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Studies on the zeolites. Part II. Thomsonite (including faroelite) and gonnardite.[1]

By MAX H. HEY, B.A., B.Sc.
With X-ray measurements by F. A. BANNISTER, M.A.
Assistant-Keepers in the Mineral Department of the British Museum of Natural History.

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THOMSONITE.

THOMSONITE was first recognized by H. J. Brooke (1820)[2] in a study of the mesotypes or 'needlestones'. The 'needlestone' of Lochwinnoch and Old Kilpatrick, Dumbartonshire, was found to be an independent species, in addition to the natrolite, mesolite, and scolecite into which A. F. Gehlen and J. N. Fuchs,[3] and J. N. Fuchs,[4] had divided the mesotype of R. J. Haüy.[5] The new species was named *thomsonite*, in honour of Dr. T. Thomson, who analysed the material a few months later.

The next year, Sir David Brewster (1821) made an optical and goniometric study of some so-called apophyllite crystals from Vesuvius, and finding them to differ from all other minerals he had studied, recognized a new species, naming it *comptonite*, in honour of Lord Compton. It is perhaps worth noting that Brewster's optical studies, made with the simplest apparatus, were of remarkable accuracy. He gave a correct account of the optics of comptonite,

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[1] Part I. General Review. Min. Mag., 1930, vol. 22, p. 422.

[2] See bibliography, p. 121.

[3] A. F. Gehlen and J. N. Fuchs, Schweigg. Journ., 1813, vol. 8, p. 358.

[4] J. N. Fuchs, Schweigg. Journ., 1816, vol. 18, p. 1.

[5] R. J. Haüy, Traité de Min., Paris, 1801, vol. 3, p. 151.

but for some reason failed to recognize the identity of thomsonite and comptonite, though he had studied the former (1821, 1822); he appears never to have published the promised details of his results.

Indeed, the identity of thomsonite and comptonite was not generally recognized, even after T. Thomson (1836) had analysed comptonite from Vesuvius, and E. Melly (1838) similar crystals from Kaaden, Bohemia. (T. Monticelli and N. Covelli, 1825, and A. Breithaupt, 1832, had united the species, but without definite reason.) It remained for C. F. Rammelsberg (1839) to bring forward the analyses, together with his own, in evidence for the union. Comptonite is still used as a varietal name to cover the short prismatic or tabular crystals so typical of the Vesuvius and Kaaden occurrences.

Mesole, analysed by J. Berzelius (1822), was united with thomsonite by W. Haidinger (1845). M. F. Heddle (1853) added further analyses and proposed the name *faroelite*, from the original and typical locality, in place of mesole, which is too much like mesolite; this emendation has been fairly generally adopted. A. Des Cloizeaux (1863) brought optical evidence for the union of faroelite and thomsonite, while E. T. Wherry (1923, 1925) would separate them, also on optical grounds. Faroelite includes the silica-rich members of the thomsonite isomorphous series, and usually is botryoidal in habit.

Besides these, the principal varieties, a number of other varieties of thomsonite have been described, and a number of minerals, originally assigned species rank, have been shown to be identical with thomsonite. Scoulerite, ozarkite, picrothomsonite, mesolitine, karphostilbite, sphaerostilbite, koodilite, lintonite, and bagotite fall into these two groups. Chalilite and harringtonite, which have been included with thomsonite by many authors, are mixtures, while sloanite appears to be really laumontite. All these, with kalithomsonite and gonnardite, are discussed later.

Composition and constitution.

The analyses of thomsonite and its varieties all indicate that it is a true zeolite in the sense defined in part I of this series; that is, it is a hydrated alumino-silicate, with the ratio $O/(Si+Al) = 2$ (and hence $Al_2O_3/(Na_2O+CaO) = 1$). The percentage of silica varies between about 36 and 42 per cent.[1] Several views of its composition

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[1] S. J. Thugutt (1912) attributes the variations in composition to impurities; this view is quite untenable.

and constitution have been put forward, but in view of the results of Prof. W. L. Bragg and his pupils, as set out in part I, it is unnecessary to discuss the constitutional formulae proposed by F. W. Clarke (1894, 1895), H. C. McNeil (1906), G. Tschermak (1917), and others.

Recently, E. T. Wherry (1923, 1925), from a study of the chemical and optical data then available, concluded that thomsonite should be divided into three species of fixed composition, namely thomsonite proper, $NaCa_2Al_5Si_5O_{20}.6H_2O$; faroelite or mesole, $Na_2Ca_2Al_6Si_7O_{26}.7H_2O$; and a third species, free of or poor in soda, of uncertain formula, for which he retains the name comptonite. He writes: 'The earliest name given to presumably soda-free material was comptonite (Brewster, 1821), although it has also been subsequently termed chalilite, picrothomsonite, sloanite, calciothomsonite, etc.' Vesuvian comptonite is certainly not soda-free, and the proper application of this and the other names are set out elsewhere. Wherry lays especial stress on the discovery of O. B. Bøggild (1922) that thomsonite is often intergrown with natrolite or mesolite, and interprets many analyses as such mixtures, or mixtures of thomsonite and faroelite. He rightly complains of the lack of chemical analyses of optically controlled material, a matter which the author has striven to remedy by sixteen new analyses[1] of material carefully separated and optically tested (table I). These, together with the new optical and X-ray data, conclusively disprove Wherry's view, and in the main support that of A. N. Winchell (1925, 1926). The latter concludes that there is an isomorphous replacement $NaSi \rightleftarrows CaAl$ in thomsonite, between extremes of about $Ca_8Na_4Al_{20}Si_{20}O_{80}.25H_2O$ and $Ca_5Na_7Al_{17}Si_{23}O_{30}.20H_2O$. He supports this view from the available chemical and optical data, and by new optical measurements and Debyeograms.

The author's results bear out Winchell's theory except in two respects. Winchell did not investigate the water content, but accepted the conclusions of S. G. Gordon (1924) in this respect. (The latter proposed an isomorphous mixture of $CaAl_2Si_2O_8.3H_2O$ and $Na_2Al_2Si_3O_{10}.H_2O$, which is not supported by the author's studies.) The author finds the water content appreciably constant at 24 molecules per unit cell. Secondly, Winchell does not consider the substitution $Na_2 \rightleftarrows Ca$ of much importance in zeolite constitution, whereas

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[1] Including one made some years ago by Mr. F. N. Ashcroft, and now published with his kind permission.

TABLE I. New chemical, optical, and X-ray data on thomsonite.

		1.	2.	3.	4.	5.	6.	7.	8.
Percentage composition.	SiO_2 ...	40·3	41·20	41·44	40·57	40·39	39·87	40·73	39·91
	Al_2O_3 ...	28·5	29·47	29·52	29·08	29·39	28·98	29·93	29·91
	CaO ...	11·2	11·87	13·02	11·85	7·09	9·70	12·17	12·29
	Na_2O ...	5·7	4·70	2·93	3·40	10·54	6·50	4·66	4·83
	K_2O ...	nil	0·27	nil	nil	nil	nil	nil	0·02
	H_2O ...	14·1	12·74	13·89	14·33	12·39	14·28	12·55	13·40
	Total ...	99·8	100·25	100·80	99·23	99·80	99·33	100·04	100·36
Oxygen % ...	...	39·63	39·86	40·10	39·86	40·20	39·64	40·46	39·90
Oxygen, atoms per unit cell.		78·91	79·68	79·06	79·65	79·64	—	—	—
Atoms per unit cell, assuming oxygen 80.	Si	21·72	21·70	21·82	21·84	21·42	21·60	21·42	21·18
	Al	18·08	18·28	18·34	18·46	18·46	18·50	18·60	18·74
	Ca ...	6·48	6·68	7·36	6·84	4·02	5·64	6·86	7·02
	Na ...	5·90	4·80	2·98	3·56	10·84	6·82	4·74	4·98
	K	—	0·18	—	—	—	—	—	0·02
	H_2O ...	25·34	22·34	24·40	25·74	21·94	25·78	22·00	24·48
	Si+Al ...	39·80	39·98	40·16	40·30	39·88	40·10	40·02	39·92
	Ca+Na ...	12·38	11·66	10·34	10·40	14·86	12·46	11·60	12·02
Sp. gr.	... ...	2·326	2·328	2·310	2·327	2·314	2·29*	2·11*	2·26*
Optical constants, for Na-light.	α	1·511	1·518	1·518	1·518	1·523	1·518	1·517	1·520
	β	1·513	1·520	1·522	1·520	1·525	1·520	1·520	1·522
	γ	1·518	1·529	1·528	1·528	1·537	1·528	1·527	1·528
	2V ...	75° ± 10	66° ± 3	—	—	—	—	60° ±	—
	$(\alpha+\beta+\gamma)/3$	1·514	1·522	1·523	1·522	1·528	1·522	1·521	1·523
Cell-sides in Å.†	a	13·07	—	13·05	—	13·07	—	—	—
	b	13·07	—	13·05	—	13·07	—	—	—
	c	13·23	—	13·23	—	13·23	—	—	—
Water left, in mols. per unit cell, after heating to:	100° C. ...	22·10	20·76	22·20	24·62	19·56	22·36	20·30	—
	300° C. ...	—	12·08	11·58	—	15·12	—	—	—
	350° C. ...	10·98	—	—	9·12	—	6·84	5·00	—

* Probably too low. † Accuracy, ±0·05 Å.

1. B.M.1914,1077. Table Mtn., Colorado. Confused aggregate of small platy crystals. Alkalis on 0·12 gram, rest on 0·10 gram.
2. B.M.1907,739. Nolsö, Faeroe Islands. Coarse fibrous, radiating, of the kind called sphaerostilbite.[1]
3. B.M.1920,502. Goble, Columbia Co., Oregon. Small waxy spherules, very finely fibrous. Analysis on 0·25 gram.[1, 2]
4. B.M.1907,761. Nolsö, Faeroe Islands. Botryoidal fibrous, faroelite.
5. B.M.1918,107. Schieferberg, Salesl, Bohemia. A fibrous crust of minute cruciform twins.[1]
6. B.M.95233. Hauenstein, Leitmeritz, Bohemia. Botryoidal fibrous, the surface showing the terminations of cruciform twins. Alkalis on 0·29 gram, water on 0·27 gram, rest on 0·53 gram.
7. B.M.1914,1537. Table Mtn., Colorado. Botryoidal fibrous.
8. Ashcroft, 2978. Carnmoney Hill, Ballyduff, Co. Antrim. Analysed by F. N. Ashcroft (priv. comm.). Fe_2O_3 0·15 %, MgO nil, SrO nil. Sp. gr. and optics by M. H. H.

[1] Water by loss on ignition. [2] Alkalis on filtrate from lime.

TABLE I (*continued*).

		9.	10.	11.	12.	13.	14.	15.	16.
Percentage composition.	SiO_2 ...	40·23	39·01	39·14	38·00	37·40	38·69	37·17	37·11
	Al_2O_3 ...	30·19	29·94	30·35	31·03	30·73	31·91	31·93	31·76
	CaO ...	10·25	12·71	12·17	13·92	14·32	14·06	13·98	14·04
	Na_2O ...	6·67	4·80	4·88	4·73	3·33	3·82	4·00	3·59
	K_2O ...	nil	nil	nil	nil	nil	nil	trace	nil
	H_2O ...	12·42	13·24	13·20	13·10	13·53	12·83	13·35	13·80
	Total ...	99·76	99·70	99·74	100·78	99·31	101·31	100·43	100·30
Oxygen % ...	...	40·39	39·86	39·97	39·73	39·60	40·11	39·68	39·53
Oxygen, atoms per unit cell.	...	80·05	80·98	80·76	80·96	78·94	81·20	80·83	80·25
Atoms per unit cell, assuming oxygen 80.	Si	21·26	20·92	20·92	20·22	20·24	20·28	19·88	19·94
	Al	18·82	18·92	19·12	19·48	19·60	19·72	20·12	20·14
	Ca	5·80	7·30	6·96	7·92	8·30	7·90	8·00	8·08
	Na ...	6·84	4·98	5·04	4·88	3·50	3·88	4·14	3·74
	K	nil	nil	nil	nil	nil	nil	trace	nil
	H_2O ...	21·86	23·66	23·56	23·26	24·40	22·44	23·80	24·68
	Si+Al ...	40·08	39·84	40·04	39·70	39·84	40·00	40·00	40·08
	Ca+Na ...	12·64	12·28	12·00	12·80	11·88	11·78	12·14	11·82
Sp. gr.	... ...	2·308	2·366	2·353	2·373	2·322	2·352	2·373	2·364
Optical constants for Na-light.	α	1·517	1·529	1·523	1·526	1·523	1·523	1·529	1·528
	β	1·519	1·531	1·525	1·528	1·525	1·526	1·531	1·531
	γ	1·526	1·544	1·532	1·540	1·532	1·539	1·542	1·545
	2V... ...	60° ±	59° ± 2	60° 6′	50° ±	47½° ±	56° ± 1	51° 39′	54° ± 2
	$(\alpha+\beta+\gamma)/3$	1·521	1·535	1·527	1·531	1·527	1·529	1·534	1·535
Cell-sides in Å.†	a	—	—	—	—	—	—	13·09	—
	b	—	—	—	—	—	—	13·09	—
	c	—	—	—	—	—	—	13·23	—
Water left, in mols. per unit cell, after heating to:	100° C. ...	20·66	23·10	23·18	23·00	22·30	21·70	23·38	21·38
	300° C. ...	11·22	—	16·16	16·14	—	14·92	16·44	—
	350° C. ...	—	10·50	—	—	7·68	—	—	9·46

† Accuracy, ±0·05 Å.

9. B.M.1918,66. Ritina-thal, Leitmeritz, Bohemia. Botryoidal fibrous. Analysis on 0·49 gram.[1, 2]

10. B.M.1914,733. Burger's quarry, West Paterson, New Jersey. Radiating lamellar.[1]

11. B.M.1914,956. Seisser Alpe, Bozen, Tyrol. Radiating massive, of prismatic needles. Analysis on 0·90 gram.[1, 2]

12. B.M.94975. Kaaden, Bohemia. Small clear isolated crystals. Analysis on 0·29 gram.[1, 2]

13. B.M.35300. Monte Catini, Val de Cecina, Tuscany. Radiated platy. 'Picrothomsonite.' MgO 0·10 % = Mg 0·08 atom per unit cell.

14. B.M.60055. Monte Somma, Vesuvius. Small platy crystals and crystal groups. Analysis on 0·28 gram.[1, 2]

15. B.M.94985. Old Kilpatrick, Dumbartonshire (probably from Bowling quarry). Stout radiating glassy prisms, very pure.

16. B.M.1914,1306. Dulce quarry, Marienberg, Aussig, Bohemia. Glassy botryoidal crust composed of fairly large crystals.

[1] Water by loss on ignition. [2] Alkalis on filtrate from lime.

Notes on the analyses:—Two evaporations were made for silica in every case, but the trace of silica in the alumina precipitate was not separated. Wherever the amount of material permitted, the Lawrence Smith method was used for alkalis and the Penfield method for water; where the material was insufficient for this, the water was determined as loss on ignition, and the alkalis in the filtrate from the lime, magnesia being neglected. One gram (about) of material was used for the main analysis, one gram for the water, and ½ gram for the alkalis, except where noted.

the author finds that material with an appreciable substitution of this type is fairly common.

Both Wherry and Winchell make use of only a limited number of selected analyses in their studies. While it is essential to exclude all doubtful analyses in a first review of the composition, it is desirable to check the conclusions thus obtained by comparison with all available data. For the first review, the author's new analyses alone will be considered. They vary in accuracy, since the amount of material available was only small in some instances, but the purity of the material was carefully controlled. For each specimen, the optical data were determined as fully as possible, and Mr. F. A. Bannister made X-ray determinations of the cell-sides of four specimens. The densities were also determined, but, owing to the finely fibrous nature of some of the material, the accuracy of this constant is not as great as could be desired in some cases. These exceptions are distinguished by asterisks; all are low values, probably due to unavoidable inclusions of air among the minute fibres. All these data are incorporated in table I.

Combining the chemical, X-ray,[1] and density determinations, we arrive at the actual number of atoms per unit cell. The oxygen content is found to be constant within the limits of experimental error, and is 80 atoms per unit cell. The water content varies irregularly about 24 molecules per unit cell, while the silicon, aluminium, sodium, and calcium contents vary greatly. (The unit cell contents tabulated in table I are based on the assumption that there are 80 oxygen atoms per unit cell, except of course the figure for oxygen, which is based on the chemical, X-ray, and density data together.) The sum of aluminium and silicon is, however, very constant at 40, so that $O/(Al+Si) = 2$; from this we can conclude

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[1] Since the cell-sides were shown to be practically constant in length, it appeared permissible to assume their values for those specimens for which they were not actually determined. The approximation of the determined atomic contents to the ideal figure is much better than is usually accepted as satisfactory, and suggests that the usual practice might be improved on.

that we are dealing with an indefinitely extended tridimensional anion. But the number of kations per unit cell (Ca + Na) is decidedly variable, and though a strong tendency towards a definite integral value, 12, is noticeable, the new analyses are sufficient to show that the $Na_2 \leftrightarrows Ca$ substitution is by no means negligible. (Analysis 72, on optically controlled material, also confirms this.) Thomsonite that has been shown to contain exactly the characteristic number of 12 kations per unit cell will be referred to, in accordance with the convention suggested in part I, as *normal*. In normal thomsonite we have a replacement of NaSi by CaAl, and departures from normality indicate substitution of Ca by Na_2 or some other pair of monovalent kations, or of Na_2 by Ca or some other divalent kation.

For comparative purposes the composition is conveniently summed up by two figures—Al, the number of aluminium atoms per unit cell of 80 oxygen atoms, and (Ca + Na), the total number of kations per unit cell. The departure of the latter figure from 12 measures the extent of $Na_2 \rightleftarrows Ca$ replacement; (Ca + Na – 12) equals the amount of Ca replaced by Na, a negative difference, of course, meaning the reverse replacement.

Turning to the remainder of the available analyses, we require some means of judging their quality. We might follow Wherry and Winchell in excluding all the older work and also any analyses that appear unreliable for other reasons; but a better means of classification is available. It is generally accepted that for the zeolites the molecular ratio $Al_2O_3/(CaO + Na_2O) = 1$; from this it follows (cf. part I) that $O/(Si + Al) = 2$, and we have already seen that the new data confirm this, and also fix the number of oxygen atoms in the unit cell at 80. Further, following H. S. Washington, W. F. Hillebrand, and other prominent chemists, we may take the maximum permissible errors for a first-class analysis as ±0·25 % in the SiO_2, 0·2 % in the Al_2O_3, and 0·1 % in the CaO and the Na_2O. From these premises it follows that for a first-class analysis of pure material, the number of silicon and aluminium atoms per unit cell (calculated on an assumption of 80 oxygen atoms per unit cell) should fall within the limits[1] 40·00 ± 0·20. To this we may add a further criterion, that the summation should fall between 99·70 and 100·80 %.

Of 107 analyses collected from the literature, only 34 satisfy these criteria; 28 are second-class analyses, for which the summation

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[1] In general terms, the tolerance in (Si + Al) is ±0·5 % for first-class and ±1·0 % for second-class analyses.

[*continued on* p. 64

TABLE II. Analytical data for thomsonite (from the literature), arranged by localities, and recalculated to atomic ratios (O = 80), giving the number of atoms per unit cell.

No.	Si.	Al.	Ca.	Na.	H_2O.	Si + Al.	Ca + Na.	Sp. gr.	Class.
17.	20·94	19·16	7·10	4·22	23·56	40·08	11·66*	2·362	A
18.	21·32	19·40	5·98	4·54	21·02	40·72	10·52	2·17	C
19.	22·36	17·32	6·44	5·72	22·26	39·70	12·16	—	B
20.	20·74	18·92	6·00	8·32	23·60	39·62	14·30†	—	C
21.	22·28	17·36	6·54	5·68	22·74	39·66	12·22	—	B
22.	22·26	17·84	6·54	4·36	26·78	40·10	10·90	—	A
23.	21·38	18·74	6·72	5·04	24·50	40·04	11·76	—	A
24.	21·50	17·42	7·34	5·80	21·40	38·94	13·14	2·276	B
24*a*.	20·96	19·04	6·36	6·34	22·10	40	12·68		
25.	20·10	19·58	10·20	0·48	24·04	39·68	10·68	2·30	B
26.	21·64	18·16	6·86	5·12	26·20	39·80	11·98	2·229	B
27.	21·34	18·36	7·30	4·94	24·72	39·70	12·24	2·229	B
28.	21·70	18·44	2·70	12·50	18·78	40·14	15·20	2·125	B
29.	21·62	18·58	6·26	5·22	21·68	40·22	11·48	2·19–2·30	B
30.	20·64	19·34	7·24	4·98	24·14	39·98	12·22	—	A
31.	20·30	19·88	7·20	4·88	23·90	40·18	12·08	2·383	A
32.	20·14	19·94	7·74	4·16	24·92	40·08	11·90	—	A
33.	19·74	20·26	7·90	4·38	24·02	40·00	12·08	—	A
34.	20·38	19·26	7·48	5·26	23·54	39·66	12·96‡	—	B
35.	18·72	20·60	10·76	1·32	25·20	39·32	12·08	—	C
36.	22·00	18·22	6·92	5·50	23·50	40·22	12·42	—	B
37.	21·60	18·36	6·36	3·98	25·80	39·98	11·60§	2·131–2·147	A
38.	21·94	17·76	6·54	5·92	23·56	39·68	12·46	—	B
39.	21·18	18·84	5·70	5·56	22·76	40·02	12·20‖	—	A
40.	21·70	18·64	6·42	4·46	24·22	40·34	10·88	—	B
41.	22·38	17·80	5·60	5·90	21·10	40·18	11·50	—	A
42.	20·36	18·88	7·62	6·62	20·88	39·26	14·22	2·34	C
43.	21·94	17·32	7·72	3·92	22·64	39·26	12·04	—	C
44.	18·60	20·88	9·54	3·86	26·00	39·48	13·40	—	C
45.	19·94	20·12	7·84	4·18	24·32	40·06	12·04	2·380	A

* Including 0·08 atom K per unit cell.

† 0·5 % Fe_2O_3 not included.

‡ Including 0·22 atom Mg.

§ Including 0·54 atom Mg and 0·72 atom K; 3·28 % Fe_2O_3 not included.

‖ Including 0·26 atom K, 0·04 atom Mn, and 0·64 atom $Fe^{\cdot\cdot}$.

TABLE II (*continued*).

ICELAND:

17. S. von Waltershausen, 1853. Berufjord. 'Karphostilbite.'
18. F. von Kobell, 1866. Reidarfjord (=Rodetfjord, =Rodefjord).

FAEROE ISLES:

19. J. Berzelius, 1822. Faeroe Is. 'Mesole.'
20. M. G. Retzius, 1825. Dalsnypen, Sandö.
21. M. F. Heddle, 1889. Nolsö (=Naalsoe).
22. C. Hersch, 1887. Nolsö.
23. J. Lemberg, 1876. Nolsö. [B.M.86707.
24. G. T. Prior, 1898. Nolsö. 'Sphaerostilbite.' Optics, see table VI.

24*a*. 24 recalculated, assuming 9 % of gyrolite as impurity. The specimen carries gyrolite.

GERMANY:

25. O. Luedecke, 1877. Pflasterkaute, Eisenach, Thuringia.

IRELAND:

26. J. Apjohn, 1844. Co. Antrim. 'Mesolitine.'
27. J. Apjohn, 1844. Newtown Crumlin, Clough, Co. Antrim. 'Mesolitine.'
28. T. Thomson, 1836. Co. Antrim. Supposed to be mesolite.
29. A. & R. Brauns, 1924. Larne, Co. Antrim. Supposed to be harringtonite.

SCOTLAND:[1]

30. L. Gmelin, 1826. Old Kilpatrick, Dumbartonshire.
31. C. F. Rammelsberg, 1853. Old Kilpatrick.
32. G. Tschermak, 1917. Old Kilpatrick.
33. J. Lemberg, 1876. Old Kilpatrick.
34. J. Berzelius, 1822. Old Kilpatrick.
35. T. Thomson, 1836. Old Kilpatrick.

Probably from Bowling quarry, as is most material labelled 'Old Kilpatrick'. There is also an inaccurate analysis by T. Thomson (1827).

36. (R. D. ?) Thomson, 1852. Isle of Skye. Optics, see table VI. B.M.95003.
37. M. F. Heddle, 1882. Old Man of Storr, Skye.
38. M. F. Heddle, 1857. Old Man of Storr.
39. M. F. Heddle, 1882. Quirang, Skye.
40. M. F. Heddle, 1857. Portree, Skye.
41. M. F. Heddle, 1857. Uig, Skye. This highly siliceous material should repay re-examination if still extant.
42. A. Lacroix, 1887. Bishopton railway tunnel, Renfrewshire. 2E 85°, $\gamma-\alpha$ 0·027. This analysis is not very satisfactory.
43. R. S. Houston, 1912. Boylestone quarry, Barrhead, Renfrewshire.
44. R. S. Houston, 1912. Boylestone quarry.
45. J. W. Young, 1873. Mugdock water tunnel, Stirlingshire.

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[1] T. Thomson's analyses (1820, 1836) of material from Lochwinnoch, Renfrewshire, are inaccurate, and this, the original thomsonite locality, is still unrepresented analytically.

TABLE II (*continued*).

No.	Si.	Al.	Ca.	Na.	H_2O.	Si+Al.	Ca+Na.	Sp. gr.	Class.
46.	20·52	18·88	8·12	4·92	26·76	39·40	13·20*	2·25	C
47.	20·62	19·14	7·66	4·66	24·12	39·76	12·32	2·389	B
48.	20·52	19·08	7·36	5·10	19·54	39·62	13·30†	—	C
49.	19·64	20·06	8·18	4·04	24·20	39·70	12·86‡	—	B
50.	19·88	19·54	8·44	3·88	24·38	39·44	13·08§	2·31	C
51.	22·84	17·02	7·42	2·40	24·84	39·86	10·16‖	—	A
52.	20·16	20·34	7·28	3·64	28·44	40·50	11·02¶	2·26	C
53.	20·54	19·00	8·64	3·60	24·04	39·54	12·24	—	C
54.	20·82	19·18	7·38	4·40	23·92	40·00	11·78	—	A
55.	18·02	21·30	7·30	7·68	21·74	39·30	16·16**	—	C
56.	20·64	17·88	10·06	3·52	32·40	38·52	13·58	2·22–2·34	C
56*a*.	21·42	18·58	7·46	3·66	25·86	40	11·12		
57.	20·82	19·68	6·74	4·18	23·00	40·50	10·92	2·31	C
58.	20·20	19·80	7·74	4·38	23·24	40·00	12·12	2·374	A
59.	22·48	17·10	7·02	4·70	23·60	39·58	11·92††	2·29	C
60.	20·60	19·34	7·64	3·98	23·78	39·92	11·98‡‡	2·35–2·40	A
61.	20·02	19·70	6·68	7·42	20·08	39·72	14·12	2·35–2·38	B
62.	20·52	19·86	7·08	3·96	22·58	40·38	11·38§§	2·37	B
63.	20·32	20·12	7·40	3·58	22·40	40·44	10·98	—	C
64.	20·44	19·72	7·74	3·64	22·92	40·16	11·38	—	A
65.	19·80	20·14	7·86	7·86	23·91	39·94	12·02‖‖	2·39	A
66.	21·48	18·68	4·70	8·70	26·18	40·16	13·40	—	A
67.	21·12	19·64	4·14	8·30	23·64	40·76	12·44	2·37	C
68.	22·44	17·78	6·68	3·68	23·00	40·20	10·38¶¶	—	A
69.	21·04	18·90	6·32	6·50	20·30	39·94	12·82	—	C
70.	22·94	17·42	4·03	7·93	26·36	40·36	11·96	—	B
71.	21·72	18·28	6·54	5·20	23·36	40·00	11·74	2·25	B

* Including 0·16 atom K.

† Including 0·84 atom K.

‡ Including 0·42 atom K and 0·24 atom Mg.

§ Including 0·46 atom K and 0·30 atom Mg.

‖ Including 0·64 atom K.

¶ Including 0·10 atom K.

** Including 0·54 atom K; after deduction of 8·34 % $CaCO_3$.

†† Including 0·20 atom K.

‡‡ Including 0·36 atom K.

§§ Including 0·34 atom K.

‖‖ Including 0·50 atom K.

¶¶ 0·40 % Fe_2O_3 not included.

ITALY: TABLE II (*continued*)

46. V. Gennaro, 1929. Bettolina, Valle di Ayas, Piedmont. Optics, see table VII.
47. P. Riva, 1914. Casarza, Liguria. Optics, see table VII.
48. S. von Waltershausen, 1853. Cyclopean Isles, Catania, Sicily.
49. E. Manasse, 1906. Monte Catini, Val de Cecina, Tuscany.
50. E. Manasse, 1906. Monte Catini.
51. J. Lemberg, 1876. Monte Catini. 'Faroelite.'
52. V. Gennaro, 1929. Val di Viu, Piedmont. Optics, see table VII.
53. G. Freda, 1885. Monte Somma, Vesuvius.[1]

TYROL:

54. J. Lemberg, 1883. Fassa-thal (=Val di Fassa).
55. K. John, 1875. Palle Rabbiosa, Monzoniberg, Fassa-thal.
56. G. Bellinzona, 1923. Monzoniberg, Fassa-thal.

56*a*. 56 recalculated, assuming 9·00 % $CaCO_3$ as impurity.

57. K. Haushofer, 1868. Pufler Loch, Seisser Alpe (=Alpe di Seis), Bozen (=Bolzano).

BOHEMIA:

58. G. Tschermak (E. Zdarek, analyst), 1917. Wesseln (=Veselí), Aussig (=Ústí nad Labem).
59. R. Görgey (B. Mauritz, analyst), 1911. Neubauerberg, Mickenhan, Böhmisch-Leipa (=Česká Lípa).
60. C. F. Rammelsberg, 1853. Seeberg, Kaaden (=Kadaň). 'Comptonite.'
61. F. X. M. Zippe, 1836. Seeberg, Kaaden. 'Comptonite.'
62. C. F. Rammelsberg, 1853. Seeberg, Kaaden. 'Comptonite.'
63. E. Melly, 1838. Seeberg, Kaaden. 'Comptonite.'
64. R. Raffelt (A. Svehla, analyst), 1882. Eulenberg (=Kočičho hradu, =Katzenberg, =Kamaitschken, =Kamaicken), Leitmeritz (=Litoměřice).
65. B. Erben, 1885. Eulenberg, Leitmeritz.
66. C. Hersch, 1887. Hauenstein, Leitmeritz.
67. C. F. Rammelsberg, 1853. Hauenstein, Leitmeritz.[2]

ASIATIC RUSSIA:

68. K. D. Glinka, 1904. Zchra-Zkaro, Borschom, Transcaucasia.
69. P. P. Pilipenko, 1915. Zavodinsky mine, Altai Mts., Siberia.

INDIA:

70. R. D. Thomson, 1834. Karanja Island, Bombay Harbour. 'Faroelite.'

AFRICA:

71. G. D'Achiardi, 1902. Schiket (or Sciket), Eritrea.

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[1] There are also two analyses of highly altered material by E. Scacchi (1888), and one very inaccurate analysis by T. Thomson (1836).

[2] Analysis 6 (table I) agrees with anal. 66; anal. 67 is perhaps of a different generation of material. The analysis by J. Freyssmuth (1818) was probably made on a mixture of thomsonite and mesolite, such as F. Cornu (1903) has shown to occur at this locality.

TABLE II (*continued*).

No.	Si.	Al.	Ca.	Na.	H_2O.	Si+Al.	Ca+Na.	Sp. gr.	Class.
72.	20·88	19·10	6·70	4·24	22·58	39·98	11·06*	2·299	B
73.	21·46	19·08	5·18	5·04	22·98	40·54	10·52†	—	C
74.	21·80	18·18	6·60	4·98	22·58	39·98	11·58	—	A
75.	21·56	18·34	6·50	5·72	21·96	39·90	12·22	—	A
76.	20·96	19·32	6·92	4·26	22·84	40·28	11·22‡	2·339	B
77.	20·08	19·86	8·10	3·90	24·90	39·94	12·00	2·24	A
78.	21·58	18·48	6·74	4·84	22·74	40·06	11·56	—	A
79.	21·46	18·72	6·74	4·54	22·62	40·18	11·28	—	A
80.	22·24	17·98	6·14	4·98	21·36	40·22	11·02	—	B
81.	21·56	18·38	6·38	5·72	22·70	39·94	11·54§	—	A
82.	21·62	18·36	6·38	5·72	20·86	39·98	12·08	—	A
83.	21·68	18·38	6·36	5·42	23·08	40·06	11·78	—	A
84.	21·44	18·66	6·26	5·10	21·90	40·10	11·36	—	B
85.	21·64	18·76	6·22	4·70	21·24	40·40	10·92	—	C
86.	22·34	17·52	6·38	5·10	25·66	39·86	11·72‖	—	A
87.	21·62	18·96	5·92	4·18	24·40	40·58	10·62¶	2·32–2·37	C
88.	24·14	16·62	4·92	3·82	22·40	40·76	9·00**	—	C
89.	21·86	18·12	6·60	5·00	24·86	39·98	11·60	2·316	A
90.	20·16	19·62	8·18	4·16	25·22	39·78	12·34††	2·24	B
91.	23·10	16·74	5·50	5·96	22·64	39·84	11·66‡‡	—	B
92.	19·80	19·44	9·28	3·68	24·50	39·24	13·10§§	2·405	B
92*a*.	19·68	20·06	8·58	3·80	24·96	39·74	12·52§§		
93.	20·32	19·84	7·34	4·70	23·32	40·14	12·06	2·35	A
94.	20·06	19·94	7·88	4·14	23·62	40·00	12·04‖‖	2·386	A
95.	20·10	19·86	6·94	4·20	23·16	39·96	11·60¶¶	—	A
96.	22·94	17·40	5·88	3·26	23·68	40·34	10·12***	—	B
97.	22·46	17·82	5·98	3·36	24·66	40·28	10·64†††	—	B
98.	20·98	19·92	6·24	3·48	27·96	40·90	10·04‡‡‡	2·3–2·4	C

* Including 0·12 atom K.

† Including 0·28 atom K.

‡ Including 0·04 atom K.

§ 0·79 % Fe_2O_3 not included.

‖ Including 0·26 atom K; 0·23 % free Fe_2O_3 deducted.

¶ Including 0·34 atom K and 0·18 atom Fe··.

** Including 0·24 atom K; 0·81 % free Fe_2O_3 deducted.

†† After deduction of 0·15 % free Fe_2O_3.

‡‡ Including 0·20 atom Mg.

§§ Including 0·12 atom K.

‖‖ Including 0·02 atom K; 0·08 % free Fe_2O_3 deducted.

¶¶ Including 0·66 atom K.

*** Including 0·98 atom K, 0·04 atom Mg.

††† Including 1·30 atom K, 0·04 atom Mg.

‡‡‡ Including 0·32 atom K.

TABLE II (*continued*).

CANADA:

72. T. L. Walker and A. L. Parsons (E. W. Todd, analyst), 1922. Amethyst Cove, King's Co., Nova Scotia. 'Faroelite.'
73. O. C. Marsh, 1882. Cape Blomidon, King's Co., Nova Scotia.
74. H. How, 1858. Port George, Annapolis Co., Nova Scotia. 'Faroelite.'
75. H. How, 1858. Port George. 'Faroelite.'
76. T. L. Walker and A. L. Parsons (E. W. Todd, analyst), 1922. Port George.

UNITED STATES:

77. J. L. Smith and G. J. Brush, 1853. Magnet Cove, Ozark Mts., Arkansas. 'Ozarkite.'[1]
78. W. F. Hillebrand, 1883. North Table Mtn., Golden, Jefferson Co., Colorado.
79. W. F. Hillebrand, 1883. North Table Mtn. A first generation.
80. W. F. Hillebrand, 1883. North Table Mtn. A second generation.
81. W. F. Hillebrand, 1883. North Table Mtn.
82. W. F. Hillebrand, 1885. North Table Mtn.[2] Optics, E. T. Wherry (1923, 1925).
83. F. W. Clarke and G. Steiger, 1902. Table Mtn., Golden, Colorado.[3]
84. H. B. Patton, 1900. North Table Mtn. A second generation.
85. H. B. Patton, 1900. North Table Mtn.[2] A third generation.
86. S. F. Peckham and C. W. Hall (Miss Linton, analyst), 1880. Grand Marais, Lake Superior, Minnesota. 'White lintonite.'
87. S. F. Peckham and C. W. Hall (Miss Linton, analyst), 1880. Grand Marais. 'Green lintonite.'
88. S. F. Peckham and C. W. Hall (Miss Linton, analyst), 1880. Grand Marais. Red and white banded pebbles.
89. G. A. Koenig, 1878. Grand Marais.
90. J. L. Smith and G. J. Brush, 1853. Grand Marais.
91. L. Sperry, 1892. Grand Marais. [see table VII.
92. S. G. Gordon, 1923. Franklin Furnace, Sussex Co., New Jersey. Optics,
92*a*. 92 recalculated, assuming 3·6 % datolite as impurity.
93. F. A. Canfield, 1911. Paterson, Passaic Co., New Jersey. [table VII.
94. A. H. Phillips, 1924. Peekskill, West Chester Co., New York. Optics, see
95. D. F. Hewett, E. V. Shannon, and F. A. Gonyer, 1928. Ritter Hot Spring Grant Co., Oregon. Optics, see table VII.
96. D. F. Hewett, E. V. Shannon, and F. A. Gonyer, 1928. Ritter Hot Spring. Optics, see table VII. Supposed to be pseudomesolite.
97. D. F. Hewett, E. V. Shannon, and F. A. Gonyer, 1928. Ritter Hot Spring. Optics, see table VII. Supposed to be mesolite.
98. F. A. Genth, 1875. Fritz Island, Schuylkill River, Reading, Berks. Co., Pennsylvania. Probably impure material.

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[1] Also an inaccurate analysis by C. U. Shepard (1852).

[2] Hillebrand worked on material from a quarry on the east face of North Table Mtn., Patton on material from a quarry on the south face. Of the author's analyses (nos. 1 and 7, table I), no. 1, judging by the appearance and composition, probably corresponds to Hillebrand's second generation, and no. 7 to Patton's second generation.

[3] Probably from North Table Mtn.

TABLE II (*continued*).

No.	Si.	Al.	Ca.	Na.	H_2O.	Si+Al.	Ca+Na.	Sp. gr.	Class.
99.	20·58	19·88	7·12	3·74	24·44	40·46	10·86	—	C
100.	21·42	18·44	6·58	5·98	22·36	39·86	12·56	—	A
101.	22·42	17·34	5·28	7·32	11·96	39·76	12·82*	—	B
102.	19·52	20·58	7·58	4·58	19·78	40·10	12·62†	—	A
103.	21·92	18·60	6·34	3·58	23·54	40·52	10·24‡	—	C

* Including 0·14 atom Mn, 0·08 atom Fe··; after deduction of 15·68 % $CaCO_3$.

† Including 0·46 atom K. ‡ Including 0·32 atom K.

limits are extended to 99·50 and 101·00 %, and the tolerance[1] in (Si+Al) to ±0·40; 25 have been accounted third-class analyses, including those made on material admittedly somewhat impure, and those with summation or differences outside the above limits; the remaining 20 have been rejected completely, some being made on impure or altered material, others being quite inaccurate, and yet others probably do not really refer to thomsonite.[2] The first three groups of analyses are set out in table II, in atoms per unit cell on a basis of 80 oxygen atoms, the percentages not being reproduced. A graphical representation of both old and new analyses is given in fig. 1, where Ca, (Ca+Na), and (Si+Al) are plotted against Al. Table II is arranged topographically, in order to call attention to the fair agreement shown by analyses of different specimens from the same locality.

This method of classification for zeolite analyses (which will be adopted throughout the present series of studies) is open to several criticisms. The most serious is that it cannot detect contamination by another zeolite, or by any other mineral for which the ratio $O/(Al+Si) = 2$, such as nepheline or felspar. But while this is perfectly true, it applies equally to any other means of classification. On the whole, the method proposed appears to be the best available. All the analyses of generally admitted accuracy, such as those of Hillebrand and Tschermak, fall into the first class, and the division

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[1] In general terms, the tolerance in (Si+Al) is ±0·5 % for first-class and ±1·0 % for second-class analyses.

[2] For these rejected analyses, see J. von Freyssmuth, 1818; T. Thomson, 1820, 1827, and 1836; R. D. Thomson, 1846; W. Hisinger, 1826 and 1838; G. Meneghini, 1852; C. U. Shepard, 1852; G. A. Kenngott, 1853 and 1854; and E. Scacchi, 1888.

TABLE II (*continued*).

WEST INDIES:

99. P. T. Cleve, 1870. Sandy Bay, Anguilla, West Indies.

AUSTRALIA:

100. C. Anderson, 1907. Invernell, Macintyre River, Gough Co., New South Wales.
101. J. C. H. Mingaye, 1916. Prospect quarry, Parramatta, Cumberland Co., New South Wales.

OCEANIC ISLANDS:

102. K. Oebbecke (A. Schwager, analyst), 1881. Rallap, Palau Islands, Pacific.

ARCTIC:

103. H. R. Gaertner and F. Machatschki, 1927. Disko Island, Greenland.[1]

closely coincides with one made independently on general considerations.

Accepting, then, this means of classification, fig. 1 shows that there is a marked tendency towards normality, which would mean a constant number of kations (12) per unit cell, but many analyses show a departure from this relation outside the limits of experimental error (which may be taken as ±0·4 for first-class and 0·8 for second-class analyses), including several of the author's on material definitely tested for purity.[2] Of course, only the latter can be taken unquestionably as evidence of the $Na_2 \rightleftarrows Ca$ substitution, for where there is no evidence of the homogeneity of the material, the departure of (Ca + Na) from 12 might be due to admixture of another zeolite, which, as already mentioned, would not disturb the (Si + Al) figure. But since the new analyses were made on material optically tested for impurity, there appears no reason why we should assume impurity without distinct reasons. (Such reasons would be the discovery that other specimens from the same locality were mixtures, or bad agreement between analyses of material from the same locality.) Classifying all the analyses by localities (table II) we find *normal* material constitutes only about half the occurrences.

Summing up, we may say that thomsonite is an alumino-silicate, with an infinite tridimensional anion, $\{(Si,Al)O_2\}_n$, in which Al replaces Si to a variable extent, from about 20·4 to 16·0 atoms Al and 19·6 to 24·0 atoms Si per unit cell of 80 oxygen atoms. The number of kations per unit cell is also variable, but shows a strong

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[1] It should be noted that some 'mesole' from this island has proved to be gyrolite (M. F. Heddle, 1901; O. B. Bøggild, 1908).

[2] Definite tests for purity are also mentioned in respect of analyses 32, 58, 81, 82, 94, 95, 96, and 97.

tendency towards a constant integral value, 12. This constancy is maintained by a replacement of type $NaSi \leftrightarrows CaAl$, and it is fully realized in the *normal* thomsonites in an appreciable number of occurrences.

The X-ray data suggest that the thomsonite unit is essentially $Na_4Ca_8Al_{20}Si_{20}O_{80}.24H_2O$, in which about 4(CaAl) may be isomorphously replaced by 4(NaSi). On this isomorphous replacement may be superimposed one of Na_2 by Ca or of Ca by Na_2. A similar case is that of nepheline,[1] where the unit is essentially $8(NaAlSiO_4)$, but there is almost invariably a noticeable replacement of NaAl by Si.

Crystallography, X-ray study, and density.

Owing to the rarity of well-crystallized material, the crystallography of thomsonite has been relatively little studied. H. J. Brooke (1820) found $mm' = 89° 20'$ for the Old Kilpatrick material, and Sir David Brewster (1821) 89° 13′ for Vesuvian comptonite, but the only accurate studies are those of W. C. Brøgger (1878, 1879, and 1890) on material from Låven, Norway, and of F. Zambonini (1908) on Vesuvian material.[2] The system is orthorhombic, and the accepted axial ratio (after Brøgger), $a : b : c = 0{\cdot}9932 : 1 : 1{\cdot}0066$.

Owing to lack of suitable material the author has been able to add no more accurate goniometric measurements, but he has been able to show that the material examined by V. von Lang (1863) and figured in most text-books (e.g. Dana, 6th edit., 1892, p. 600, fig. 3) as natrolite of an unusual habit is actually thomsonite. The specimen von Lang studied is identified beyond question as B.M. 34108, and consists mainly of massive fibrous natrolite, with natrolite crystals of the usual habit in the cavities; but on one side is a small cavity lined with small thomsonite crystals,[3] of the same habit (*a b c m y r x p*, Dana's letters) as those studied by Brøgger from Låven. Measurements gave results agreeing with von Lang's figures, but the latter did not observe the plane *x* on the crystals he studied. The correct orientation is : *a* of von Lang = *c* of Brøgger, $b = b$, $c = a$, $h = r$, $a = m$, $n = y$, $z = p$.

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[1] F. A. Bannister, Min. Mag., 1931, vol. 22, p. 569.

[2] For other measurements see H. J. Brooke (1822), W. Phillips (1837), J. D. Dana (1837), R. P. Greg and W. G. Lettsom (1858), O. Luedecke (1877), A. Hahn (1891), O. Bøggild (1909), and P. C. Habert (1897).

[3] Microchem., found Ca, Na; Ax. pl. and $Bx_a \perp$ prism, elongation ±, 2E 80–90°, +, $n > 1{\cdot}525$.

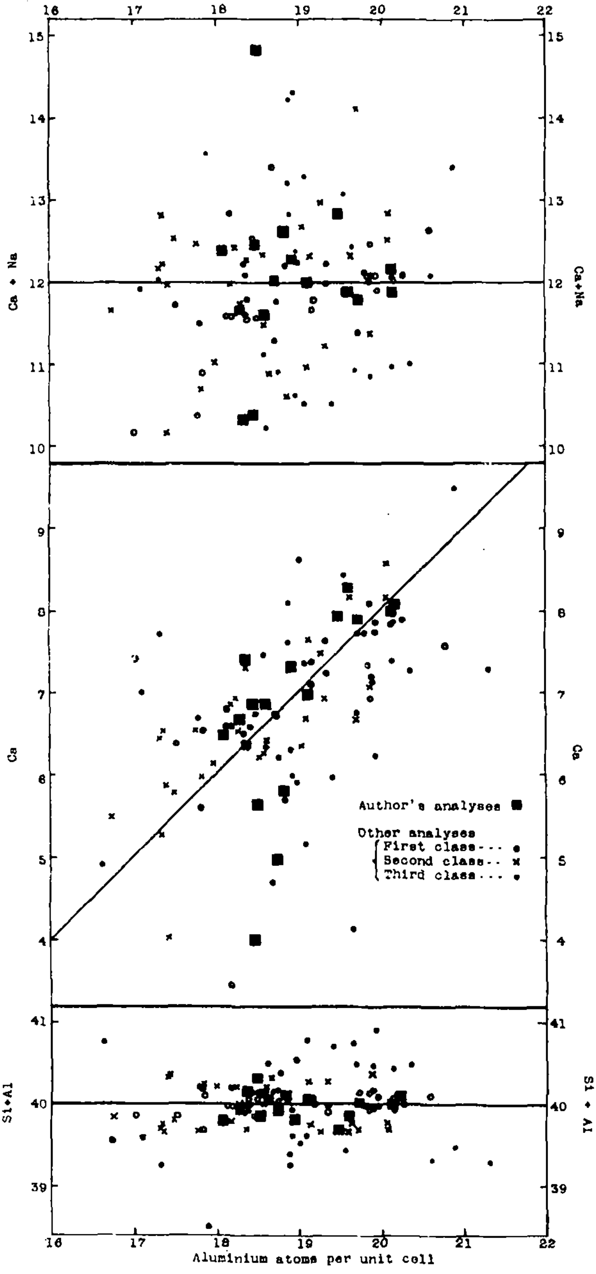

FIG. 1. A graphical representation of the chemical data for thomsonite. Against the number of aluminium atoms per unit cell there are plotted the number of calcium atoms per unit cell, the number of kations per unit cell, and the number of silicon plus aluminium atoms per unit cell.

Thomsonite has a perfect cleavage parallel to *b*(010) and a good one parallel to *a*(100). In the faroelites, the facility of these cleavages becomes reversed. According to M. F. Heddle (1858), faroelite has also a third cleavage, which he took as *m*(110), at 63° 40′ to *a* ; this should clearly be indexed (120).

The habit of thomsonite is in general prismatic parallel to the *c*-axis, the more silica-rich material only occurring as fine fibres, with the exception of one occurrence on Table Mtn., Colorado, where it forms a confused aggregate of small thin plates[1] parallel to *b*(010). The silica-poor material generally crystallizes as short prisms, or as plates tabular to *b*(010) with a remarkable development of planes vicinal to *b* and *a* : the base *c*(001) appears to be always replaced by a pair of vicinal planes at about 2½° to one another. But at Old Kilpatrick and Oberstein we find radiating bundles of long, fairly stout (1–3 mm.) prisms, which are rarely free.

According to A. Des Cloizeaux (1862), thomsonite is pyro-electric, though he gives no details ; it would follow that it cannot belong to the holohedral class of the orthorhombic system (D_{2h}), but must be either digonal holoaxial or didigonal polar (D_2 or C_{2v}). Tests by Kundt's or Brewster's method on a long prism of the Old Kilpatrick material gave indefinite results, but Dr. W. A. Wooster kindly undertook to investigate the matter. I supplied him with three plates of the Old Kilpatrick material (no. 15, B.M. 94985) cut perpendicular to the three principal axes, which had been soaked in water for a month to restore any water that might have been lost during grinding and polishing. The same plates were used in the optical studies; they were about 1 mm. thick and 2–4 mm. across. They proved too small for piezo-electric study, but Dr. Wooster successfully demonstrated a distinct, though small, pyro-electric effect, on which he reports as follows.

'. . . The electric axis of thomsonite is the *b*-axis. The method used was as follows. The crystal was suspended in liquid air by a long thin glass fibre, and, when cold, a metal plate, also immersed in the liquid air, was made to approach the crystal. If an electric charge developed by the cooling is present on the crystal then it is, of course, attracted to the conductor, and by the movement of the fibre the existence of a pyro-electric effect can be found.' This

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[1] W. Cross and W. F. Hillebrand, 1882. One of the analysed specimens, no. 1 (B.M. 1914, 1077) is very similar. Cross and Hillebrand also observed the dome *r*(101) on these crystals.

ingenious method, which was demonstrated to this Society on March 17, 1931, by A. J. P. Martin (Min. Mag., 1931, vol. 22, p. 519), ensures that there will be no disturbance by effects due to dehydration.

This result definitely assigns thomsonite to the didigonal polar class (C_{2v}), with the *b*-axis as the polar axis. Attempts to gain confirmatory evidence from etch-figures met with no success, nor does the facial development offer any assistance, but this symmetry is in agreement with the X-ray data, as found by J. Wyart (1931) and by W. H. Taylor.[1] Both these workers find that the halving of certain spacings (namely, ($hk0$) for ($h+k$) odd and ($0kl$) for ($k+l$) odd) leads to a choice of two possible space-groups, C_{2v}^{10} and D_{2h}^{12}. If the former is correct, then the *b*-axis is the polar symmetry-axis, in agreement with the pyro-electric data. Thomsonite may therefore safely be referred to the space-group C_{2v}^{10}. Dr. Taylor's work is still in progress, and he hopes to find the parameters of the various atoms in the structure. Mr. F. A. Bannister's X-ray studies have been directed to a different end, the tracing of any variation in the X-ray photographs with the chemical composition, but his photographs are all in agreement with the above space-group.

Mr. Bannister's X-ray measurements of the cell-sides, included in table I,[2] show that they, and the axial ratios, are practically constant throughout the range of Si/Al-ratios studied. The X-ray data indicate that Brøgger's axial ratio is not exact, owing probably to the development of vicinal faces. The true axial ratio, for Old Kilpatrick material (Al 20·12 atoms per unit cell), appears to be $a:b:c = 1{\cdot}000:1:1{\cdot}011$ and does not vary measurably with composition. No departure of the ratio a/b from unity could be detected,[3] but the different intensities of *a* and *b* reflections excludes tetragonal symmetry, without need for an appeal to optical or other evidence. This is not in exact agreement with J. Wyart's measurements; he found *a* 13·02, *b* 13·14, *c* 13·22 Å., all ±0·06, the axial ratio $a:b$ being 0·9901 : 1. The difference is, however, just on the limits of Wyart's

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[1] Private communication. The material used by Dr. Taylor was from the same specimen (no. 15, B.M. 94985) from Old Kilpatrick as was analysed and employed for most of the present work.

[2] Cell-sides of thomsonite associated with gonnardite, see table XV.

[3] A direct comparison of *a* and *b* was obtained for four orders between the 6th and 16th by two rotation photographs, one about an axis in the plane (100) and tilted 5° to the axis [001], and one similarly placed in the (010) plane, giving the *a* and *b* diffractions on the equatorial line respectively.

experimental error, and he does not appear to have considered the question of this axial ratio particularly.

X-ray work at first suggested that the true c-axis should be half the length assumed by Brøgger. Oscillation and rotation photographs of long exposure, however, show that no major alteration in the axial ratio as usually accepted is necessary. On these photographs, all taken about the c-axis of single crystals from no. 15, several first and third layer-line diffractions corresponding to the type (hkl), where l is 1 or 3, are now found to be definitely present but extremely weak. Their presence on the single crystal photographs of thallium-thomsonite and dehydrated thomsonite can also be observed, although these photographs were not of long exposure. It is impossible to be sure whether these weak reflections are present on fibre photographs, but the true unit cell probably contains eighty oxygen atoms for all the thomsonites examined.

From a review of the relative cell-volumes derived from the chemical and specific gravity data on the assumption that there are exactly eighty oxygen atoms per unit cell, it appears probable that there is a small increase in the cell-volume with increase of silica, amounting to about ½ % per additional Si per unit cell; this is just on the limit of experimental error as regards the X-ray measurements.

If we assume that this is the true variation in cell-volume, we obtain the specific gravity curve shown in fig. 2. It will be seen that many of the older values, especially for silica-rich material, are low compared with the present curve; there can be little doubt that this is partly due to the finely fibrous nature of such material, and the consequent difficulty of entirely avoiding inclusions of air. There is no evidence of any effect of $Na_2 \rightleftarrows Ca$ substitution on the density, but special experiments have shown that dehydration lowers it by about the amount that would be expected if there is no lattice-shrinkage. If the temperature has been allowed to exceed about 270° C., lattice-shrinkage sets in and higher densities are found. The reality of lattice-shrinkage Mr. Bannister has demonstrated by X-ray measurements (compare below, under dehydration).

The X-ray work was first directed towards measuring the edges of the true unit-cell as accurately as the material available would permit. These measurements were carried out upon analysed material found to differ most in chemical composition and density. An interesting fact that emerged early on in the course of this work was the impossibility of isolating single crystals of thomsonite from

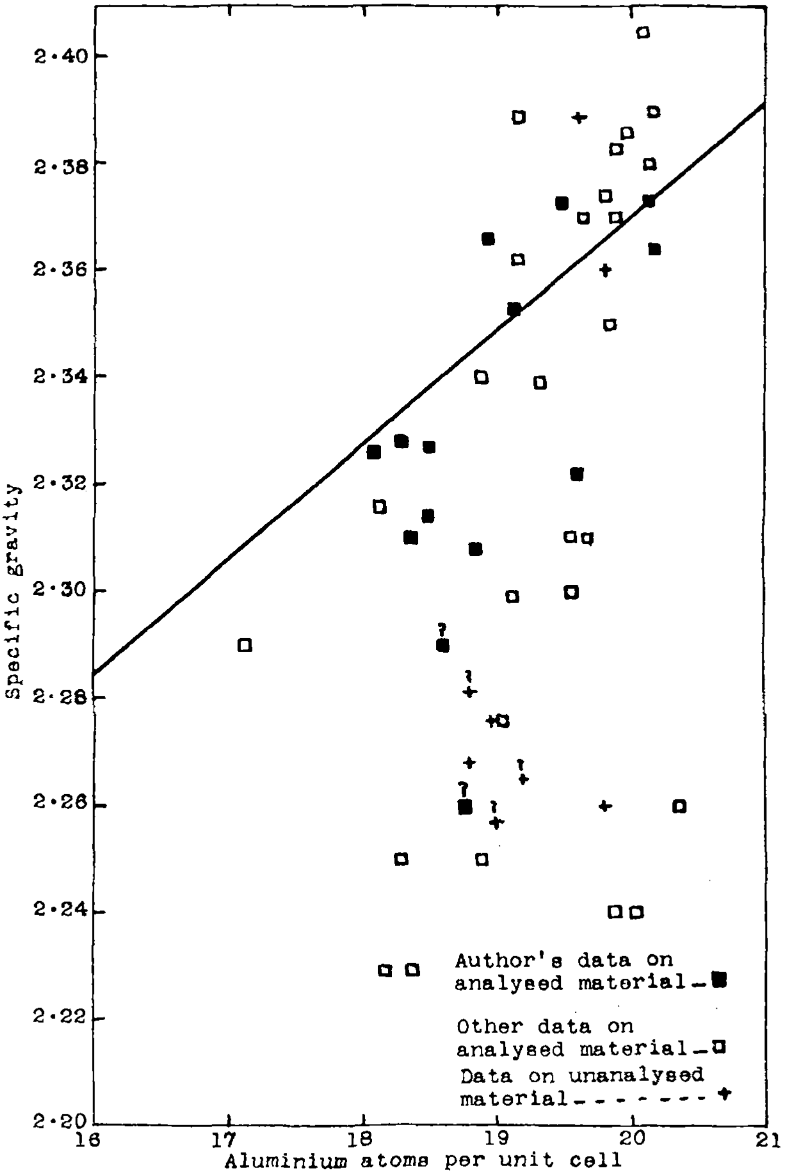

FIG. 2. The variation of the specific gravity of thomsonite with its composition. The composition of the unanalysed specimens estimated by the aid of fig. 4.

the greater number of specimens examined. Only such material as is found by chemical analyses to have a low silica content offers single crystals suitable for investigation by the oscillation or rotation methods (e.g. no. 15, Old Kilpatrick). Several small plates selected from no. 1 (Table Mountain), although giving normal optic pictures perpendicular to the *b*-face, do not yield Laue photographs typical for thomsonite along the *b*-axis. Each plate yields a fibre photograph with well-defined layer-lines, and measurements from these indicate that the apparent plates are small sheaves of sub-parallel crystals, the extreme members having their *a*-faces at about 2° and their *b*-faces at about 6° to one another.

The other specimens examined, viz. no. 5 (Salesl) and no. 3 (Goble) also yielded only fibre photographs. The results indicate that the unit-cell dimensions vary only within the errors of measurement and it has not been possible to trace any certain relationship between this variation and change in chemical composition. The fibre and powder photographs of the various specimens investigated were all indexed carefully with a view to tracing also any change in the intensities of spots. A direct comparison between the fibre photographs and the single crystal rotation photograph about the *c*-axis of the Old Kilpatrick material could only be effected for the equatorial diffractions (*hk*0), for which the coincidence was perfect. No intensity changes were observed on any of these photographs. It must be remembered, however, that such intensity comparisons to be of any value in structure work should be quantitative and carried out upon single crystals.

X-ray measurements have also been carried out upon dehydration and base-exchange products of thomsonite. The parent material in each case was Old Kilpatrick material no. 15 (B.M. 94985), and where possible rotation photographs of single crystals of the products were taken in order to make direct comparison of spacings and intensities possible. The most interesting results were obtained with the thallium product which contained small undistorted crystals giving good photographs. The cell-dimensions appear to be unaltered, and although chemical analysis of the thallium derivative[1] shows that 10 % of the heavy metal has replaced 1 % of sodium the intensity changes capable of visual observation are restricted to the appearance of the (103) and (114) spots, the disappearance of the (112) spot, and a few other minor changes in intensity

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[1] See table XIII below.

TABLE III. Indices and intensities of spots on a rotation photograph about the *c*-axis of a crystal of Old Kilpatrick thomsonite (no. 15). Cell-sides *a* 13·09, *b* 13·09, *c* 13·23 Å.

First two indices (*hk*).	Third index, *l*,				First two indices (*hk*).	Third index, *l*,			
	0.	2.	4.	6.		0.	2.	4.	6.
10	–	*s*	*vs*	–	61	–	*vvw*	*vvw*	*w*
11	–	*w*	–	–	62	*m*	*w*	*w*	*m*
20	*s*	*s*	*vs*	–	54	–	*w*	–	*vw*
21	–	*s*	*vw*	–	63	–	*vw*	*m*	*w*
22	*vs*	*vw*	*vvw*	*w*	70	–	*m*	*vvw*	*vvw*
30	–	–	–	*w*	71 55	*vvw*	*m*	*s*	*w*
31	*s*	*vs*	*s*	*w*	64	*s*	*w*	*vw*	*vvw*
32	–	*s*	*vw*	*m*	72	–	*w*	*vw*	*vw*
40	*m*	*w*	*vw*	*m*	73	*s*	*vvw*	–	*vvw*
41	–	*vs*	*w*	*m*	65	–	*m*	*w*	*vvw*
33	*vvw*	*w*	*s*	–	80	*w*	*w*	*s*	*m*
42	*m*	*vs*	*s*	*w*	81 74	–	*m*	*w*	*m*
43 50	–	*m*	*vvw*	*w*	82	*w*	–	*vvw*	*vvw*
51	*w*	–	*vw*	*w*	66	*vw*	*vvw*	*vvw*	–
52	–	*vw*	*m*	*m*	83	–	–	*vvw*	–
44	*vw*	–	–	–	75	*vvw*	*vw*	*vw*	–
53	*w*	*w*	–	*m*	84	*s*	*w*	*w*	*vw*
60	*vs*	*vvw*	*vw*	*w*					

Notes:—Indices of the following types are all very weak: (*hk*1), (*hk*3), and (*hk*5), but the following could be indexed with certainty: (221) *vvw*, (301) *vvw*, (311) *vw*, (321) *vvw*, (551) or (771) *vvw*, and (113) *vvw*.

Cu—K_α radiation was used with a nickel filter. *s* = strong, *m* = medium, *w* = weak, *vs* = very strong, &c.

A series of oscillation photographs would be necessary to distinguish (*hkl*) from (*khl*) reflections; the intensities given above all correspond to the sum of both diffractions.

of diffraction from planes of low indices recorded in table IV. Planes of high indices do not appear to be affected. Silver-thomsonite did not yield sufficiently uniform material to render the powder photograph obtained very reliable for intensity comparison. No obvious changes could be detected. Old Kilpatrick thomsonite dehydrated at 270° C. and 400° C. respectively, both yielded identical rotation photographs. The cell-height *c* remains unaltered, but the *a* and *b* spacings have shrunk to 12·92 Å. Intensity changes again are restricted to planes of low indices (see table IV), the most noteworthy being the disappearance of (112), the appearance of (103), and the strengthening of (750) and (550) or (710). In all the photographs of these products the first and third layer-lines are very weak but definitely present.

TABLE IV. Indices and intensities of spots on rotation photographs about the *c*-axis of (*a*) a crystal of thallium-thomsonite, and (*b*) a partially dehydrated crystal of thomsonite.

(*a*) Thallium-thomsonite. Cell-sides *a* 13·04, *b* 13·04, *c* 13·23 Å.

(220)	*s*	(432) (502)	*w*	(424)	*m*
(400)	*w*	(522)	nil	(434) (504)	nil
(510)	*vw*	(532)	*vw*	(226)	*vvw*
(440)	nil	(602)	nil	(306)	*vvw*
(530)	*vw*	(612)	nil	(316)	*vvw*
(600)	*s*	(103)	*m*	(326)	*w*
(640)	*vs*	(104)	*s*	(406)	*w*
(321)	*vvw*	(114)	*m*	(426)	*vw*
(102)	*w*	(204)	*s*	(436) (506)	*vw*
(112)	nil	(214)	nil	(516)	nil
(202)	*w*	(224)	nil	(526)	*w*
(212)	*w*	(314)	*m*	(536)	*w*
(222)	nil	(324)	nil	(616)	*vw*
(312)	*m*	(334)	*m*	(626)	*w*

(*b*) Partially dehydrated thomsonite. Cell-sides *a* 12·92, *b* 12·92, *c* 13·23 Å.

(310)	*m*	(640)	*vs*	(112)	nil
(400)	*s*	(730)	*vs*	(312)	*m*
(620)	*w*	(750)	*w*	(322)	*m*
(710) (550)	*w*	(211)	*vvw*	(103)	*vw*
		(311)	*w*	(113)	*w*

The above intensities include all spots whose intensities appear appreciably different from those of untreated thomsonite (table III). All other spots have nearly the same intensities in all three photographs. As in table III, the intensities correspond to the sum of (*hkl*) and (*khl*) diffractions.

The changes consequent on partial dehydration, though small, indicate that the view of G. Tammann, F. Rinne, E. Löwenstein, and W. H. Taylor,[1] that the zeolites are mixed crystals of a hydrate and an anhydrous substance, the water having definite lattice positions, is probably correct. But careful measurements of intensities of X-ray diffractions, made on a zeolite the structure of which has been fully worked out, will be needed finally to elucidate this point. For on the view of E. Mallard, G. Friedel, F. M. Jaeger, W. Hartwig, and others[1] that the water is merely adsorbed, and holds no definite lattice positions, its presence or absence must still influence the relative intensities of different diffractions, since it

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[1] Literature, see part I of this series (loc. cit.), and also F. M. Jaeger, Proc. Sect. Sci. Akad. Wetensch. Amsterdam, 1927, vol. 30, p. 479; Trans. Faraday Soc., 1929, vol. 25, p. 334; and W. Hartwig, Zeits. Krist., 1931, vol. 78, p. 173.

cannot strictly be distributed uniformly, but only in the space unoccupied by other atoms. This factor has been overlooked by W. Hartwig (loc. cit., p. 188) in his discussion of the structure of analcime; the removal of the water *must* influence the relative intensities, though the effect may be very small.

Accepting J. Wyart's (1931) space-group determination, it is readily possible to build up a structure on the lines of that proposed for natrolite by L. Pauling,[1] which shall accommodate the Si and Al, Ca and Na atoms of the formula $Na_4Ca_8Al_{20}Si_{20}O_{80}.24H_2O$ in structurally different positions and agree with the results obtained in the study of the absorption of alcohol vapours by partially dehydrated thomsonite; it also fits in with the optical properties of thomsonite along the lines of W. A. Wooster's [2] data on birefringence and its relation to crystal-structure. But the publication of such speculative structures can serve no useful purpose; moreover, W. H. Taylor hopes to work out the structure from intensity measurements.

Optics.

The optical properties of thomsonite were first studied by Sir David Brewster (1821), who wrote of the Vesuvian material:

> Comptonite has two axes of double refraction, one of which is parallel, and the other perpendicular, to the axis of the prism. It consequently gives the double system of coloured rings. The *inclination* of the resultant axes, or diameters of no polarisation, is nearly 56°; or they are inclined about 28° each to a line at right angles to the faces *a*, *e* [*b*, *b′*], which is the *principal axis* of the crystal. The action of this axis is *positive*, like that of topaz. The plane passing through these resultant axes is perpendicular to the axis of the prism. The index of refraction is 1·553, when the ray passes through *a* and *f* [*b* & *m′*], and no separation of the images can be seen; but through *a* and *mn* [*b* & *c*], the images may be distinctly separated.

Or in modern nomenclature: Biaxial, optic axial plane parallel to $c(001)$, $Bx_a \perp b(010)$, positive, 2V 56° approx., n 1·553, $\gamma - \alpha$ moderate, $\beta - \alpha$ not noticeable. These results have been fully confirmed by subsequent investigators apart from small numerical corrections. A. Des Cloizeaux (1862) found by an indirect method, α 1·497, β 1·503, γ 1·525 for Kaaden material, values as much too low as Brewster's were too high. The first accurate determination of the refractive indices of a thomsonite (A. Scheit, 1912) is comparatively recent; that the indices

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[1] L. Pauling, Proc. Nat. Acad. Sci. U.S.A., 1930, vol. 16, p. 453.

[2] W. A. Wooster, Zeits. Krist., 1931, vol. 80, p. 495.

are variable was first noticed by S. G. Gordon (1923), while A. H. Phillips (1924) implies that the variation is connected with the Ca/Na-ratio, the indices increasing with increase in lime. The author's results clearly show that this is not correct. The refractive indices vary with the Si/Al-ratio, increasing with increase in Al, but are little affected by replacement of Ca by Na_2 or vice versa. In *normal* thomsonites, of course, the refractive index does follow the calcium content, since there is a replacement of NaSi by CaAl, but only by plotting the refractive index against the number of aluminium atoms per unit cell can we get good agreement for all the specimens. So plotted (fig. 4), the agreement is very good except with a few specimens where the replacement of Ca by Na_2 (or vice versa) has been very considerable, such as no. 5 (B.M. 1918,107); these show evidence that the refractive index is increased by this replacement, by about 0·0025 per calcium atom (per unit cell) replaced.

Only one of the analysed specimens was suitable for a detailed optical study by precision methods, namely the Old Kilpatrick material no. 15 (B.M. 94985). From one of the long prisms from this specimen two 60°-prisms and three plates (perpendicular to the three principal axes) were cut, and soaked in water for a month, to restore any water lost during the grinding and polishing. With these, measurements were made of the refractive indices, optic axial angles, and birefringences for light of a series of wave-lengths (isolated by a monochromator). The results are combined in table V, and shown graphically in fig. 3. An attempt to fit a three- or four-constant dispersion formula to the refractive index curves gave no satisfactory results. Perhaps better success will be obtained when the dielectric constants of thomsonite have been measured. A determination of these constants for the zeolites should furnish results of considerable interest, but unfortunately could not be undertaken. Combining the experimental results for refractive index, birefringence, and optic axial angle, we conclude that the birefringences remain practically constant (for Old Kilpatrick material) at 0·00252 $(\beta-\alpha)$ and 0·0134 $(\gamma-\alpha)$ throughout the range of wave-lengths studied.

The remainder of the analysed specimens were not sufficiently well crystallized to furnish prisms and plates. The refractive indices were therefore measured for sodium-light by the immersion method (accuracy about ± 0·003). The optic axial angles were determined, where possible, on a crystal attached to a cover slip with celluloid varnish, and immersed in a liquid of refractive index equal to the

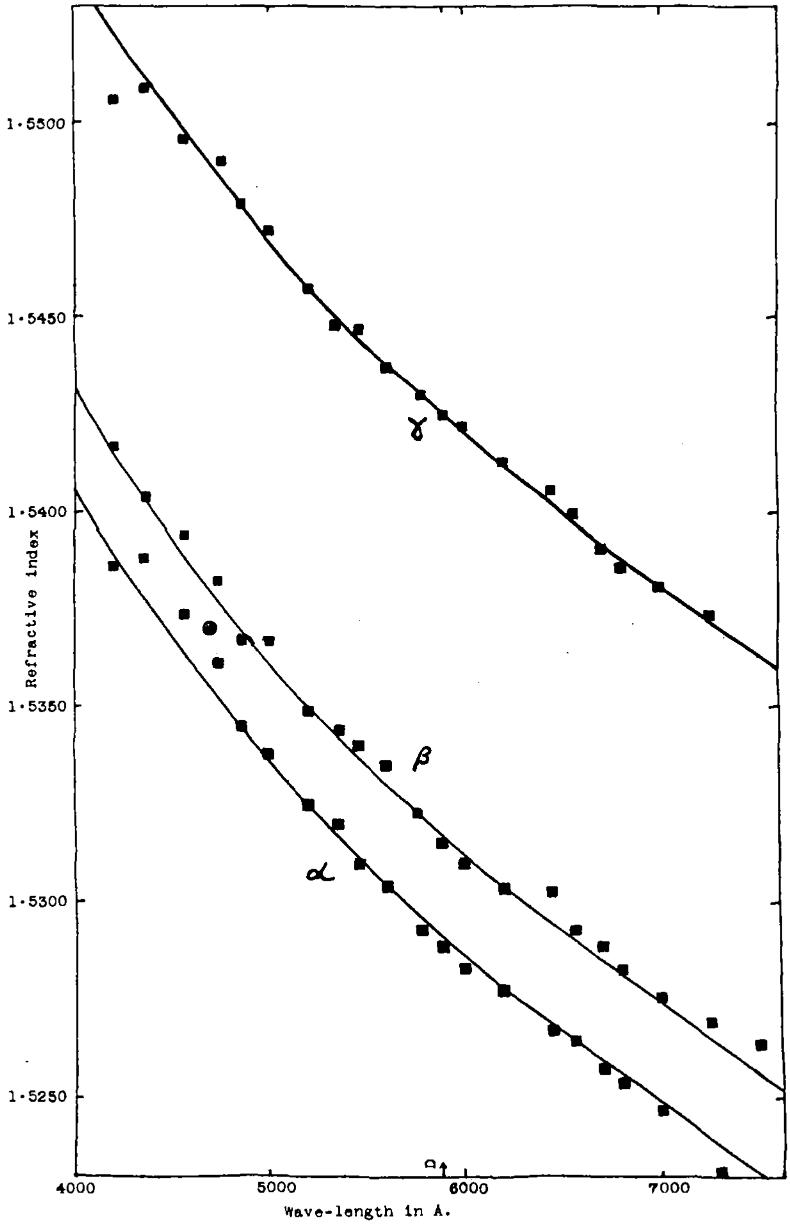

FIG. 3. The variation of the refractive indices of thomsonite (from Old Kilpatrick) with the wave-length of the light.

TABLE V. Optical data for the Old Kilpatrick thomsonite, analysis no. 15 (B.M. 94985).

Wave-length (in Å).	α.	β.	γ.	$\gamma-\alpha$.	$\beta-\alpha$.	2V.
7500	—	1·5264	—	—	—	—
7250	1·5230	1·5270	1·5374	0·0144	—	—
7000	1·5247	1·5276	1·5381	0·0134	—	—
6800	1·5254	1·5283	1·5386	0·0132	—	—
6708 (Li)	1·5258	1·5289	1·5390	0·0132	—	51° 18′
6563 (H)	1·5265	1·5293	1·5400	0·0135	—	—
6438 (Cd)	1·5268	1·5303	1·5406	0·0138	—	—
6200	1·5278	1·5304	1·5413	0·0135	—	—
6000	1·5283	1·5310	1·5422	0·0139	—	—
5893 (Na)... ...	1·5289	1·5314	1·5425	0·0136	—	51° 39′
5780 (Hg yellow)...	1·5293	1·5323	1·5430	0·0137	—	—
5600	1·5304	1·5335	1·5437	0·0133	0·00251	—
5461 (Hg green) ...	1·5310	1·5340	1·5447	0·0137	—	—
5350 (Tl)	1·5321	1·5344	1·5448	0·0127	—	51° 42′
5200	1·5325	1·5349	1·5457	0·0132	—	—
5000	1·5338	1·5367	1·5472	0·0134	—	—
4861 (F)	1·5345	1·5367	1·5479	0·0134	—	—
4750	1·5361	1·5382	1·5490	0·0129	—	51° 58′
4555 (Cs)	1·5374	1·5394	1·5496	0·0122	—	—
4356 (Hg violet) ...	1·5388	1·5404	1·5508	0·0120	0·00259	—
4200 (Rb)... ...	1·5386	1·5417	1·5506	0·0120	—	52° 47′

Determinations of the refractive indices were made with two prisms, one giving γ and β and the other γ and α. Differences for the two values of γ never exceeded 0·0008, and averaged 0·0004. The birefringence $\beta-\alpha$ and the optic axial angle 2V were measured directly.

index β of the crystal. With several specimens, single crystals could not be isolated for this purpose, fine fibres which appeared under the microscope to be single proving aggregates with parallel *c*-axes.

The optical data thus obtained for the analysed specimens are included in table I, while the optical constants and specific gravities of a number of unanalysed specimens are given in table VI. Neglecting the values given by H. Michel (1911) and A. Ondřej (1921), who appear merely to have copied Des Cloizeaux's values, the optical data available from the literature[1] is given in table VII.

Since it has been found that the refractive indices of thomsonite vary with the Si/Al-ratio, it is possible to deduce the approximate number of aluminium atoms per unit cell from the mean refractive

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[1] Isolated data (optic axial angles and birefringences) are also given by Sir David Brewster, 1821; A. Des Cloizeaux, 1857, 1862, 1867; V. von Lang, 1857; A. Lacroix, 1886; A. Hahn, 1891; L. Gentil, 1895; G. Cesàro, 1907, 1908; and J. E. Hibsch and A. Scheit, 1911.

index, provided there is not too extensive a $Na_2 \leftrightarrows Ca$ substitution. Fig. 4 provides a graphical means for this estimation, which has been applied to all the unanalysed specimens included in tables VI and VII; the aluminium contents deduced are given in the tables, together with any analytical values obtained on other specimens from the same locality. The generally good agreement between the aluminium contents deduced from the refractive indices and those found analytically on other specimens from the same locality adds further confirmation of the correctness of the refractive index graph.

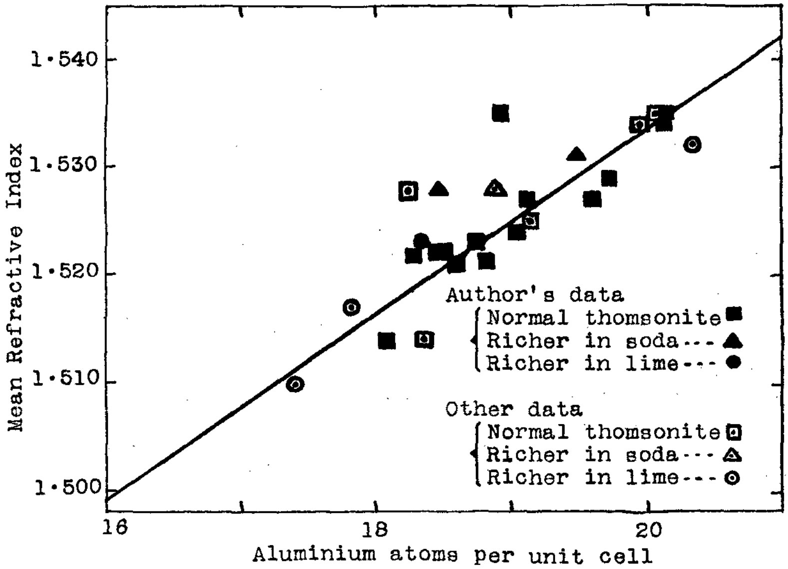

FIG. 4. The variation of the mean refractive index, $(\alpha+\beta+\gamma)/3$ (for sodium-light), of thomsonite with the chemical composition.

From the new results shown in table I, the mean refractive index curve for sodium-light, fig. 4, was first drawn. The birefringence curves, fig. 5, were then drawn, utilizing the accurate measurements for the Old Kilpatrick material to fix one point in each line; the other birefringence data on analysed material (mostly very approximate) were next plotted, and the further course of the lines (assumed straight) among the points estimated as well as possible, with the aid of the few optic axial angle measurements available. A quantity of birefringence data for unanalysed material was then incorporated, by means of the mean refractive index and fig. 4; the agreement

[*continued on* p. 84

TABLE VI. New optical data on thomsonite. Unanalysed material, arranged by localities; data by the immersion method, accuracy about ± 0·003. With estimates of the probable composition, Al (number of aluminium atoms per unit cell), made by means of fig. 4. For comparison, where other material from the same locality has been analysed, the analytically determined Al-figure (Al′) is also given.

Locality and British Museum No.		α.	β.	γ.	Sp. gr.*	Al.	Al′.
FAEROE ISLES:	B.M.						
Hestö	32681	1·518	1·520	1·525	—	18·6	—
Eide, Osterö	1907,765	1·521	1·523	1·530	—	19·0	—
Dalsnypen, Sandö ...	1918,46	1·523	1·525	1·530	2·27	19·2	18·92
Nolsö	27377	1·521	1·524	1·528	2·17	18·9	18·5†
Nolsö	86707	1·521	1·523	1·528	2·28	Anal. 24	
NORWAY:							
Låven (= Lamö), Langesundfjord ‡	34108	1·526	1·528	1·536	—	19·6	—
GERMANY:							
Oberstein, Oldenburg ...	1916,71	1·528	1·531	—	—	20·2	—
IRELAND:							
Co. Antrim. 'Scoulerite'	60060	1·514	—	—	2·10	18·0	—
Co. Antrim. 'Antrimolite'	20056	1·522	1·524	—	—	19·0	—
Collinward, Belfast ...	1909,408	1·514	1·516	1·520?	—	18·0	—
SCOTLAND:							
Old Kilpatrick. 'Gibsonite'	95488	1·527	1·529	1·535	—	19·6	—
Hartfield Moss, Renfrewshire. 'Gibsonite' §	95485	1·524	1·526	1·532	—	19·2	—
Isle of Skye	95003	1·523	1·526	1·533	—	Anal. 36	
Isle of Skye	86651	1·521	1·524	1·528	—	18·9	—
Storr Mt., Isle of Skye	95004	1·518	1·520	1·526	—	18·6	—
FRANCE:							
Chaux de Bergonne, Gignat, Puy-de-Dôme ‖ ...	1930,166	1·514	1·516	1·525	—	18·2	—
ITALY:							
Cyclopean Isles, Sicily	94992	1·522	1·524	1·535	—	19·2	19·08

* These values are not very accurate, the material being too finely fibrous.

† Mean of several analyses ranging from 17·84 to 19·04.

‡ Thomsonite crystals measured by V. von Lang as natrolite.

§ A new locality for thomsonite.

‖ A thin layer on gonnardite. 2V 62° approx. Several other specimens (B.M. 83037, 1908,372, 1918,58) all give the same results.

TABLE VI (*continued*).

Locality and British Museum No.		α.	β.	γ.	Sp. gr.*	Al.	Al .
BOHEMIA:							
Neubauerberg, Böhmisch-Leipa ...	B.M. 1918,338	1·521	1·523	1·529	2·28	18·8	17·10
Steinberg, Schlössl, Hammer-Unterwiesenthal†	1914,1534	1·522	1·524	1·530	—	19·0	—
Böhmisch-Leipa ...	94991	1·523	1·525	1·528	2·25	18·8	—
Jakobs-Bergel, Rübendörfel, Leitmeritz ...	1916,487	1·524	1·526	1·533	—	19·4	—
Malik's quarry, Radobil, Leitmeritz ...	1916,486	1·526	1·529	1·541	—	19·4	—
Kamnitz. 2V 60° appr.	28013	1·526	1·530	1·540	2·360	19·8	—
Wostray Hill, Schreckenstein ...	1919,23	1·528	1·530	1·540	—	20·0	—
Eulenberg, Schüttenitz	1914,428	1·529	1·532	1·542	—	20·1	20·14 19·72
INDIA:							
Sirur, Ahmadnagar district, Bombay ‡ ...	94976	—	1·525	—	—	19·4	—
JAPAN:							
Maze, Echigo §... ...	1918,204	—	1·521	1·525	—	18·6	—
NORTHERN RHODESIA:							
Palm Grove, Victoria Falls §	1929,1706	—	--	1·518	—	18·0	—
CANADA:							
Amethyst Cove, Nova Scotia	1924,1149	1·521	1·523	1·527	2·27	18·9	19.10
Johnson Asbestos mine, Thetford mines, Quebec	1926,106	1·520	1·523	1·532	—	19·0	—

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* These values are not very accurate (except that for Kamnitz), the material being too finely fibrous.

† There is some confusion regarding this locality, which is often given as Hammer-Unterwiesenthal, Annaberg, Saxony; actually, Hammer-Unterwiesenthal is in Saxony, but Schlössl is in Bohemia, and the zeolite occurrence would also appear to be on the Bohemian side of the frontier.

‡ This specimen is mentioned by F. R. Mallet (Man. Geol. India, Calcutta, 1887, part 4, p. 119). A re-examination shows that it consists mainly of compact fibrous okenite, on which are scattered small spherules of faroelite and a few crystals of epistilbite. The locality is a new one for okenite and epistilbite. The okenite, which has refractive indices α 1·531, γ 1·536, was analysed, and gave: SiO_2 53·18 %, Al_2O_3 1·81, CaO 27·58, H_2O 17·51, total 100·08. At 100° C., 5·08 % water was lost, and at 300° C., 13·40 %.

§ A new locality for thomsonite.

TABLE VI (*continued*).

Locality and British Museum No.		α.	β.	γ.	Sp. gr.	Al.	Al′.
CANADA (*continued*):							
Bagot, Ontario. 'Bagotite'	54508	—	1·525	—	—	19·4	—
UNITED STATES:							
Franklin Furnace, New Jersey	B.M. 1918,285	—	—	1·541	—	20·2	20·06
Grand Marais, Lake Superior. Green 'Lintonite' ...	53910, 53912	—	1·525	—	—	19·4	—
AUSTRALIA:							
Flinders, Melbourne ...	83472	—	1·526	—	—	19·0	—
OCEANIC ISLANDS:							
Kerguelen Island, Indian Ocean	50324	1·516	1·518	1·524	—	18·3	—

TABLE VII. Optical properties of thomsonite. Data from the literature, classified by localities. With unanalysed specimens, an estimate of the probable composition, Al (number of aluminium atoms per unit cell), has been made by means of figs. 4 and 6; and for comparison, where other specimens from the same locality have been analysed, the analytically determined Al-figure (Al′) is also given. With analysed specimens, the number of the analysis in table II is given.

Locality, observer, &c.	α.	β.	γ.	2V.	Al.	Al′.
FAEROE ISLES:						
Faeroe Isles. P. Riva, 1914... ...	<1·514	—	—	—	—	18·5*
Faeroe Isles. S. G. Gordon, 1923 ...	1·517	1·520	1·530	—	18·7	,,
Faeroe Isles. A. N. Winchell, 1926†	1·513	1·520	1·528	—	18·4	,,
Faeroe Isles. V. Gennaro, 1929 ...	1·517	1·520	1·530	—	18·7	,,
NORWAY:						
Låven, Langesundfjord. J. Koenigsberger and W. J. Müller, 1921 ...	1·523	1·528	1·538	—	19·6	—
SCOTLAND:						
Old Kilpatrick. E. T. Wherry, 1923	1·520	1·525	1·540	—	19·4	20·00‡
Old Kilpatrick. S. G. Gordon, 1923	1·529	1·531	1·541	50°	20·0	,,
Old Kilpatrick. A. N. Winchell, 1926	1·526	1·528	1·540	—	19·7	,,
Old Kilpatrick. V. Gennaro, 1929...	1·529	1·531	1·541	—	20·0	,,
FRANCE:						
Chaux de Bergonne, Gignat, Puy-de-Dôme. E. S. Larsen, 1921§ ...	1·514	1·515	1·520	52°	18·0	—

* Mean of several analyses from 17·84 to 19·04.

† Winchell also gives for other Faeroe specimens, β 1·512, 1·517, 1·520, and 1·521.

‡ Mean of several analyses ranging from 19·26 to 20·60.

§ Supposed to be gonnardite.

TABLE VII (*continued*).

Locality, observer, &c.	α.	β.	γ.	2V.	Al.	Al′.
ITALY:						
Casarza, Liguria. P. Riva, 1914 ...	1·517	1·520	1·538	46°	Anal.	47
Vesuvius. P. Riva, 1914	>1·514	1·525	—	—	19·2	19·72
Vesuvius. A. Des Cloizeaux, 1862...	—	—	—	53°	20·0	,,
Bettolina, Valle di Ayas, Piedmont. V. Gennaro, 1929	1·523	1·525	1·537	45°	Anal.	46
Val di Viu, Piedmont. V. Gennaro, 1929	1·530	1·531	1·541	44°	19·8	20·34
	1·528	1·530	1·538	—	Anal.	52
TYROL:						
Fassa-thal. A. Des Cloizeaux, 1862	—	—	—	2E 93½°	19·2	19·18
BOHEMIA:						
Birkigt. A. Scheit, 1914*	1·5235	1·525	1·537	42°	19·4	—
Jakuben. A. Scheit, 1912† ...	1·521	1·523	1·534	49°	19·0	—
Seeberg, Kaaden. S. G. Gordon, 1923	1·527	1·529	1·540	55°	19·8	19·70‡
ALGIERS:						
Cape Djinet. L. Gentil, 1895 ...	—	—	—	50°	20·2	—
UNITED STATES:						
Franklin Furnace, New Jersey. V. Gennaro, 1929	1·530	1·532	1·542	—	20·2	—
Franklin Furnace, New Jersey. S. G. Gordon, 1923	1·530	1·532	1·542	48°	Anal.	92
Fritz Island, Schuylkill River, Pennsylvania. S. G. Gordon, 1923 ...	1·516	—	1·527	—	18·8	19·92
Peekskill, New York. A. H. Phillips, 1924	1·527	1·531	1·543	—	Anal.	94
Ritter Hot Spring, Oregon. D. F. Hewett et al., 1928	1·522	1·524	1·529	Small	19·0	Anal. 95§
Ritter Hot Spring, Oregon. D. F. Hewett et al., 1928	1·526	1·529	1·533	Small	19·5	
Ritter Hot Spring, Oregon. D. F. Hewett et al., 1928‖ ...	—	1·517	—	Small	Anal.	97
Ritter Hot Spring, Oregon. D. F. Hewett et al., 1928¶ ...	—	1·510	—	Small?	Anal.	96
Table Mtn., Colorado. E. T. Wherry, 1923, 1925	1·512	1·513	1·518	—	Anal.	82
Table Mtn., Colorado. N. A. Winchell, 1926	1·512	1·520	1·528	—	18·4	18·45

* Sp. gr. 2·389. 2V calc., not directly measured.

† Sp. gr. 2·389. 2V calc., not directly measured. $\gamma-\alpha$ 0·01318, $\beta-\alpha$ 0·00226.

‡ Mean of 5 analyses ranging from 19·34 to 20·12.

§ Analysis on a mixture containing these two constituents.

‖ Supposed to be mesolite. Optically positive. Elongation β.

¶ Supposed to be pseudomesolite. Elongation γ. $\gamma-\alpha$ very small.

TABLE VII (*continued*).

Locality, observer, &c.	α.	β.	γ.	2V.	Al.	Al′.
UNITED STATES (*continued*):						
Table Mtn., Colorado. A. N. Winchell, 1926 … …	1·513	1·520	1·526	—	18·4	18·45
Table Mtn., Colorado. A. N. Winchell, 1926* … …	1·513	1·519	1·526	—	18·3	,,
Italian Mtn., Colorado. C. W. Cross and E. V. Shannon, 1928 … …	1·515	1·520	1·538	50–60°	18·5	—
OCEANIC ISLANDS:						
Bennet Island, New Siberian Isles, Arctic Ocean. H. G. Backlund, 1920† … … … …	1·525	1·527	1·539	51° 12′	19·6	—

* Winchell also gives for other Table Mtn. specimens, β 1·513, 1·516, and 1·520.

† $\gamma - \alpha$ 0·0145, directly measured.

resulting was quite good considering the approximate nature of the data. Lastly, the optic axial angle curve, fig. 6, was calculated from the birefringence curves. Most of the earlier optic axial angle determinations were made on unanalysed material, and are only of use in making a rough estimate of the composition of the specimen studied. Owing to the approximate nature of most of the data, no influence of the $Na_2 \rightleftarrows Ca$ substitution on the birefringence or optic axial angle could be traced.

The birefringence curves show that γ decreases much more rapidly than β or α with increase in silica, and at a certain point, about 17·8 atoms Al per unit cell, the optic sign should change and become negative, the *a*-axis becoming the acute bisectrix; with a still further increase in Si, the material should become uniaxial for successive wave-lengths, at about 17·2 atoms Al, and then the axial plane should become parallel to *b* (010), the elongation becoming positive. The negative axial angle should rapidly increase, become positive (the acute bisectrix then becoming the *c*-axis), and a second, positive uniaxial point should appear about 16·8 atoms Al per unit cell; beyond this the axial plane will become parallel to *a* (100) and the positive acute bisectrix the *c*-axis, till about Al 16·4, when the optic sign changes once more, the negative acute bisectrix now being the *b*-axis.

It must be emphasized that, owing to the approximate nature of most of the data, the exact course of the birefringence and optic axial angle curves remains doubtful. The figures given for the uniaxial points and points of change of optic sign above are very approximate, but there appears no reason to doubt the general course of the curves.

Although there are several analyses which show aluminium contents below 17·8 atoms per unit cell (nos. 19, 21, 22, 38, 41, 43, 51, 56, 59, 68, 70, 86, 88, 91, 96, 97, and 101) they are all (except nos. 96 and 97) on material which was never optically examined, and as remarked above, such material may have been mixed with other zeolites, which might lower the aluminium content. Hence a judgement of the frequency of occurrence of such high-silica material must take account of the probability of contamination, as shown by comparison with other analyses and optical studies of material from the same locality. Such comparison leads us to doubt the purity of all but the two optically examined specimens.

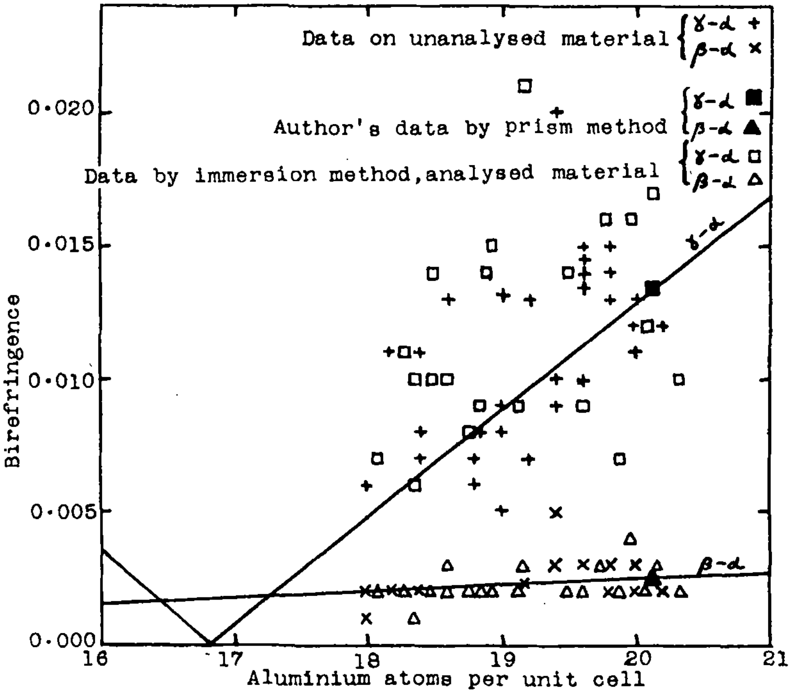

FIG. 5. The variation of the birefringence of thomsonite with the chemical composition. The composition of the unanalysed specimens estimated by the aid of fig. 4.

These two were examined by D. F. Hewett, E. V. Shannon, and

F. A. Gonyer (1928), under the names of mesolite and pseudomesolite, (analyses 97 and 96), but there is little doubt that both were really thomsonite. Their 'mesolite' had a refractive index of 1·517, and a Si/Al-ratio of 1·24, differing considerably from the corresponding values for true mesolite (the latter, as will be shown in a later paper in this series, are very constant at *n* 1·505 and Si/Al 1·5); but the agreement with the properties predicted from figs. 4 and 6 for thomsonite with 17·82 atoms Al per unit cell is very good. The optical properties of the 'pseudomesolite' (*n* 1·510, Si/Al 1·3, 17·40 Al atoms per 80 oxygen) also fit well to the curves, while their agreement with the original description of pseudomesolite by N. H. Winchell (1900) is by no means so good. It is unfortunate that both of these specimens were much too finely fibrous for conoscopic observation.

Dehydration and vapour-pressure.

We now come to the properties most characteristic of the zeolites as a group—the base-exchange capacity and the peculiar dehydration curves. The work which has been done on the zeolites in general in these respects, and the various conclusions that have been drawn were discussed in part I. With regard to the dehydration curves, it is sufficient here to say that the methods adopted by all workers up to the present are inadequate to give a complete account of the relations without the expenditure of an inordinate amount of time. Isothermal and isobaric dehydrations both require rather complex apparatus if accurate results are to be obtained, and are very slow, owing to the long time taken for the establishment of equilibrium. Most workers have employed the isobaric method, but few have regulated the water vapour-pressure accurately,[1] and no one has attempted to trace the full vapour-pressure surface of any zeolite by a series of isobaric or isothermal curves.

The author has sought to make a complete determination of the pressure-composition-temperature or vapour-pressure surface (*pct*-surface) practicable, by reducing the time required to attain equilibrium and so allowing a larger number of observations to be completed per diem. Moreover, the speeding-up will also mean a saving of material, for with shorter exposure to high temperatures,

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[1] Dehydrations in an oven without any attempt to control the water vapour-pressure approximate (very roughly) to isobaric curves. They unfortunately constitute most of the data for nearly all zeolites.

lattice-shrinkage, necessitating fresh material, will be slower to appear.

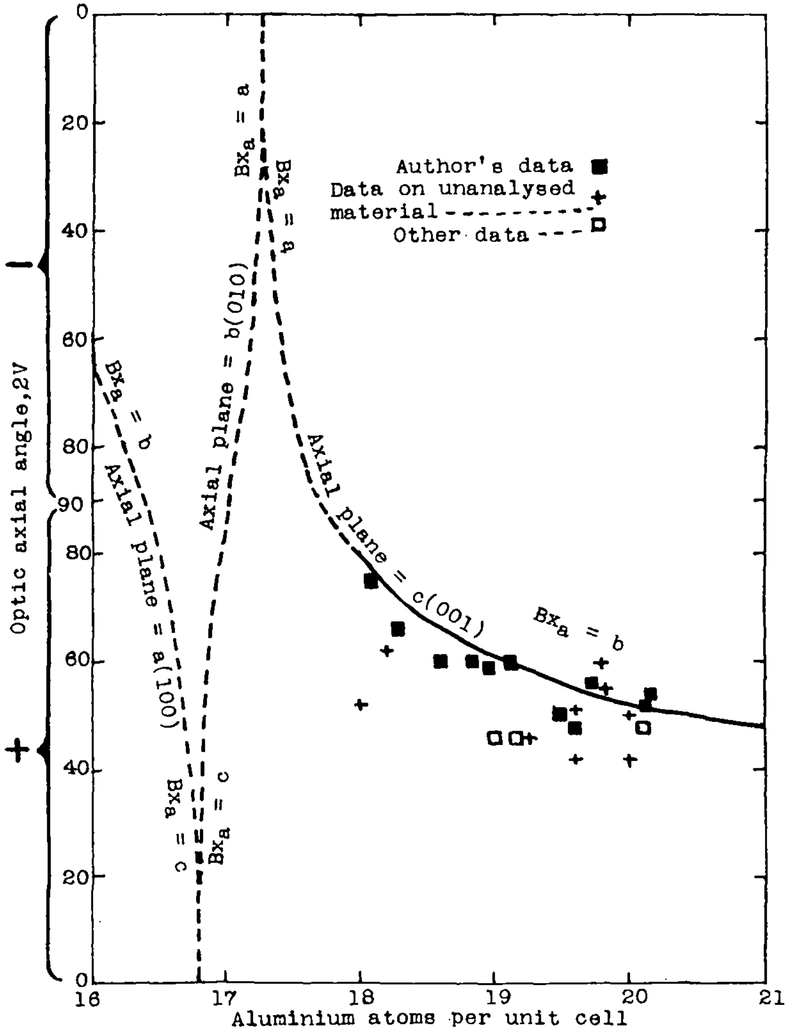

FIG. 6. The variation of the optic axial angle (2V) of thomsonite with the chemical composition. The composition of the unanalysed specimens estimated by the aid of fig. 4.

The shortening of the equilibrium time is accomplished very simply by the use of isohydric curves. If a quantity of zeolite of known composition is heated in a limited space, very little water-vapour need be given off to establish the equilibrium vapour-pressure in this space, and hence there will be a great saving of time. In practice, with the form of apparatus now to be described, equilibrium is reached at moderate temperatures as rapidly as a steady temperature can be established, and some ten observations per diem are possible.

The apparatus is shown diagrammatically in fig. 7. The zeolite is contained in the cylindrical bulb *Z*, and is heated in an electrical thermostatic air-oven (temperature control ±1° C.), the bulb being enclosed within two wide glass tubes to assist in maintaining a steady temperature. The temperature is measured by the thermometer *T*, which is a fused-silica nitrogen-filled instrument (largely avoiding errors due to alterations in the size of the bulb under long continued heating); it is completely within the two guard-tubes, so no stem-correction is needed. As a check, a second thermometer *t* is placed with its bulb just outside the guard-tubes; this generally reads a little different from the first, but no observations were made if the difference exceeded 5° C., except for the very highest temperatures.

From the zeolite bulb, a thick-walled capillary tube leads to the small manometer *m*, which is enclosed in an auxiliary oven *O*; the temperature of this oven is maintained just high enough to avoid condensation of water in *m* by means of an electric heater in series with a rheostat (the temperature used is 10–20° C. above that at which water has a vapour-pressure equal to that present in *Z*). The auxiliary oven *O* has a pair of glass windows *W* to allow of illumination and observation of the manometer *m*. A bulb *B* is arranged above *m*, so that after a series of measurements, the manometer can be evacuated, the mercury being tipped into *B* by inclining the apparatus, *Z* gently heated, and water pumped off. The amount of water removed is determined by weighing this part of the apparatus before and after the dehydration. To allow of this procedure, a tap *H* is arranged above *B*, followed by a rubber connexion to the remainder of the apparatus. One charge of zeolite is generally used for three or four series of observations, after which the bulb *Z* is broken open, the zeolite removed, and the water content checked by the Penfield method. The agreement between the final water content so found directly and that deduced from the initial content and the several weights pumped off is generally very good.

The range of manometer *m* is ± 30 mm. and small pressures are read directly on it (with *B* evacuated and *H* closed), using a reading telescope and a stainless steel scale; the accuracy attainable over this range (0 to 30 mm.) is ±0·3 mm. Greater accuracy (±0·05 mm.) is attainable for pressures below 0·7 mm. by the use of an eyepiece scale in the telescope.

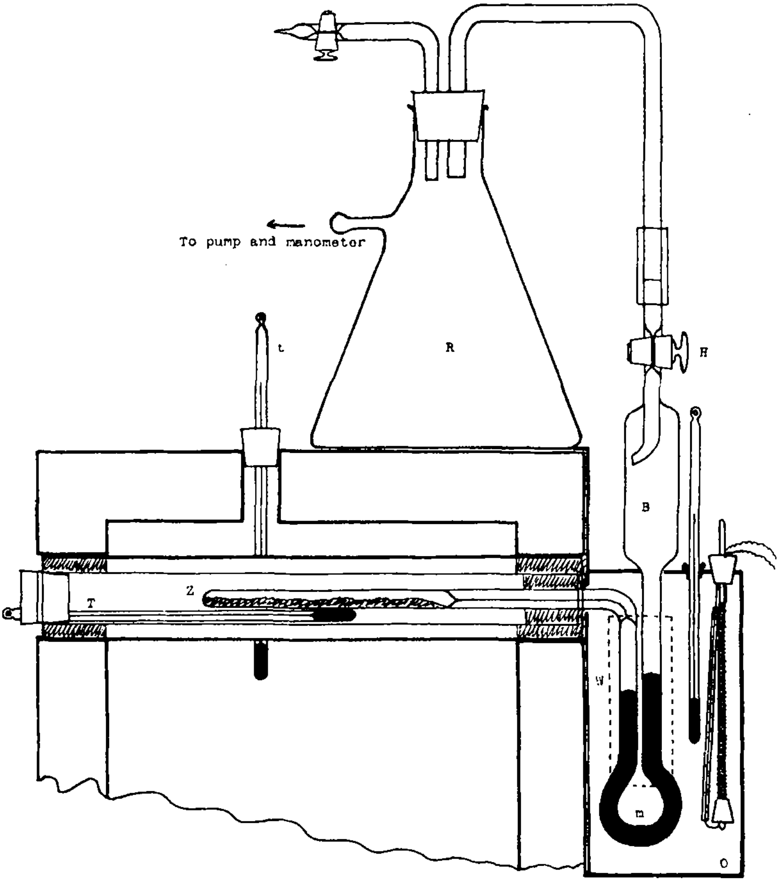

FIG. 7. Sketch of the apparatus employed for the study of the vapour-pressure and dehydration relations of thomsonite.

For higher pressures, from 30 mm. to atmospheric, the manometer *m* is used as a null instrument. For this purpose, the tap *H* is connected up through a steadying air reservoir *R* with a vacuum-pump and a large manometer; by manipulation of the taps connecting *R* to the pump and the atmosphere, the manometer *m* is balanced, and the pressure read on the large manometer.[1]

The results obtained in one series of observations with this apparatus really trace an isochore, since the volume of the system of zeolite and vapour is constant so long as the manometer *m* is used as a null instrument. But they approximate closely to an isohydric curve, since the free vapour-space is small. Moreover, if the volume of the apparatus from *Z* to the surface of the mercury in *m* is measured, the amount of water-vapour lost by the zeolite is readily calculated. With the dimensions of apparatus used, the vapour-space is about 2 c.c.; and with 1 gram of zeolite, the water-loss is negligible below 30–40 mm. pressure, and at 1 atmosphere and 140° C. only amounts to 0·1 %.

With other zeolites, it may in some cases be necessary to supplement the isohydric curves with isobaric dehydration curves, for reasons which will be set out when the occasion demands this addition, but with thomsonite the isohydric curves proved sufficient to determine the *pct*-surface fully. Nevertheless one isobaric dehydration curve, for a water vapour-pressure of 16 mm., was made (table XI), and up to 270° C. agrees very well with the isohydric curves, showing that the new method is satisfactory. But above about 280° C. the isobaric curves show a greater loss of water at a given temperature than corresponds to the *pct*-surface drawn from the isohydric curves (cf. fig. 8 b). This is due to the lattice-shrinkage, the effect of which is more noticeable in the isobaric curve, which involves longer heating than the isohydric method. Even the latter cannot be entirely free from the effects of the shrinkage reaction, which may be reasonably expected to raise the vapour-pressure for any given temperature and degree of hydration. A tendency for the vapour-pressure to rise with time at temperatures above 300° C., and a certain degree of irreversibility were noticed in the isohydric experiments, the pressure being higher for a given temperature if the temperature had previously been above 300° C. for some time.

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[1] In a new modification of this apparatus, which has been used in the study of natrolite, arrangements are made for connecting the reservoir *R* to the pressure side as well as to the vacuum side of the pump; the range of measurement is thereby extended to about 1400 mm. (nearly 2 atmospheres).

TABLE VIII. Vapour-pressure of the Old Kilpatrick Thomsonite (B.M. 94985). p = pressure in mm. Hg; t = temperature in °C.; c = water content of the thomsonite in mols. per unit cell; in the notes column, + indicates equilibrium after a rise of temperature, – after a fall of temperature, ? doubtful equilibrium.

c.	*p.*	*t.*	Log *p.*	Notes.
		Series 1.		
23·80	0·3	20°	$\bar{1}$·5	?
,,	2·0	40	0·30	? +
,,	10·4	54	1·02	+
23·78	35·3	69	1·55	+
,,	29·0	67	1·46	–
,,	25	60	1·40	? +
,,	26·1	55	1·42	? +
,,	47·7	76	1·68	+
23·74	126	105	2·10	? +
,,	100	87	2·00	–
,,	117	94	2·07	+
23·68	130	104	2·11	? +
,,	171	118	2·23	? +
,,	204	124	2·31	? +
23·64	270	135	2·43	? +
23·66	216	124	2·33	? –
23·60	367	147	2·56	? +
23·56	515	168	2·71	? +
,,	493	167	2·69	? +
23·54	560	185	2·75	? +
		Series 2.		
20·24	1·0	118°	0·00	+
,,	2·7	132	0·43	+
,,	4·5	141	0·65	+
,,	1·2	117	0·08	–
,,	7·6	160	0·88	?
,,	39·5	184	1·60	+
,,	48	186	1·68	+
,,	41	183	1·61	–
,,	89·0	203	1·95	+
,,	19·5	160	1·29	–
,,	6·2	149	0·79	–
,,	15·0	158	1·18	+
20·22	104	206	2·02	+
,,	137	213	2·14	+
,,	172	220	2·24	+
		Series 3.		
19·44	98	232	1·99	–
,,	108	239	2·03	+
,,	128	248	2·11	+
19·40	242	266	2·38	+
		Series 3 (*continued*).		
19·36	375	285	2·57	+
19·34	460	291	2·66	+
19·32	560	300	2·75	+
19·30	694	308	2·84	+
,,	760	312	2·88	+
19·36	390	287	2·59	–
19·42	160	252	2·20	–
19·46	3·7	164	0·56	+
,,	6·0	179	0·78	+
,,	7·9	184	0·90	+
,,	30	204	1·48	+
,,	6·5	178	0·81	–
,,	5·8	170	0·76	+
,,	25	198	1·40	+
,,	47	213	1·67	+
19·44	69	224	1·84	+
		Series 4.		
15·94	4·2	247	0·62	+
,,	4·7	252	0·67	+
,,	1·6	220	0·20	+
,,	1·8	231	0·25	+
,,	3·6	251	0·56	+
,,	5·1	259	0·71	+
,,	7·0	267	0·84	+
,,	3·8	250	0·58	–
,,	2·5	238	0·40	–
,,	0·35	207	$\bar{1}$·54	+
,,	22	283	1·34	+
,,	14	274	1·15	–
,,	35	302	1·54	+
,,	27	293	1·43	+
,,	42	312	1·62	+
,,	33	301	1·52	–
,,	21	281	1·32	–
,,	27	298	1·43	+
,,	33	305	1·52	+
,,	42	312	1·62	+
,,	52	319	1·72	+
,,	67	326	1·83	+
15·92	82	335	1·91	+
,,	99	341	2·00	+
,,	115	348	2·06	+

TABLE VIII (*continued*).

c.	p.	t.	Log p.	Notes.
	Series 4 (*continued*).			
15·94	69	332	1·84	−
,,	51	321	1·71	−
,,	33	304	1·52	−
,,	28	293	1·45	−
,,	24	287	1·38	−
,,	22	283	1·32	−
,,	21	277	1·32	−
,,	17	267	1·23	−
,,	15	261	1·18	−
,,	13·5	271	1·13	+
,,	9·5	263	0·98	−
,,	8·0	260	0·90	−
,,	27	294	1·43	+
,,	22	282	1·32	−
	Series 5.			
10·10	0·35	255	$\bar{1}$·54	+
,,	0·5	260	$\bar{1}$·70	+
,,	1·3	281	0·11	+
,,	2·0	289	0·30	+
,,	0·9	270	$\bar{1}$·95	−
,,	1·3	277	0·11	+

c.	p.	t.	Log p.	Notes.
	Series 5 (*continued*).			
10·10	1·5	284	0·18	+
,,	2·2	292	0·34	+
,,	2·8	297	0·45	+
,,	3·2	301	0·51	+
,,	4·2	312	0·62	+
,,	4·6	320	0·66	+
,,	5·9	329	0·77	−
,,	6·3	334	0·80	+
,,	8·0	340	0·90	+
	Series 6.			
7·68	0·15	290	$\bar{1}$·18	−
,,	0·15	310	$\bar{1}$·18	+
,,	0·2	316	$\bar{1}$·30	+
,,	0·3	328	$\bar{1}$·47	+
,,	0·35	337	$\bar{1}$·55	+
,,	0·6	358	$\bar{1}$·78	+
,,	0·3	334	$\bar{1}$·47	−
,,	0·35	342	$\bar{1}$·55	−
,,	0·4	348	$\bar{1}$·60	+
,,	0·25	329	$\bar{1}$·40	+
,,	0·2	312	$\bar{1}$·30	+

The vapour-pressure observations (for Old Kilpatrick thomsonite) are set out in table VIII, and shown graphically in figs. 8 a, b, c, and d. In fig. 8 a the observed data are plotted, the logarithms of the vapour-pressures being taken as ordinates, while the abscissae are the reciprocals of the absolute temperatures: the reasons for this method of plotting are, first that the range of pressures involved is too great for a direct plot of the pressure, and secondly that theoretical considerations show that the method of plotting used should give straight lines for the isohydric curves. To make the graph clearer, a scale of temperatures in degrees Centigrade and scale of pressures have been added. For comparison, the vapour-pressure curve of water has been included. The isohydric curves drawn in this diagram are, of course, sections of the *pct*-surface perpendicular to the *c*-axis. Sections perpendicular to the *p*-axis constitute the isobaric curves, shown in fig. 8 b.

Fig. 8 b has been drawn indirectly, using interpolated data from fig. 8 a, and shows the isobaric curves for 0·1, 1, 10, 100 and 1000 mm. Hg pressure, deduced from the isohydric experiments. The directly measured isobaric dehydration values, for a pressure of 16 mm. Hg, are included, and will be seen to agree quite satisfactorily. The third possible main section of the *pct*-surface, perpendicular to the *t*-axis, gives the isothermal curves, such as G. Tammann (1897) obtained for several zeolites at room-temperature (for thomsonite he could get no dehydration within the limited range of pressure available). Fig. 8 c shows the isothermal curves for thomsonite, for several temperatures, as deduced from the isohydric experiments.

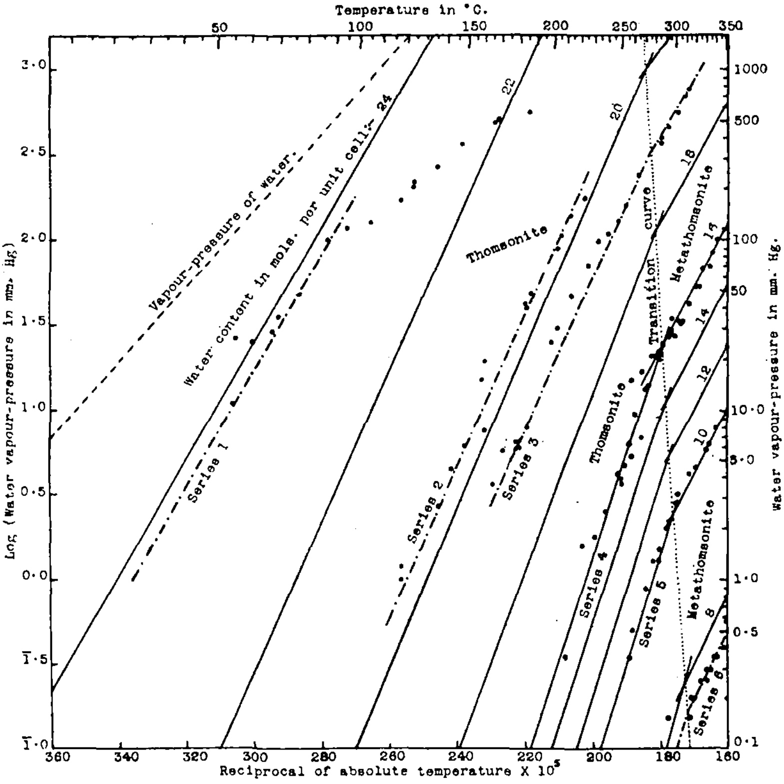

FIG. 8 a. Isohydric vapour-pressure curves of Old Kilpatrick thomsonite, showing the transition to metathomsonite.

As above mentioned, theoretical considerations lead us to expect the isohydric curves, when plotted as ($\log p : 1/T$)-curves (where T is the absolute temperature) to be straight lines. The well-known equation $d(\log_e p)/dT \approx Q/RT^2$ gives by integration, $\log_e p = A' - Q/RT$, which with $\log p$ and $1/T$ as co-ordinates is the equation of a straight line. In this equation, A' is an integration constant, while Q is the molecular heat of hydration of the substance. Accordingly, the equation only represents a straight line provided Q does not vary with temperature. As a rule, the variation of Q with temperature is negligible in most reactions; that it is so in the present case is shown by the good agreement of the experimental data with a set of straight lines (cf. fig. 8 a). Only one set of points departs from a straight line relation, namely the last nine of series 1; these have been included to point out one source of error which must always be guarded against in this kind of work. If it should happen that the zeolite gets displaced from its proper position well up towards the closed end of the tube Z (fig. 7), that part of it nearest the outside of the oven may become cooled below the temperature of the rest; it will then tend to absorb the water-vapour given off by the rest of the zeolite, and the observed pressure will be much lower than should correspond to the observed temperature. The last nine points of series 1 are vitiated by this cause.

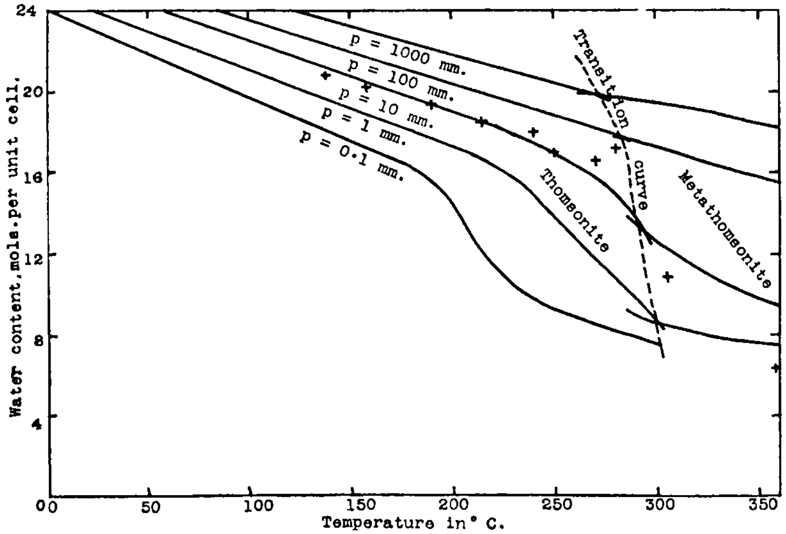

FIG. 8 b. Isobaric dehydration curves of Old Kilpatrick thomsonite, calculated from the vapour-pressure data, together with some direct measurements for a pressure of 16 mm. Hg.

Not only does this method of plotting serve to enable us to draw a more accurate line through the points (since a straight line can be drawn more accurately than a curve), but it also enables us to obtain

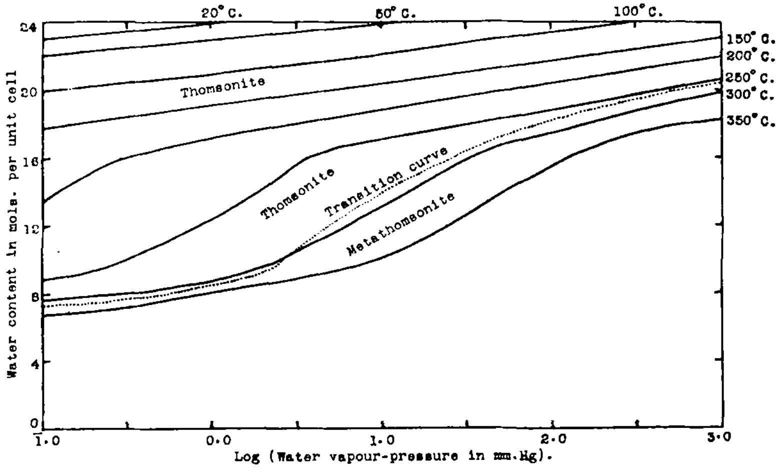

FIG. 8 c. Isothermal dehydration curves of Old Kilpatrick thomsonite, calculated from the vapour-pressure data.

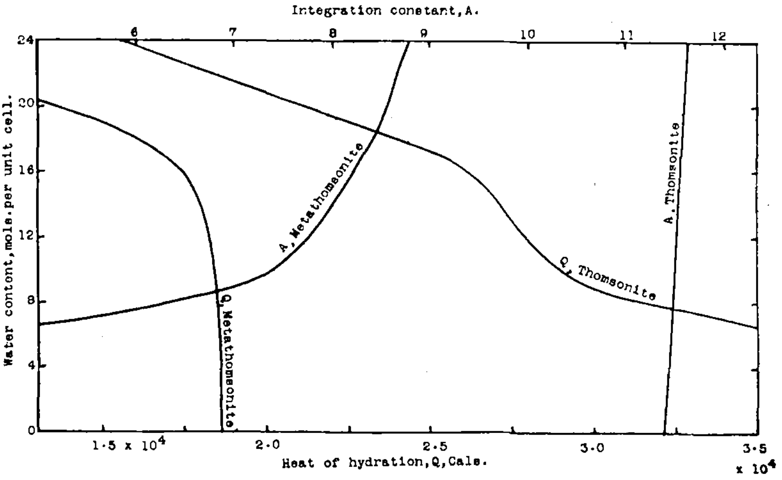

FIG. 8 d. The variation of the molecular heat of hydration Q of Old Kilpatrick thomsonite and metathomsonite with the degree of hydration, calculated from the vapour-pressure data. Also the variation of the constant A in the equation $\log_{10}p = A - Q/RT(\log_e 10)$.

an indirect measure of the heat of hydration of thomsonite at various degrees of hydration. The values obtained from the various isohydric curves are given in table IX, and shown graphically in fig. 8 d. It should be explained that the molecular heat of hydration (Q) at a given degree of hydration (c) is the heat evolved when an infinite amount of thomsonite of the given degree of hydration is allowed to absorb one molecular weight in grams (18 grams) of water-vapour. A check of the heat of hydration by direct experiment is unfortunately impossible, since partially dehydrated thomsonite re-combines too slowly with water.

TABLE IX. The heats of hydration of thomsonite and metathomsonite, for various degrees of hydration, deduced from the isohydric curves. And the integration constants (A) of the equation $\log_{10}p = A - Q(RT\log_e 10)$.

Mols. water per unit cell.	Thomsonite.		Metathomsonite.	
	Q, cals.	A.	Q, cals.	A.
23·80	$1{\cdot}59\times10^4$	11·62	—	—
20·26	2·10	11·58	—	—
19·46	2·15	11·58	$1{\cdot}37\times10^4$	8·50
15·94	2·61	11·55	1·74	8·16
10·10	2·88	11·50	1·83	7·42
7·70	7·16	11·48	1·84	6·13

Further, since the constants Q and A' at any given degree of hydration fully determine the isohydric curve, the most accurate method of interpolation for arbitrary degrees of hydration is to plot the values of Q and A' (or better, of Q and A, where A is the integration constant in the equation $\log_{10} p = A - Q/(RT\log_e 10)$, and equals $A'/\log_e 10$) against the degree of hydration, c; smooth curves are drawn through the points derived from the experimental isohydric curves, and values of Q and A taken from these curves. The experimental data for both Q and A are included in table IX, and shown graphically in fig. 8 d. And the isohydric curves drawn as full lines in fig. 8 a have been interpolated in this manner (the dot-and-dash lines in fig. 8 a are the experimental isohydric curves).

It will be seen that the vapour-pressure surface of thomsonite consists of two continuous portions which cut along a line. In all three sets of sections, this is expressed as discontinuities in the gradient of the curves. And the values of A and Q, for the same degree of hydration, are quite different for the two portions of the *pct*-surface. The only interpretation that can be given of such a vapour-pressure surface is that thomsonite is dimorphous. The ordinary form to which all the above optical studies apply is stable at ordinary temperatures, but on raising the temperature, transforms into a second form, at a temperature depending on its water content; the greater the water content the lower the transition temperature. The transition is readily reversible, as is seen by the rapid return of the vapour-pressure to the values characteristic of the low-temperature form on cooling below the transition point; nevertheless, the transition point can be overrun, as we see from the series 4 of table VIII. The numerous observations in the neighbourhood of 270–300° C. in this series form the main evidence of transition; continuous lines might be drawn for the other series, but experience of the accuracy of the method suggests that the lines drawn represent the facts best. But an attempt to determine the optics of the high-temperature form (and confirm its existence at the same time) by rapidly cooling thomsonite that had been heated above the transition point met with no success; the transformation is clearly too facile for this method of observation.

The direct observation of the transition is made more difficult by the fact that the transition temperature is so high and is raised by loss of water. Several attempts to overcome these difficulties were made, but without success. A cleavage plate of the Old Kilpatrick material, immersed in paraffin wax in an electrically heated cell on the stage of a horizontally placed microscope, showed no appreciable alteration in the optic axial figure till the temperature reached 240° C.; at this temperature the brushes become hazy, raising a hope that the transition was commencing, and patches with comparatively low birefringence appeared at two parts of the crystal, but the cell cracked and put an end to the experiment. There is reason to believe that the crystal lost an appreciable amount of water in spite of the wax-bath, which it was hoped would prevent this; moreover, the temperature of the crystal-plate was probably by no means uniform, and may have exceeded the transition temperature locally.

In view of these failures, it cannot be said that the transition, and the existence of a dimorphous, high-temperature form of thomsonite is definitely proved, but there is considerable evidence for it; and it is probable that the high-temperature form is identical with the metathomsonite of F. Rinne (1897). In his valuable studies of the effect of dehydration on the optical properties of the zeolites, Rinne found that if thomsonite is heated (and, if necessary, cleared in oil, the oil merely filling the cracks with a liquid of about the same refractive index as the mineral and so improving the transparency) the interference-figure remains unaltered, except for a small increase in the optic axial angle and a considerable fall in the birefringence, unless the temperature was very high. In the latter case, there is a permanent change, the optic axial plane becoming parallel to *a*, and the new product was named metathomsonite.

TABLE X. Effect of dehydration on the refractive indices of Old Kilpatrick and Nolsö Thomsonites.

	Old Kilpatrick, No. 15.						
Water content in mols. per unit cell. ...	23·80	22·52	21·32	20·28	18·48	16·86	16·54*
α	1·524	1·526	1·519	1·516	1·508	1·504	1·504
β	1·531	1·528	1·521	1·518	1·510	1·506	1·506
γ	1·542	1·536	1·532	1·528	—	—	1·512

	Nolsö, No. 2.		
Water content in mols. per unit cell. ...	22·34	16·34	16·16
α	1·518	—	1·503
β	1·520	1·509	1·505
γ	1·529	—	—

* Shrinkage 1·5 % by volume.

The author's experiments fully confirm this; in addition, a few observations have been made on the effect of dehydration on the refractive indices and on the density, but much more study will be needed before the changes can be properly described. Since oils and other organic liquids consisting of large molecules are not absorbed by dehydrated thomsonite, the immersion method can safely be used to determine the changes in refractive index. It should, perhaps, be emphasized that the data all relate to the low-temperature form, whatever temperature the dehydration was carried out at, since the transition is so readily reversible. As table X and fig. 9 show, the dehydration causes a fall in the refractive index, amounting in the initial stages to about 0·0025 per molecule of water removed per unit cell (or probably somewhat less), while the density also decreases slightly. But if the temperature has exceeded about 270–300° C. both refractive index and density tend to increase again owing to lattice-shrinkage. The reality of lattice-shrinkage is shown by Mr. Bannister's experiments, discussed below. Lattice-shrinkage, too, is probably responsible for the discrepancy between the ready reversibility of the thomsonite⇄metathomsonite transition as observed by the author and the apparent permanence of metathomsonite when prepared by the method of Rinne.

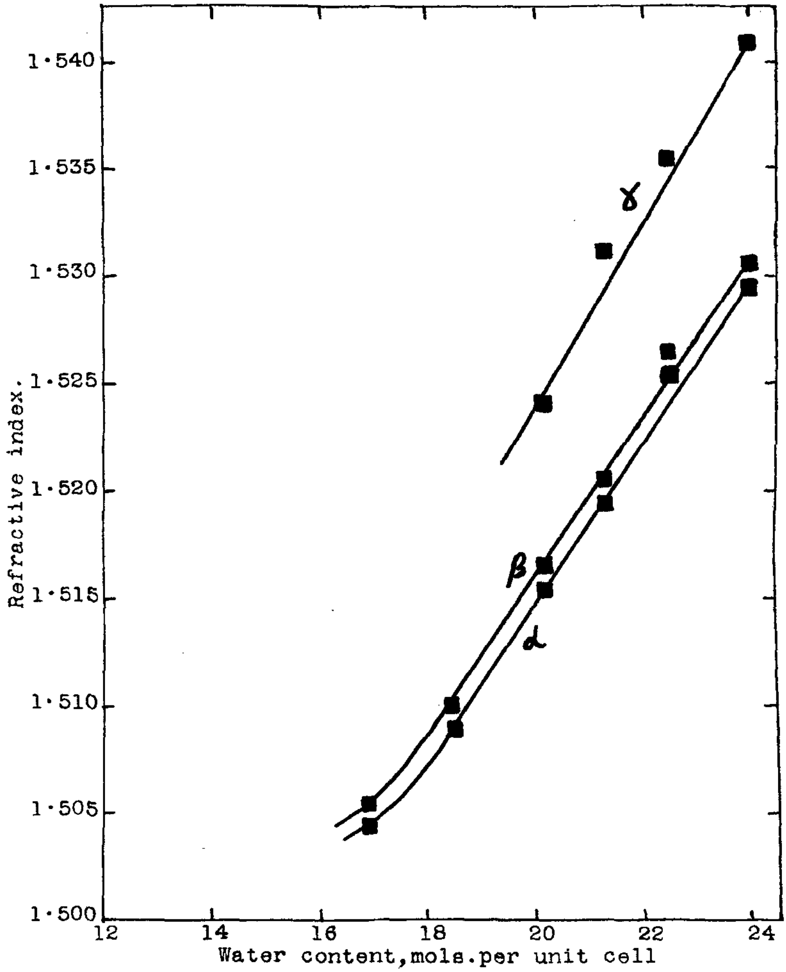

FIG. 9. Effect of dehydration on the refractive index of Old Kilpatrick thomsonite.

It will be noticed that the heat of hydration of dehydrated thomsonite or metathomsonite increases with increasing dehydration. This behaviour is precisely what would be expected if the partially dehydrated material is regarded as a mixed crystal, and indicates that when thomsonite is dehydrated, the more water is lost, the more firmly is the remaining water held. Further, the heat of hydration of metathomsonite is always less than that of thomsonite of the same degree of hydration; this is again in accordance with theory, as metathomsonite is the high-temperature form.

If the isohydric curve for fully hydrated thomsonite drawn in fig. 8 a is correct, it will, on extrapolation, cut the vapour-pressure curve of water at about 150° C., corresponding to about 4½ atmospheres pressure. This can only mean that at higher temperatures thomsonite must lose a certain amount of water, even when in contact with liquid water; the amount of water lost will be just that necessary to bring the vapour-pressure down to that of water. Such behaviour was fully to be expected as a possibility. Unfortunately, the experimental difficulties in the way of a direct confirmation are very great.

The above data relate entirely to the Old Kilpatrick material, and we still require to know what the effect of change of the Si/Al- and kationic ratios on the vapour-pressure is before we have an adequate picture of the vapour-pressure relations of thomsonite. Time did not permit of a proper investigation of this matter, but an isobaric dehydration of the more silica-rich Nolsö specimen, B.M. 1907,739 (analysis 2), was made for a pressure of 16 mm. (table XI, and fig. 10).

TABLE XI. Isobaric dehydration of Old Kilpatrick (no. 15) and Nolsö (no. 2) thomsonites for a water vapour-pressure of 16 mm. Hg.

	Water content, mols. per unit cell.	
Temperature.	Old Kilpatrick.	Nolsö.
138° C.	20·72	18·78
158	20·28	16·92
190	19·40	16·88
215	18·48	16·34
240	18·04	16·40
250	16·86	16·16
270	16·54	16·08
280	17·24	14·02
305	10·72	12·42
360	6·30	4·86

Further, the loss at 100° C. and at 300° C. or 350° C. was determined for all the analysed specimens (included in table I). The vapour-pressure is unknown, but is the same for all dehydrations at any one temperature, so that these data are available for comparison purposes.

There are also F. Zambonini's measurements (1905, 1908) on material of unknown composition, from Table Mountain, Colorado, for water vapour-pressures of about 10·0 mm. and 0·1 mm., and the measurements of A. Damour (1858), B. Erben (1885), C. Hersch (1887), G. D'Achiardi (1902), T. L. Walker and A. L. Parsons (1922), and H. R. Gaertner and F. Machatschki (1927), which, unfortunately, are all for no definite vapour-pressure. Lastly, there are a few isolated determinations of the 'hygroscopic' water lost at 100° C. or 110° C., again for an unknown water vapour-pressure.

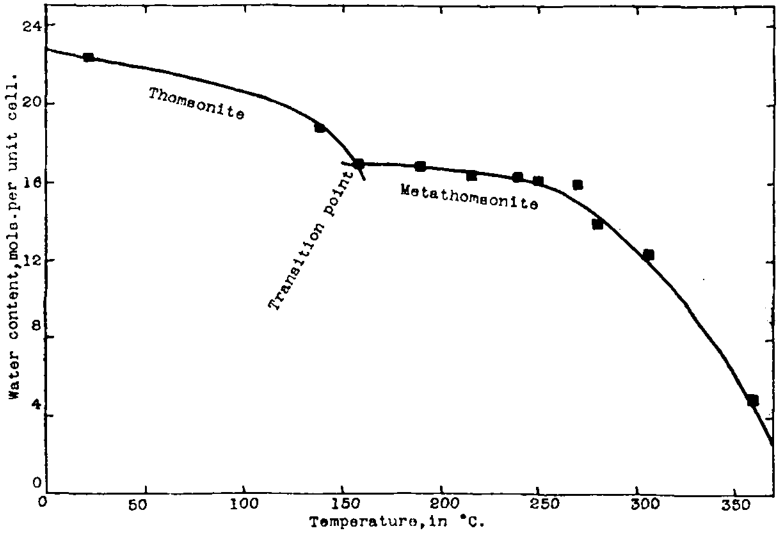

FIG. 10. Isobaric dehydration curve of Nolsö thomsonite, B.M. 1907,739 (analysis 2, table I), for a water vapour-pressure of 16 mm. Hg.

If we assume that where no water vapour-pressure data are given, the determinations were made in air of average moisture content—say 5 to 10 mm., and that Zambonini's measurements were made on 'normal' material with about 18·4 atoms Al per unit cell (the average composition of Table Mountain material), it becomes possible to compare these various measurements, but the resulting information is meagre; we can only say that the influence of a change of the Si/Al-ratio on the *pct*-surface is small, but takes the form of a lessened affinity for water and correspondingly increased vapour-pressure in the silica-rich material.

One deduction of some importance can, however, be made. It will be noticed that the author's 16 mm. isobaric curve for the Nolsö material (Al 18·28) shows a distinct break at about 160° C. and H_2O 17·0 mols. per unit cell; this break has been confirmed by repeated measurements, and there seems no reason to doubt that it represents the transition point between low- and high-temperature forms—between thomsonite and metathomsonite. With this material also, attempts to confirm the transition optically have not been successful, presumably because it is so readily reversible. The point of interest, however, is that this transition takes place at a much lower temperature (160° C. as against 290° C.) than does that of the less siliceous Old Kilpatrick material for a similar water content. In other words, increase in the Si/Al-ratio lowers the transition temperature for a given water content. With sufficiently silica-rich material, the transition temperature should be depressed to room-temperature—in other words, the high-temperature form should become the stable type. It is possible (see below) that gonnardite may represent such material.

The other available dehydration data fit in satisfactorily with these conclusions, except those of T. L. Walker and A. L. Parsons. Their curve shows several breaks, but none about the expected point. But since they did not approach the equilibrium from both sides, and did not take any precautions to regulate the water vapour-pressure, their curve can only be regarded as approximate. Further careful study of the vapour-pressure (*pct*-) surface of thomsonite is necessary before we can be certain of the existence and conditions of stability of the high-temperature form.

The dependence of the reversibility on the thermal history has been one of the most puzzling things about the dehydration phenomena of zeolites. F. Rinne suggested[1] that this was due to a second reaction, a shrinkage of the lattice, the velocity of which is, at least for moderate temperatures, less than the velocity of dehydration. To test this matter, Mr. Bannister has made X-ray photographs of the Old Kilpatrick thomsonite before dehydration, after dehydration for 3 hours at 270° C. in vacuo (when 4·1 % water was lost, 1·0 % of it being regained on standing over water for 3 weeks), and after the

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[1] F. Rinne, Ber. Sächs. Akad. Wiss. Leipzig, Math.-phys. Kl., 1920, vol. 72, p. 12. [M.A. 4–372.]

dehydrated material had been heated to 360° C. in a sealed tube (to prevent further loss of water) for 120 hours. After being thus 'shrunk' the dehydrated material only regained 0·63 % water in 5 weeks in saturated air; and its cell-spacings were then *a* 12·93, *b* 12·93, and *c* 13·23 Å., as compared with 13·09, 13·09, and 13·23 for the fully hydrated material, and 12·92, 12·92, and 13·23 for that which had been dehydrated with as little shrinkage as possible. The percentage shrinkage involved is 2·5 %, and this agrees fairly well with the densities. The calculated density for material after loss of 4·1 % H_2O, without lattice-shrinkage, is 2·29. Found, for the fully shrunk material, 2·35 (shrinkage 3 %), and for that dehydrated with minimum shrinkage, 2·33 (shrinkage 2 %). The shrinkage was apparently almost complete in 3 hours at 270° C.

Another interesting property of the zeolites is their capacity for absorbing other vapours in place of water; this property has been investigated by several workers. O. Weigel has put forward[1] a formula relating the quantity of any vapour absorbed with its van der Waal's constants, but has since shown[2] that this does not represent the whole truth, and that the polarity of the molecules presented for absorption is also an important factor. He had already recognized the possible effect of the size of the molecules, and had shown that while chabazite will absorb methyl and ethyl alcohols and formic acid, it cannot absorb ether, acetone, or benzene. Weigel had also applied such observations to estimate the size of the channels in the zeolite structure. His conclusions in this direction are, however, open to one important criticism, in that the occurrence or non-occurrence of absorption depends not only on size, but on polarity, and the polarity of the compounds he employed was very variable.

In studying the absorption of vapours by thomsonite with the hope of throwing some light on its structure, the author has employed only alcohols, so as to avoid the variable polarity factor. The following were used: ethyl, *n*- and *i*-propyl, and *n*-, *i*- and *t*-butyl alcohols, the formulae of which are in order: $CH_3.CH_2OH$, $CH_3.CH_2.CH_2OH$, $(CH_3)_2.CHOH$, $CH_3.CH_2.CH_2.CH_2OH$, $(CH_3)_2.CH.CH_2OH$, and $(CH_3)_3.COH$.

The results (table XII) were somewhat unexpected; their exact significance is not yet really clear, and the following suggestions are

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[1] O. Weigel, Sitzungsber. Gesell. Naturwiss. Marburg, 1924, p. 107. [M.A. 4–373.]

[2] O. Weigel and E. Bezner, ibid., 1927, p. 57. [M.A. 5–78.]

largely speculative. The small absorption of *n*-propyl and the three butyl alcohols may probably be set down to surface absorption (including condensation in cleavage cracks and other fissures); a similar percentage may reasonably be subtracted from the figures for ethyl and *i*-propyl alcohols as due to the same cause. After correction we have the data set out in columns 3 and 5 of table XII. The optical properties of these alcohol-treated specimens were not determined.

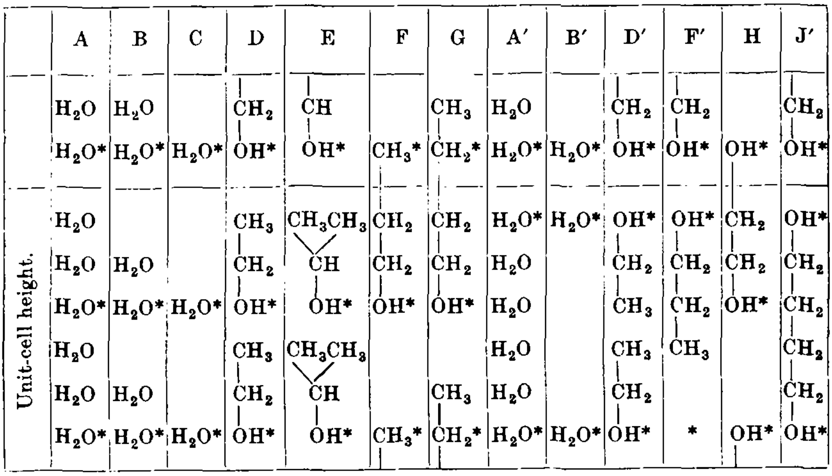
TABLE XII. Absorption of alcohol vapours by dehydrated Old Kilpatrick thomsonite (9·28 % water, = 16·6 mols. per unit cell).

	Absorbed, %.		Absorbed, mols. per unit cell.		
Alcohol.	Found.	'Corrected'.	Found.	'Corrected'.	Length of molecule, in Å.
Ethyl ...	6·0	4·3	4·2	3·0	6–6·5
n-Propyl ...	2·5	0 ?	1·4	0 ?	8·5–9
i-Propyl ...	6·0	4·3	3·2	2·4	6–6·5
n-Butyl ...	1·1	0 ?	0·4	0 ?	11 approx.
i-Butyl ...	2·4	0 ?	1·0	0 ?	8·5–9
t-Butyl ...	0·8	0 ?	0·4	0 ?	6–6·5

Now from the fact that the cleavages of thomsonites are *a*(100) and *b*(010) we may reasonably conclude that these planes are probably closer packed than *c*(001), and that the channels along which water and other absorbed vapours (and also migrating kations in base-exchange) must mainly pass are probably parallel to the *c*-axes (or rather, that the channels parallel to the *c*-axis probably have a greater diameter than those in other directions). The removal of 4 % water allows the absorption of 4·3 % ethyl or *i*-propyl alcohols, indicating that somewhat less than one water molecule must be removed to allow of the entry of each carbon atom or oxygen of the alcohol molecule—a result we should rather expect since the carbon atom is just a little smaller than oxygen. This accounts approximately for the amounts of ethyl and *i*-propyl alcohols absorbed; the non-absorption of the other alcohols must be due to the greater size of their molecules. Now the overall length of the three-atom chains of ethyl, *i*-propyl, and *t*-butyl alcohols is about 6 to 6·5 Å., while *n*-propyl and *i*-butyl alcohols contain four-atom chains, some 8·5 to 9 Å. long, and *n*-butyl alcohol has a five-atom chain about 11 Å. long.

The coincidence of the chain length of the largest molecules absorbed with the *c*-dimension of the thomsonite pseudo-unit cell

(6·6 Å.) suggests that they are actually arranged parallel to this axis, and that the non-absorption of *n*-propyl and *n*-butyl alcohols, with their longer chains, is simply due to their excessive length. Since one ethyl or *i*-propyl alcohol molecule appears to replace three water molecules, it appears likely that the 24 water molecules of the unit cell are distributed over four channels, six to each. These channels would be long enough to accommodate one *n*-butyl, or even one *n*-amyl alcohol molecule per cell per channel, and the probable explanation of the non-absorption of the higher alcohols by a partially dehydrated thomsonite with 16 mols. water per unit cell would appear therefore to be a matter of polarity. If we suppose that there are two principal foci of residual affinity per 13·23 Å. in each channel parallel to the *c*-axis, a rational explanation of the observations can be found. The two foci must either be closely adjacent (*A′–J′*, fig. 11), or separated by about 6·6 Å., half the unit cell height (*A–H*, fig. 11). The foci of residual affinity are probably to be identified with the neighbourhood of pairs of Al-atoms in the structure (cf. the pairs of Al-atoms in the chain proposed for the structure of natrolite by L. Pauling (loc. cit.)).



	A	B	C	D	E	F	G	A′	B′	D′	F′	H	J′
	H_2O	H_2O		CH_2	CH		CH_3	H_2O		CH_2	CH_2		CH_2
	H_2O^*	H_2O^*	H_2O^*	OH*	OH*	CH_3^*	CH_2^*	H_2O^*	H_2O^*	OH*	OH*	OH*	OH*
Unit-cell height.	H_2O			CH_3	CH_3CH_3	CH_2	CH_2	H_2O^*	H_2O^*	OH*	OH*	CH_2	OH*
	H_2O	H_2O		CH_2	CH	CH_2	CH_2	H_2O		CH_2	CH_2	CH_2	CH_2
	H_2O^*	H_2O^*	H_2O^*	OH*	OH*	OH*	OH*	H_2O		CH_3	CH_2	OH*	CH_2
	H_2O			CH_3	CH_3CH_3			H_2O		CH_3	CH_3		CH_2
	H_2O	H_2O		CH_2	CH		CH_3	H_2O		CH_2			CH_2
	H_2O^*	H_2O^*	H_2O^*	OH*	OH*	CH_3^*	CH_2^*	H_2O^*	H_2O^*	OH*	*	OH*	OH*

FIG. 11. A diagrammatic representation explaining the behaviour of thomsonite towards alcohols. The vertical columns represent channels in the thomsonite structure parallel to the *c*-axis, while the asterisks indicate foci of residual affinity.

In unit-cell height, six carbon or oxygen atoms, with their attendant hydrogen, can be accommodated in a chain. Fig. 11 shows presumptive arrangements of alcohol and water molecules in the channels on these assumptions, the foci of residual affinity being represented by asterisks (*). In accordance with modern chemical theory, it is assumed that the oxygen of water or hydroxyl has residual affinity which can neutralize that of the anhydrous thomsonite lattice, while the carbon atoms of a saturated alcohol chain have no such residual affinity available. It will be seen that full hydration with 24 molecules of water per unit cell (six per channel), or partial hydration with even as little as 8 mols. water per unit cell, will bring oxygen atoms near all the foci of residual affinity, and satisfy it pretty completely (*A–C* and *A′–C′*, fig. 11).

So also will saturation with ethyl alcohol (*D*, *D′*), *i*-propyl alcohol (*E*), methyl alcohol, or any combination of these alcohols and water. But *n*-propyl alcohol (*F*, *F′*) or *n*-butyl alcohol (*G*) or higher alcohols have so long a chain that the position near one focus of residual affinity must be occupied by a carbon atom, which is incapable of saturating residual affinity. Hence, even if the remaining positions be filled with water, the resulting arrangement will be less stable than one containing the same amount of water but no alcohol—i.e. a partially dehydrated thomsonite with more than 8 mols. water per unit cell left should not absorb alcohols with a chain longer than 6–6·5 Å.

Probably an unshrunk thomsonite with less than 8 mols. water per unit cell would absorb the longer-chain alcohols freely, since they would satisfy at least a part of the free residual affinity, but to dehydrate thomsonite to this extent without lattice-shrinkage (which practically inhibits the absorption of water, let alone alcohols) is a matter of great difficulty, if not, indeed, impossible.

It might be possible to distinguish between the two possible arrangements of the foci of residual affinity above mentioned. It will be seen from *H* and *J′* (fig. 11) that if the foci are spaced at half the unit-cell height, or approximately so, ethylene glycol (and no other $\alpha:\omega$-dihydric alcohol) should be absorbed by partially dehydrated thomsonite, while if they are closely adjacent, tetramethylene glycol only should be absorbed. As the glycols are non-volatile, the procedure adopted with the alcohols could not be used; absorption had to be from the liquid instead of the vapour phase. Tetramethylene glycol was not, unfortunately, available, but a quantity of thomsonite was partially dehydrated, its refractive indices measured, then portions were immersed in ethylene and trimethylene glycols for several days, and the refractive indices redetermined. The partially dehydrated material had α 1·508, β 1·510; after immersion in trimethylene glycol for a week these indices were unaltered,[1] while another portion immersed in ethylene glycol was decidedly affected, the indices rising to α 1·518, β 1·520. Thus it is evident that ethylene glycol is absorbed, but trimethylene glycol is not, and the foci of residual affinity are probably spaced at half the unit-cell height (fig. 11, *A–H*). The birefringence, position of the optic axial plane, and magnitude and sign of the optic axial angle appeared unaffected by the absorption of ethylene glycol.

With regard to the cross-section of the channels we can make but slight inferences. As we are dealing with molecular dimensions we cannot treat the channels as of uniform cylindrical or other simple cross-section. But any suggested structure for thomsonite must be such as to leave spaces into which *i*-propyl alcohol molecules can enter, but which are still too small for the *t*-butyl alcohol; this implies that the cross-section of the channels must, in places at least, reach some 15 to 25 Å^2.

Base-exchange.

Although not so characteristic as the peculiar dehydration relations, the property of base-exchange is common to most, if not all, of the zeolites. But the velocity of reaction of different zeolites under similar conditions varies enormously from species to species. Thus the reaction between chabazite and *N*-KCl solution is appreciable after a few minutes, and equilibrium is attained in a few days at room-temperature, while under the same conditions thomsonite gives no appreciable reaction in six months.

As a result of its low reactivity, thomsonite has been little studied as regards base-exchange. J. Lemberg (1883, 1887) treated it with various solutions at temperatures up to 200° C. in sealed tubes, but there is little doubt that his products were largely mixtures, and his reactions complicated by solution and recrystallization (compare H. Schneiderhöhn, 1915). H. C. McNeil (1906) treated thomsonite with fused sodium chloride, but the author finds on repeating this experiment that the product is anhydrous, cannot be rehydrated by

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[1] Benzyl alcohol is not absorbed by partially dehydrated thomsonite, and is miscible with the glycols; it therefore served for washing the treated material free from glycol before the refractive index determination.

boiling with water for a week, and has a complex optical structure. Its appearance under the microscope suggests a mixture of albite and sodalite.

The author first attempted to obtain base-exchange in thomsonite by digestion with aqueous solutions ($AgNO_3$, $CaCl_2$, $NaCl$, KCl, Na_2CO_3, K_2CO_3, and sodium acetate) at the ordinary temperature; little or no appreciable reaction could be detected after twelve months except with the K_2CO_3. At this point the supply of material ran short; it was impossible to take more from the Old Kilpatrick specimen, B.M. 94985, without spoiling it. My thanks are, therefore, especially due to Miss J. M. Sweet for the presentation of another Old Kilpatrick specimen, which made it possible to continue the work. Dr. C. E. Marshall kindly maintained a further series of experiments at 100° C. for 700 hours, but again with little effect; the products were analysed and examined optically, the results being included in table XIII.

Attention was therefore turned to the method of F. W. Clarke and G. Steiger (1902), who treated thomsonite with dry ammonium chloride in a sealed tube at 300° C., and with molten silver and thallous nitrates. Since the temperature during these experiments was comparatively low, the loss of water was not serious. The products they showed to be homogeneous, crystalline, and probably orthorhombic (straight extinction); analysis proved that a simple base-exchange was involved. The author's experiments were therefore directed principally to making certain that the silver- and thallous-thomsonites did really retain the thomsonite structure, and determining their optical constants and specific gravities. Attempts were also made to extend the method to other kations.

The salts tried, with the composition and properties of the products, are shown in table XIII. The X-ray investigations of the silver- and thallous-thomsonite, carried out by Mr. F. A. Bannister, have already been mentioned. They show clearly that the essentials of the thomsonite structure are retained in these derivatives.

Much more work will be needed in this field, but the few experiments made show that the base-exchange reactions of thomsonite are extremely sluggish, and that the tendency for the number of kations per unit cell to be maintained at 12 is very strong, and militates against any exchange of monovalent for divalent ions or vice versa. Substitution of the sodium by other monovalent ions is more easy, but reaction is still slow; it is curious that silver and thallium should be more readily introduced from the salt fusion than lithium, potassium, or ammonium (the analytical data for the ammonium-thomsonite probably err on the generous side as to its ammonia content). Substitution of calcium by barium or copper is also very slight. The substitutions did not go far enough to trace their effect on the optical properties and density except with the potassium,

TABLE XIII. Experiments on base-exchange, with Old Kilpatrick thomsonite.

Treatment ...		None.	Digested one year at room-temperature with a solution of:			700 hrs. at the boil with a strong solution of:			
			Na_2CO_3.	K_2CO_3.	$AgNO_3$.	NaAc.*	KCl.	$CaCl_2$.	$BaCl_2$.
Percentage composition.†	SiO_2	37·17	37·86	37·01	38·21	37·53	37·86	35·64	36·42
	Al_2O_3	31·93	32·98	30·00	32·22	32·58	32·62	31·47	31·32
	CaO	13·98	15·18	14·95	10·89	13·55	13·61	13·43	15·80
	Na_2O	4·00	2·92	1·12	5·85	4·26	2·89	4·24	4·56
	R_2O ‡	—	—	3·25	0·10	—	1·82	—	0·16
	H_2O	13·35	13·28	14·30	13·44	13·07	12·96	12·80	12·93
	Total	100·43	102·22	100·63	100·71	100·99	101·76	97·58	101·19
Oxygen % ...		39·68	39·88	38·70	39·87	39·90	39·67	39·69	39·39
Oxygen, atoms per unit cell.§		80·83	—	79·12	—	—	—	—	79·83
Atoms per unit cell, assuming oxygen 80.	Si ...	19·88	19·78	20·30	20·28	19·90	19·94	19·6	19·50
	Al ...	20·12	20·32	19·40	20·16	20·38	20·24	20·4	19·80
	Ca ...	8·00	8·50	8·78	6·20	7·42	7·68	8·0	9·04
	Na ...	4·14	2·96	1·20	6·02	4·38	2·96	4·6	4·72
	R‡ ...	—	—	1·80	0·02	—	1·22	—	0·04
	H_2O	23·80	23·14	26·16	23·80	23·12	22·76	23·4	22·54
Sp. gr. ...		2·373	—	2·381	—	—	—	—	2·36
Refract. Indices, Na-light.	α ...	1·529	1·528	1·529	1·529	1·529	1·530	1·529	1·529
	β ...	1·531	1·531	1·531	1·532	1·531	1·532	1·530	1·531
	γ ...	1·542	1·543	1·544	1·544	1·542	1·544	1·542	1·540
Cell-sides, in Å.‖	a ...	13·09	—	—	—	—	—	—	—
	b ...	13·09	—	—	—	—	—	—	—
	c ...	13·23	—	—	—	—	—	—	—
Water left at 120° C., mols. per unit cell.		—	22·86	25·66	23·08	—	—	—	22·04

* Sodium acetate.

† The first ten analyses by Mr. S. E. Ellis of the Mineral Department.

‡ R indicates the kation of the salt employed in the base-exchange treatment.

§ Assuming no change in the cell-sides except where measured cell-sides are quoted.

‖ Accuracy, ±0·05 Å.

TABLE XIII (*continued*).

Treatment ...		700 hrs. at the boil with:	Fusion at 200° C. for 48 hrs. with:*			Fusion at 250° C. for 48 hrs. with:*		
		$CuCl_2$.	KCNS.	NH_4NO_3.	$AgNO_3$.	$TlNO_3$.	$LiNO_3$.	$NaClO_3$.
Percentage composition.	SiO_2 ...	37·15	37·34	38·32	35·33	34·19	38·58	37·81
	Al_2O_3 ...	30·72	33·59	33·10	31·58	28·67	33·00	32·61
	CaO ...	13·52	13·67	14·43	13·95	12·52	14·43	14·21
	Na_2O ...	4·50	2·92	2·64	0·70	1·74	2·59	4·06
	R_2O† ...	0·49	0·46	1·52‡	10·37	11·74	0·46	—
	H_2O ...	12·67	13·24	11·14	9·87	10·14	11·85	11·99
	Total ...	99·05	101·22	101·15	101·80	99·00	100·91	100·68
Oxygen % ...		39·66	39·95	40·88	37·89	36·55	40·64	40·42
Oxygen, atoms per unit cell.§		80·03	81·34	82·08	81·12	80·42	82·43	81·95
Atoms per unit cell, assuming oxygen 80.	Si ...	20·10	19·66	19·74	19·52	20·14	20·00	19·86
	Al ...	19·60	20·86	20·14	20·56	19·90	20·16	20·18
	Ca ...	7·84	7·72	7·98	8·24	7·92	8·02	7·98
	Na ...	4·72	2·98	2·64	0·74	1·98	2·60	4·12
	R† ...	0·20	0·32	1·80‡	2·96	1·96	0·92	—
	H_2O ...	22·86	24·92	19·18	18·20	19·92	20·46	20·98
Sp. gr.		2·35	2·371	2·385	2·52	2·595	2·362	2·361
Refract. Indices, Na-light.	α ...	1·529	1·523	1·523	1·582	1·568	1·523	1·525
	β ...	1·530	1·525	1·525	1·588	1·572	1·527	1·527
	γ ...	—	1·537	1·537	1·600	1·586	1·533	1·535
Cell-sides, in Å.‖	a ...	—	—	—	13·02	13·04	—	—
	b ...	—	—	—	13·02	13·04	—	—
	c ...	—	—	—	13·23	13·23	—	—
Water left at 120° C., mols. per unit cell.		22·36	20·18	16·64	17·90	16·36	15·56	19·02

* The fusion was continued (intermittently) for 48 hours, and after leaching and thorough washing the residue was treated for 48 hours with water at 100° C. and then air-dried. But in the $AgNO_3$ experiment the water treatment was omitted.

† R indicates the kation of the salt employed in the base-exchange treatment.

‡ Probably rather too high.

§ Assuming no change in the cell-sides except where measured cell-sides are quoted.

‖ Accuracy, ±0·05 Å.

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thallous, and silver salts, but it is clear that the effect of the other substituents cannot be very great (comparison must be made, in each case, with untreated thomsonite of the same water content). Thallium and silver both cause a marked increase in the refractive index and density, as was to be expected, while the cell-size remains unaffected; the birefringence is slightly increased, while the optic axial angle remains practically unaffected. Potassium appears to have little if any effect on the refractive index, but raises the density appreciably. The considerable base-exchange obtained by digestion with K_2CO_3 solution for a year is notable in view of the slight reaction obtained with a KCNS fusion. No attempt could be made to trace the effect of base-exchange on the vapour-pressure.

The minor varieties of thomsonite.

The two principal varieties of thomsonite, distinguished by composition and optical properties, have already been discussed, namely comptonite and faroelite or mesole. Besides these, several other names appear as synonyms or varieties of thomsonite—some rightly, some wrongly—most of which were originally regarded as independent species. We may conveniently deal with these in order of date. The principal varieties will also be noted in the list for completeness.

Thomsonite, H. J. Brooke, 1820.

Comptonite, Sir David Brewster, 1821.

Mesole, J. Berzelius, 1822; name changed to *faroelite*, M. F. Heddle, 1857.

Gibsonite. This mineral name appears to occur but thrice in the literature. A. Dufrénoy (1847), who is the authority for the references by T. Egleston (1887) and A. H. Chester (1896), states that A. Des Cloizeaux had seen specimens in the collection of T. Allan, to which W. Haidinger had given the name of gibsonite, and adds that they seemed to resemble prehnite. The name never appears to have been published by Haidinger or Allan, but the specimens seen by Des Cloizeaux are now in the British Museum, together with the MS. catalogue in which Allan attributes the name to Haidinger. There are two specimens from Hartfield Moss, Paisley, Renfrewshire, on which the gibsonite occurs as small brick-red spherules formed of aggregated plates, and one from Calton Hill, Edinburgh, on which there are red pseudomorphs of gibsonite after analcime. These specimens Allan acquired and catalogued before 1828. A later note in the catalogue by R. P. Greg, to whom the collection passed after Allan's death, states that 'the Gibsonite has become known now only as Cluthalite'—the red ferruginous variety of analcime described by T. Thomson in 1836. This identification was, however, erroneous, as a new examination of the specimens by optical means has shown. The original gibsonite specimens are in fact all fibrous or platy thomsonite, coloured red by ferruginous inclusions, and slightly more siliceous than the coarsely prismatic Old Kilpatrick thomsonite, as is shown by their lower refractive indices (see table VI). Precisely similar material has been found on several other specimens from Renfrewshire and Dumbartonshire in the British Museum collection. In some cases it accompanies analcime or prehnite, or both, and may also be associated with the more usual coarsely prismatic thomsonite, which it appears to precede in crystallization. Gibsonite, therefore, is shown by the original specimens to have been neither analcime nor yet prehnite, and the name may well be retained to distinguish the pink fibrous thomsonite of Renfrewshire and Dumbartonshire, which does not appear to have been recognized hitherto, from the well-known coarsely prismatic thomsonite.

Sphaerostilbite of F. S. Beudant, 1832, was a botryoidal fibrous variety of stilbite if Beudant's analysis was correct. But the description corresponds rather to the more coarsely fibrous kinds of faroelite, especially as the mineral is stated to gelatinize with acids. A. Des Cloizeaux (1862) referred the species to faroelite, suggesting that there had been confusion between the specimen described and that analysed. As pointed out by G. T. Prior (1898), who brought chemical and optical data (anal. 24) to show that typical 'sphaerostilbite' specimens were faroelite, the same view was probably also held by M. F. Heddle, but obscured by a misprint (mesolite for mesole) in R. P. Greg and W. G. Lettsom's Mineralogy of Great Britain and Ireland (1858).

Chalilite of T. Thomson, 1836, from Co. Antrim, was united with thomsonite by A. Des Cloizeaux (1862) and more definitely by J. D. Dana (1868), on the ground that it passes over without apparent break into scoulerite (q.v.). But A. Lacroix (1886) could recognize no crystallized mineral in it. The author has had the opportunity of examining a fragment of Thomson's original specimen, which he gave to Greg and which is now in the British Museum collection (B.M. 95007), and is able to confirm Lacroix's finding. It is a confused granular mass, largely isotropic, in which nothing definite could be recognized. This is in agreement with Thomson's analysis (1836) which certainly does not indicate a zeolite, even impure. The analysis of K. von Hauer (1853) is completely different from Thomson's.

Harringtonite, another of Thomson's Co. Antrim species (1836), presents a more difficult problem. A. Lacroix (1885) examined a portion of one of Thomson's specimens and found it to be mainly isotropic, with a few natrolite crystals. On the other hand, the British Museum specimens appear to be all mixtures of thomsonite and mesolite in about equal proportions. That Thomson included material of very variable composition under this name is evident from a comparison of his original analysis (1836) with five others made in his laboratory (R. D. Thomson, 1846). This mineral has generally been classed with mesolite on the basis of A. Kenngott's goniometry (1850) and K. von Hauer's analysis (1854); but A. and R. Brauns examined a supposed harringtonite which was nearly or quite pure thomsonite (1924; anal. 29). A specimen studied by O. B. Bøggild (1922) was an intergrowth of thomsonite and natrolite. Since all the analyses of T. Thomson and R. D. Thomson may be interpreted as mixtures of thomsonite and mesolite, it appears that such a mixture really constituted the original material, and that the particular specimen examined by Lacroix was not typical.

Scoulerite, which forms red-brown to white pisolitic masses, also from Co. Antrim, was described by T. Thomson (1840) as a variety of thomsonite. His analysis, showing 6 % soda, was apparently never published in full. The optical study of A. Lacroix (1886) shows that this mineral is a mixture of thomsonite with more or less of an isotropic substance, a result the author has been able to confirm. The thomsonite has a refractive index about 1·513, while the isotropic substance, n about 1·485, is apparently a clay, perhaps allophane.

Mesolitine which J. Apjohn (1844) described, and of which he gives two analyses (anals. 26 and 27), was recognized as thomsonite by L. J. Spencer in 1903. Apjohn's description of this, another Co. Antrim mineral, shows that it was identical with scoulerite; thus two analyses of the latter made four years after its original description by Thomson have passed unnoticed owing to their being assigned to a new species.

Koodilite of A. Dufrénoy (1847) was afterwards referred by him (1859) to thomsonite.

Ozarkite of C. U. Shepard, 1846, occurring as white veinlets and patches in elaeolite from the Ozark Mts., Arkansas, was shown by J. L. Smith and G. J. Brush (1853; anal. 77) to be thomsonite. Optical tests on the specimens in the British Museum confirm this.

It was first recognized as a zeolite by J. D. Whitney (1850), and classed by him as a massive scolecite.

Picrothomsonite. In 1852, G. Meneghini and E. Bechi examined zeolites from Monte Catini, Val de Cecina, Tuscany, and found high proportions of magnesia which no subsequent author has been able to confirm. On this basis they set up several species or varieties, including picrothomsonite. The British Museum specimen (B.M. 35300), although small, is not uniform, One fragment tested showed the presence of considerable magnesia, while the analysed portion (anal. 13) contained but little. The name is usually applied to all the thomsonite from Monte Catini, but might better be reserved for those that carry magnesium—if any such really exist.

Sloanite is another species set up by Meneghini and Bechi (1852); their original analysis is very poor, but the description suggests laumontite, and this the British Museum specimen (B.M. 31348, presented by the Chevalier Sloane in 1860) unquestionably is. E. Manasse (1906) analysed a specimen of natrolite under this name, and Dana (Syst. Min., App. II, 1909, p. 74) says this analysis proves sloanite to be a variety of thomsonite, a slip several authors have copied.

Karphostilbite of S. von Waltershausen (1853) is clearly a pure thomsonite, as his analysis (anal. 17) shows.

Lintonite. The white, green, and red and white banded zeolite pebbles from Grand Marais, Lake Superior, Minnesota, have been examined by several workers, some regarding them as mesolite, some as thomsonite (anals. 86–91). S. F. Peckham and C. W. Hall (1880), who regarded them all as thomsonite, gave the varietal name of lintonite to the white and green pebbles (anals. 86 and 87). N. H. Winchell (1898) distinguished three minerals in the pebbles, intermixed in variable proportions; mesolite is in many the principal constituent, in others thomsonite, while the green ones consist largely or wholly of lintonite, which he raises to species rank on optical grounds. The lintonite he regards as monoclinic, finding extinction-angles of up to 19°, and a refractive index lower than that of thomsonite. E. T. Wherry (1925) considered the pebbles were mixtures of thomsonite and mesolite. The British Museum specimens (B.M. 53910, 53912) afford no evidence of the distinctive nature of lintonite, the green pebbles being very granular in structure, and apparently composed wholly of thomsonite[1] (β 1·525 about,

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[1] S. J. Thugutt (1912) found green pebbles to be mixtures of thomsonite and mesolite; but his method of diagnosis is not wholly reliable.

indicating Al about 19·4 atoms per unit cell); the red and white banded ones are mixtures of thomsonite and mesolite. The composite nature of the pebbles accounts for the very variable Si/Al-ratio shown by the analyses.

Bagotite of T. Egleston (1887), from Bagot, Ontario, was found by L. J. Spencer (1897) to be identical with lintonite. This material (B.M. 54508) also has a refractive index (β) about 1·525 (whence Al = about 19·4 atoms per unit cell).

Kalkthomsonit and *Natronthomsonit* were names applied by C. F. Rammelsberg (1895) to the hypothetical end-members of the $Na_2 \rightleftarrows Ca$ substitution (then formulated as of fixed Si/Al-ratio). For this purpose they (or their translations calcio-thomsonite and sodio-thomsonite) may well be retained.

Gonnardite of A. Lacroix[1] has presented some difficulties, which cannot be said to be satisfactorily cleared up yet, but there is reason to believe it is identical with a highly siliceous metathomsonite (the polymorph derived from thomsonite at a sufficiently high temperature, cf. p. 119).

Winchellite is a varietal name proposed by D. S. Martin (1912) for the mesolite pebbles (really thomsonite and mesolite mixtures) from Grand Marais, Lake Superior. (Compare L. J. Spencer, 1919.)

Calciothomsonite was the name applied by S. G. Gordon (1923) to a silica-poor normal thomsonite (Anal. 92). This seems quite undesirable, especially as its German equivalent had already been used by C. F. Rammelsberg (1895) for the lime end-member of the $Na_2 \rightleftarrows Ca$ substitution. In 1924, Gordon used the same name in the latter sense, applying it to the hypothetical end-member, $CaO.Al_2O_3.2SiO_2.3H_2O$, in his theory of the composition of thomsonite. (Compare L. J. Spencer, 1925.)

Kalithomsonite, of S. G. Gordon,[2] has been shown by X-ray examination by Mr. Bannister to be a distinct species, and not a variety of thomsonite; it is hoped to publish details of this work shortly. The *c*-spacing is 17·5 Å., and the symmetry tetragonal.

GONNARDITE.

In 1871, F. Gonnard described what appeared to be a perfectly normal mesole (faroelite) from Chaux de Bergonne, Gignat, Puy-de-

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[1] A. Lacroix, Bull. Soc. Franç. Min., 1896, vol. 19, p. 426.

[2] S. G. Gordon, Proc. Acad. Nat. Sci. Philadelphia, 1924, vol. 76, p. 261. [M.A. 2-385.]

Dôme, with an analysis by F. Pisani. The same material was examined optically by A. Lacroix (1896) who showed that it was of composite structure. The interior of the spherules Lacroix found to consist of a finely fibrous mineral with positive acute bisectrix parallel to the elongation of the fibres, and a small optic axial angle —indeed, it might possibly be uniaxial; the outside layer of the spherules consisted of fibres with a negative elongation, apparently forming continuations of the fibres of the inner layer. To the inner layer Lacroix gave the name of gonnardite.

No further work seems to have been done on the new mineral (which was widely regarded as doubtful), until E. S. Larsen (1921) determined the optical properties of material which he accepted as gonnardite; these showed that the material he examined was a thomsonite, although he did not recognize the fact.

When the study of the variation of the optical properties of thomsonite with its composition revealed that a highly siliceous thomsonite should (over a small range of composition, about 8·5 atoms Al per unit cell) have a positive acute bisectrix parallel to the c-axis, it appeared probable that gonnardite was merely such a highly siliceous variety of thomsonite. But an examination of all the gonnardite specimens in the British Museum showed that the inner layer had a negative elongation—the reverse of what Lacroix had described; the outer layer has β parallel to the elongation of the fibres. Professor Lacroix very kindly sent a specimen (B.M. 1930,166) from his collection, but the result was the same, and an error of sign would appear to have crept into his original description.

A study of the optical properties of the outer layer showed that it was thomsonite. The refractive index and optic axial angle were in good agreement with Larsen's figures for his supposed gonnardite, and indicate an aluminium content about 18·0 to 18·2 atoms per unit cell.

The inner layer, the true gonnardite, has a small birefringence, about 0·002 and a refractive index in the neighbourhood of 1·508; its negative acute bisectrix is parallel to the fibres, and the optic axial angle is less than 50°; it may indeed be uniaxial. It is difficult to ascertain the composition, for a complete separation of the gonnardite from the faroelite proved impossible on an adequate scale to supply material for analysis. However, the gonnardite occupies about three-quarters of the diameter, hence about half the volume of the spherules. The analysis of the whole yielded the results under I in table XIV below; II is Pisani's analysis; the atomic proportions per 40 oxygen atoms are set out under I *a* and II *a*. Since the outer layer has the properties of a thomsonite (presumably normal) with Al 9·1 atoms approx. per 40 oxygen atoms (III), the approximate composition of the gonnardite (on a basis of 40 oxygen atoms) is probably as given under IV.

TABLE XIV. Composition of the gonnardite-thomsonite spherules from Chaux de Bergonne, Gignat, France.

	SiO_2.	Al_2O_3.	CaO.	Na_2O.	H_2O.	Total.	
I.	41·85	27·02	9·29	7·25	14·36	99·77%	(M.H.H.)
II.	42·3	28·1	10·0	6·7	14·1	101·2	(F. Pisani, 1871).

	Si.	Al.	Ca.	Na.	H_2O.	O.	Sp. gr.
Ia.	11·27	8·58	2·68	3·78	12·90	40	2·259
IIa.	11·17	8·75	2·83	3·43	12·42	40	2·256–2·357
III.	10·9	9·1	3·1	2·9	12·0	40 ...	Thomsonite.
IV.	11·6	8·3	2·1	4·0	13 or 14	40 ...	Gonnardite.

The water content of gonnardite must remain doubtful. Owing to the finely fibrous nature of the material, it is very possible that some absorbed water might have been present; and as the gonnardite was only some 50 per cent. of the analysed specimen, any error due to this or other causes would be doubled. It is therefore quite possible that the water content may be 12 mols. per 40 oxygen atoms, the same as in thomsonite, or indeed anything from say 10 to 16 mols. Considerations based on the X-ray results suggest 14 mols. (See below.)

The X-ray measurements, combined with the density and chemical data given above, lead to a cell containing 40·0 oxygen atoms, but this exactness can only be fortuitous, as the above density is probably high owing to presence of faroelite. The density of 2·19 obtained with the small gonnardite fragment mentioned below and used for X-ray study is certainly too low; it leads to an oxygen content of 38·8 atoms per unit cell. Hence we may reasonably conclude that the true oxygen content is 40 atoms per unit cell, the same as in the pseudo-cell of thomsonite.

In composition, therefore, gonnardite agrees with a highly siliceous thomsonite, but its optical properties cannot be reconciled with such an interpretation. In the hope of gaining more information on its relation to thomsonite Mr. Bannister undertook an X-ray study.

A powder photograph of the faroelite-gonnardite mixture obtained by powdering one of the spherules was first taken, but showed very little difference from a thomsonite photograph. After some trouble, a small fragment of gonnardite practically free from faroelite was obtained; it consisted of a group of fibres with their *c*-axes nearly parallel. From this fragment an excellent fibre photograph was obtained, which gave the cell-sides $a = b = 13{\cdot}35$, $c = 6{\cdot}65$ Å., all $\pm 0{\cdot}05$. The *c*-dimension cannot be accurately measured from this photograph, nor can it be certain that additional, very weak layer-lines are not present half way between those observed. These additional lines, if present, would necessitate a doubling of the *c*-dimension, making it comparable with that of thomsonite, which gives additional lines of this kind. It is therefore an open question whether the *c*-dimension of gonnardite may not be 13·3 Å., but the half value will be taken here.

TABLE XV. Indices and intensities of lines on a fibre photograph of gonnardite, B.M. 83037. Cell-sides $a = b = 13{\cdot}35$, $c = 6{\cdot}65$ Å, all $\pm 0{\cdot}05$.

(220)	*vs*	(631) ?	*vw*
(310)	*s*	(640)	*s*
(400)	*w*	(730)	*s*
(420)	*m*	(800)	*vvw*
(510)	*s*	(820)	*m*
(440)	*m*	(840)	*m*
(331)	*vvw*	(633) ?	*w*
(600)	*s*	(10·0·0)	*w*
(620)	*w*	(10·4·0)	*m*

Cu–K_α radiation was used and the fibre rotated during exposure. A powder photograph of a larger quantity of material shows the presence of thomsonite. A few weak lines on the above photograph were also due to admixed thomsonite. They were indexed as follows: (412) *w*, (204) *w*, (600) *vvw*. For the admixed thomsonite $a = b = 13{\cdot}05$, $c = 13{\cdot}23$ Å., all $\pm 0{\cdot}05$.

The spacings and intensities (table XV) are very similar to those of thomsonite, except for the presumptive halving of the *c*-axis, but the departure is distinctly greater than would be expected for a silica-rich thomsonite with about 16·0 atoms Al per unit cell. Together with the optical data, this seems to furnish fairly conclusive evidence that gonnardite, while very closely related to thomsonite, is rightly to be regarded as a separate species, related to thomsonite, perhaps, in some such manner as are α- and β-quartz. Such a substance is known in metathomsonite, the high-temperature form of thomsonite, and the author's experiments (see above) indicate that with a sufficient increase in silica, metathomsonite might become stable at the ordinary temperature. The optical orientation of metathomsonite, too, agrees with that of gonnardite (cf. p. 98).

Owing to its finely fibrous nature, nothing is known of the crystallography of gonnardite; the optical properties indicate a symmetry at least orthorhombic, and possibly tetragonal or hexagonal. Here the X-ray work again comes to our aid, excluding the hexagonal system, and showing that the symmetry is either orthorhombic or tetragonal. Owing to the limitations imposed by the nature of the material and the fact that the axial ratio $a:b$ is unity within the experimental error, a and b reflections cannot be distinguished, but we can say $(hk0)$ is halved for $(h+k)$ odd. Little else can be deduced from the photographs.

If the identification of gonnardite as a metathomsonite is correct, our experience of similar transitions suggests that the symmetry elements of thomsonite should be preserved in gonnardite, and probably some others added; the system would still be orthorhombic.

The X-ray data also offer us further evidence as to the probable water content of gonnardite. The unit cell is 4 per cent. bigger than would be expected for the half-cell of a thomsonite of the same Si/Al-ratio—an additional 32 Å^3., and as the other atomic contents are the same, this additional volume will all be available for the absorption of extra water. The volume of a water molecule is about 15 or 20 Å^3., suggesting that two additional molecules per unit cell are likely to be taken up, and this is quite within the probable limits indicated by the analyses. The most probable unit-cell formula of gonnardite is therefore $(Ca, Na)_6(Al, Si)_{20}O_{40}.14H_2O$, with Al about $8\frac{1}{4}$, and, to correspond, Ca about $2\frac{1}{4}$.

The small amount of material available precluded any attempt to study the vapour-pressure or base-exchange of gonnardite.

SUMMARY.

A chemical, optical, and X-ray study of a considerable number of thomsonite specimens has led to the conclusion that thomsonite and faroelite form a continuous isomorphous series. The variation in the optical properties with the chemical composition has been traced, and the relations of a number of varieties and supposed varieties of thomsonite to the typical mineral examined. The true symmetry of thomsonite is shown to be didigonal polar (C_{2v}). The unit cell is shown to contain 80 oxygen atoms, and (in the fully hydrated

mineral) 24 water molecules; the silicon and aluminium contents are variable, but the sum of silicon and aluminium is constant and equal to 40. The sodium and calcium contents are also variable, but the sum of calcium and sodium is normally 12. Thomsonite appears to be essentially $Na_4Ca_8Al_{20}Si_{20}O_{80}.24H_2O$, with two isomorphous replacements, $CaAl \rightleftarrows NaSi$ and $Ca \rightleftarrows Na_2$; the most siliceous member observed to date appears to have about 17·4 atoms Al per unit cell, while the $Ca \rightleftarrows Na_2$ substitution leads to variations in the sum of calcium and sodium from about 10·3 to about 14·8 atoms per unit cell; but material with this sum between 11·5 and 12·5 is most common. There is a very marked pseudo-cell, with *c*-axis half that of the true cell, and, to correspond, half the atomic contents.

It has been shown that the refractive indices of thomsonite vary considerably, falling with increase in the Si/Al-ratio, and the mean refractive index ranged, in the specimens studied, from 1·517 to 1·535. There is reason to believe that the usual diagnostic criterion for thomsonite is not invariably true, and that thomsonites may occur in which the optic axial plane is parallel, instead of perpendicular to the elongation. There is a considerable probability that such material has been observed but passed unrecognized, at least once.

Apparatus has been designed and applied to measure the vapour-pressure of thomsonite at various temperatures and degrees of dehydration, and it appears very probable that a dimorphous high-temperature form (probably to be identified with the metathomsonite of F. Rinne) exists, the transition being readily reversible. A few experiments on the absorption of vapours other than water by dehydrated thomsonite, and on the base-exchange reactions of thomsonite, have also been made. Thomsonite has been recognized from four new localities.

Chemical, optical, and X-ray studies of gonnardite have shown it to be an independent species, though very closely related to thomsonite. It is suggested that it may be identical with the high-temperature form of thomsonite (the metathomsonite of F. Rinne). Larsen's optical data for gonnardite are shown to refer to thomsonite, while there is an error of sign in the description of Lacroix. The space-group of gonnardite could not be determined, but its unit-cell, slightly larger than the pseudo-cell of thomsonite, appears to contain approximately $Ca_2Na_4Al_8Si_{12}O_{40}.14H_2O$.

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