

ART. XXV.—*The Minerals of Litchfield, Maine*; by F. W. CLARKE.

IN Kennebec County, Maine, along and near the boundary between the towns of Litchfield and West Gardiner, are scattered many boulders of an *elæolite* rock. For many years these have yielded to collectors of minerals superb specimens of blue sodalite, yellow cancrinite and zircon; but although the parent ledge appears at several points, it seems nowhere to have been opened. In addition to the minerals already mentioned, the boulders contain albite, lepidomelane, a black mineral resembling columbite, a flesh-colored mineral which has been called indiscriminately *elæolite* or cancrinite, and a massive alteration product known to local collectors under the provisional name of "white sodalite." Although specimens from the locality are widely distributed in cabinets, some of the minerals seem to have been but partially described; and I have therefore thought it worth while to study them somewhat closely. The supposed columbite I have not examined, for want of material; the zircon I have omitted, since it has been sufficiently studied by Gibbs;\* but the sodalite and cancrinite, although they had been well analyzed by Whitney.† I have included in my investigation, for reasons which will appear below.

## ELÆOLITE.

This species occurs abundantly in Litchfield and West Gardiner in characteristic, dark gray, cleavable masses of strong greasy luster. Since it is the typical mineral of its group, and as I can find no published analysis of it from this locality, the following results may have value as a matter of record.

H <sub>2</sub> O	.....	·86
SiO <sub>2</sub>	.....	43·74
Al <sub>2</sub> O <sub>3</sub>	.....	34·48
CaO	.....	<i>trace</i>
MgO	.....	<i>trace</i>
K <sub>2</sub> O	.....	4·55
Na <sub>2</sub> O	.....	16·62
		<hr/>
		100·25

The specimen analyzed contained minute inclusions of black mica, but not enough of them to notably affect its composition. The analysis agrees fairly well with the published analyses of *elæolite* from other places.

\* Poggend. *Annalen*, lxxi, 559.† Poggend. *Annalen*, lxx, 431.

## CANCRINITE.

This mineral is one of the most abundant and characteristic at the locality, and varies considerably in appearance. Two analyses of it were made by Whitney, one of the yellow variety, the other of a greenish modification. I have myself seen nothing to answer to the latter description, but have selected three typical samples for investigation. They may be briefly described and indicated as follows:

- A. Bright orange yellow, with strong luster and cleavage, transparent in thin fragments.
- B. Dirty pale yellow, less lustrous, highly cleavable, also transparent in thin fragments.
- C. Bright yellow, granular. The commonest variety.

For ease of comparison I have tabulated the analyses side by side with Whitney's; indicating his yellow cancrinite by "D," and his greenish variety by "E." The carbonic acid determinations were made for me by Mr. R. B. Riggs, who used the Gooch tubulated crucible, and collected the gas evolved directly in a potash bulb.

	A.	B.	C.	D.	E.
SiO <sub>2</sub> .....	36.29	35.83	37.22	37.42	37.20
Al <sub>2</sub> O <sub>3</sub> .....	30.12	29.45	28.32	27.70	27.59
Mn <sub>2</sub> O <sub>3</sub> .....	trace	trace	trace	} .86	.27
Fe <sub>2</sub> O <sub>3</sub> .....	"	"	"		
CaO .....	4.27	5.12	4.40	3.91	5.26
Na <sub>2</sub> O .....	19.56	19.33	19.43	20.98	20.46
K <sub>2</sub> O .....	.18	.09	.18	.67	.55
MgO .....	----	----	.07	----	----
H <sub>2</sub> O .....	2.98	3.79	3.86	2.82	3.28
CO <sub>2</sub> .....	6.96	6.50	6.22	5.95	5.92
	<hr/> 100.36	<hr/> 100.11	<hr/> 99.70	<hr/> 100.31	<hr/> 100.53

It will at once be observed that cancrinite "A," which, from its appearance, was presumably the purest type of the mineral, is the highest of all in carbonic acid and lowest in water. It is also the highest in soda and alumina. Whitney's two analyses show more potash than mine, but in other respects run fairly near "C," which, as I have said, represents the commonest, and probably the least pure variety. But in order to understand the variations better, we must consider the flesh-colored mineral referred to in my introductory paragraph, which, as I have said, has been called indiscriminately *elæolite* or *cancrinite*, according to the fancy of the collector. It sometimes occurs in specimens of considerable size, is lustrous and cleavable, and to the eye appears perfectly homogeneous. An analysis gave the following results; the carbonic acid, as in the other cases, being determined by Mr. Riggs.

SiO <sub>2</sub> .....	38·93
Al <sub>2</sub> O <sub>3</sub> .....	32·52
CaO .....	2·47
Na <sub>2</sub> O .....	17·02
K <sub>2</sub> O .....	3·23
H <sub>2</sub> O .....	2·83
CO <sub>2</sub> .....	2·95

99·95

These figures plainly indicate that the mineral is a mixture of elæolite and cancrinite, but do not show whether the mixture is mechanical, or due to isomorphism. To determine this point, Mr. J. S. Diller kindly undertook a microscopic examination of the material, comparing it in thin sections with the elæolite and cancrinite "B," from the specimens of which portions were previously analyzed. He found the mineral to be a merely mechanical commingling of the two species, in nearly equal proportions, and later he succeeded in separating them by means of Sonstadt's solution. This fact, considered together with the apparent homogeneity of the material, renders it probable that the variations in composition of the cancrinite are due to small admixtures of elæolite; and that Whitney's specimens were rather more so contaminated than mine. Still, the entire series of cancrinite analyses are fairly concordant, and confirmatory of each other. In discussing the formula of the mineral, however, analysis "A" will be given preference.

## SODALITE.

On account of its beauty, and its intense blue color, this mineral, as it occurs at Litchfield, is a favorite among collectors. It is now somewhat scarce, at least in large or compact specimens, and it ought to be carefully searched for in place. It often occurs intermingled with cancrinite, forming beautifully mottled masses, and also is associated intimately with the white, massive alteration-product to be described later. The following analysis was made, partly for comparison with Whitney's, and partly to aid in the study of the accompanying white mineral.

	Clarke.	Whitney.	Whitney.
SiO <sub>2</sub> .....	37·33	37·30	37·63
Al <sub>2</sub> O <sub>3</sub> .....	31·87	32·88	30·93
Fe <sub>2</sub> O <sub>3</sub> .....	—	}	1·08
Na <sub>2</sub> O .....	24·56		25·48
K <sub>2</sub> O .....	·10	·59	<i>undet.</i>
Cl .....	6·83	6·97	<i>undet.</i>
H <sub>2</sub> O .....	1·07	—	—
	101·76	101·60	
Deduct O=Cl .....	1·54		
	100·22		

In my analysis iron was not looked for, because the ignited alumina, which should have contained it if present, was perfectly white. Otherwise the analyses agree tolerably well.

#### HYDRONEPHELITE, A NEW SPECIES.

Intimately associated with the sodalite is the white alteration product mentioned in the last paragraph. So close is the association, in fact, and so similar in occurrence are the two minerals, that the latter has been called white sodalite by the local collectors. Like the sodalite it is found in seams, and yields specimens as much as two centimeters in thickness; it is white, lusterless, and has the fracture of sodalite; and probably it originated from the alteration of the latter. Two specimens of it were analyzed, which were received from two different collectors, with the following results:

	A.	B.
H <sub>2</sub> O .....	13·12	13·30
SiO <sub>2</sub> .....	38·90	39·24
Al <sub>2</sub> O <sub>3</sub> .....	33·98	33·16
CaO .....	·05	<i>trace</i>
Na <sub>2</sub> O .....	13·21	13·07
K <sub>2</sub> O .....	1·01	·88
Cl .....	<i>trace</i>	----
	<hr/> 100·27	<hr/> 99·65

The alumina carried a trace of iron, and a doubtful trace of manganese was also indicated. Hardness, 4·5. Fusible easily to a white enamel. Soluble in hydrochloric acid, and gelatinizing upon evaporation. Fracture irregular, resembling that of the sodalite. In general, the mineral may be said to have the appearance of a slightly altered feldspar, minus the distinct cleavage.

These analyses left little doubt in my mind that I had a new mineral to deal with, and one belonging to the zeolite family. Such minerals are well-known derivatives of the nephelite group, and thomsonite and natrolite have especially been often noted. In composition the new product differs distinctly from natrolite, but agrees in ratios approximately with thomsonite; forming, as far as chemical evidence alone goes, the soda end of a series passing through rauite, up to ozarkite; the last named mineral being the nearest towards the lime end of the series. A comparison of the analyses of these elæolite derivatives is worth making, on account of its suggestiveness. The ozarkite was analyzed by Smith and Brush, the rauite, from Brevig, by Paykull.\*

\* Ber. der Deutsch. Chem. Gesell., vii, 1334.

	Ozarkite.	Rauite.	Hydronephelite.
H <sub>2</sub> O-----	13·80	11·71	13·12
SiO <sub>2</sub> -----	36·85	39·21	38·90
Al <sub>2</sub> O <sub>3</sub> -----	29·42	31·79	33·98
Fe <sub>2</sub> O <sub>3</sub> -----	1·55	·57	----
CaO-----	13·95	5·07	·05
Na <sub>2</sub> O-----	3·91	11·55	13·21
K <sub>2</sub> O-----	----	----	1·01
	<hr/> 99·48	<hr/> 99·90	<hr/> 100·27

Inasmuch, however, as massive minerals, and especially those which are produced by processes of alteration, are always subject to doubt, I requested Mr. Diller to assist me with a microscopic examination of the new substance. He very kindly acceded to my request, and I subjoin an abstract of his results.

"A section was carefully prepared so as to show both the sodalite and the white, lusterless mineral associated with it, in such a way as to reveal their relations. The extremely irregular line of contact between the sodalite and its secondary products is well defined in transmitted light, but is even more distinct between crossed nicols from the fact that the sodalite, being isotropic, remains dark in all positions, while the other minerals are more or less brilliantly colored. The secondary products, which have clearly resulted from the zeolitization of the sodalite, are two in number. One of them forms very much the larger portion, probably nearly 90 per cent of their total amount, and the other is imbedded in the first in the form of distinct grains. Under the microscope in transmitted light the predominating mineral, which is doubtless a zeolite as shown by your analyses, is more or less deeply clouded like decomposed feldspar. Between crossed nicols it breaks up into flaky grains which vary considerably in the intensity of their color. Some remain dark, others range through light and medium tints of red and yellow, according to the position of the section. The isotropic grains in converging light are proved to be distinctly uniaxial and positive, and the anisotropic ones as far as can be determined exhibit parallel extinction. It is evident therefore that the zeolite must be either quadratic or hexagonal in the system of its crystallization. Some of the grains show an indistinct striation approximately parallel to the vertical axis, but a distinct cleavage could not be discerned. In basal sections three sets of fractures could be rarely made out with sufficient distinctness to suggest that the mineral is probably hexagonal. The mode of its occurrence indicates clearly that it has resulted from the zeolitization of the sodalite; a phenomenon which has been observed in many rocks.

"The small grains of the other secondary mineral are so intermingled with the uniaxial zeolite as to indicate that both are derived from the sodalite. They are easily distinguished from the zeolite in which they are imbedded. In transmitted light they are perfectly clear and transparent, with so high an index of refraction as to appear to rise above the surrounding mass. The grains are entirely without crystallographic boundaries, but are traversed by distinct cleavage lines. Between crossed nicols they are much more brilliantly colored than the associated zeolite, and if the section is rotated they become dark when the cleavage lines make a prominent angle ( $15^{\circ}$ – $33^{\circ}$ ) with the principal sections of the prisms. The mineral is certainly biaxial, and in all probability belongs to one of the two inclined systems of crystallization, but its definite determination is not practicable under the circumstances."

In view of the presence of an impurity in the new zeolite, Mr. Diller suggested a re-analysis of it, to be made on carefully purified material. The purification, by means of Sonstadt's solution, he kindly undertook, determining at the same time the specific gravity of the mineral. The crude material gave him a sp. gr. of 2.263, while the zeolite was a little lighter and the imbedded grains a little heavier. After purification the coarsely powdered zeolite was carefully picked over under the microscope until Mr. Diller felt confident that the sum of all impurities could not exceed one per cent. The mineral, then dried at  $100^{\circ}$ , gave me the following analytical results:

$\text{H}_2\text{O}$ .....	12.98
$\text{SiO}_2$ .....	38.99
$\text{Al}_2\text{O}_3$ .....	33.62
$\text{CaO}$ .....	.07
$\text{Na}_2\text{O}$ .....	13.07
$\text{K}_2\text{O}$ .....	1.12

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99.85

These figures confirm the previous analyses, and show that the impurity which vitiated them must have been small in amount and similar in composition to the new zeolite. The latter, I think, may be considered as fairly well established; and its formula may be written  $\text{Al}_3(\text{SiO}_4)_3\text{Na}_2\text{H} \cdot 3\text{H}_2\text{O}$ ; which requires, water, 13.76; soda, 13.54; alumina, 33.41, and silica, 39.29. This composition, and the manifest relations of the mineral to nephelite, the parent member of the group, naturally suggest for it the name *hydronephelite*, which seems to be both appropriate and descriptive. Chemically, as I have already observed, the species approximates to a soda thomsonite; but optically it appears to be quite different. This fact suggests the desirability of a careful microscopic re-examination of all

the other massive zeolitic alterations of *elæolite* which have been on analytical grounds referred to the *thomsonite* series. *Hydronephelite*, indeed, is directly derived from *sodalite*, but the latter itself probably originated from *elæolite*; so that the new species may quite properly be considered along with the other zeolites which were previously mentioned. The fact that it contains more potassium than the *sodalite*, is noteworthy, and calls for an explanation which I am unfortunately not prepared to offer.

*Albite and Lepidomelane.*

The *albite* of Litchfield, which appears to be associated with other undetermined feldspars, is mostly in obscure masses. Occasionally a fragment is found with a translucent cleavage surface one or two centimeters broad. Such a specimen was partially analyzed, giving  $H_2O$  0.52,  $SiO_2$  66.39,  $Al_2O_3$  19.69,  $K_2O$  0.99,  $Na_2O$  10.17. These figures serve only for complete identification of the species.

The *lepidomelane* exists abundantly in the *elæolite* rock, but mostly in small black scales. Sometimes tolerably large plates of it are found, black and brilliant, decidedly brittle, and apparently affected by alteration. An analysis gave the following results. The iron determinations were made by Mr. Riggs.

$H_2O$ .....	4.62
F .....	none
$TiO_2$ .....	"
$SiO_2$ .....	32.09
$Al_2O_3$ .....	18.52
$Fe_2O_3$ .....	19.49
$FeO$ .....	14.10
$MnO$ .....	1.42
$MgO$ .....	1.01
$K_2O$ .....	8.12
$Na_2O$ .....	1.55
	<hr/>
	100.92

This analysis is noteworthy on account of the extremely low percentage of silica, which is approached, so far as I can ascertain, only in an analysis by Rammelsberg of a black mica from Brevig. The ratio between silicon and oxygen is nearly 1:5, which agrees with no known formula. My results make it extremely probable that the mica is a mixture and that it has undergone an alteration tending toward the ultimate development of some chloritic species. Still it deserves, as also do the feldspars of the locality, a more thorough examination.

In attempting to discuss the formulæ of cancrinite, sodalite, and hydronephelite, certain points should be carefully borne in mind. First, the three species must be considered, not independently, but relatively to each other; for all the evidence indicates for them a common origin. That origin is from the first member of the group, elæolite or nephelite; the empirical formula for which has been finally fixed by Rauff.\* In partially rational form it may be written  $\text{Na}_6\text{Al}_6(\text{SiO}_4)_7(\text{SiO}_3)_2$ ; ignoring the small replacement of sodium by potassium which has been shown by synthetic investigations to be non-essential. Not only does the mode of occurrence and association of the minerals point to community of origin, but the same conclusion is emphasized by the experiments of Lemberg† upon the artificial alteration of silicates. When elæolite from Fredriksvårn was digested 180 hours with a solution of sodium carbonate, a partial transformation into a *soda* cancrinite was effected; while a digestion of six months with a caustic soda solution containing sodium chloride gave a product identical in composition with sodalite. Many such experiments were tried by Lemberg, yielding a large class of similar results. His method of procedure probably did not give absolutely pure or definite compounds, and yet his researches furnish evidence of great value in discussing the chemical structure of many minerals.

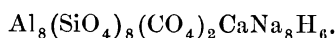
If we compare the published analyses of cancrinite from different localities, we shall find that they vary in two ways. First, there are variations which are probably due to small admixtures of elæolite, such as I have shown to occur at Litchfield; and secondly, the ratio between the lime and the carbonic acid ranges between rather wide limits. In the cancrinite from Miask, the two are about equivalent; while the Litchfield mineral contains only half enough calcium to saturate the carbonic acid. The lime and soda, however, vary reciprocally; so that when one is high, the other is low; and, furthermore, the experiment quoted from Lemberg goes to show that a cancrinite may exist containing no lime whatever. If this conclusion be correct, then the carbonic acid of the mineral must be represented as linked with aluminum; a supposition which finds some justification in the existence of the rare species dawsonite. The function of water in cancrinite remains doubtful; if it be regarded as water of crystallization, the formula of the residue becomes less easy to write intelligibly; but if it forms a part of the atomic structure, it is almost necessary to represent the carbonic acid as orthocarbonic, in the group  $\text{CO}_4$ . This mode of consideration, as will appear later, leads to a

\* Zeitschr. für Kryst., ii, 445.

† Zeitschr. der Deutsch. Geol. Ges., xxxv, 557, 1883.



simple general formula for cancrinite, covering all variations in composition except such as are due to impurity, and correlating the mineral with the allied species sodalite and nosean. For the Litchfield mineral the following special formula may be written, giving the theoretical composition in the column below:



	Found.	Calculated.
$\text{SiO}_2$ .....	35.83 to 37.22	35.9
$\text{Al}_2\text{O}_3$ .....	28.32 to 30.12	30.6
$\text{Na}_2\text{O}$ .....	19.33 to 19.56	18.6
$\text{CaO}$ .....	4.27 to 5.12	4.2
$\text{CO}_2$ .....	6.22 to 6.96	6.6
$\text{H}_2\text{O}$ .....	2.98 to 3.86	4.1
		<hr/> 100.0

In this case the water as found is slightly lower, and the soda slightly higher, than the calculated values; which is probably ascribable to the mutual replaceability of sodium and hydrogen.

The formula commonly accepted for sodalite, and the one which is certainly the simplest, is that deduced by Bamberger\* from his analysis of the mineral from Tiahuanuco. Written empirically, this formula is  $\text{Na}_4\text{Al}_4(\text{SiO}_4)_4\text{Cl}$ , which requires considerably less chlorine than has ordinarily been found in the species. In Bamberger's analysis, as finally corrected, he obtained 5.54 per cent, as against nearly seven per cent in Whitney's determinations. The difference he ascribes to silica in the chloride of silver as weighed by other analysts; and yet in my own estimation every care was taken to eliminate such impurity, and my results confirm the older figures. Still, both figures have theoretical interest, as will be seen further on; and I am inclined to believe that the Bolivian mineral was more nearly typical than that from Litchfield. To the latter we may assign the empirical formula  $\text{Na}_9\text{Al}_7(\text{SiO}_4)_7\text{Cl}_2$ , which is directly derivable from the formula for nephelite, and which agrees quite sharply with the analyses.

We now have three empirical formulæ ready for comparison side by side, as follows:

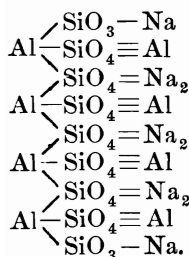
Nephelite .....	$\text{Al}_8(\text{SiO}_4)_7(\text{SiO}_3)_2\text{Na}_8$
Cancrinite (Litchfield) .....	$\text{Al}_8(\text{SiO}_4)_8(\text{CO}_4)_2\text{CaNa}_8\text{H}_6$
Sodalite " .....	$\text{Al}_7(\text{SiO}_4)_7\text{Cl}_2\text{Na}_9$

These may easily be put into structural form by an application of the principle suggested in a former paper,† that orthosilicates containing aluminum are to be represented as substi-

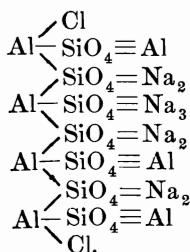
\* Zeit. für Kryst., v, 581.

† Clarke and Diller, this Journal for May, 1885.

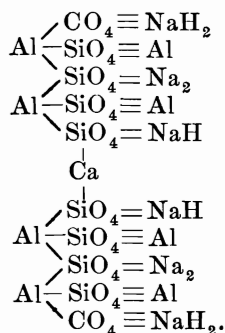
tution derivatives of the normal salt  $\text{Al}_4(\text{SiO}_4)_3$ . The latter contains the fundamental nucleus  $\text{Al}(\text{SiO}_4)_3$ , which appears to be capable of a sort of polymerization, and which forms the basis of the subjoined symbols.



*Nephelite.*



*Sodalite.*

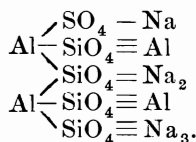


*Cancrinite.*

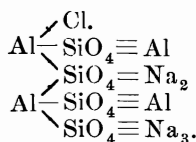
Now although these formulæ fit the analyses, and express a structural similarity of type, two of them are capable of a further generalization. Remembering the reciprocal variations between soda and lime in different cancrinites, and the fact that a soda cancrinite is quite possible, we may write the following general formula for that species;  $\text{Al}_4(\text{SiO}_4)_4 \text{CO}_4 \text{Na}_5 \text{H}_3$ ; which requires

$\text{SiO}_2$ .....	35.8	
$\text{Al}_2\text{O}_3$ .....	30.4	
$\text{Na}_2\text{O}$ .....	23.1	Partly replaceable by $\text{CaO}$ .
$\text{CO}_2$ .....	6.6	
$\text{H}_2\text{O}$ .....	4.1	

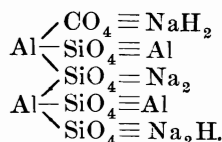
Comparing this with Bamberger's sodalite formula, and with the generally accepted formula for nosean, we have this remarkable series of structural expressions.



*Nosean.*

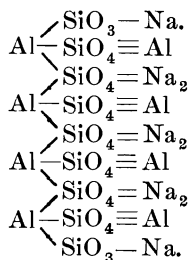
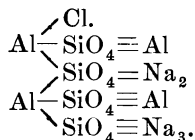
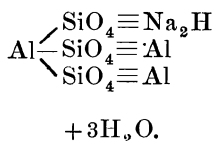


*Sodalite.*



*Cancrinite.*

The formula of hauynite of course reduces to the same type, and so also, probably, does that of microsommite. Hydro-nephelite has a still simpler formula, which, however, includes three molecules of water of crystallization. It may be advantageously compared in series with its parent minerals, in which case we have the following set of structures:

*Nephelite.**Sodalite.**Hydronephelite.*

These formulæ express with decided clearness, the natural order of transition from one species to another. The alteration of a mineral necessarily involves the passage from a less stable to a more stable condition; and in this instance we observe precisely that state of affairs. From a quite complex and therefore easily disturbed molecule, through an intermediate, simpler compound, we pass to one which is simplest of all, and hence, presumably the most stable. I do not deny that such formulæ are subject to criticism, and that possibly the advance of knowledge may brush them to one side; and yet I feel justified in claiming that they have some real value in the coördination of observed facts, and that, through their singular suggestiveness, they assist in the prosecution of research.

Laboratory U. S. Geological Survey, Washington, 1886.