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Studies on the zeolites. Part V. Mesolite.¹

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[Read March 23, 1933.]

MESOLITE was first recognized as a separate species by J. N. Fuchs (1816), being one of the three species into which he subdivided R. J. Haüy's species mesotype (1801). Its individuality has been repeatedly doubted, many workers regarding the mesotype group as an isomorphous series with natrolite and scolecite as the endmembers, and mesolite merely an intermediate isomorphous mixture. The frequent occurrence of intergrowths of mesolite with natrolite or scolecite is largely responsible for this confusion, these intergrowths frequently passing for single minerals, and being adduced as evidence of the occurrence of all intermediate compositions in the supposed isomorphous series. Faroelites, and mixtures containing faroelite, have also been analysed as mesolite from time to time.

Doubts as to the individuality of mesolite have persisted even after the work of R. Görgey (1908, 1909) and H. L. Bowman (1909), but the present work provides strong evidence of the correctness of their conclusions: natrolite, mesolite, and scolecite are three distinct and separate species, isostructural, but not isomorphous, differing in symmetry, and with a limited degree of isomorphous replacement.

¹ Part IV. Ashcroftine (kalithomsonite of S. G. Gordon). Min. Mag., 1933, vol. 23, p. 305.

With such confusion over the status of mesolite itself, it is not remarkable that there has been much confusion over its varieties. *Poonahlite* was described by H. J. Brooke (1831) from Poonah, Bombay, India, and there has been considerable confusion as to its proper place. Many authors have classed it with scolecite, but H. L. Bowman (1909), who deals fully with its history, proved its identity with mesolite. *Harringtonite* of T. Thomson (1836) has been widely regarded as mesolite, but in the author's opinion¹ it was really a compact and intimate mixture of thomsonite and mesolite.

Another of T. Thomson's County Antrim minerals, Antrimolite (1836), has also been assumed to be mesolite. Thomson's original analysis is quite uninterpretable, while one by W. Parry made in R. D. Thomson's laboratory (1846) would appear to be a mesolite, as were two specimens analysed by M. F. Heddle (1857). But an examination of several specimens in the British Museum, obtained between 1837 and 1850, and including some purchased from the dealer (P. Doran) who supplied Thomson, suggests that appearance was the main factor in the classification of this material; antrimolite probably consisted simply of any fibrous zeolite or mixture of zeolites, coating scalenohedra (generally amber-coloured) of calcite, and so attaining somewhat of a stalactitic appearance. Mesolite, scolecite, natrolite, thomsonite, and mixtures of these have all been found as such specimens, and unless Thomson's original material can be re-examined. there is no evidence that any of these has a better right to the name of antrimolite than any other. Verrucite of J. Apjohn (1845), also from Co. Antrim, is shown by his analysis to be simply mesolite.

Pseudomesolite of A. N. Winchell (1900) cannot be united with mesolite if Winchell's results are correct, for it would appear to be monoclinic, fibrous, and elongated perpendicular to the symmetryaxis, with probably the same optic orientation as scolecite, but very low birefringence, optically positive, and a composition nearly identical with that of mesolite. The species certainly calls for further examination by modern methods. The optical properties of O. Luedecke's 'mesolite' from Pflasterkaute, Eisenach (1881), suggest pseudomesolite; but his analysis was probably made on a mixture. E. Rimann² has also examined material, the optical properties of which suggest pseudomesolite; a re-examination of either of these specimens would be of considerable interest.

¹ Part II of this series, Min. Mag., 1932, vol. 23, p. 113.

² E. Rimann, Centr. Min., 1909, p. 767.

Metamesolite of A. Cavinato (1927) is merely dehydrated or nearly dehydrated mesolite, and, as will be shown below, has no claim to species rank.

The material examined under the name of mesolite by E. Hussak¹ differs notably from mesolite in composition, cleavage, and optical properties, and may be a distinct species. It would undoubtedly repay re-examination.

Chemical composition.

Five new analyses of mesolite were made on carefully selected and optically tested material, with the results shown in table I. These specimens were used for all the other work described below.

					1.	2.	3.	4.	5.
	, SiO ₂	•••			46.05	47.13	46.98	46 ·15	46.1
	Al ₂ O ₃	•••			26.84	26.02	26.43	27.04	26.6
ege.	CaO	•••			10.00	10.09	10.06*	9.73	10.6
iti	Na ₂ O				3.82	4.50	4.57	4.64	$5 \cdot 1$
Ser.	K ₂ O	•••		•••	0.79	0.11	0.05	0.06	
Per	H ₂ O	•••	••••		12.30	12.17	11.94	12-36	12-0
ΤŬ	Total				99.80	100.02	100.03	99.98	100.4
$ \begin{array}{c} SiO_2 & \dots & \dots & \dots & 46\cdot05 \\ Al_2O_3 & \dots & \dots & \dots & 26\cdot84 \\ CaO & \dots & \dots & \dots & 10\cdot00 \\ Na_2O & \dots & \dots & \dots & 3\cdot82 \\ K_2O & \dots & \dots & \dots & 12\cdot30 \\ H_2O & \dots & \dots & \dots & 12\cdot30 \\ Total & \dots & \dots & \dots & 12\cdot30 \\ Total & \dots & \dots & \dots & 12\cdot30 \\ Oxygen \% & \dots & \dots & 11\cdot21 \\ Oxygen, atoms per \\ unit cell. & & & \\ Si & \dots & \dots & \dots & 12\cdot30 \\ Al & \dots & \dots & \dots & 12\cdot30 \\ Ca & \dots & \dots & \dots & 10\cdot6 \\ Na & \dots & \dots & 11\cdot6 \\ K & \dots & \dots & \dots & 12\cdot7 \\ \end{array} $	41.40	41.53	41.30	41.3					
	Oxygen unit	i, ato cell.	ms	per }	240.5	241.7	242.6	240-8	241-1
	/ Si				70-9	$72 \cdot 2$	72.8	70-8	71-1
9	Al	•••			49 ·8	47.9	48·3	48 ·8	48-6
Ň.	Ca			•••	16.9	16.9	16.7	16.3	17.4
ten ten	Na .	•••			11.6	13.6	13.7	14.1	15.3
C NE	K.	•••			1.6	0-2	0.1	0.1	
<u>8</u> 8	H ₂ O .	•••			64.5	63.3	61.8	64.5	61.5
Ā	Si + A1		•••		120.7	120.1	$121 \cdot 1$	119.6	119.7
1	Ca+Na	4 K	•••		30.1	30.7	30.5	30.5	32.7
	Sp. gr. Refract	(D ¹⁹)† vive in	· dex	 (B)	$2.258 \\ 1.507$	$2.258 \\ 1.507$	$2.258 \\ 1.5074$	$2 \cdot 256 \\ 1 \cdot 507$	$2 \cdot 257 \\ 1 \cdot 507$
				W27	•				

TABLE I. New chemical analyses of mesolite.

* SrO tested for and found absent.

[†] Corrected to vacuum.

1. B.M. 33868. Bhore Ghaut, Syhadree Mts. (= Siahadri Mts. or Western Ghats), Bombay, India. Dull crystals.

2. B.M. 31746. Iceland. Radiating, coarsely prismatic.

3. B.M. 49969. Syhadree Mts., Bombay, India. Clear large crystals.

4. B.M. 95279. Kvivig (= Qvivig), Strömö, Faeroe Islands. Clear crystals.

5. B.M. 1914,754. Burger's quarry, West Paterson, New Jersey, U.S.A. Long thin needles.

¹ E. Hussak, Bol. Comm. Geog. Geol. Estado São Paulo, 1890, no. 7, p. 244; abstract in Zeits. Kryst. Min., 1892, vol. 21, p. 405. As mentioned above, a number of the older analyses of 'mesolites' were undoubtedly made on impure material mixed with natrolite, scolecite, or faroelite, and these must be rejected in considering the composition of mesolite. There is no satisfactory criterion to ensure their rejection but optical study, and this unfortunately involves rejection of most of the analyses hitherto published. In only seven analyses from the literature is an optical control of the purity of the material mentioned.

It was at first supposed, on the strength of the remarkable similarity between X-ray rotation photographs of natrolite, mesolite, and scolecite about the axis of elongation of the crystals, that the unit cell of mesolite must contain 80 oxygen atoms. It therefore seemed that the correct unit cell formula must be $Na_4Ca_6Al_{16}Si_{24}O_{80}.22H_2O$ with, normally, a small replacement of Ca by 2Na, rather than the formula $Ca_2Na_2Al_6Si_9O_{30}.8H_2O$, proposed by R. Görgey (1908, 1909) and accepted by H. L. Bowman (1909), T. L. Walker and A. L. Parsons (1922), A. N. Winchell (1925), and A. Cavinato (1926, 1927). But it seemed curious that the atomic proportions calculated from the five new analyses and the seven selected analyses from the literature should be in better agreement with Görgey's formula than with the new one.

Further work showed that the true unit cell of mesolite contains 240 oxygen atoms, one of the cell-sides (a) having approximately treble the length of the corresponding cell-side (b) of natrolite. Görgey's formula is therefore correct and represents one-eighth of the unit cell contents, the true unit cell formula being Ca₁₆Na₁₆Al₄₈Si₇₂ O₂₄₀.64H₂O. The atomic cell-contents, calculated from the densities, analyses, and cell-sides are given for the five new analyses in table I, and for the analyses from the literature in table II. There is often a small replacement of Na by K (up to 1.6 atoms per unit cell), and there is commonly a small replacement of 2Na by Ca, rarely the reverse. The Si/Al ratio does not seem to depart notably from 1.5; any small departure is usually by way of excess alumina, but there is no evidence suggestive of $NaSi \rightleftharpoons CaAl$ substitution. The water content is perhaps open to some doubt. The five new analyses agree in suggesting 64H₂O, but many of the analyses from the literature lead to higher figures, 68 or 72H₂O per unit cell.¹ The study of the

¹ The space-group, C_2^3 (face-centred aspect), indicates that the water content must be a multiple of 4 mols. per unit cell, and the very close approximation to the orthorhombic space-group \dot{C}_{29}^{19} suggests that the water content is most probably a multiple of 16 mols. per unit cell. vapour-pressure (see below) tends to confirm the figure $64H_2O$, and it is probable that many of the higher values are due to the use of finely fibrous material, to water absorbed owing to fine grinding, or to deliberate saturation by standing over water. The vapour-pressure work on mesolite and on natrolite¹ indicates that such saturation is not justified with these zeolites, complete hydration only requiring a considerably lower vapour-pressure than that of water.

				~	-	-			
			a.	<i>b</i> .	с.	d.	е.	f.	g.
Si			70.8	71.8	71.5	$72 \cdot 1$	71.6	71.4	71.8
Al	•••		48.9	49.0	48.9	48 ·1	48·2	48.0	50.2
Са	•••		18.8	16.5	16.5	15.5	16.2	16.0	$15 \cdot 1$
Na	•••		12.5	12.7	14.0	15.9	16.0	18.5	11.1
к		•••			0.4	0.2	0.8	—	-
H ₂ O	•••		69.3	69.0	65.7	64.5	64.8	67.8	85.5
Si + A1			119.7	120.8	120.4	$120 \cdot 2$	119.8	119.4	122.0
Ca + Na +	-K		31.3	29.2	30.9	31.6	33.0	34.5	$26 \cdot 2$
Class of a	analysis		Α	В	A	Α	Α	в	С
Sp. gr.			_		2.260	2.26-	2.257	2.272	2 ·18
						2.27			

 TABLE II.
 Analyses of optically-tested mesolite, from the literature.

 Re-calculated in atomic proportions per 240 oxygen atoms.

a. A. Cavinato, 1926. Iceland.

b. A. Cavinato, 1927. Berufjord, Iceland.

c. T. L. Walker and A. L. Parsons (E. W. Todd, analyst), 1922. Cape d'Or, Cumberland Co., Nova Scotia.

d. H. L. Bowman, 1908. Poonah, Bombay, India. Var. poonahlite.

e. T. L. Walker and A. L. Parsons (E. W. Todd, analyst), 1922. Gates mountain, Annapolis Co., Nova Scotia.

f. R. Görgey (E. Ludwig, analyst), 1909. Faeroe Islands.

g. E. E. Schmid, 1871 and 1880. Iceland. Optically examined by O. Luedecke, 1881.

Only those analyses for which optical tests of the purity of the material are mentioned are included in table II (where they are classified as was done for thomsonite), but the majority (38) of the remaining analyses are in good agreement with the new formula, and it seems reasonable to explain the others (10) as analyses of mixtures.²

¹ Part III of this series, Min. Mag., 1932, vol. 23, p. 243.

² In all, 64 analyses were collected from the literature, of which 18 were accounted as first-class, 15 second-class, 22 third-class, and 9 rejected as worthless. Potash was estimated or shown to be absent in only 5 of the first-class, 3 second-class, and 3 third-class analyses. The method of classification, as was explained in Part II of this series, cannot detect admixture with another zeolite. No method yet proposed for the evaluation of analyses would.

M. H. HEY ON

Crystallography, density, and pyroelectricity.

Owing to the confusion of the three members of the mesotype group, and the invariable occurrence of mesolite as supplementary twins, the latter was long regarded as orthorhombic. Optical studies by several workers have shown that mesolite crystals are invariably twinned, but there is a conflict of evidence as to the true symmetry of the mineral, some classing it as anorthic, others as monoclinic. The author's optical studies, and X-ray work by F. A. Bannister, point to monoclinic symmetry.

The habit of mesolite is very simple, long prisms r(301) and $R(\overline{3}01)$ with a pyramid¹ o(311) and $p(\overline{3}11)$ or $O(3\overline{1}1)$ and $P(\overline{3}\overline{1}1)$ being almost invariably the sole forms observed. The orientation used is based on that of G. Cesàro (1909), modified in accordance with the X-ray results, and depends on optical study. In each individual of the twin, that pair of prism-faces which show a blue colour between crossed nicols are taken as r(301) and $r'(\overline{301})$; the neighbour to r(301) across the acute normal-angle of the prisms is taken as $R(\overline{3}01)$, whereby the free end of the crystal, carrying the pyramids, may fall either to the right (positive end of the b-axis), or to the left (negative end of the b-axis), which two kinds of crystal may be termed rightand left-handed respectively. This is in accord with the symmetryclass, digonal polar (C_2) . The crystals are invariably twinned, consisting of right- and left-handed sectors, as revealed optically, of which either may predominate; or the right-handed sectors may predominate at one end, and the left-handed at the other end of the same crystal. There is no external indication of the twin structure revealed by optical work. The crystals are, indeed, pseudo-orthorhombic, and have commonly been set up with the elongation as c-axis, the prism being indexed m(110) and the pyramid o(111); the above orientation is in better accord with the true axial ratio, as shown by X-ray work, and with the symmetry, and will be used here. The old c-axis becomes the new b-axis, and the old b-axis the new *a*-axis. The transformation determinant is 030/001/100.

Mesolite crystals from four of the analysed specimens have been measured, but unfortunately only one specimen yielded crystals with

¹ In a previous paper (Min. Mag., 1933, vol. 23, p. 367), the pyramid was given indices of the type (111), in the belief that the cell-dimensions of mesolite were very nearly the same as those of natrolite; further X-ray work has shown that this supposition was incorrect. The a- and c-axes of the present setting were interchanged in the former setting.

pyramid faces that reflect well. The angle rR(301): ($\overline{3}01$), measured on natural and on cleavage prisms, gave a mean value of 88° 33', the measurements ranging from 88° 28' to 88° 40'. The other results are given in table III. The crystals show a rather irregular development of the pyramid, one face being often much larger than the others. The faces of the pyramid were replaced by an oscillating vicinal combination of two faces in the zone [011], (or [011]), resulting in striations parallel to this zone, as found by R. Görgey (1909).

TABLE III. Goniometric measurements on mesolite (specimen no. 3).

Interzonal angles:	Crystal 1.	Crystal 2.	Crystal 3.	Crystal 4.	Calc.
$[rR]:[op] = [010]:[01\overline{1}]$	70° 26′	70° 28′	70° 29′	70° 23′	70° 26'
[rR]:[o' p'] = [010]:[011]	70° 28′	70° 33′	70° 6′ (?)	70° 15′	70° 26′
Interfacial angles:					
ro(301):(311) (vicinal)*	63° 36' 64° 3' to 64° 26'	63° 56′ to 65° 29′ 63° 57′ to 65° 31′	63° 39' to 64° 16'	64° 2′ to 64° 42′	63° 36′
$R_p(\overline{3}01):(\overline{3}11)$ (vicinal)*	64° 53' to 66° 7' 63° 25' to 64° 6'	64° 7′ to 65° 1′ 63° 42′ to 65° 51′	} _	63° 40′ and 64° 22′	63° 36′

* Each face o(311) or $p(\overline{3}11)$ is in general replaced by a pair of vicinal faces in the zone $op[01\overline{1}]$. The replacement is oscillatory, and on the goniometer the signal-image is spread out into a band, brightest at its two ends; the cited angles are for the ends of this band.

If the zones containing the vicinal pyramids be taken as the true [011] and [011], which seems a priori reasonable, since the interzonal angle [010]: [011] between them and the prism-zone is nearly the same for all recorded measurements,¹ the axial ratio may be calculated from this interzonal angle (70° 26') and the cleavage angle rR (88° 33'), provided the interaxial angle, β , is known. The latter cannot be obtained from the goniometric data, except that it must be very near 90° as the crystals are so markedly pseudo-orthorhombic; the X-ray work indicated a value for β of 90° 0′ ± 20′. Taking this figure, the axial ratio computed from the goniometric data is a:b:c = 8.658: 1: 2.813, for Cesàro's setting, compared with a ratio 8.640: 1: 2.811 from the X-ray measurements. In Dana's setting (c-axis = elongation, a-axis bisects the acute normal-angle of the prism, pyramid = (111)), these ratios become 0.9750: 1: 0.3465 (goniometric) and

¹ See W. Phillips (1823), O. Luedecke (1881), R. Görgey (1909), and A. Cavinato (1926).

0.976:1:0.347 (X-ray). The goniometric value is probably the better.

Five new determinations of the specific gravity of mesolite (see table I) show no appreciable differences; and they are confirmed by the most reliable of the older values. After allowing for the changes in specific gravity and in cell-size due to the potash content (compare natrolite, Part III of this series, loc. cit.), the specific gravity of fully hydrated potash-free mesolite of ideal composition may safely be taken as D^{19}_{4} 2.256, corrected to vacuum.¹

Mesolite has been examined for pyroelectricity by several workers, but some of the earlier work is useless owing to uncertainty as to which member of the mesotype group was really used. Positive results were obtained by J. N. Fuchs (1816), Sir David Brewster (1821), and A. Des Cloizeaux (1862), and negative by P. Riess and G. Rose (1843).

An application of the liquid-air method of testing to several twinned crystals from the specimen B.M. 49969 showed that there is a small but definite pyroelectric effect, the elongation (*b*-axis) being the polar axis. The experiments were carried by Dr. W. A. Wooster of Cambridge, whom we wish to thank for his kind assistance in this matter.

No piezoelectric tests on mesolite have been published, though it is possible that the negative result recorded for scolecite by G. Greenwood and D. Tomboulian² may be due to the use of mesolite, the two minerals being often confused.

Parallel growths of mesolite and natrolite have been recorded from various localities by several workers.³ The author has observed mesolite tipping or in extension of natrolite needles from Neubauerberg, Böhmisch-Leipa (fully described in Part III of this series, loc. cit., p. 254) and precisely similar material (save in the length of the needles) from Neuschloss, Böhmisch-Leipa, and from the Dulce quarry,

¹ The specific gravities cited in Parts II, III, and IV of the series are all compared to water at 4° C., but are not corrected to vacuum. The correction may be made by subtracting 0.001 throughout.

² G. Greenwood and D. Tomboulian, Zeits. Krist., 1932, vol. 81, p. 30. [M.A. 5-131.]

³ Mindelberg, Aussig, Bohemia, G. Cesàro, 1909; Friedrichsthal, Bensen, Böhmisch-Leipa, R. Görgey, 1909; Neubauerberg, Böhmisch-Leipa, A. Scheit, 1911; Kara-dagh, Crimea, Miss O. A. Brinken, 1914; Fichtenberg, Salesl, Bohemia, J. E. Hibsch, 1917; Weilberg, Siebengebirge, Rhine, R. Brauns, 1922. Some of these authors have not recognized the mesolite as such.

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Marienberg, Aussig, Bohemia. He has also observed mesolite intimately intergrown with scolecite, from Co. Antrim (so-called antrimolite), and from Arendal, Norway; with natrolite, from Richmond, Melbourne, Australia, from Killyflugh and other places in Co. Antrim, and from Talisker, Skye; and with thomsonite from Co. Antrim (socalled antrimolite), from Nordtunga, Iceland, and from the Faeroe Islands. Such admixtures have been described from several other localities, in particular by O. B. Bøggild (1922).

In the course of a re-classification of the fibrous zeolites in the British Museum collection mesolite has been recognized on specimens from several localities which appear to be new for this mineral, at least no mention of them could be found in a survey of the literature; the localities are: Arendal, Norway; Monte Somma, Vesuvius, Italy; Mezzavalle, Predazzo, Fleimsthal, Tyrol; Richmond, Melbourne, Australia; Disko Island, Greenland; Eide, Österö, Faeroe Islands; Tjörnevig, Strömö, Faeroe Islands; and Dulce quarry, Marienberg, Aussig, Bohemia.¹

X-ray work.

Apart from a Lauegram published by A. Cavinato (1926) as evidence of monoclinic symmetry, the only X-ray work on mesolite appears to be that of W. H. Taylor, C. A. Meek, and W. W. Jackson (1933), who found that short-exposure rotation photographs of mesolite and natrolite are practically identical, and concluded that the two species are isostructural.

Using very thin needles and giving a fairly long exposure, Mr. F. A. Bannister has been able to find very definite differences between natrolite and mesolite rotation photographs about the axis of elongation. Although there is a very marked similarity between the two, many lines are found to be present in the latter which are absent in the former. At first it appeared that the cell-sizes of the two zeolites were the same, the mesolite lacking the systematic halvings shown by natrolite, but when an attempt was made to index the mesolite photographs, and especially when an attempt was made to index a pair of oscillation photographs about the same axis, over a range of 5° from the positions in which r(301) and $R(\overline{3}01)$ respectively are

¹ S. J. Thugutt (1912) described mesolite from the last locality, but his method of diagnosis was not reliable, and J. E. Hibsch (1915) failed to confirm it. The British Museum material is quite unlike that described by Thugutt, being tiny cappings on natrolite needles.

parallel to the X-ray beam,¹ many diffractions could not be satisfactorily indexed. Rotation photographs of mesolite were therefore taken about the *b*- and *c*-axes, when it became clear that the cell-side *a* has approximately treble the length of the corresponding cell-side (*b*) of natrolite. It was then possible to index the photographs satisfactorily, and also to index oscillation photographs about the *a*- and *c*-axes.

When the photographs had been indexed, it was found that all planes of the type (hkl) with (h+k), (h+l), or (k+l) odd are halved, indicating a face-centred lattice. There are probably no other systematic halvings. Diffractions from several planes of the types (h0l), with (h+l) not a multiple of 4 were observed, e.g. (604), (406), and (10.0.20). All these reflections are at most weak. No reflections from planes of the type (0kl) with (k+l) not a multiple of 4, or of the type (hk0) with (h+k) not a multiple of 4 were observed, but the number of such planes which could be sought for was limited.

Lauegrams show no evidence of departure from orthorhombic symmetry, but the optical data prove that the symmetry must be monoclinic, and in conjunction with the pyroelectric effect and the systematic halvings, point to the space-group C_2^3 . Apart from the optical properties there is no evidence of any departure from the spacegroup C_{2*}^{19} , with b-axis polar.

The cell-sides are $a\ 56.7\pm0.1$, $b\ 6.54\pm0.02$, $c\ 18.44\pm0.04$ Å., and the interaxial angle, β , is 90° 0′ ± 20′. The number of oxygen atoms per unit cell has been calculated from the cell-sides, specific gravity, and chemical data for each of the analysed specimens, with the results included in table I.

When base-exchange products were being examined, it was desirable to be able to distinguish readily between a mesolite and a natrolite. A rotation photograph about the c-axis of mesolite (b of natrolite) would do this easily, but when the rather imperfect crystals of base-exchange products are in question, the setting of this axis vertical is difficult, and it proves simpler to take a rotation photograph about the axis of elongation. On such photographs of mesolite, using Cu-K radiation, if given adequate exposure, using a small crystal, diffractions are observed on the equator at 2

¹ No differences could be detected between these two photographs, made on a carefully selected untwinned fragment. This emphasizes the marked pseudoorthorhombic nature of mesolite.

² For the cylindrical co-ordinates, ξ and ζ , used for rotation photographs, see J. D. Bernal, Proc. Roy. Soc. London, Ser. A, 1926, vol. 113, p. 117. [M.A. **3**-333.]

 ξ 0.39, 0.41, 0.63, and 0.64, due to the planes (804), (14.0.2), (14.0.6), and (20.0.4) or (22.0.4) respectively¹; on the first layer-line at ξ 0.09, 0.16, 0.21, 0.25, 0.29, 0.31, 0.32, 0.37, and 0.39, due to (111), (511), (711), (113), (513), (11.1.1), (713) (13.1.1), and (11.1.3) respectively; and on the second layer-line at ξ 0.18, 0.20, and 0.27, due to (222), (422), and (822). A rotation photograph of natrolite about the elongation (in this case the *c*-, not the *b*-axis) shows no trace of diffractions in these positions, which do not correspond to integral planes of the natrolite lattice. In the base-exchange products, the actual positions of the spots are a little altered, but their positions in relation to the spots common to mesolite and natrolite remain unaltered, and their presence or absence is readily decided, determining the product as a mesolite or natrolite respectively. (See table IX.)

It is hoped to discuss the relation between the structures of natrolite, mesolite, and scolecite as soon as work on the last species, at present in progress, is completed.

Optics.

The optical behaviour of mesolite has been studied by several workers, with rather variable results. G. Cesàro (1909), C. Anderson (1907), and A. Cavinato (1926) found straight extinction on the prismfaces, and on artificial faces in the prism-zone, while most other workers² have found distinctly inclined extinction, the extinctionangles given ranging from 1° to 11° on the prism-faces. Cut sections parallel to the *a* and *c* axes of crystals from one of the analysed specimens have been examined, but no results have been found not consistent with monoclinic symmetry. Certain apparent anomalies observed have been described and explained in a previous paper.³

With the exception of A. Des Cloizeaux (1862) and E. Hussak (1890, loc. cit.), all workers are agreed that at the ordinary temperature the optic axial plane of fully hydrated mesolite is perpendicular to the elongation (b-axis), the positive bisectrix lies in the obtuse normalangle of the prism, and at room-temperature the optic axial angle is very large, positive (at least for the red end of the spectrum), and

² A. Des Cloizeaux, 1862; O. Luedecke, 1881; A. Lacroix, 1897; A. N. Winchell, 1900; H. L. Bowman, 1908; R. Görgey, 1909; J. E. Hibsch, 1917.

³ M. H. Hey, A possible source of error in the determination of symmetry from optical extinction-angles. Min. Mag., 1933, vol. 23, p. 367.

¹ These are in addition to the diffractions given by natrolite in the same region.

very sensitive to change of temperature or of wave-length. The birefringence is very low, so that the needles appear isotropic under the microscope—a useful means of identification.

Plates of mesolite cut perpendicular to the elongation show a division into sectors which do not extinguish simultaneously. The usual appearance is that shown in fig. 1, two small sectors twinned on and bounded by a(100) or c(001) or both (as β is nearly 90° 0', these alternatives cannot be distinguished) being based on an opposite pair of prism-faces of the main crystal. Measurements of the principal extinction-angle (a: c-axis) gave a value of 8.0°, independent of wavelength, and not affected by moderate changes of temperature. The optic axial angle, 2V, was measured on crystals immersed in a liquid of the same refractive index as the crystals, with the results discussed below, a check on the principal extinction-angle being obtained at the same time.

Refractive index determinations were made for a series of wavelengths on a cut prism from specimen no. 3. Only the index β was measured, at a temperature of 18-20° C. (table IV, and fig. 2).

TABLE IV.	Refractive indices (β) of mesolite (specimen no. 3) for
	light of various wave-lengths.

	λ (Å.)			β.	λ (Å.).		β.
7000	•••		•••	1.5037	5461 (Hg green)		1.5092
6708	(Li)	•••		1.5049	5350 (Tl)		1.5098
6563	(H)	•••		1.5056	5200		1.5110
6250	•••			1.5062	5000		1.5119
6000	•••			1.5071	4861 (H)	•••	1.5126
5893	(Na)		•••	1.5074	4550 (Cs)		1.5147
5708	(Hg yell	ow)	•••	1.5081	4358 (Hg violet)		1.5150

Mean probable error ± 0.0010 in the absolute values of β ; in the relative values for any two wave-lengths, ± 0.0002 .

As R. Görgey (1909) showed, the optic axial angle of mesolite is highly sensitive to temperature. He found that at 10° C. the mineral is optically positive for all colours, but with rise of temperature $2V_{\gamma}$ increases and the mineral becomes negative for all colours at about 30° C.; $2V_{\alpha}$ continues to fall rapidly, and at about 35° C. a uniaxial point appears, after which the optic axial plane becomes parallel to a(100), a being the acute bisectrix for all colours, while γ is now parallel to the elongation. The extinction-angle on b(010) is not noticeably altered during these changes. There is no loss of water by the mineral during the changes, which are perfectly reversible on cooling. The author has been able to confirm and supplement Görgey's results, though with some quantitative differences which are probably due to small differences in composition. The cut plates used for these studies were made from crystals from specimen no. 3; one plate was cut approximately perpendicular to the *c*-axis, and one perpendicular to the *a*-axis. The optical behaviour of the latter at about 19° C. has already been described.¹ The former, 0.94 mm. thick, proved



FIG. 1. Optic orientation and twin-structure of mesolite, as seen on a section perpendicular to the *b*-axis (elongation) of a crystal from specimen no. 3 (Syhadree Mts.).

Fig. 2. Variation of the refractive index (β) of mesolite (specimen no. 3) with the wave-length of light.

less suitable for accurate study, since the three portions in twin position, of which it is composed, overlap very considerably, resulting in too low values for the birefringence and for the temperatures of uniaxiality. The use of cut plates with the twinned portions overlapping may in some measure account for Görgey's lower figures for the temperatures of uniaxiality, but it is believed that the principal cause is the different composition of his material, which was much richer in sodium than that employed in the present work. It was noticed (loc. cit.) that the cut plate perpendicular to the *a*-axis showed growthbanding of more and less birefringent portions with different optic

¹ M. H. Hey, Min. Mag., 1933, vol. 23, p. 367. A different setting was used, and the plate is there described as approximately perpendicular to the *c*-axis, the present *a*- and *c*-axes being interchanged.

axial angles. Variations in composition during the growth of the crystal appear to be the only reasonable explanation for this. Unfortunately, time did not permit of a detailed examination of the other analysed specimens, nor were they so suitable for the preparation of cut plates.

Each of these cut plates was examined mounted on the Miers stagegoniometer, on a Dick microscope, and immersed in a small cell containing a mixture of xylene and chlorbenzene adjusted to have about the same refractive index as the crystal. A thermocouple built into the cell was used to determine the temperature, which was varied by dropping in hot liquid from a reservoir, the level in the cell being kept steady by a syphon and constant-level device.

The results of the measurements of the birefringence and optic axial angle of mesolite at various temperatures, made with these plates and apparatus, are shown in figs. 3 and 4 and tables V and VI. Only the plate perpendicular to the *a*-axis proved satisfactory for birefringence measurements, plates perpendicular to the *c*- and *b*-axes showing overlapping of the twin portions. On this plate $\beta - \alpha$ (above 55-60° C. $\gamma - \alpha$) was measured at several temperatures and found to

TABLE V. Birefringence $\beta - \alpha$ ($\gamma - \alpha$ above 55-60° C.) of mesolite (specimen no. 3) for several temperatures. Observed values and values calculated from a smoothed curve. Probable error about ± 0.00010 .

Temp.	•••	$22 \cdot 2^{\circ}$	$37 \cdot 2^{\circ}$	$52 \cdot 3^{\circ}$	67·3° C.
$\beta - \alpha$ obs.		0-00095	0.00102	0.00118	0.00129
$\beta - a$ cale.		0.00095	0.00107	0.00120	0.00131

TABLE VI. Optic axial angles $(2V_{\alpha})$ of mesolite (specimen no. 3) for various temperatures and wave-lengths. Calculated values from the equations deduced for the birefringences. Optic axial plane parallel b(010) except for these values marked *, for these parallel c(001).

<i>t</i> .		λ6708 Å.	6250.	5893.	5461.	5350.	5000 Å.
0° C		108°	107 1 °	107°	106 <u>‡</u> °	106°	105 <mark>‡</mark> °
10		101	100	99	98 1	98	97
20		92	91	90	89 1	89	88
30		82	80	79	77 .	77	76
40		71	68	66	64	63	61
45	•••	63	60	57	54	53	51
50	•••	55	50	46	42	41	38
55		42	35	31	25	23	15
60		20	0	17*	25*	28*	33*
65		29*	36*	40*	44*	45*	49*
70° C		46*	59*	53*	56*	58*	61*
Uniaxial at		62° C.	60°	59°	57.5°	57°	56° C.

increase appreciably with rise of temperature, while being practically independent of the wave-length of the light. The equation $\beta - a = (7 \cdot 7 + 0 \cdot 08t)10^{-4}$, where t is the temperature in °C., expresses satisfactorily the variation in this birefringence.

Next, the birefringences $\gamma - \beta$ and $\gamma - \alpha$ ($\beta - \alpha$ above 55-60° C.) were



FIG. 3.

F1G. 4.

FIG. 3. Variation of the optic axial angle of mesolite (specimen no. 3) with the temperature and with the wave-length of light.

FIG. 4. Variation of the optic axial angle of mesolite (specimen no. 3) with the temperature, for light of wave-lengths λ 6708 and 5000 Å. Measured values and calculated curves.

calculated from the measured optic axial angles and the appropriate values of $\beta - \alpha$. The equations

$$\gamma - a = \{21 \cdot 1 - 0 \cdot 16t + 0 \cdot 82 \times 10^{-3} (\lambda - 5000)\}10^{-4}$$
 and
 $\gamma - \beta = \{13 \cdot 4 - 0 \cdot 24t + 0 \cdot 82 \times 10^{-3} (\lambda - 5000)\}10^{-4}$

express satisfactorily the variations in these birefringences with temperature (t in °C.) and wave-length (λ in Å.).

From the equations the optic axial angles were calculated, for a series of temperatures, for each of the wave-lengths used (table VI). The agreement between the measured and calculated values is satisfactory (compare fig. 4, where the curves for λ 6708 and 5000 Å. are shown; the other curves, not reproduced to avoid confusion, agree equally well), and hence there seems no point in reproducing all the measured values.

Measurements on the plate cut perpendicular to the c-axis gave confirmation of the general behaviour of the optic axes, but owing to the overlapping of the twin portions, the quantitative results were of no use.

Vapour-pressure.

An investigation of the vapour-pressure (pct) surface of mesolite was made with the apparatus and by the methods employed for thomsonite and natrolite and described in Parts II and III of this series (loc. cit.). A series of isohydric curves interpolated from the experimental curves is shown in fig. 5, and isobaric curves computed from them in fig. 6; the latter will be seen to give fairly good agreement with the results of previous workers,¹ plotted in the same figure. Values of the heat of hydration,² Q, and of the integration constant, A, of the equation $\log_{10} p = A - Q/(RT \log_e 10)$, for various degrees of hydration, are plotted in fig. 7, and tabulated in table VII. The agreement between the observed pressures and those calculated from the values of Q and A interpolated from the figures of table VII by means of fig. 7 was throughout at least as good as for natrolite.

TABLE VII. The constants of the vapour-pressure surface of mesolite for a series of water-contents (c); heat of hydration, Q, and the integration constant, A, of the equation $\log_{10} p = A - Q/(RT\log_{e} 10)$.

с.	Q, cals.	<i>A</i> .	<i>c</i> .	Q, cals.	A.
64 ·0	$1{\cdot}05{ imes}10^4$	8.15	34.0	$2{\cdot}60\! imes\!10^4$	10.83
63.4	1.24	8.20	31.0	2.67	10.88
62.5	1.43	8.67	28.0	2.74	10.95
61.6	1.91	10.42	25.0	2.82	11.03
61·Ô	1.97	10.52	22.0	2.90	11.17
58 ·0	2.14	10.69	19.0	2.19	11.34
55.0	2.20	10.75	16.0	3.06	11.51
52.0	2.25	10.76	13.0	3.13	11.68
49·0	2.30	10.77	10.0	3.20	11.85
46 ·0	2.34	10.78	7.0	3.28	12.02
43 ·0	2.39	10.78	4.0	3 <i>·</i> 36*	12.19*
41 ·0	2.45	10.78	0.0	$3.44* \times 104$	12.36*
37.0	$2{\cdot}52\! imes\!10^4$	10.79			

* Extrapolated.

¹ Isobaric curves: F. Zambonini, 1908; A. Cavinato, 1927. Rough isobaric curves: L. Pelacani, 1908; T. L. Walker and A. L. Parsons, 1922.

² Heat of hydration for one gram-molecule of water absorbed from the vapour phase and exclusive of the external work involved. For further explanation and definition of this and other quantities involved, and the symbols used see Parts II and III, loc. cit. There seems no necessity for reproducing all the actual measurements as was done for natrolite; the results tabulated for that zeolite (part III; loc. cit., table V) show the order of accuracy attained, and the vapour-pressure to be expected for any composition and



FIG. 5. Isohydric vapour-pressure curves of mesolite (specimen no. 1, from Bh ore Ghaut).

temperature (or the composition or temperature, the other variables being given) may readily be found by the aid of figs. 5, 6, and 7, and the values of Q and A in table VII.

The vapour-pressure surface found for mesolite leaves no possible doubt as to its interpretation. There are no discontinuities anywhere in the surface or in its curvature. Hence we have throughout a single solid phase. Mesolite on dehydration undergoes no transition, but loses its water continuously, and Cavinato's metamesolite (1927) is not to be regarded as a separate species, but merely as the

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FIG. 6. Isobaric dehydration curves for mesolite calculated from the vapourpressure measurements. Together with dehydration data from the literature.



FIG. 7. The variation, with the water content, of the molecular heat of hydration (Q) of mesolite, and of the integration constant (A) of the equation $\log_{10} p = A - Q/RT(\log_4 10)$.

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end-member of the isomorphous series of variously hydrated mesolites.

This is confirmed by a study of the effect of dehydration on the optical properties of mesolite (table VIII and fig. 8). As with thomsonite and natrolite, the refractive indices fall rapidly in the initial stages of dehydration, to rise again in the later stages. Unfortunately, partially dehydrated mesolite absorbs water-vapour from the air too rapidly to allow of accurate X-ray measurements of



FIG. 8. The variation of the mean refractive index of mesolite (specimen no. 1) with the water content.

its cell-size being made, so it cannot be decided how far the rise in refractive indices in the later stages of dehydration is due to latticeshrinkage, and how far to increasing polarization of the various atoms as the water is removed. When mesolite is almost completely dehydrated, absorption of water-vapour is much slower, but X-ray study then shows that the crystal-structure has largely broken down. No definite changes of cell-size or intensities of diffraction could be detected.

 TABLE VIII.
 The variation of the mean refractive index of mesolite (specimen no. 1) during dehydration.
 Yellow light.

(specimen no. 1) during u	ienyaration.	reno	w ngno.		
Water content, mols. per unit cell	64	59	49	43	30
Refractive index	1.507	1.503	1.500	1.498	1.505
Since partially dehydrated n	nesolite ab	sorbs	water	so ra	pidly,

a check on the heat of hydration was possible. The same method and apparatus were used as for natrolite.¹ Partially dehydrated mesolite (2.5381 grams mesolite, dehydrated to a loss of weight of

¹ Part III of this series, Min. Mag., 1932, vol. 23, p. 275.

0.1725 grams, leaving 33.7 mol. H_2O per unit cell) was thrown into water in a crucible, the water-equivalent of the crucible and contents being 15.18 grams ¹; the temperature rose 4.6° C. in three minutes, and then began to fall; extrapolated to zero time, the rise is 4.9° C., whence the mean heat of hydration of mesolite by water-vapour, over the range 64 to 34 mols. per unit cell, and exclusive of the external work, is found to be 18,000 cals. per gram-mol. of water absorbed. The value computed from the vapour-pressure measurements is 17,000 cals., in reasonable agreement considering the approximate nature of the calorimetry and the difficulty of ascertaining exactly the slope of the isohydric curves.

An X-ray examination of a partially dehydrated mesolite containing about 30 mols. H_2O per unit cell indicated a considerable breakdown of the lattice, the photograph being very weak. A very unexpected and inexplicable result is that the cell size was found to be a 56.8, b 6.53, c 18.56 Å.—a larger cell than that of the fully hydrated mesolite.

Base-exchange.

The base-exchange reactions of mesolite are of considerable interest, since they furnish a possible means of testing the assumption, commonly made, that natrolite, mesolite, and scolecite are isostructural.

J. Lemberg (1876) treated mesolite with hot potassium chloride solution, and obtained a product approximating in composition to $K_2Al_2Si_3O_{10}.2H_2O$, and by treating this with sodium chloride solution, a product having approximately the composition of natrolite was obtained. Lemberg, however, did not examine his products optically, so that their identity with natrolite and potassium-natrolite was never fully established. Moreover, under his experimental conditions it was possible that the mesolite might dissolve up and natrolite crystallize out rather than a simple base-exchange occur.

G. Steiger (1905) treated mesolite with a fusion of thallous nitrate, at 240–280° C., and obtained a product approximating to $Na_{0.4}Tl_{10.6}$ $Ca_{2.5}Al_{16}Si_{24}O_{80}.17.3H_2O$. He did not examine this product optically, nor did he compare it with thallous-natrolite, which he had previously prepared. The composition is practically the same as

¹ The water-equivalent is, of course, the water-equivalent of the crucible, plus that of the (fully hydrated) mesolite originally taken, plus the weight of water put in the crucible, minus the weight of water lost by the mesolite (and re-absorbed during the experiment). The specific heat of mesolite was assumed to be 0.24, near that of natrolite.

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that of the thallous-mesolite described below, which suggests that Steiger's product was also a mesolite.

The author has already shown¹ that a four-months digestion of natrolite with calcium chloride results in a considerable baseexchange, which does not, however, go so far as to produce mesolite. Mesolite (no. 1, from Bhore Ghaut) treated with cold saturated solutions of sodium carbonate, sodium chloride, or calcium chloride for $2\frac{1}{2}$ years, showed no appreciable base-exchange, and optical examination revealed no trace of natrolite or scolecite.²

More success was attained in base-exchange experiments with salt fusions. With sodium chlorate at 270° C., $\frac{1}{3}$ of the calcium was replaced in 48 hours, while fusions of silver, thallous, and ammonium nitrates replaced part of the calcium and all or nearly all the sodium. Potassium and lithium nitrate fusions readily replaced all the sodium and nearly all the calcium. The products were all analysed and examined optically, and X-ray rotation photographs were taken of those which had not broken up into powder. The results are shown in table IX.

The lithium compound was homogeneous, and agreed in all respects (chemical, optical, and X-ray) with the lithium derivative prepared from natrolite.³ Its X-ray rotation photograph showed no trace of the extra spots, above mentioned, which serve as a useful criterion for the distinction of mesolite and natrolite. Hence this compound prepared from mesolite is actually lithium-natrolite, and its preparation provides the first decisive proof that natrolite and mesolite are indeed isostructural.

The potassium compound agreed in chemical composition and optical properties with potassium-natrolite, and is certainly identical with the latter, though the proof could not be made so conclusive as for the lithium compound, since the material prepared from natrolite was a very fine powder so that only Debyeograms could be taken. But that prepared from mesolite was more coarse-grained and gave

¹ Compare Part III of this series, loc. cit., pp. 277 ff.

² Complete chemical analyses of these products were made, but there seems no point in reproducing them, as no change is revealed.

³ The latter was formerly described (Part II of this series, loc. cit., and W. H. Taylor *et al.* (1933)) as having a 17.95, b 18.55, c 6.47 Å. These values are inaccurate; new measurements give a 17.84, b 18.63, c 6.47 Å., in better agreement with the compound prepared from mesolite (see table IX). More accurate measurements were also made of the ammonium-natrolite previously described, and the cell-sides were found to be a 17.96, b 18.35, c 6.55 Å., all ± 0.02 Å.

						Treated	l with a sa	alt fusion	L:	
				270° C.	200° C.	270° C.	180° C.	180° C.	270° C.	220° C.
			Un-	70 hrs.	48 hrs.	70 hrs.	48 hrs.	48 hrs.	96 hrs.	48 hrs.
			treated.	NaClO ₃ .	KCNS.	LiNO ₃ .	NH ₄ NO ₃ .	AgNO ₃ .	AgNO ₃ .	TINO ₃ .
					ſ	¶			٩	
	SiO_2	•••	46.05	47.15	43 ·82	50.70	49.96	38.88	39-30	28.60
e d	Al ₂ O ₃	•••	26.84	28.26	24.82	29.50	27.86	22.87	22.47	18.13
tio	CaO	•••	10.00	6.74	2.49	2.08	8.55	5.59	6.03	2.90
ent	Na ₂ O	•••	3.82^{-1}	8.14	nil	nil	nil	1.76	nil	0.58
n p	R ₂ O*	•••			18-51	6.36	6·37‡	20.86	27.21	[42.75]
Å ö	H ₂ O	•••	12.30	10.49	10.32	10.46	7.58‡	9.47	5.91	7.04
- 1	Total		99.80	100.78	99.96	99.10	100.32	99.43	100.92	100.00]
Oxyg	en %	•••	41.21	42 ·11	39.71	45·34	43.97	35.23	34.79	26.35
Oxyg per u	en, atoms nit cell.	\$	240·5	242 ?	82.8 ?	80.2	241.7	$245\pm\$$	83·0±	_
- -	(Si	•••	23.6	23.7	24 ·0	24 ·0	$24 \cdot 1$	$23 \cdot 2$	23.9	$23 \cdot 1$
86 86	Al	•••	16.6	16.7	16.0	16.5	15.9	16.9	$16 \cdot 1$	17.3
pel xyi	Ca		5.6	3.6	1.5	1.0	4.4	3.7	3.9	2.5
808	Na		3.9†	7.9	nil	nil	nil	2.1	nil	0.9
n d	R*				12.9	$12 \cdot 2$	7.1	6.8	8.6	9.8
at A	0,H/		21.5	17.6	18.9	16.5	12.2	19.8	12.0	19.0
Sp. g	r		2.259	2·22 ?	2.22?	2.19	2·144	2.63-	$2.8 \pm$	$2 \cdot 35 \pm$
								2.7	5	
Mean	refractiv	7 e }								
indez light	t for Na	a-} 1	1.50	74 1.499) 1.483	1.491	1.511	1.54	2 1.552	2 1.588
Biref	ringence	· • •	0.00	2 0.00 4	l±0.010	0.013	l variabl	e, 0·004	4 ± 0.002	very
							very sm	all		small
Optio elong	c orientat ation =	ion	;} β	γ	γ	γ	_	a	a	γ
	. (a	••••	56.7	56.3	19.27	17.61	56.4	56.7	18.30	
dę	< {b		6.54	6.54	19.77	18.65	6.53	6.53	18.90	-
ບ. <u>ສ</u>	.5 (_c		18.44	18.58	6.51	6.49	18.38	18.42	6.51	—
	r*		0.98	0.98	1.33	0.68	1.44	1.5 ?	1.5 ?	1.7 ?
ť.	<i></i>									
r le Bole	ន្លី (110° C).	21-4	16.2	2.6	16.2	11.5	19.5	11.7	8.3
Wate in n	{§∫ 300° ().	7.0	0.5	0.9	3.2	nil?	3.9	2.4	5.0

TABLE IX. Experiments on base-exchange, with Bhore Ghaut mesolite (specimen no. 1).

* R indicates the kation of the salt employed in the base-exchange, and r is its radius in Å.

† Also 0.79 % K₂O, or 0.53 atoms K per 80 oxygen.

 \ddagger The estimation of NH₃ and H₂O in this material was made by the method described in Part III of this series, loc. cit., p. 278, footnote.

|| By difference.

 \P The optics, water contents, and X-ray data show these products are natrolites and not mesolites.

§ Taking the sp. gr. as 2.69; if it is taken as 2.63, the oxygen content comes to 239 atoms per unit cell.

fairly good rotation photographs, on which the critical spots were definitely absent.

Two silver compounds were prepared, one by the action of silver nitrate at 180° C. for 48 hours, the other by treatment for 96 hours at 270° C.; neither was completely homogeneous, as shown by the variable refractive indices and densities. The product treated for the shorter time and containing least silver, was shown by X-ray photographs to be still a mesolite, the critical diffractions being all present; in agreement with this, its water content was higher (19.8 mols. per 80 oxygen) than that of natrolite (16 mols.). The compound richer in silver, on the other hand, was found to be a silver-natrolite, a result confirmed by its low water content. The silver-mesolite had a higher birefringence than this silver-natrolite, and a lower refractive index and density. The silver-natrolite prepared from mesolite still contained a considerable amount of calcium, and on comparing it with the lime-free compound prepared from natrolite (which also contains more silver) it will be seen that the birefringence is higher, the refractive index and density lower than those of the latter; there were also distinct intensity differences in the X-ray photographs.

By the action of an ammonium nitrate fusion, a compound was prepared which an X-ray photograph showed to be an ammoniummesolite. This material had a very low birefringence and was zoned, the outside of the crystals having a positive elongation and the centre a negative one, with an isotropic zone between.

Thallous nitrate gave a base-exchange product consisting of a fine powder, of which no X-ray examination could profitably be made; judging from its high water content, it is a thallous-mesolite.

By the action of a sodium chlorate fusion on mesolite, it was hoped that natrolite would be produced; the product actually obtained had a higher birefringence and lower refractive index than mesolite, and its optical orientation was $\gamma = b$ (axis of elongation of the needles), but an X-ray photograph showed that it was still a mesolite. It is probable that the optical changes are due rather to its lower water content than to the replacement of calcium by sodium; even after soaking in water it only contained 18 mols. water per 80 oxygen. It may be noted that the decrease in water content is approximately equal to the increase in sodium, but this can hardly be more than an accidental relation, for in the transformation of mesolite into natrolite, twelve sodium atoms are taken up, but only six water molecules are lost (calculated on a basis of 80 oxygen). Probably the effect of the introduction of sodium is simply to increase the water-vapour pressure at any given temperature and water content.

A study of the effect of base-exchange on the vapour-pressure of a suitable zeolite should prove of considerable interest. Time did not permit of this being carried out, but for a rough comparison, each of the base-exchange products was dehydrated at 110° C. and at 300° C.; no attempt was made to regulate the water-vapour pressure, but as all the specimens were heated together in the same oven, it is approximately the same for all, so permitting of comparison. The water-vapour pressure was probably about 1–3 mm. Hg.

The introduction of extra sodium into mesolite (in place of calcium) is seen to lead to a greater loss of water both at 300° C. and 110° C., indicating a marked increase in the vapour-pressure, as suggested above. Ammonium (replacing all the sodium and part of the calcium) also caused an increase in the vapour-pressure, as was to be expected from its large ion. Thallium, with a still larger ion, caused a still greater loss of water at 110° C., but the loss at 300° C. was less than for the ammonium-mesolite, and of the same order as that of mesolite itself. Silver, on the other hand, with an atomic radius probably intermediate between those of ammonium and thallium, had little effect on the water loss at either temperature. The reason for this behaviour is quite obscure.

The base-exchange products prepared from natrolite¹ were not studied in respect of their dehydration. But the potassium-, lithium-, and silver-calcium-natrolites now prepared from mesolite were included in the above series of dehydration experiments. At 110° C. natrolite itself will retain about 15 mol. water per unit cell, at 300° C. nil.¹ Replacement of sodium by lithium with a smaller atomic radius is seen to favour retention of the water, as might be expected, while the larger potassium atom favours its loss. Indeed, the effect of potassium is so great that while natrolite itself only loses most of its water at about 230°C. at 1 mm. Hg, potassiumnatrolite has lost nearly all of it at 110°C. Silver, although its atomic radius is probably greater than that of potassium,¹ only causes a small increase in the loss of water at 110° C., and at 300° C. the loss is probably less than for natrolite itself. Ammonium, an ion somewhat larger than potassium, evidently has an even greater effect

¹ Part III of this series, loc. cit.

on the affinity of natrolite for water than has potassium, for the ammonium-natrolite was anhydrous.¹ The large thallium ion, on the other hand, had a less effect than ammonium (contrary to the case with mesolite), for the author obtained thallous-natrolite hydrated; ¹ Steiger's thallous-natrolite, though prepared in the same manner, was, on the other hand, anhydrous, so the thallium ion must decrease the affinity of natrolite for water very markedly.

Summary.

Mesolite is an independent species, isostructural, but not isomorphous with natrolite and scolecite. Five new analyses have been made. The unit cell, of dimensions a 56.7, b 6.56, c 18.44 Å., β 90°0′, contains Na₁₆Ca₁₆Al₄₈Si₇₂O₂₄₀.64H₂O. The space-group is C³₂, but there is a marked approximation to the orthorhombic space-group C_{2m}^{19} . Goniometric measurements are in excellent agreement with the X-ray data, and show a marked development of vicinal pyramids. The optical properties, vapour-pressure, and base-exchange of mesolite have been studied in considerable detail. It has been found possible to make a ready distinction between mesolite and natrolite by X-ray methods, and it has been shown that by the action of fusions of potassium thiocyanate, lithium nitrate, and silver nitrate on mesolite, compounds identical with similar derivatives prepared from natrolite may be obtained; these have the natrolite structure. The vapourpressure measurements have been checked by a direct determination of the heat of hydration of partially dehydrated mesolite. Mesolite has been observed on specimens from eight localities new for the species.

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