A STATISTICAL MODEL FOR THE CRYSTAL STRUCTURE OF MULLITE

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A complete structure analysis is presented for mullite $(1.71Al_2O_3 \cdot SiO_2)$; the space group is confirmed as Pbam, the lattice constants being a = 7.549 ± 0.008; b = 7.681 ± 0.008; c = 2.884 ± 0.003 A. The pattern is deduced from a $\sigma(x,y)$ projection, and is refined by means of difference syntheses $F_0 - F$ and least-squares methods; R = 0.095.

The structure is very similar to that of sillimanite (Taylor); the main difference is that only 84.2% of the positions for oxygen atoms joining $(Al, Si)O_4$ tetrahedra are filled, which is in accordance with the chemical composition. This results in displacement of 15.8% of the Si and Al atoms from the positions they take in sillimanite; they go to tetrahedral positions that remain vacant in sillimanite.

The formula should be put as

$$Al\,[Si_{2-x}Al_xO_{11} {\scriptstyle\frac{11}{2} - \frac{x}{2}}],$$
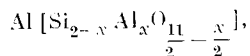
in which $1.25 \leq x \leq 1.40$ for mullites in the range $\frac{3}{2}$ to $\frac{2}{1}$.

1. Introduction

The diffraction patterns of mullite and sillimanite are very similar, so the structures are very similar, although the two differ greatly in physical and chemical properties; this makes the pair of great interest in chemical crystallography. Moreover, mullite has some exceptional properties, especially heat resistance and the power to form a wide range of solid solutions with the oxides of various elements; this makes it a principal component of porcelain, fireclay, and aluminous refractories generally, and these uses make a knowledge of the structure of some practical importance.

In 1928, Taylor [1] deduced the structure of sillimanite by trial and error, and from this the structure of mullite, because the rotation patterns are similar. He and Hey [2] gave a revised structure for sillimanite in 1930, and one for mullite in 1932 [3], the latter being such as not to conflict with the laws of classical structural crystallography. This model has infinite chains of AlO_6 octahedra, which meet along their edges; the chains are joined via double sets of SiO_4 tetrahedra (Fig. 1), in which half of the Si atoms are replaced by Al. These double sets (chains) are not the same as those in amphibole but are (Belov [4]) a new type of silicate radical $[SiAlO_5]^{3-}$ (the sillimanite radical).
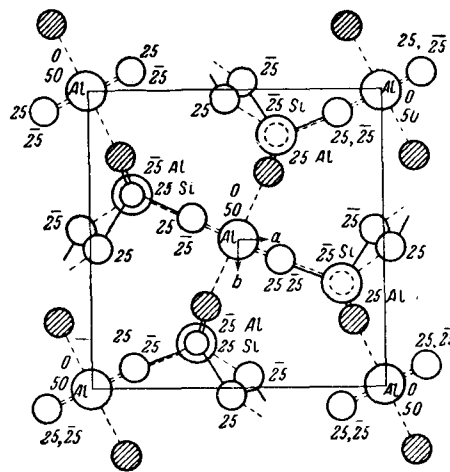

Fig. 1. Structure of sillimanite [2].

The space group of sillimanite is Pbnm; Z = 4; the measured density is 3.24 g/cm³; and the lattice constants are a = 7.43; b = 7.58; c = 5.74 A (these were later revised [5-8]).

Mullite is more troublesome, for Bowen and Greig [9] determined its composition as $3Al_2O_3 \cdot 2SiO_2$, which,

with the measured density of 3.15 g/cm^3 and the con-
stants a = 7.48, b = 7.63, c = 2.87 A [1], gives integers
for the numbers of Si and Al atoms in the unit cell but
not an integer for the O atoms. The results was inex-
plicable in terms of classical structural concepts, so
Taylor [3] proposed a model having a unit cell of sides
2a, 2b, and c (in relation to sillimanite). The general
array was as in sillimanite, but the $3Al_2O_3 \cdot 2SiO_2$
formula forced him to replace four Si by Al and to ex-
clude two of the four O atoms that previously joined the
SiO_4 tetrahedra (Fig. 2). This model did not agree with
the observed diffraction pattern; moreover, it implied
that each of these two O atoms is surrounded by four Al
atoms all in one plane, which would make the O quadri-
valent.

This feature formed the basis of a revision by War-
ren [10], who in 1933 proposed a model having a unit
cell of dimensions 2a, 2b, and c/2. This model was
dominated by the structure elements of sillimanite, but
some of the Al atoms were located in tetrahedral holes
that remain vacant in sillimanite (Fig. 3). This gave
the normal valency for oxygen, but it did not agree
with the diffraction pattern.

These attempts to fit mullite to classical models
were entirely justified, for little was then known about
structures with randomly filled equivalent positions.

This possibility was demonstrated in 1937 by Eitel
[11], who expressed the view that mullite must have a
defect structure (one with certain positions randomly
filled). He also explained the incongruent melting in
terms of the defect structure.

None of the later papers contains a fresh discussion
of the structure, although much fresh evidence accumu-
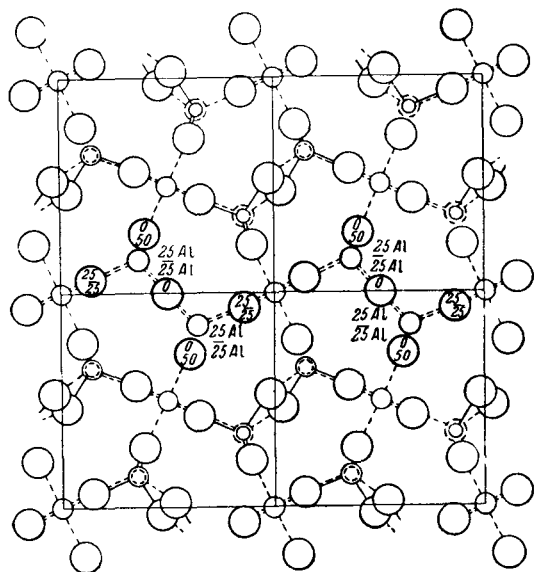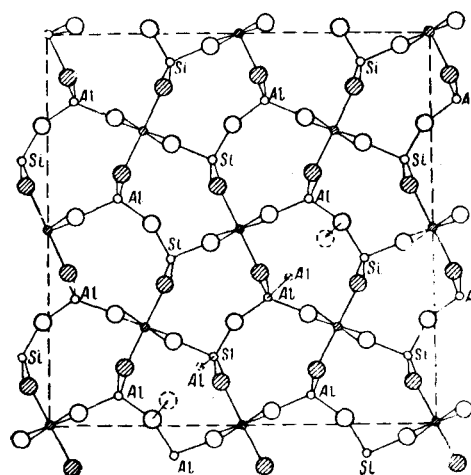lated. Kurilenko [12] and Scholze [5] found that the

Fig. 3. Structure of mullite [10].

odd layer lines on the [001] rotation pattern of mullite
are made up of diffuse spots and are very weak relative
to the normal layer lines; these diffuse spots do not cor-
respond to integral h and k. Agrell and Smith [8] con-
firmed this in 1960; they found that some specimens
give sharp layer lines, but not ones the same as those of
mullite. These they called S-mullites (for sharp) to
distinguish them from D-mullites (diffuse).

The diffuse lines clearly imply that the structure is
disordered.

It has been found [6-8, 13-22] that the $3Al_2O_3$
$2SiO_2$ composition (this is termed $^3/_2$) is not the only
one; the range $^3/_2$ to $^2/_1$ occurs. Some [14, 23, 24] con-
sider that there are mullites containing more SiO_2 even
than in $^3/_2$, but this view is not generally accepted [6-8,
15, 16, 21]. The lattice constants increase linearly with
the Al_2O_3 content [7, 8], with a increasing most rapidly,
although the thermal history of the material complicates
the picture [21].

The $^2/_1$ mullite is that with the most Al_2O_3; it has
been given the name praguite [14] on the basis that its
powder pattern differs slightly from that of standard
melted mullite ($^3/_2$), but no account was taken of Rooks-
by and Partridge's work [13], which deals with differences
in the powder patterns of α-mullite ($^3/_2$) and β-mullite
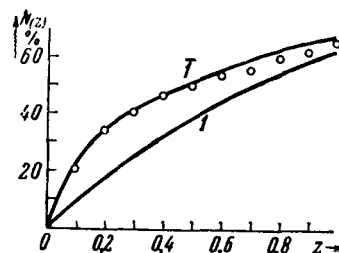($^2/_1$).

Fig. 2. Structure of mullite [3].

Fig. 4. Theoretical and measured
intensities for 1 and $\bar{1}$ for mullite.

This short survey is far from exhausting the problems that demand a review of the structure; the most important ones here are the type of melting [14-16, 18 to 20, 25-27], the solid solutions with oxides [7, 8, 28 to 30], and the thermal conversion of sillimanite to mullite [28].

2. Structure Analysis

We used a monocrystal of electrically melted mullite of composition 25.61% SiO_2, 73.42% Al_2O_3, 0.84% Fe_2O_3, and 0.13% TiO_2.* Copper radiation was used to record Weissenberg patterns of hk0, hk1, hk2, h0l, and 0kl types; molybdenum radiation was used to record precession patterns for hk0, h0l, and 0kl reflections. These gave a = 7.549 ± 0.008, b = 7.681 ± 0.008, c = 2.884 ± 0.003 A, which values agree well with earlier ones [5,6,8]. The rotation patterns on [001] showed no odd layer lines, but the oscillation photographs (angle of oscillation 20°, exposure 60 hr) showed diffuse lines as before [5, 8, 12]. Our specimen was a D-mullite [8]; the systematic absences correspond to space group Pbam and Pba2. These groups do not fit the 010, 030, and 032 spots, but these were very weak, extraordinarily sharp, and dependent on the type of pattern. Ewald's construction revealed them as false spots resulting from double reflections from plane pairs 121 and $\bar{1}\bar{1}\bar{1}$, 220 and $\bar{2}10$ (precession), and $\bar{1}21$ and 111.

Suspension in Clerici solution gave the density as 3.150 ± 0.002 g/cm³, which corresponds to a unit cell containing the following numbers of atoms: 1.352 Si, 4.570 Al, 0.034 Fe, 0.005 Ti, and 9.621 O. The sum Si + Al + Fe + Ti is very close to six, as Taylor [1] expected and as Agrell and Smith found [8]. Further work was based on the composition 1.36 Si, 4.64 Al (which includes the Fe and Ti), and 9.68 O; the calculated density is 3.171 g/cm³. This makes the material a $^{1 \cdot \pi}\!/_1$ mullite.

The intensities were measured photometrically to 7% on integrated Weissenberg patterns; only the very weak spots were estimated visually. In both cases, we used separately recorded scales, which were developed along with the patterns. The values were corrected for the Lorentz polarization, and absorption factors, and then were converted to absolute terms by Wilson's method. This gave us 71 independent structure factors of hk0 type (including five zero ones), 70 of hk1 type (12 zero), 53 of hk2 type (four zero), and 17 of h0l type (one zero). These were used in F_{hk0}^2 and F_{h0l}^2 to construct P(u, v) and P(u, w), and all were used in a statistical test for a center of symmetry [32] (Fig. 4). The P(u, v) was readily interpreted in terms of Taylor's model, while P(u, w) showed that the z can be only 0 and ½. This also fits the structure of sillimanite, in which z can be only 0, ¼, ½, and ¾, but the lattice constants must be doubled. Of course, 17 factors are too few to give a proper P(u, w), so these z were adopted only as a work-

ing hypothesis, which was confirmed in the later stages by means of the hk1 and hk2 factors.

Figure 4 shows that the experimental points lie close to the curve corresponding to a center of symmetry; this is not a complate proof that $\bar{1}$ is present (and that the space group is Pbam), but it does imply that most of the atoms are related in pairs by such centers.

A weighted combination of 0.34 Si with 0.66 Al [denoted by (Al,Si) in what follows] was used in accordance with the $^{1 \cdot \pi}\!/_1$ composition; this has tetrahedral coordination, and only the Al has octahedral. The O atoms joining the SiO_4 and AlO_4 tetrahedra were placed on two-fold axes (coordinates 0 ½ ½), the repeat distance along c being equal to the lattice constant. The other O atoms, together with the Al, have the same repeat distance in sillimanite, and Al differs little from Si in scattering power, so this model fits our diffraction patterns, the density being as found and the space group being Pbam. This enabled us to deduce the signs and so to construct an electron-density projection, which revealed all atoms well. Here R was 0.28.

This made it clear that the structure has the general lines of that of sillimanite [1] and that the finer details of both structures must be examined. To this end we referred the σ(x,y) pattern to the absolute scale and deduced the number of electrons for each position. This revealed that: a) The numbers are closer to those for neutral atoms than to those for ions; b) the peak for O_{III} is diagonally elongated (Fig. 5a) and has an electron number less than 8; c) the (Al, Si) peak corresponds to less than 13.3 electrons (0.34 Z_{Si} + 0.66 Z_{Al} = 13.3); and d) the low peak near (Al, Si), which we denote by (Al, Si*), corresponds to 2-3 electrons (height of peak about 5 el/A²), and cannot be interpreted as a false peak resulting from termination of the Fourier series, for there are no peaks as high as this (apart from the atoms already interpreted) in the entire pattern. Also, this peak lies at the middle of a tetrahedral hole, which is not filled in sillimanite.

These facts led to the following deductions: a) Mullite is not solely ionic; b) the O_{III} position, which we assumed to be filled by two O atoms at 0 ½ ½, is randomly filled by only 1.68 atoms, in accordance with the $^{1 \cdot \pi}\!/_1$ composition and with the need to eliminate some of the O when Al replaces Si (relative to sillimanite; there is also the deformation of O_{III} to consider); and c) the coordination tetrahedran around (Al, Si) is left unclosed if the atom O_{III} is lost. The result of the last would be to displace the central atom to a hole not filled in sillimanite. Then, if O_{III} is 84.2% filled in

*Preliminary results have been given already [31];these are all confirmed, except those on praguite, for the initial chemical analysis was incorrect.
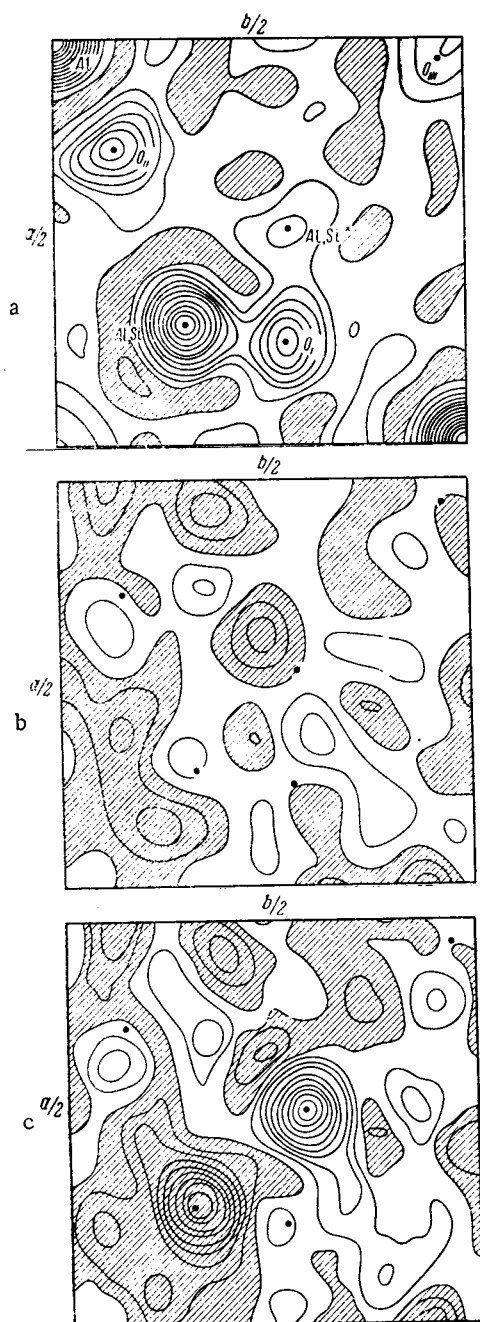
Fig. 5. Fourier syntheses for mullite:
a) $\sigma(x, y)$, lines at intervals of 2.5 el/A^2,
negative regions hatched; /) final F_0 - F
synthesis for hko, lines at intervals of
0.5 el/A^2 model 5 of Table 3); c) F_0 - F
synthesis without allowance for the ran-
dom filling of the (Al,Si) and (Al,Si) *
positions (model 6 of Table 3), lines at
intervals of 0.5 el/A^2.

this 1.71/1 mullite, the (Al,Si) position should also be
84.2% filled with the (Al,Si*) position 15.8% filled. This
gave a great improvement in R in further structure refine-
ment. The $\sigma(x,y)$ was refined by means of F_0 - F syntheses and

least-squares methods until all signs remained constant
and F_0 - F became free from gradients at the positions
for the atoms. Geometrical considerations indicate that
the distorted O_{III} peak consists of two almost coincident
ones of coordinates 0.025 and 0.475 (the fourfold posi-
tion in group pgg). No further refinement by means of
temperature factors and least squares was attempted, for
the structure factors were few in number. Then the
value for R = $\Sigma \|F_b| - |F\| / \Sigma |F_b|$, in the final stage,
came out as 0.091 (or 0.084 if the zero reflections are
omitted). Figure 5a shows the final electron-density
projection; Fig. 5b shows the final F_0 - F synthesis.

The P(u, w) projection and the resemblance to sil-
limanite gave the z as Al ~ 0, Al, Si $\sim \frac{1}{2}$, Al, Si* $\sim \frac{1}{2}$,
$O_I \sim 0$, $O_{II} \sim \frac{1}{2}$, $O_{III} \sim \frac{1}{2}$; these values were checked
(apart from the use of the few F_{h0l}) by comparing the
F_{hk1} and F_{hk2}; in addition, $\sigma_1(x,y)$ and $\sigma_2(x,y)$ weighted
projections were constructed. The peaks in $\sigma_*(x,y)$ are
reproduced in $\sigma_2(x, y)$ and become minima in $\sigma_1(x,y)$
at the points where z = $\frac{1}{2}$ was expected. The (Al,Si*)
peak was confirmed on all projections; the R for the
F_{hk1} was 0.097 (0.083) and that for F_{hk2} was 0.097
(0.093), which correspond precisely with the R for the
F_{hk0}. The F_0 - F syntheses for F_{hk1} and F_{hk2} confirmed
all the coordinates.

The best agreement between the observed and cal-
culated structure factors was obtained by using weight-
ing factors for the (Al, Si), (Al, Si*), and O_{III} positions,
which indicates that these positions are randomly filled.
Of course, the defect structure still fits space group Pbam.

Figure 6 shows the results of the structure analysis
in terms of coordination polyhedra projected along c;
Fig. 7 shows an axial view of the structure. The loss of
O_{III} and the displacement of (Al, Si) are clear. Table 1
gives the atomic coordinates for mullite and, for com-
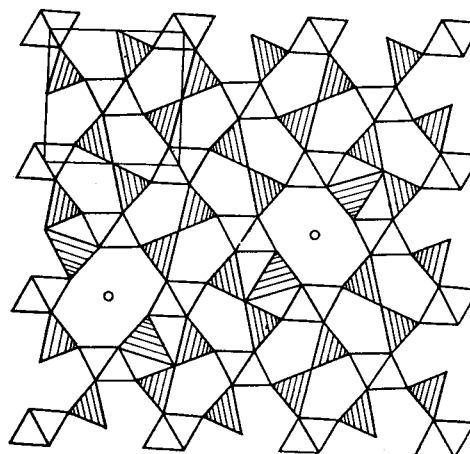parison, those of sillimanite [2]. The probable errors

Fig. 6. Projection of the structure of mullite on
on the xy plane represented with coordination
polyhedra.

TABLE 1. Atomic Coordinates for Mullite (Present Work) and for Sillimanite [2]

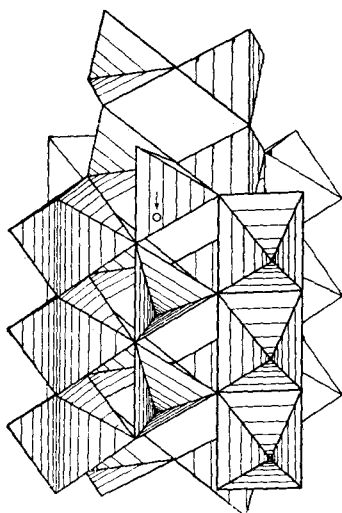| Atom	Mullite			Sillimanite			Symbol [2]
	x	y	z	x	y	z	
Al	0	0	0	0	0	0	Al_I
Al, Si	0.352±0.001	0.161±0.001	1/2	0.36	0.15	1/4 3/4	Al_{II} Si
Al, Si*	0.233±0.005	0.287±0.005	1/2	—	—	—	—
O_I	0.374±0.003	0.279±0.003	0	0.39	0.28	0	O_{IV}
O_{II}	0.138±0.003	0.078±0.003	1/2	0.15	0.07	1/4 3/4	O_I O_{II}
O_{III}	0.025±0.003	0.475±0.003	1/2	0.03	0.47	1/4	O_{III}

Fig. 7. Crystal structure of mullite represented in coordination polyhedra.

in the coordinates were deduced by Vainshtein's method [33]. Table 2 gives the observed and calculated structure factors.

3. Discussion

The essential difference between sillimanite and mullite is that the latter has its (Al, Si), (Al, Si*), and O_{III} positions only randomly filled. Consider now the O_{III} position; the peak is interpreted in terms of two atoms, not of one distorted by a very anisotropic temperature factor, for sillimanite† gives a similar peak (although here we have four O atoms in positions having coordinates of 0.03, 0.47, $1/4$, and the equivalent positions in group Pbnm [2]). The O atoms lie around a twofold screw axis with a repeat distance of 5.77 A, in accordance with the regular sequence of Si and Al atoms in the tetrahedral positions. Mullite has its Si and Al in the ratio of 34:66, so the O atoms form an irregular array near the screw axis. The array has no definite repeat distances, and so cannot contribute to

the odd layer lines (which it does for sillimanite). The array contains only 1.68 oxygen atoms in the fourfold positions 0.025 and 0.475, as appears in $\sigma(x,y)$, in accordance with the $^{1 \cdot 71}/_1$ composition; the above interpretation is then the most probable. If the filling of that position has any degree of order, we would expect to find diffuse layer lines; if the order is such as to make the 5.77 A repeat distance dominant, we should get sharp odd layer lines. The explanation of the S and D mullites probably lies here.

The most important result from the present work is that the (Al, Si) and (Al, Si*) positions are filled randomly to the extent of 84.2 and 15.8%, respectively. This has been checked by means of the R for various interpretations of Fourier syntheses made with various weighting factors; Table 3 gives the results, which show that model 5 is the most probable. The $F_0 - F$ synthesis (Fig. 5c) confirms this; here, the F were calculated from model 6 of Table 3. Similar syntheses for the F_{hk1} and F_{hk2} gave exactly the same result.

The defect structure explains the isomorphous series from sillimanite to $^2/_1$ mullite [34]. Our work indicates that the members of this series differ only in the number of O atoms that have left the O_{III} position and in the corresponding number of (Al, Si) atoms that have moved to (Al, Si*). This needs a final structure analysis on $^2/_1$ mullite to confirm it; the reason why $^2/_1$ mullite is an end-member of the series may well be that the structure becomes unstable if more than 20% of the O_{III} positions are unfilled.

It is convenient to denote mullites by the Al_2O_3/SiO_2 ratio as above, as others have done already [6]; S and D can also be used [8]. For example, our specimen is a 1.71/1-D mullite. Other components $(Fe_2O_3, Cr_2O_3, TiO_2,$ and so on) must be denoted specially.

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† Work on sillimanite was in hand in our laboratory at the same time; the results at present (R = 0.071) confirm Hey and Taylor's structure, but with improved atomic coordinates. The (Al,Si)* peak does not appear here.

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TABLE 2. Comparison of Measured and Calculated Structure Factors

hk0	F_o	F	hk0	F_o	F	hk1	F_o	F
20	7.9	10.1	28	17.5	14.2	56	5.8	7.5
40	33.0	−30.4	38	4.2	−4.2	66	0	−1.0
60	37.4	36.5	48	15.2	15.2	76	0	−5.2
80	36.4	35.8	58	16.1	15.4	17	20.2	20.9
11	19.8	18.0	19	23.0	21.5	27	14.8	−16.1
21	39.7	38.8	29	11.8	10.5	37	0	−0.7
31	4.8	7.4	39	6.1	−8.1	47	7.5	8.7
41	9.5	−9.5				57	12.7	10.6
51	6.5	8.2				67	10.9	12.8
61	21.8	−23.2	R_{hk0}=0,091 (0,084)			08	19.8	20.5
71	8.1	9.3				18	7.6	6.5
81	15.8	14.3	hk1	F_o	F	28	5.9	5.9
91	11.6	12.8				38	4.9	−6.5
02	0	2.5	20	28.4	25.9	48	0	0.5
12	27.5	−31.0	40	27.8	26.5	58	4.4	−4.6
22	33.8	34.0	60	6.6	−7.8			
32	11.8	−13.6	80	0	1.3			
42	33.2	33.2	11	26.3	26.2	R_{hk1}=0,097 (0,083)		
52	34.2	31.1	21	8.9	4.0			
62	12.7	13.3	31	19.2	17.7	hk2	F_o	F
72	4.3	−4.7	41	8.5	7.9	20	3.4	5.0
82	0	1.7	51	18.0	19.0	40	13.6	−17.5
92	15.7	−14.6	61	0	0.4	60	28.4	27.8
13	22.4	22.5	71	16.8	15.6	80	28.1	30.7
23	35.1	−31.8	81	14.0	−12.6	11	10.6	9.3
33	10.5	−11.7	91	4.0	6.3	21	24.0	23.6
43	2.6	0.3	02	4.6	0.3	31	11.4	10.9
53	15.5	14.1	12	34.2	33.6	41	7.8	−7.4
63	13.5	16.3	22	5.2	3.5	51	6.8	5.0
73	26.4	27.1	32	19.9	20.1	61	16.6	−17.1
83	5.0	−6.5	42	26.1	23.8	71	7.6	7.6
04	14.0	14.2	52	37.3	−34.8	81	9.6	11.6
14	8.6	−10.4	62	10.1	11.5	02	0	0.5
24	24.6	21.8	72	0	0.5	12	19.2	−18.8
34	0	1.3	82	6.1	3.0	22	23.8	22.9
44	28.9	28.5	92	18.9	17.6	32	10.2	−9.7
54	3.8	−3.8	13	0	−2.0	42	23.3	23.0
64	0	0.2	23	0	−0.7	52	26.5	23.9
74	9.7	12.3	33	50.1	50.0	62	11.0	10.3
84	11.2	10.8	43	0	0.5	72	6.3	−3.9
15	6.6	−4.3	53	17.9	19.6	13	20.6	21.5
25	25.1	−24.3	63	0	0.8	23	17.1	16.1
35	19.3	20.4	73	8.7	−8.2	33	7.5	7.5
45	13.7	12.3	83	4.8	5.8	43	0	0.3
55	6.6	6.0	04	41.1	39.1	53	13.1	12.3
65	9.6	8.3	14	8.6	−9.7	63	10.4	12.7
75	4.0	3.5	24	8.2	9.5	73	19.7	20.6
85	18.0	−16.9	34	12.0	−11.0	04	6.9	9.7
06	23.6	21.5	44	15.4	−17.2	14	2.4	−1.6
16	15.3	14.2	54	15.3	13.6	24	18.1	16.7
26	10.3	11.9	64	23.2	22.3	34	2.6	0.9
36	4.6	5.0	74	5.1	−4.6	44	23.9	23.9
46	8.7	9.2	84	19.4	18.0	54	4.4	−4.0
56	3.2	−3.1	15	33.1	34.9	64	0	0.9
66	23.6	20.6	25	36.0	36.4	74	6.6	10.7
76	4.7	−3.1	35	4.9	−5.9	15	8.4	6.2
17	9.3	−11.0	45	12.2	−11.7	25	20.1	−18.7
27	11.9	12.8	55	8.1	6.9	35	16.6	17.4
37	36.9	34.2	65	17.6	−17.4	45	11.5	10.0
47	9.1	−9.7	75	24.7	24.4	55	6.9	6.0
57	16.7	18.6	06	5.2	−5.1	65	6.8	6.6
67	7.8	−9.4	16	0	−0.1	06	19.1	18.0
08	0	1.3	26	15.3	13.4	16	11.4	10.8
18	20.0	−18.6	36	6.0	7.3			
			46	21.3	21.1			

TABLE 2 (continued)

$hk2$	F_O	F	$hk2$	F_O	F	$h0l$	F_O	F
26	8.2	9.1	$R_{hk2}=0.097$ (0.093)			22	4.0	5.0
36	4.5	3.5				42	18.4	—17.5
46	7.5	7.5	$h0l$	F_O	F	62	22.8	27.8
56	3.7	—2.5				82	27.2	30.7
17	6.7	—8.8	01	33.0	—29.1	03	17.7	—14.9
27	10.0	9.7	21	30.4	25.9	23	20.1	16.6
37	27.4	28.5	41	25.9	26.5	43	19.7	17.5
47	6.0	—8.0	61	8.2	—7.8	$R_{h0l}=0.093$ (0.090)		
08	0	0.8	81	0	1.3			
18	13.5	—12.9	02	70.0	76.5	$R_{hkl}=0.095$ (0.086)		

TABLE 3. R for Several Interpretations of σ (x, y), σ_1 (x, y), and σ_2(x, y).

№	Filling of position , %							R	
	Al	Al, Si	Al, Si*	$O_{III}^{0;1/2}$	O_{III} 0.025; 0.175, 1/2	O_I	O_{II}	with zeros	without zeros
1	100	100	—	100	—	100	100	0.239	0.212
2	100	84.2	15.8	100	—	100	100	0.172	0.154
3	100	84.2	15.8	84.2	—	100	100	0.142	0.126
4	100	84.2	15.8	—	50	100	100	0.107	0.097
5	100	84.2	15.8	—	42.1	100	100	0.095	0.086
6	100	100	—	—	42.1	100	100	0.206	0.192

Work on sillimanite was in hand in our laboratory at the same time; the results at present (R = 0.071) confirm Hey and Taylor's structure, but with improved atomic coordinates. The (Al,Si) peak does not appear here.

The formula of mullite may be related to that of sillimanite, Al[SiAlO$_5$]; mullite has Al replacing some Si, and [SiAlO$_5$]$^{3-}$ is derived from the hypothetical [Si$_2$O$_5$]$^{2-}$, so the formula may be put as

$$\mathrm{Al}\,[\mathrm{Si}_{2-x}\mathrm{Al}_x\mathrm{O}_{11\,-\,\frac{x}{2}}\,],$$
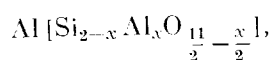
in which 1.25 ≤ x ≤ 1.40 for mullites in the range 3/2 to 2/1.

Table 1 gives the interatomic distances computed from the coordinates; these agree generally with those given in the literature, and a more detailed analysis is not justified until further structure factors are available from the later stages of the present work on sillimanite and mullite. I observe merely that the mean distance between (Al, Si) and the O atoms at the corners of the tetrahedra is 1.72 ± 0.03 A, which agrees well with the 1.72 ± 0.02 A calculated by Smith's method [35] for Al:Si = 66:34 in the tetrahedra.

4. Conclusions

A structure analysis has been performed on 1.71/1-D mullite; it has been found that the structure is very similar to that of sillimanite, the difference being that some of the O atoms are missing from the O$_{III}$ positions, and that the Al and Si atoms are corresponding-ly displaced from the (Al, Si) position into the (Al,Si*) tetrahedral position, which is not filled in sillimanite. The structure is a combination of Taylor's model (which describes the general features), Warren's (for the displacement of the Al and Si), and Eitel's (for the random defect structure).

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