plane, $b_0 \parallel \beta$, has only been determined by GOSSNER and KRAUSS (1933). An indexed powder diagram has only been given by NEUMANN and BERGSTØL (ASTM Powder Diffraction File).

Precession photographs were taken in order to confirm the orientation of the unit cell and to make a provisional determination of β .

Plate 1, fig. 1 shows the $h0l$ level, fig. 2 the $h1l$ level, and plate 2, fig. 1, $h0l$ level (black spots) and $h1l$ level (white spots) of the reciprocal lattice planes parallel to the optical axial plane. In correspondence with the choice of the direction of the mica-like cleavage as $\{001\}$, the reciprocal vector perpendicular to this cleavage was taken as c^* . In order to follow GOSSNER and KRAUSS (1933) the direction which make an angle with c^* about 87° was taken as a^* .

The $hk0$ level precession photograph shown in plate 2, fig. 2 was taken after turning the crystal 90° around a^* .

The precession photographs verify the orientation of the optical indicatrix in relation to the unit cell stated by GOSSNER and KRAUSS (1933). However, parameters comparable to those obtained by GOSSNER and KRAUSS (1933), (with $\beta = 93^\circ$) can be chosen only if the c_0 axis is doubled. A much smaller unit cell is therefore proposed (plate 2, fig. 1).

For the final determination of the unit cell parameters, GUINIER ($\text{CuK}\alpha$) photographs were taken by E. K. ANDERSEN (Mineralogical Geological Institute of the University of Copenhagen). Si was used as internal standard.

In order to minimize the differences between $\sin^2\theta$ observed and $\sin^2\theta$ calculated the method of least squares approximation was used. These calculations were carried out on the GIER computer made available to the Copenhagen Observatory by the Carlsberg Foundation. A program (REFBASE-1) made by E. S. LEONARSEN was used.

Table 1 presents $\sin^2\theta$ measured, d_{hkl} and hkl for both unit cells.

Table 1. $\text{Sin}^2\theta$, d_{hkl} and hkl , of *Naujakasite*. GUINIER exposure, $\text{Cu}_{K\alpha}$ radiation, $\lambda = 1.5418 \text{ \AA}$.

Int	Setting of GOSSNER and KRAUSS (with double e_0)			New setting
	$\text{sin}^2\theta$ measured	d_{hkl}	hkl	hkl
5	0.01174	7.11	-2 0 2	-2 0 1
5	0.01254	6.88	2 0 2	2 0 0
2	0.02249	5.13	1 1 5	1 1 1
4	0.02391	4.98	-2 0 6	-2 0 2
5	0.03310	4.23	-3 1 1	-3 1 1
10	0.03719	3.99	0 2 0	0 2 0
5	0.03743	3.98	3 1 3	3 1 0
3	0.04161	3.78	-3 1 5	-3 1 2
3	0.04211	3.75	4 0 0	-4 0 1
6	0.04358	3.69	0 2 4	0 2 1
5	0.04537	3.62	1 1 9	1 1 2
7	0.04695	3.56	-4 0 4	-4 0 2
4	0.04890	3.48	-2 0 10	-2 0 3
6	0.05008	3.44	4 0 4	4 0 0
3	0.05275	3.35	2 0 10	2 0 2
5	0.06098	3.12	-2 2 6	-2 2 2
6B	0.06343	3.06	2 2 6	2 2 1
3	0.07103	2.89	4 0 8	4 0 1
6	0.07603	2.79	5 1 1	-5 1 1
4	0.07732	2.77	-5 1 3	-5 1 2
2	0.07920	2.74	4 2 0	-4 2 1
2	0.08124	2.70	1 1 13	1 1 3
5	0.08434	2.65	-4 2 4	-4 2 2
1	0.08600	2.63	-2 2 10	-2 2 3
6	0.08733	2.61	4 2 4	4 2 0
1	0.08974	2.57	-1 3 3	-1 3 1
3	0.09217	2.54	2 0 14	2 0 3
5	0.09552	2.49	-4 0 12	-4 0 4
1	0.10106	2.42	-1 1 15	-1 1 4
1	0.10760	2.35	-3 3 1	-3 3 1
7	0.11611	2.26	-3 3 5	-3 3 2
5	0.11996	2.22	1 3 9	1 3 2
1	0.13488	2.10	6 2 2	-6 2 1
5	0.14884	1.997	0 4 0	0 4 0
2	0.15194	1.976	-5 3 3	-5 3 2
4	0.16847	1.877	8 0 0	-8 0 2

Three lines are omitted, 5.61 (1), 3.43 (2) and 2.92 (1); they correspond to the three strongest lines in analcite, 5.61 (80), 3.43 (100) and 2.925 (80).

Table 2 shows the determined unit cell parameters for both settings.

Transformation matrix of axis and face indices is: new to old: 100/
010/104.

Table 2. *Determined unit cell parameters.*

Setting of GOSSNER and KRAUSS. (with double c_0).	New setting
$a_0 = 15.039 \pm 0.002 \text{ \AA}$ $b_0 = 7.991 \pm 0.001 \text{ \AA}$ $c_0 = 38.461 \pm 0.007 \text{ \AA}$ $\beta = 92.69 \pm 0.02$	$a_0 = 15.039 \pm 0.002 \text{ \AA}$ $b_0 = 7.991 \pm 0.001 \text{ \AA}$ $c_0 = 10.487 \pm 0.002 \text{ \AA}$ $\beta = 113.67 \pm 0.02$
Vol. = 4617 \AA^3	Vol. = 1154 \AA^3
Space groups C 2/c or Cc	Space groups C 2/m, Cm or C2

It should be noted that the main objection, already mentioned by GOSSNER and KRAUSS (1933), the a_0 and b_0 axes, relating naujakasite with the micas persists in the new setting.

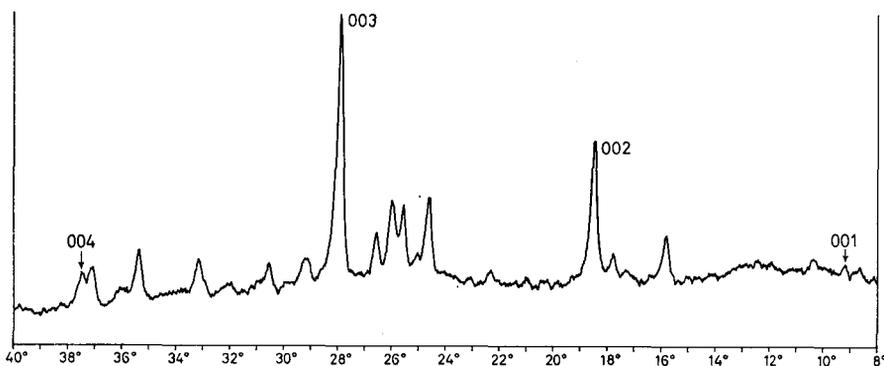


Fig. 2. The X-ray diffractometer diagram of an orientated preparation of naujakasite.

In fig. 2 is shown the X-ray diffractometer diagram of an orientated preparation of naujakasite (made by sedimentation of a suspension of particles less than 2μ on an object glass).

Chemistry

In order to get the purest material possible for chemical analysis arfvedsonite, analcite and steenstrupine were separated from the naujakasite using a FRANTZ isodynamic magnet and heavy liquids, respectively,

The hand picked naujakasite crystals were ground down to 200 to 250 mesh and separated in four fractions on the magnet. Fraction no. 1

consists of nearly pure arfvedsonite, fraction no. 2 of naujakasite still containing arfvedsonite crystals and analcite, fraction no. 3 of naujakasite almost entirely free of arfvedsonite and analcite, and fraction no. 4 of analcite and naujakasite almost entirely free of arfvedsonite, but with minor amounts of steenstrupine enclosed in the naujakasite. From fraction no. 3 the purest obtainable naujakasite was isolated by means of heavy liquids, utilizing the fact, that the crystal fragments containing the analcite are lighter than the pure naujakasite.

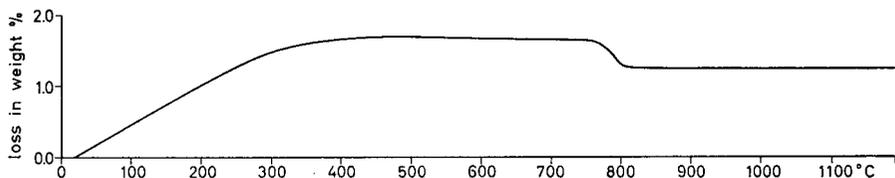


Fig. 3. Thermogravimetric curve of naujakasite.

Part of the difficulty in obtaining a reasonable formula for naujakasite is probably due to insufficient separation. The material cannot be ground down to such a grain-size that the impurities will not be found in the grains. Volumetric estimations on the final obtained fraction shows that arfvedsonite constitutes not more than a few percent. The volume percent of analcite is more difficult to estimate because of the great number of very thin layers which can only be seen oblique to the mica-like basal cleavage; not more than 10 percent and almost certainly *much* less seems a fair estimate.

Due to the small amount yielded by the separations, the determinations of the weight percent of H_2O and FeO (see table 3) had to be carried out by thermogravimetric analysis prior to the chemical analysis. The results of the thermogravimetric analysis are shown on fig. 3. The low temperature release ($< 500\text{ C}^\circ$) of all the water indicates that the water is present as H_2O molecules in the mineral. The smooth curve shows that all the water is released continuously, i.e. the water is all present in the same way in the mineral.

The results of the chemical analysis are presented in table 3 and that published by BØGGILD (1933) is given for comparison. In addition an analysis by E. I. SEMENOV (pers. comm.) is presented.

The main differences between the analyses of CHR. DETHLEFSEN and I. SØRENSEN are the higher Na_2O and lower H_2O content in the new analysis.

As expected beforehand a material of this kind will not give any simple formula. The content of the unit cell has been calculated. The number of oxygen atoms per unit cell was found to be 51.23. The nearest numbers divisible by two, as demanded by the space groups, are 52 or

Table 3. *Chemical Analyses of Naujakasite.*

	Tuperssuatsiait					Naujakasik	
	Weight percent	Weight percent *	Molecular ratio	Content of the unit cell Z_E		Weight percent	Weight percent
				$Z_0 = 51.23$	$Z_0 = 54$		
SiO ₂	51.28	50.65	0.843 ~8	15.32	16.15	50.95	46.00
TiO ₂	0.03	0.03	0.000			not anal.	
Al ₂ O ₃	20.90	20.64	0.202 } ~2	7.34	7.74	20.63	19.15
Fe ₂ O ₃	7.23	2.18	0.014 } ~2	0.51	0.54	2.76	8.29
FeO		4.46	0.062 } ~1	1.13	1.19	5.25	not anal.
MnO	1.11	1.09	0.015 } ~1	0.27	0.29	0.57	0.80
CaO	0.2-0.4	0.2-0.4	0.004-0.008 } ~1	0.11**	0.12	0.55	0.00
MgO						0.10	
Na ₂ O	18.60	18.37	0.296 } ~3	10.76	11.34	14.51	16.78
K ₂ O	0.50	0.49	0.005 } ~3	0.18	0.19	0.80	1.00
P ₂ O ₅	0.07	0.07	0.001	0.04	0.04	not anal.	
H ₂ O		1.71	0.095 ~1	3.45	3.64		
H ₂ O (100°)						1.02	
H ₂ O (ign.)						2.60	2.87
Total	99.92- 100.12					99.74	
Analyst	I. SØRENSEN					Chr. DETH- LEFSEN In O. B. BØG- GILD (1933)	E. I. SEME- NOV personal comm.

* The weight percent arrived at by incorporating the results of the thermogravimetical analysis.

** Average.

50. Recalculation of the number of metal atoms, using the above number of oxygen atoms, yields values which in reasonable combinations are not whole numbers.

BØGGILD (1933) proposed to disregard the iron which he considered to be due to the inclusions of arfvedsonite, and arrived at the formula $\text{HNa}_3\text{Al}_2\text{Si}_4\text{O}_{13}$. Calculations using the idealized formula of arfvedsonite, $\text{Na}_3\text{Fe}_4^{\text{II}}(\text{Fe}^{\text{III}}, \text{Al})(\text{OH}|\text{Si}_4\text{O}_{11})_2$ show that 4.46 weight percent FeO corresponds to about 15 weight percent (about 12 volume percent) arfvedsonite; it will be seen therefore, that the iron cannot be neglected. It should be noticed also that the ratio FeO/Fe₂O₃ is not that of the idealized formula of arfvedsonite.

Calculations of the number of Si, Si+Al and Si+Al+Fe^{III} atoms, accepting that less than 10 weight percent of analcite may be mixed with the analysed material, easily show that no whole or nearly whole number divisible by two can be obtained. Further calculations, accepting that

additional arfvedsonite, up to 10 weight percent, may be mixed with the analysed material lead to the same result. The modifications in the number of atoms in fact are small.

Error in the H_2O determination cannot explain the discrepancies either, since $Z_{H_2O} = 4.00$, which would correspond to a water content of 1.99 (only 1.71 was found) did not yield a whole number of Si, Si + Al and Si + Al + Fe^{III} atoms divisible by two.

Error in the determination of the density attributable to the thin layers of air and/or analcite along the basal cleavage may explain part of the discrepancies ($\Sigma Z_0 = 54.00$ demands, density = 2.661 g/cm³).

The formula, $(Na, K)_6 (Fe^{II}, Mn, Ca) (Al, Fe^{III})_4 Si_8 O_{26} \cdot H_2O$; $Z = 2$, corresponding to $\Sigma Z_0 = 54.00$ is tentatively proposed. Expressed in terms of the nearest whole numbers, the calculated numbers of different atoms 12, 2, 8, 16, 52 and water molecules 2 are compatible with the space groups $C_{2/m}$, C_2 and C_m , but rather large deviations from whole numbers are still found. The symmetry of the H_2O molecule compared with the symmetry of the twofold position of the space group $C_{2/m}$ makes it seem probable that C_2 and C_m should be preferred. In this connection it should be mentioned that no pyroelectric effect could be detected.

Until a detailed structural analysis of the mineral has been carried out the relation of naujakasite to other minerals cannot be determined.

In addition to the main constituents, traces of Y, La, Ce, Cr, Ti, Ga, Zr, Sn, Pb, U and Th were determined with the X-ray fluorescence.

Alteration

Naujakasite alters easily through several stages to analcite. In the early stages a vaguely delimited group of alteration products are formed which have fairly constant properties inconsistent with any known mineral. In the following provisional description the name *hydro-naujakasite* will, for the sake of convenience, be applied to the whole group.

The habit, the dimensions and the cleavage pattern of the crystals are the same as that of naujakasite.

A powder preparation (150 to 200 mesh) shows the most characteristic and most easily observable difference between naujakasite and hydro-naujakasite. The direction perpendicular to the mica-like cleavage, which in naujakasite was indicated by an optic-axis figure in a biaxial negative crystal, is now indicated by an acute bisectrix figure in a biaxial positive crystal. When examined by means of the universal stage the mineral is seen to be monoclinic, with the obtuse bisectrix parallel to

b. $2V_\alpha = 100^\circ - 120^\circ \perp (001)$: $\gamma = +$ or $- 2-3^\circ$. The principal indices of refraction for $\lambda = 589$ nm.

$$n_\alpha = 1.528 \pm 0.002$$

$$n_\beta = 1.529 \pm 0.002$$

$$n_\gamma = 1.531 \pm 0.002$$

GUINIER diagrams of hydro-naujakasite consist of two separate sets of lines. (1) Sharp, weak lines corresponding to spacings which match the spacings observed in naujakasite. (2) Broad, weak lines the corresponding spacings of which are different from the spacings in naujakasite. The number of lines of the first set varies from specimen to specimen. The fact that the first set of lines may represent either remnants of naujakasite preserved in the hydro-naujakasite, or lattice spacings in the hydro-naujakasite inherited from the naujakasite and the apparently random variation from specimen to specimen of the position and the intensity of the second set of lines, are the reasons that the unit cell parameters have not been determined, and that no diagram of hydro-naujakasite is presented. Precession photographs perpendicular to the $\{001\}$ cleavage and perpendicular to the optical axial plane of hydro-naujakasite show the a^*-c^* plane of naujakasite. This indicates that the optical axial plane of the hydro-naujakasite is perpendicular to the optical axial plane of the naujakasite.

Chemically the alteration seems to be nothing but a hydration.

A detailed description of the hydro-naujakasite will be given in a later publication.

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Plate 1

Fig. 1. Precession photograph showing the $h0l$ level of the reciprocal lattice.

Fig. 2. Precession photograph showing the $h1l$ level of the reciprocal lattice.

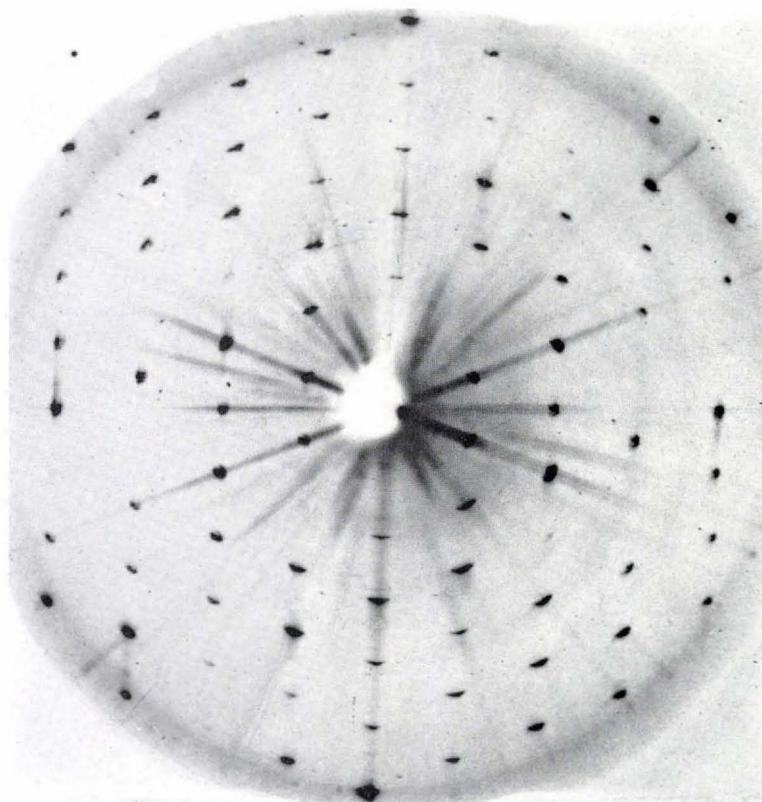


Fig. 1

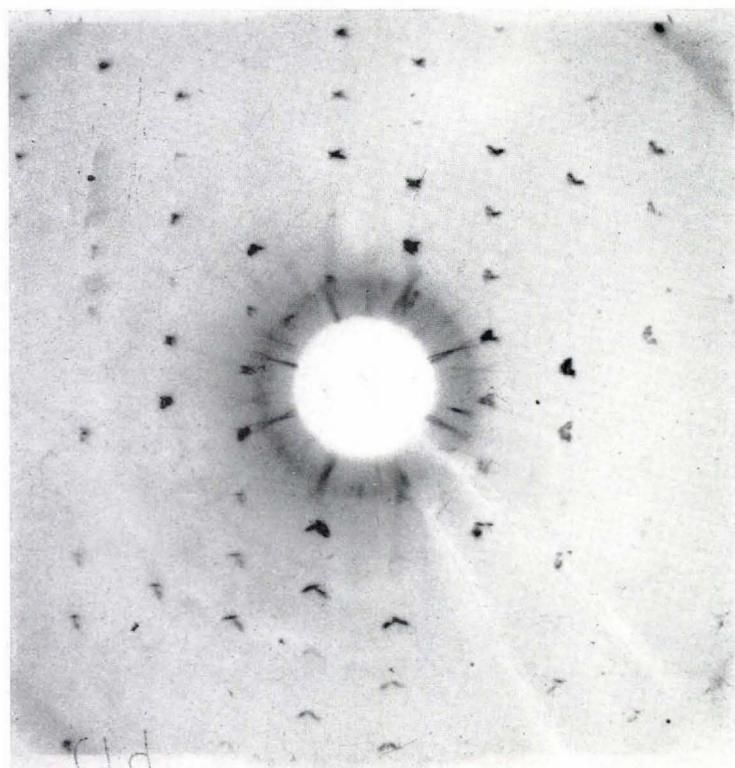


Fig. 2

Plate 2

Fig. 1. Photo montage showing the $h0l$ level (black spots) and the $h1l$ level (white spots). Dashed lines show a^*-c^* -axis in the setting of Gossner and Krauss (1933). The full line shows the a^* -axis and Dashed line the c^* axis in the new setting.

Fig. 2. Precession photograph showing the $hk0$ level of the reciprocal lattice.

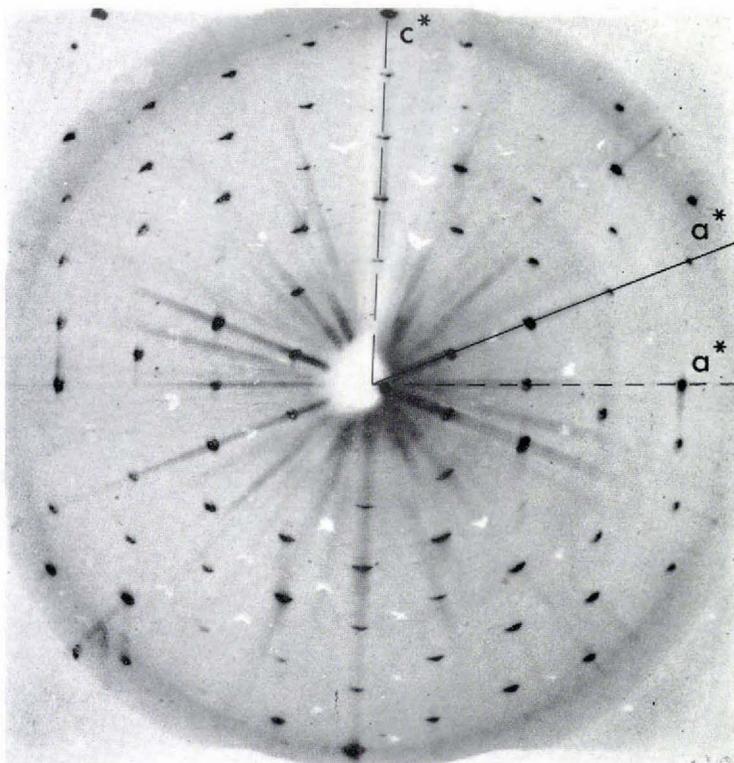


Fig. 1

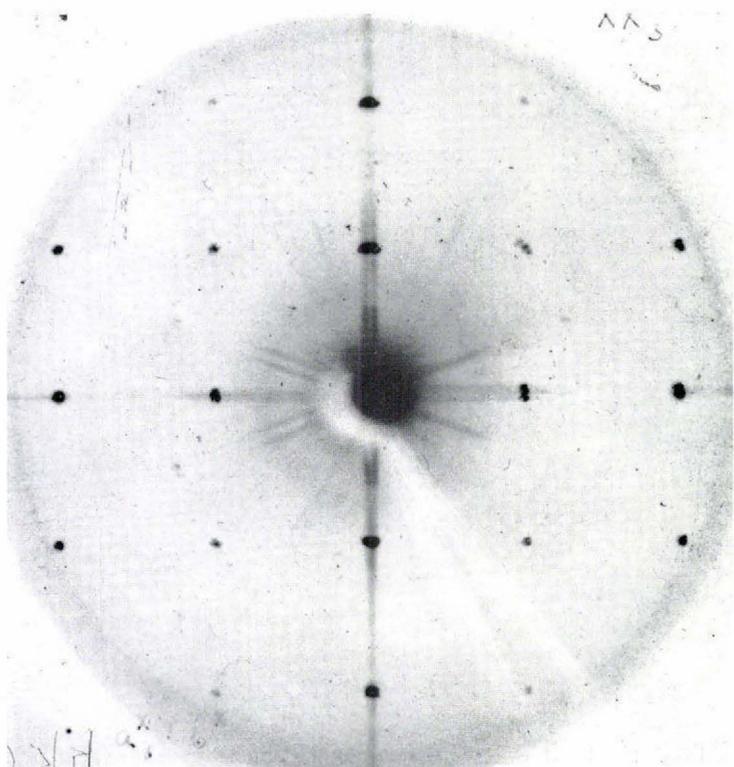


Fig. 2