

The Crystallography and Structure of Eitelite, $\text{Na}_2\text{Mg}(\text{CO}_3)_2$

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Abstract

Properties of eitelite from the type locality and of the more abundant, newly recognized, eitelite from the Mapco Shrine Hospital #1, well core, Duchesne County, Utah, are identical with those of synthetic $\text{Na}_2\text{Mg}(\text{CO}_3)_2$. The lattice is rhombohedral ($a_{rh} = 6.168$, $\alpha = 47^\circ 14'$), with the cell content, $\text{Na}_2\text{Mg}(\text{CO}_3)_2$. Dimensions of the hexagonal cell are a 4.942(2) Å, c 16.406(7). Both natural and synthetic crystals have a relatively simple habit, involving several first order rhombohedra and the pinacoid. Etch figures on (0001) and (000 $\bar{1}$) indicate centrosymmetry.

The structure found in the space group $R\bar{3}$ accounts nicely for the excellent basal cleavage. Parallel to this cleavage there are sheets of $(\text{Mg}(\text{CO}_3)_2)^{--}$ in which Mg is in almost perfect octahedral coordination between oxygens of 6 different CO_3 groups. Adjacent Mg-centered octahedra do not share oxygens but do share CO_3 groups. Two Na atoms in 6-fold coordination lie at different levels between successive magnesium carbonate sheets, being alternately attached to the sheet above or the one below.

Introduction

The crystalline phase $\text{Na}_2\text{Mg}(\text{CO}_3)_2$ was made by Sainte-Claire Deville (1851, pp. 89–90) by the action of a solution of sodium bicarbonate on “magnèsie blanche” at 60 to 70°. He noted that it is decomposed upon heating before fusion and stated that it is probably rhombohedral. DeSchulten (1896) also produced crystals of this phase incidental to experiments on the crystallization of a sodium magnesium chlorocarbonate, the analogue of northupite. He recognized its uniaxial negative character and reported a density of 2.729. Eitel and Skaliks (1929) reviewed earlier work on $\text{Na}_2\text{Mg}(\text{CO}_3)_2$ and prepared the phase by crystallization under a CO_2 pressure of 1240 kg/cm², at which pressure the melting point was reported to be 677°. They also reported cell dimensions and suggested a structure referred to as uncertain (“Die Bestimmung dürfte kaum als sehr sicher gelten”) in Strukturbericht II, 395–396 (1937).

Milton, Axelrod, and Grimaldi (1955) found the phase in crystals a few millimeters in diameter together with reedmergnerite, leucosphenite, shortite, searlesite and crocidolite in the Green River formation (Eocene) around 2800 feet in cuttings from the Carter Oil Company, Kermit Poulson No. 1 Well, Utah. They recognized the fine basal cleavage, reported the correct optical properties, and named the mineral eitelite.

Material

Type material was available in very small quantity for the present study. The results of X-ray observations and a set of precession patterns made on the type material in 1961 by E. C. T. Chao (U. S. Geological Survey) agree completely with newer observations on natural and synthetic material.

Abundant eitelite has recently become available in a core from the Mapco Shrine Hospital #1 Well, Duchesne County, Utah. Sections of core from the interval 4241.1–4249.1', thin sections, and separates from this interval and notes thereon by J. R. Dyni and Charles Milton were available. Eitelite occurs throughout most of this part of the core. The most abundant associated carbonates are trona, nahcolite, and shortite; northupite is less common. Magnesioriebeckite is found with trona in places. The enclosing rock is referred to as “brown marlstone”; the organic material in it, as “kerogen.” In a few places, eitelite constitutes up to 50–60 percent of the rock and its crystals may reach dimensions up to 17 mm; elsewhere it is in small scattered crystals in the marlstone with shortite.

Synthesis

$\text{Na}_2\text{Mg}(\text{CO}_3)_2$ was synthesized by the method used by Sainte-Claire Deville (1851). “Magnesium carbonate” (Mallinckrodt, AR 5950), an ultrafine powder which was found by X-ray examination to be the equivalent of hydromagnesite, was treated with a saturated solution of sodium bicarbonate at 74°C for several days, evaporation losses being replaced at intervals. In this way several crops of $\text{Na}_2\text{Mg}(\text{CO}_3)_2$, mostly in separate crystals, were

obtained. All of the largest crystals (*ca* 1 mm) have a mosaic structure and are partly turbid due to inclusion of reactants. However, most smaller crystals (*ca* 0.1 mm) are clear and well developed.

Morphology

All well-developed crystals have easily recognizable rhombohedral habits. On the synthetic crystals the most important forms are $\{0001\}$, $\{10\bar{1}1\}$, and $\{01\bar{1}2\}$. Occasional faces of other $\{h0\bar{h}l\}$ forms could not be certainly identified. One of the larger synthetic crystals (Fig. 1, left) has the pseudo-octahedral habit which DeSchulden (1896) recognized as arising from a combination of the $\{0001\}$ pinacoid and $\{01\bar{1}2\}$, rhombohedron (Fig. 2, right). Many of the larger, turbid, crystals have this habit. The smaller and more perfect crystals invariably show the $\{10\bar{1}1\}$ rhombohedron in addition and may be slightly elongated parallel to *c* (Fig. 2, left) or nearly equidimensional.

The larger eitelite crystals are firmly embedded in the marlstone and can rarely be removed intact. Though the faces are mostly dull and the edges are rounded, rhombohedra, often in stepped development, can generally be recognized. The base, as a growth form, is subordinate on natural crystals. Measurement of several typical small eitelite crystals (Fig. 1, right) and of many fragments of larger crystals by 2-circle goniometer confirms $\{10\bar{1}1\}$ and $\{01\bar{1}2\}$ as the dominant forms on natural crystals also. Moreover, faces of pairs of forms such as $(10\bar{1}1)$ and $(01\bar{1}1)$ were frequently recorded, suggesting twinning on (0001) , but no evidence of such twinning was found in those crystals examined by X-rays. Though many poor reflections were recorded, the only additional form

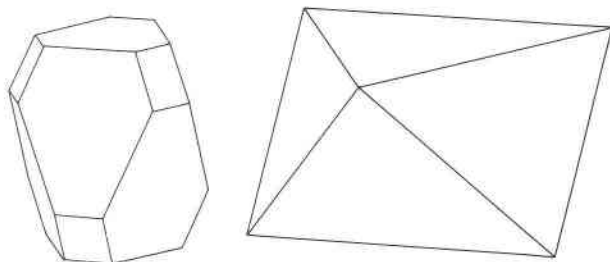


FIG. 2. Typical habits of synthetic $\text{Na}_2\text{Mg}(\text{CO}_3)_2$ crystals. Right: pseudo-octahedral habit, $\{0001\}$ and $\{01\bar{1}2\}$. Left: typical habit of smaller crystals, $\{0001\}$, $\{01\bar{1}2\}$ and $\{10\bar{1}1\}$.

confirmed was $\{10\bar{1}4\}$, which was ideally developed on one crystal.

Lattice and Symmetry

The cell dimensions and related data for eitelite and for dolomite are given in Table 1. Cell dimensions for eitelite, obtained by least squares from six high 2θ $K\alpha_1$ reflections on a doubly quartz calibrated $\text{Cu } h0\bar{h}l$ Weissenberg pattern, $\sigma = 0.04$ percent, are in excellent agreement with an earlier unpublished determination by E. C. T. Chao. No indication of variations in cell dimensions was found in single crystal patterns from natural and synthetic crystals, nor in powder or diffractometer patterns. The report of marked variations in *c* by Eitel and Skaliks (1929, p. 280) remains unexplained. All X-ray patterns can be indexed on a rhombohedral lattice. The Laue symmetry is $\bar{3}$. This restricts the permitted space groups to $R\bar{3}$ and $R3$.

Eitelite shows excellent $\{0001\}$ cleavage and thin cleavage flakes with perfect surfaces are easily obtained from the larger crystals. Such flakes are very slowly attacked by distilled water. After an hour at room temperature, no change can be seen at a magnification of 100x, but countless tiny etch figures, equilateral triangles 0.01 to 0.02 mm on edge, are visible after four or five hours (Fig. 3). The etch figures on opposite sides of each cleavage flake are in the relation of centrosymmetry.¹ The edges of the etch figures are parallel to the *a* axes, so that the morphology and etch figures alone would be compatible with the symmetry $\bar{3}2/m$; however, the evidence of the etch figures combined with the Laue symmetry established the space group as $R\bar{3}$.

¹ The etch figures are trigonal pyramidal depressions. Attempts to determine the indices of the bounding surfaces by goniometry and by observations on mounted flakes by the Jones (1960) technique failed.

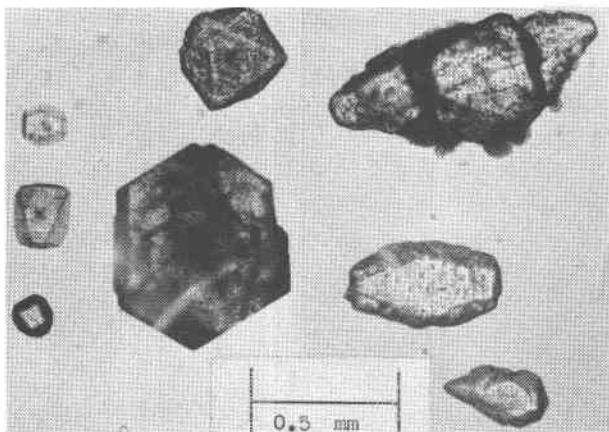


FIG. 1. Left: Crystals of $\text{Na}_2\text{Mg}(\text{CO}_3)_2$, grown at 74°C . Right: Crystals of Eitelite from Mapco Shrine Hospital #1 Well, Duchesne County, Utah.

TABLE 1. Crystallographic Data for Eitelite and Dolomite

	Eitelite	Dolomite
Hexagonal Unit Cell		
a_h	4.942(2) Å	4.815 Å *
c_h	16.406(7) Å	16.119 Å *
c/a	3.320	3.348
Rhombohedral Unit Cell		
Space Group	$\bar{R}3$	$\bar{R}3$
a_{rh}	6.168 Å	6.050 Å
α	47° 14'	46° 54'
Content	$\text{Na}_2\text{Mg}(\text{CO}_3)_2$	$\text{CaMg}(\text{CO}_3)_2$
Volume	116.67 Å ³	107.88 Å ³
Refractive Indices for Sodium Light		
ϵ	1.450(2)	1.500 **
ω	1.605(2)	1.679 **
$\omega - \epsilon$	0.155	0.179

* From Steinfink and Sans (1959, p. 680).

** From Dana's System of Mineralogy, 7th ed., vol. 2, p. 211 (1951).

Physical Properties

Clear crystals of both natural and synthetic eitelite are perfectly colorless. The refractive indices of synthetic crystals, determined in Na light by the Jones (1960) technique are ϵ 1.450(2) and ω 1.605(2), in exact agreement with the reported indices of eitelite from the type locality (Milton *et al.*, 1955). The mean index of eitelite, calculated from the specific refractive energies tabulated by Larsen and Berman (1934, Table 2) by the Gladstone and Dale relation, is 1.551, just equal to $\sqrt[3]{\omega^2\epsilon}$. Bragg (1924) devised a method for the discrete calculation of the several principal indices of birefringent carbonates. It is possible to make a comparable calculation for eitelite or other double carbonates by a modification of the Gladstone and Dale relation, using as input specific refractive energies calculated for the ordinary and extraordinary rays of the constituent carbonates. In the present case, optical properties and calculated density for pure MgCO_3 were used to calculate the appropriate specific refractive energies for ϵ - MgCO_3 and ω - MgCO_3 . Calculations for Na_2CO_3 were based on data for γ - Na_2CO_3 given by Winchell and Winchell (1964, p. 90). This is biaxial negative with small $2V$. An average value, 1.540, of β (1.535) and γ (1.546) was used for ω . The density of γ - Na_2CO_3 was calculated from cell dimensions given by Dubbeldam and

DeWolff (1969). The results compared with those of Bragg for calcite are:

	Eitelite		Calcite	
	obs	calc	obs	calc
ϵ	1.450	1.457	1.486	1.488
ω	1.605	1.610	1.658	1.631

and show the validity of the procedure used.

The density of eitelite was determined by Berman balance on four clean fragments from one crystal with an aggregate weight of 68.5 mg. Duplicate determinations on each of the four fragments gave a weighted average of 2.737 ± 0.010 . The calculated density of $\text{Na}_2\text{Mg}(\text{CO}_3)_2$ is within the limits of error of this determination. However, other Berman balance determinations, varying slightly, were consistent in indicating a density slightly higher than that calculated.

The hardness of eitelite was determined by microscopic observations on clean cleavage surfaces of the reference minerals, calcite and fluorite. All observations showed that eitelite is not as hard as fluorite but distinctly harder than calcite, so that the hardness on the Mohs scale can be given with confidence as $3\frac{1}{2}$.

In addition to the conspicuous {0001} cleavage of eitelite reported by Milton *et al.* (1955), cleavage transverse to this was noted on nearly all broken fragments. Goniometric measurements on half a dozen fragments on which secondary cleavages are conspicuous showed that breaks parallel to many first order rhombohedron directions are involved, ranging from {02 $\bar{2}$ 1} to {01 $\bar{1}$ 5}, but no prismatic cleavage, though this may be simulated by repeated steps of other steep cleavages.

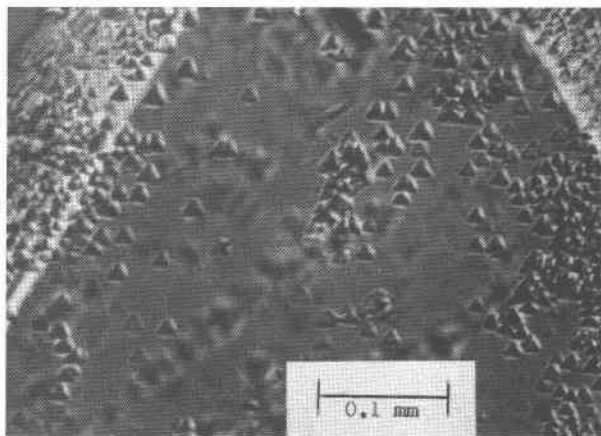


FIG. 3. Etch figures produced on cleavage surface of eitelite by distilled water during 5 hours at room temperature.

Powder Pattern

The unindexed ASTM pattern 4-737 for $\text{Na}_2\text{CO}_3 \cdot \text{MgCO}_3$, described as made from a "Lab. Prep.," closely matches the pattern for eitelite and may be used in its identification. Inquiries addressed to the Wyandotte Chemical Company for information regarding the material represented by this card were fruitless. An indexed powder pattern for eitelite from the Mapco Shrine Hospital #1 well core will be submitted to the editors of the Powder Diffraction File.

Structure Determination

The data for eitelite and dolomite (Table 1) show similarity between many constants for the two substances. A simple substitution of 2Na for Ca is obviously impossible. However, the 7.79 \AA^3 greater volume of the eitelite cell is sufficient to accommodate one Na^+ ion. From the cell content and space group alone, it follows that (using a hexagonal reference system) 3 Mg can be placed in $3a, 0,0,0$; that 6 Na and 6 C must be placed in $6c, 0,0,z$, and 18 oxygens in $18f, x,y,z$; a five-positional-parameter structure. If the CO_3 group is considered to be planar and a C-O distance is arbitrarily assigned, the number of parameters is reduced to three, $z(\text{Na})$, $z(\text{CO}_3)$ and an angular parameter for CO_3 which fixes both x and y for oxygens.

Preliminary tests showed that a sequence Mg — CO_3 — Na — Na — CO_3 — Mg along the trigonal axis, analogous to that in dolomite, could not lead to a reasonable structure nor explain the most conspicuous features of the intensity distribution. A trial structure was then found by a systematic procedure. Among the seven orders of $(000l)$ observed, three reflections ($l = 6, 12$ and 15) are markedly stronger than the others. Half of the eight possible distributions of signs for three F 's can be discarded as involving only a change in origin from 000 to $\frac{1}{2} \frac{1}{2} \frac{1}{2}$. One of the remaining four, that with all signs alike, can be eliminated as involving a very high peak at the origin or center, a condition incompatible with a structure containing no heavy atoms. The three remaining one-dimensional F_o summations based on the three strongest $(000l)$ reflections, reindexed with $l = 2, 4$ and 5 , were made with 1/60th Lipson and Beevers strips, yielding electron densities at intervals of 1/180th projected onto c over the range of z 0 to 1/6, repeatable by reflection to 1/3 and thereafter by translation. One of these summations, executed and plotted within an hour or so, yielded a moderate

peak at the origin assigned to Mg, a strong peak at 0.074, corresponding to $.667 + .407$ for CO_3 , and another, slightly lower than the origin peak, at 0.132, corresponding to $.333 - .201$ for Na. Many sets of F_c for the first seven orders of $000l$ to $l = 21$, over a range of values of z' about ± 0.010 about the indicated values, yielded a minimum of R at $z(\text{CO}_3) = 0.409$ and $z(\text{Na}) = 0.195$, within .001 of the values finally assigned to each.

Assuming a fixed C-O distance, the x and y parameters of oxygen depend only on the orientation of the CO_3 group. At first the C-O distance was chosen as 1.288, the "predicted" value (Sass *et al.*, 1957, p. 569); later the value 1.283, found by Steinfink and Sans (1959, p. 681) for dolomite, was used. The orientation of the CO_3 group need be examined only over a range of ϕ values of 30° . Among the $hki0$ reflections observable on Mo precession patterns, 4150 and 4510 as well as 5270 and 5720 (the latter pair observable by β radiation) are particularly sensitive to the orientation of the CO_3 group. The relations of these, $I_{(41\bar{5}0)} \gg I_{(4\bar{5}10)}$ and $I_{(52\bar{7}0)} \ll I_{(5\bar{7}20)}$, are compatible only with values of ϕ in the range 15° to 20° . Again, calculation of F 's for all nine observable independent $hki0$'s and comparison with observed F 's yielded a minimum R in the vicinity of 18° , corresponding to x and y for oxygen 0.293–0.295 and 0.092–0.101. Table 2 shows the observed and final calculated F 's for the two groups of reflections from which the trial parameters were chosen.

With approximate values thus found, F 's were calculated for half a dozen combinations of parameters over a small range for all possible reflections in the groups $h0hl$, $hh2hl$, $213l$, $2\bar{3}1l$, $314l$, $3\bar{4}1l$, $415l$, and $451l$ to $l = \pm 13$. Scattering factors were taken from

TABLE 2. Comparison of F_o and F_c . Values for Two Groups of Reflections Used in Selection of Trial Parameters for Eitelite

	F_o	F_c^*	F_o	F_c^{**}
0006	29.6	-29.1	11.0	20.9
00012	19.8	18.7	30.0	22.3
00015	22.6	22.2	22.0	24.7
00018	9.7	-11.8	41.0	18.4
	R = 3.8%		14.0	9.1
			33.0	9.5
			60.0	10.2
			52.0	6.8
			25.0	13.5
			R = 7.6%	
	* B = 0.7		** B = 0.4	

f-curves for Mg^{++} , Na^+ , O^- and C, constructed from data on pages 202–203, Vol. III, ITXRCr. Observations of intensities for the corresponding reflections were made on appropriate zero layer precession patterns, the series $000l$ from $l = 3$ to 21 being common to all of these patterns. After the elimination of (0003) and (21 $\bar{3}$ 4), for which observations of Cu and Mo patterns, corroborated by calculations, showed marked influence of the Renninger effect, there remained 100 independent reflections for testing the structure and adjustment of parameters. L. and P. corrections were made by means of a zero-layer Waser chart. For the equant, 0.1 mm diameter crystal used for the Mo precession patterns, μR is about 0.1; thus an absorption correction is not necessary.

Intensities were estimated visually by comparisons with standard scales, including one prepared by counted oscillations of an eitelite crystal. However, no standard could be prepared from the test crystal by precession methods, and comparison of several sets of observed F 's showed that discrepancies between them correspond to R values of 4 to 20 percent.² Accordingly, tests for adjusting the parameters were terminated when the R factor for 100 reflections of all types was brought to 13.6 percent after application of suitable temperature factors. For the $000l$'s a B value of 0.7 was indicated, whereas a value of 0.4 was indicated for the $hki0$'s. In lieu of full determination of the anisotropy of the temperature factor an average value of 0.05 for B was used for all remaining reflections.³

The parameters finally chosen are: Mg in $3a$ at 000 ; Na in $6c$ at $00z$, $z = 0.196$; C in $6c$ at $00z$, $z = 0.410$; oxygen in $18f$ at xyz , $x = 0.293$, $y = 0.092$, $z = 0.410$. These results are limited by the quality and extent of the data which did not suffice to detect differences in z for carbon and oxygen as might be expected from the surroundings of the CO_3 groups, Mg on one side, Na on the other. In the case of dolomite, Steinfink and Sans (1959, Table 1) found z for C to differ by 0.0005 from that of oxygen, the carbon being displaced 0.008 Å towards the level of the Mg's with respect to the level of the oxygens. To detect such a difference in eitelite, to establish fully the thermal anisotropy and to improve the param-

eters, refinement with better and more extensive data would be required.

Description of the Structure

In spite of the limitations just mentioned, the essential features of the structure seem well established. Figure 4 (top) shows the orientation of a CO_3 group. Due to centrosymmetry there is another CO_3 group on each trigonal axis turned an equal angle in the opposite sense. In the section parallel to $(\bar{1}2\bar{1}0)$ in Figure 4, all atoms within one rhombohedral cell are shown plus a few additional atoms in adjacent cells to account for the peaks appearing on the electron density curves on each side. These curves are based on summations of observed and calculated F 's for $000l$ from $l = 3$ to 21, comparable to the summations with more limited data used in the preliminary selection of parameters. Though there are minor differences in the shapes of the curves and the peak

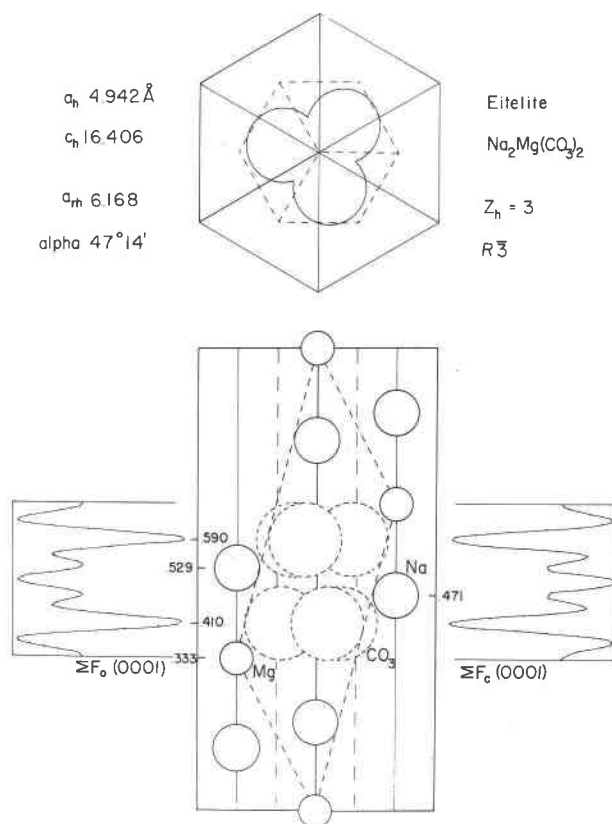


FIG. 4. Structure diagrams for eitelite.

Top: Projection onto (0001), showing orientation of CO_3 group at $z = .410$.

Below: Partial $(\bar{1}2\bar{1}0)$ section, flanked by curves for electron density projected onto c from $z = .333$ to $.667$.

Dashed lines in both parts outline primitive rhombohedral cell.

²This case would have been ideal for the use of Mo Weissenberg patterns, both to achieve better comparison of test and standard spots and to extend the range of $000l$'s for the one-dimensional summation, but limited access to equipment prevented this.

³A tabulation of F_o and F_c is available from the author.

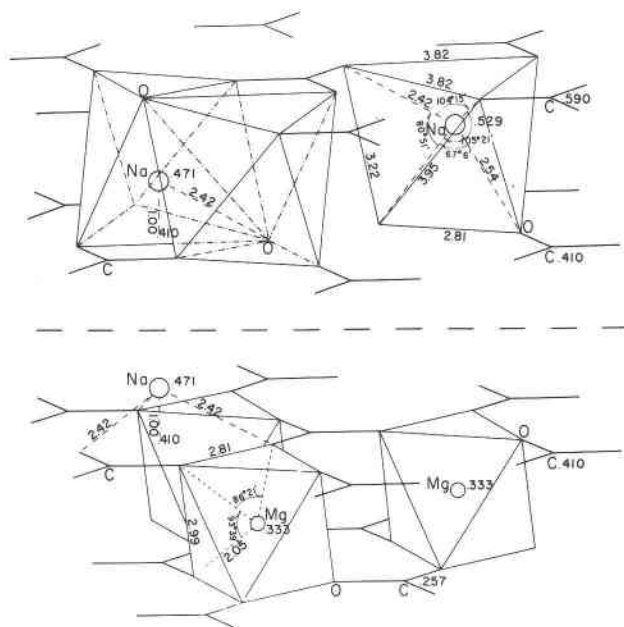


FIG. 5. Details of eitelite structure with interatomic distances and bond angles.

Below: Magnesium carbonate sheet between the levels of CO_3 groups at $z = .257$ and $.410$.

Above: Coordinations of Na ions between two magnesium carbonate sheets in the interval $z = .410$ to $.590$.

heights, the positions of the peaks are identical on both curves and correspond closely with the z parameters represented in the drawing.

From this picture it can be seen that the sequence of ions within the height $c/3 = 5.469 \text{ \AA}$, is $\text{Mg}^{++} - \text{CO}_3^{--} - \text{Na}^+ - \text{Na}^+ - \text{CO}_3^{--} - \text{Mg}^{++}$. The basal cleavage may be presumed to occur at the levels $1/6$, $3/6$ and $5/6$, between the adjacent Na^+ at slightly differing heights. The structure can then be thought to contain charged magnesium carbonate sheets parallel to the base, flanked by Na ions above and below, as represented by the sequence $\text{Na}^+ - (\text{CO}_3 - \text{Mg} - \text{CO}_3)^- - \text{Na}^+$.

The lower part of Figure 5 shows the $(\text{CO}_3 - \text{Mg} - \text{CO}_3)^-$ sheet that lies between levels $.257$ and $.410$ plus the adjacent Na^+ above at $.471$. The upper part shows the interval from $.410$ to $.590$ within which, at levels $.471$ and $.529$, lie Na^+ ions alternately attached to the lower magnesium carbonate sheet and to the next one above. The CO_3 groups at level $.410$ are common to the two parts of the picture and the Na^+ ion shown in the lower part is repeated in the upper part. Together these two parts show the atomic sequence over a z of $.333$,

after which the entire sequence is repeated by the rhombohedral translation.

Magnesium is in almost ideal octahedral coordination. The arrangement of Mg's and CO_3 's corresponds closely to that of Mg's and OH's in brucite in which Mg-O is $2.102(3) \text{ \AA}$ (Zigan & Rothbauer, 1967). In eitelite, neighboring Mg-centered octahedra share CO_3 groups at the corners but do not share O atoms at the corners so that there are no O-O edges shared between Mg-centered octahedra. In dolomite, Mg-O is 2.095 (Steinfink and Sans, 1959, p. 681). The structure of MgCO_3 has not been refined, but the Mg-O distance can be calculated with some confidence from cell dimensions and an assumed C-O distance of 1.283 . Mg-O in magnesite thus calculated is 2.10 using the cell dimensions given for synthetic material by Goldsmith and Graf (1958, Table 5) or 2.07 using the dimensions given in CRYSTAL DATA, 1963. The Mg-O distance found for eitelite, 2.053 , is but slightly below that found for dolomite or calculated for magnesite.

The more complicated structure of the interval $z = .410$ to $.590$ is shown in the upper part of Figure 5. The Na's at level $.529$ lie on the same trigonal axes as the Mg's at $.333$, whereas the Na's at level $.471$ lie on the same trigonal axes as the Mg's at $.667$ in the next sheet above. Na's are repelled by the Mg's directly below or above them to the far side of the interval between successive magnesium carbonate sheets. The irregular coordination polyhedron around each Na shares its smallest triangular face (equilateral with edge 2.81) with an Mg-centered octahedron. The distance from an Na to the oxygens at the corners of this triangle is 2.54 , and the vertical distance from the level of the CO_3 groups is 1.96 , whereas the distance to the nearest oxygens on the side to which the Na may be considered attached is 2.42 ; the vertical distance to the level of the CO_3 groups on that side is 1.00 , as indicated in the drawing.

Each of the Na-centered polyhedra at level $.529$ shares three lateral edges with Na-centered polyhedra at level $.471$. These edges (one being shown as a dot-dashed line between O's in Figure 5, top) are the longest O-O distances of the Na-centered polyhedra. Baur (1972, p. 723) has just emphasized that "Ionic structures with shared polyhedral edges and faces can only be stable if their geometry allows shortening of the shared polyhedral edges. When adjustment stresses force a shared edge to be long this is a particularly destabilizing feature of the crys-

tal structure." The latter is the case in eitelite, the "destabilization" being manifest as the {0001} cleavage which breaks the longer Na-O bonds. Alternatively, one might consider that the Na ions are strongly bonded only to the one side and that the irregular coordination polyhedron is a composite of two trigonal pyramidal coordinations, one involving strong bonds and one involving much weaker bonds; the long shared edges are a consequence of this situation.

Acknowledgments

Professor Charles Milton kindly provided the eitelite specimens as well as information that he and Mr. J. R. Dyni of the U. S. Geological Survey had obtained from study of the cores; he also lent the X-ray diffraction patterns of eitelite from the type locality. Thanks are due to Mr. J. Hampel for all of the photographs in this report. This study was supported by an NSF grant to Professor Milton and by research funds of the University of California, Berkeley.

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