Just why an occasional log in a coal bed should undergo calcification instead of becoming converted into coal is difficult to understand. Apparently the change took place after the log had become buried in the upper part of the peat deposit which formed the coal bed and the latter had been buried under a load of sediment sufficiently great to exert the pressure necessary to flatten the log. Waters charged with calcium and magnesium carbonates, or possibly sulfates, then permeated the cells and filled them with dolomitic calcite and to a large extent replaced the walls themselves, leaving just sufficient carbonaceous matter to emphasize the outlines of the original cellular structure.   The pyrite was brought in in solution at a somewhat later stage and has replaced portions of the calcite. The above described occurrence, in many respects, bears a close resemblance to the calcareous concretions or "coal balls" so rich in well-preserved plant remains, which are occasionally found associated with coal seams, and the process of formation was undoubtedly very similar.

Dr. O. E. Jennings, of the Carnegie Museum, is at present working on the identification of the specimen. A preliminary examination reveals conifer-like structures indicating that the log may represent one of the cordaites.

A NEW THEORY OF THE COMPOSITION
OF THE ZEOLITES.   PART II

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2.   THE COMPOSITION OF NATROLITE, MESOLITE, SCOLECITE, GISMONDITE AND LAUMONTITE

The so-called "natrolite group" of zeolites furnishes one of the most striking examples of the apparent validity of valence-control of isomorphous replacement. It is well known that this "group" consists of the following minerals, the composition being given as heretofore commonly accepted:

Natrolite $Na_2Al_2Si_3O_{10}.2H_2O$ $a:b:c = 0.9786:1:0.3536$

Mesolite $\begin{cases} Na_2Al_2Si_3O_{10}.2H_2O \\ 2CaAl_2Si_3O_{10}.3H_2O \end{cases}$ $a:b:c = 0.9747:1:0.3122$

Scolecite $CaAl_2Si_3O_{10}.3H_2O$ $a:b:c = 0.9763:1:0.3433$

At first glance the case seems very convincing. However, so far as similarity of crystal form is concerned, the "group" might well include also several other zeolites, as follows, the composition being expressed as heretofore commonly accepted:

Edingtonite $BaAl_2Si_3O_{10}.3H_2O$	$a:b:c=0.9872:1:0.3367$	
Thomsonite $(Na_2,Ca) Al_2Si_2O_8.2.5H_2O$	$a:b:c=0.9932:1:0.3355$	
Gismondite $CaAl_2Si_4O_{12}.4H_2O$	$a:b:c=0.9566:1:0.3188$	
Laumontite $CaAl_2Si_4O_{12}.4H_2O$	$a:b:c=1.082\ :1:0.5896$	

In fact, Tschermak in his recent comprehensive treatise[1] on the zeolites groups all these zeolites together and calls attention to certain properties they have in common, especially the property of gelatinizing with acids.

Furthermore, in the "natrolite group" the similarity in crystal form is far from complete, since natrolite is orthorhombic, mesolite is triclinic, and scolecite is monoclinic.

It is now generally agreed that two substances are not isomorphous unless they have similar composition as well as similar crystal form. In a general way there can be no doubt that the minerals of the "natrolite group" (and also edingtonite) are similar in chemical composition; but are they sufficiently similar to form a truly isomorphous group? The final test of complete isomorphism is the ability to form mix-crystals or crystal solutions. It is therefore important to learn to what extent natrolite, mesolite and scolecite form mix-crystals. For this purpose a careful selection of all available analyses of these minerals has been made on the same basis as in previous cases, namely, excluding (1) old analyses, (2) those on impure material, (3) those obviously inaccurate, (4) those judged inaccurate because the ratio $Al_2O_3:CaO+Na_2O$ is not almost exactly 1:1. The "superior" analyses selected in this way are the following:

Natrolite: Doelter's[2] Nos. 24, 26, 29, 35, and 37.
     67. T. L. Walker: *U. Toronto Studies, Geol. Ser.*, **14,** 62, (1922).
     68. A. H. Phillips: *Am. Jour. Sci.*, **XLII,** 472, (1916).
     69. G. Tschermak: *Sitz. Akad. Wiss. Wien*, **CXXVI,** 544, (1917).
     70. F. N. Ashcroft: *Mineral. Mag.*, **XVII,** 305, (1916).
Mesolite: Doelter's Nos. 10, 11, 25, 28, and 37.
     38, 39. T. L. Walker: *U. Toronto Studies, Geol. Ser.*, **14,** 57, (1922).
     40. U. Panichi: *Rend. Accad. Lincei, Rome*, **XX,** 421, 518, (1911); *Zeit. Kryst.*, **LIV,** 108, (1915).

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[1] *Sitz. Akad. Wiss. Wien.*, **CXXVI,** 541, (1917) and **CXXVII,** 177, (1918).
[2] Hdb. Mineralchemie, **II,** 2, 311, (1917).

41. "Pseudomesolite," A. N. Winchell: *Am. Geol.*, **XXVI**, 277, (1900).
Scolecite: Doelter's Nos. 9, 16, 24, 25, 27, 28, 29, 39 and 40.

41. T. L. Walker: *U. Toronto Studies, Geol. Ser.*, **14**, 67, (1922).

42. E. G. Radley: *Trans. Roy. Soc. Edinb.*, **LI**, 1, (1915).

In order to study these analyses (and those of laumontite and gismondite) by the same method applied to those of thomsonite it is desirable to adopt more siliceous molecules for the corners of the square, as follows:

	Lime	Soda
More siliceous	$Ca_2Al_4Si_{12}O_{32}$	$Na_4Al_4Si_{12}O_{32}$
Less siliceous	$Ca_4Al_8Si_8O_{32}$	$Na_8Al_8Si_8O_{32}$

It is evident that the lower half of this square is the same as the upper half of the square used for thomsonite.

As before, various points in the square represent various proportions of the four molecules, and these may be read directly from the diagram. Also various points represent varying ratios so far as Al_2O_3:SiO_2 and CaO:Na_2O are concerned, but a constant unit ratio between Al_2O_3 and $CaO+Na_2O$.

The method of finding the point which represents an analysis is much the same as in the preceding case, but if:

$x = Al_2O_3$ molecules in the less siliceous molecules

$y = Al_2O_3$ molecules in the more siliceous molecules, then

$x+y =$ total number of Al_2O_3 molecules in the analysis, and

$2x+6y =$ total number of SiO_2 molecules in the analysis.

Since there are two Al_2O_3 molecules in the less siliceous silicate molecules and one Al_2O_3 molecule in the more siliceous silicate molecules:

$x/2 \div x/2+y =$ percentage of less siliceous molecules.

All of the "superior" analyses of natrolite, mesolite and scolecite are plotted on Fig. 2. A mere glance at the resulting distribution of points is sufficient to show that three minerals are represented, which have little, if any, ability to form mix-crystals, since the points representing the analyses fall in three small groups rather than forming a series. The only analysis suggesting any important amount of solid solution is mesolite 37 which was made by Glinka[3] to show changes from fresh massive state to weathered condition. It is highly probable that the fresh material was not pure mesolite, but an intergrowth of mesolite and natrolite, which is common in this condition according to Böggild.[4] Aside from scolecite 9 which Zambonini[5] reported to be a scolecite closely associated with

[3] *Zeit. Kryst.*, **XLVI**, 287, (1909).

[4] *Kgl. Danske Vid. Sels. Meddel.*, **IV**, 35, (1922).

[5] *Mineral. Vesuv.* 296, (1910).

thomsonite, the scolecite and natrolite analyses which show any notable variation from the theoretically pure minerals, vary, not toward each other or toward mesolite, but toward thomsonite. As no one supposes that thomsonite can form mix-crystals with scolecite or with natrolite, these analyses probably represent other cases of intergrown zeolites. However, the main point is that the diagram shows no evidence cf an isomorphous series from natrolite through mesolite to scolecite, but may be taken as strong evidence that no such series exists.
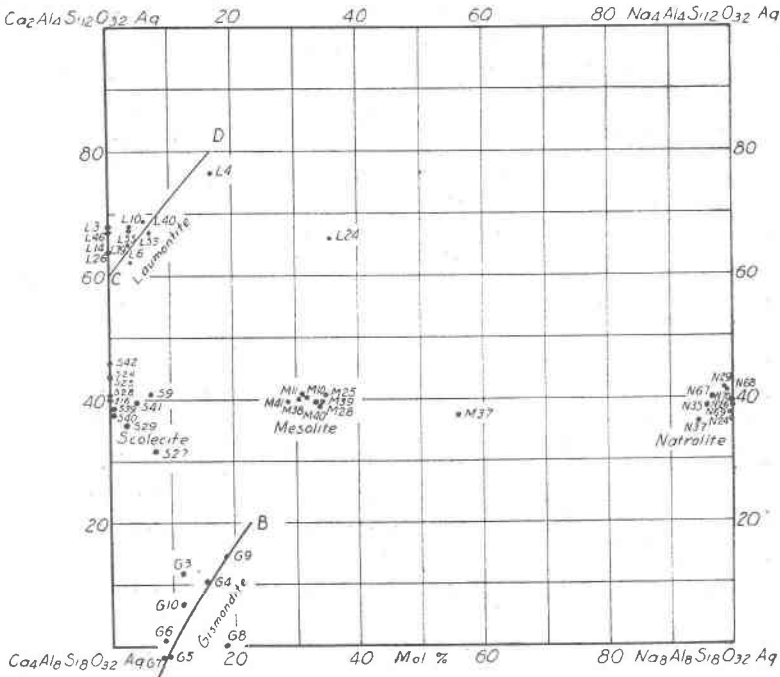

Figure 2

The Composition of Natrolite and Related Zeolites

The composition of natrolite is definite; in the diagram it is 40 per cent $Na_2O.Al_2O_3.6SiO_2 + 60$ per cent $2Na_2O.2A_2O_3.4SiO_2$, which is easily reduced to the ordinary formula, $Na_2Al_2Si_3O_{10}$. The composition of scolecite is also definite; from the diagram it is 40 per cent $CaO.Al_2O_3.6SiO_2 + 60$ per cent $2CaO.2Al_2O_3.4SiO_2$, which is equivalent to the usual formula, $CaAl_2Si_3O_{10}$. The composition of mesolite is equally definite; from the diagram it·is

33 per cent $Na_2Al_2Si_3O_{10}+67$ per cent $CaAl_2Si_3O_{10}$, which is equivalent to $Na_2Ca_2Al_6Si_9O_{30}$. Accepting the ordinary view regarding the water this formula may be written: $Na_2Ca_2Al_6Si_9O_{30}$. $8H_2O$. There is no more evidence that mesolite is composed of one molecule of natrolite with two molecules of scolecite than there is that natrolite, for example, is composed of two $Na_2O.Al_2O_3.6SiO_2$ molecules with three $2Na_2O.2Al_2O_3.4SiO_2$ molecules. Therefore, the formula of mesolite should be written $Na_2Ca_2Al_6Si_9O_{30}.8H_2O$ and not as one natrolite plus two scolecite molecules.

In regard to the other zeolites of a form somewhat similar to that of natrolite, the case of thomsonite has been discussed in a preceding section, and edingtonite is so rare that only one modern analysis has been made and therefore no information is available regarding variations in composition. Gismondite and laumontite remain to be studied.

There are ten analyses of gismondite on record;[6] two of these are very old and give unsatisfactory ratios of $Al_2O_3:CaO+K_2O$; the others are shown[7] on Fig. 2. A glance at their distribution is sufficient to show that gismondite is not of fixed composition, but is a series which varies in composition along the line shown. As in other zeolites which are not of fixed composition, the variation is like that in the feldspars, the number of $Ca+K$ atoms and of $Al+Si$ atoms remaining constant for a given number of oxygen atoms. In this case there are 11 $Ca+K$ atoms for 80 atoms of oxygen.

Accordingly, the composition of gismondite may be expressed as an isomorphous series from $KCa_{10}Al_{21}Si_{19}O_{80}.40H_2O$ at A to $K_4Ca_7Al_{18}Si_{22}O_{80}.36H_2O$ at B.

Thugutt analyzed a zeolite, which he called "*zeagonite*," and found it much richer in potash than ordinary gismondite, although otherwise much the same as the latter. It seems probable that this mineral was merely gismondite after a partial exchange of Ca for 2 K such as may occur under suitable conditions; as shown by Lemberg,[8] Zoch[9] and others, especially since Walker[10] has shown that potassium mercuric iodide solution, used in separating

[6] C. Doelter: Hdb. Mineralch., **II**, 3, 30, (1921).

[7] For gismondite the right hand molecules contain chiefly K_2O rather than Na_2O.

[8] *Zt. Dtsch. Geol. Ges.*, **XXVIII**, 546; **XXXIX,** 582, etc., (1887).

[9] Chemie der Erde, **I**, 219, (1915).

[10] *U. Toronto Studies, Geol. Ser.*, **14,** 48, (1922); *Am. Min.*, **7,** 100, (1922).

minerals, causes such a change in gmelinite and presumably in other zeolites. Thugutt later discovered that the "zeagonite" first analyzed is a mixture of phillipsite and levynite, but considered homogeneous "zeagonite" to be a natural dehydration product of phillipsite. Several writers have discussed the close relationships between "zeagonite," gismondite and phillipsite; the close chemical relationship between gismondite and phillipsite is evident by a comparison of Fig. 2 and the figure for phillipsite.

There are fifty-three analyses of laumontite listed by Doelter[11] and Shannon[12] has given the composition of a sample from Montana, while Walker[13] has published an analysis of the Nova Scotia mineral. However, the number of modern analyses (since 1880) giving a satisfactory $Al_2O_3:CaO+Na_2O$ ratio is only twelve, including Doelter's Nos. 3, 4, 6, 10, 14, 24, 26, 39, 40, 46, and 53, as well as Walker's analysis. These are plotted on Fig. 2. With the single exception of No. 24 (Fersmann's analysis of "leonhardite" from Simferopol, Russia) they show a variation which is chiefly along the line CD, that is, the variation is like that in the feldspars, the $Ca+Na$ atoms and the $Al+Si$ atoms being constant for a given number of oxygen atoms. In fact, there are 7 atoms of $Ca+Na$ for 80 oxygen atoms and the variation can be expressed as from $Ca_7Al_{14}Si_{26}O_{80}.25H_2O$ at C to $Na_2Ca_5Al_{12}Si_{28}O_{80}.25H_2O$ at D, taking approximately the accepted ratio of water. Since the analysis of "leonhardite" just mentioned contains considerable potash the abnormal position of No. 24 may be due to a change in composition of the zeolite while immersed in potassium mercuric iodide, a change which Walker[14] has shown to occur promptly in gmelinite, and one to be expected in other zeolites.

(To be continued)

[11] Hdb. Mineralch., **II**, 3, 37, (1921).

[12] *Am. Min.*, **6**, 6. (1921).

[13] *U. Toronto Studies, Geol. Ser.*, **14**, 54, (1922).

[14] *Loc. cit.* p. 48, and *Am. Min.*, **7**, 100, (1922).