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Neutron and X-Ray Crystallographic Studies on Indium Oxide Hydroxide

M. S. LEHMANN, F. KREBS LARSEN, F. REIBKE POULSEN, A. NØRLUND CHRISTENSEN and S. E. RASMUSSEN

Department of Inorganic Chemistry, University of Aarhus, DK-8000 Aarhus C, Denmark

Crystals of indium oxide hydroxide large enough for neutron crystallographic studies were grown by hydrothermal techniques. The crystal structure was determined using both neutron and X-ray techniques and the position of the hydrogen atom was determined unambiguously. A dominant feature in the structure is a short linear hydrogen bond with an O–O distance of 2.54 Å and an O–H distance of 1.08 Å.

The space group was determined as $P2_1nm$ with $a=5.26$ Å, $b=4.56$ Å, $c=3.27$ Å. The X-ray data were refined to a conventional R-value of 5.1 % and the neutron data to 4.2 %.

In a previous X-ray investigation [1] of indium oxide hydroxide, InOOH, a short oxygen-oxygen distance of 2.58 Å was found. This investigation stimulated an attempt by Schwarzmann, Glemser and Marsmann [2,3] to determine the hydrogen positions in indium oxide hydroxide from infrared and proton resonance spectra on InOOH and InOOD. The interpretations of Schwarzmann *et al.* were criticized by Christensen, Rasmussen, Nevald and Frank.[4] The system $In_2O_3-H_2O-Na_2O$ has been investigated extensively in this laboratory.[5] During crystal growth experiments on this system crystals of indium oxide hydroxide which were larger than the one used in Ref. 1 were obtained. A crystal large enough for making neutron crystallographic measurements was grown. The X-ray photographs from the larger crystals showed that the space group extinctions given in Ref. 1 were not correct. The structure reported earlier is, however, correct in principle. The improved structural description including an unambiguous location of hydrogen positions is reported below.

EXPERIMENTAL

Chemistry. Hydrothermal experiments were performed in pressure bombs lined with pure gold. Experimental conditions are given in Table 1. The volume of the pressure bomb was 260 ml in the experiments numbered 1 and 2. In all other experiments the

Table 1. Experimental conditions for crystal growth by transport reactions in the system $In_2O_3-H_2O-Na_2O$.

Expt. No.	Temp.[a] °C	Pressure atm	Time h	NaOH M	Initial conditions	Product, Crystal size mm	
						Upper part of bomb	Lower part of bomb
1	295	80	272	2	In_2O_3	InOOH 0.4 mm needles	InOOH
2	380	250	100	4	InOOH	InOOH	In_2O_3
3	378	2900	148	1	$In(OH)_3$	InOOH+In_2O_3	
4	377	2800	148	2	$In(OH)_3$	InOOH+In_2O_3	
5	362	2900	148	3	$In(OH)_3$	InOOH $0.8\times0.3\times0.3$ mm³	
6	366	2800	148	4	$In(OH)_3$	InOOH+In_2O_3	
7	355	3400	504	2	$In(OH)_3$	InOOH	InOOH+$In(OH)_3$
8	355	2800	240	2	$In(OH)_3$	$In(OH)_3$	In_2O_3
9	350	2800	504	2	$In(OH)_3$	InOOH $0.4\times0.5\times0.7$ mm³	InOOH
10	350	2800	504	2	$In(OH)_3$	InOOH	In_2O_3

[a] Temperature in the lower part of the pressure bomb.

volumes of the bombs were 7 ml. The materials used in the charges were in all cases crystalline products obtained in previous hydrothermal experiments.

X-Ray technique. A Buerger-Supper equi-inclination diffractometer was used. It was a version automated by a Pace control unit and equipped with a Picker radiation analyzer and scintillation counter. Mo$K\beta$ radiation monochromated by reflection from a LiF crystal was employed since the InOOH crystal gave strong reflections at Bragg-angles so high as to make the $\alpha_1\alpha_2$ separation troublesome. The monochromator device will be described in a forthcoming publication.[5] The pulse height analyzer was set to transmit 99 % Mo$K\beta$ radiation. A total of 3100 reflections were measured within a hemisphere of radius $\sin\theta/\lambda = 1.4$ Å^{-1}. In general four symmetry related reflections were measured in each layer line. The ω-scan technique was used with a scan angle of 2.0 degrees for all layer lines except $l=8$, for which it was 2.5 degrees. Background measurements were taken for 15 sec before and after each scan.

The diffractometer data were reduced to relative structure factors using an ALGOL program [7] which evaluated intensities, calculated averages over symmetry related reflections, applied Lp-corrections and found standard deviations. 758 symmetry independent reflections were obtained. Of these 748 had an intensity greater than twice their standard deviation as estimated from the counting statistics. Since the crystal was irregularly shaped and very small and furthermore surrounded by Canada balsam, no absorption correction was attempted.

Neutron technique. The neutron diffraction data were collected on an automatic Hilger-Ferranti four-circle diffractometer located at the DR 3 reactor at the Danish Atomic Energy Commission Research Establishment, Risø.

The wavelength of the monochromatic neutron beam was 1.022 Å, and the neutron flux at the specimen was 10^6 n/cm²/sec. The reflections were measured by the ω-2θ scan technique and the counts for each step of size 0.04 degrees were recorded. The data collection time for one reflection was approximately 2 h. The setting for reflection was the symmetrical A-setting (Furnas and Harker).[8]

A single crystal (from experiment No. 9, Table 1) with a volume of 0.14 mm³ and linear dimensions in the range 0.4–0.7 mm was used in measuring a total of 250 reflections. The reflections were recorded during two periods of measurement, 73 in one period and 177 in the other. The 250 reflections measured were chosen partly to give the expected

strong reflections and partly to give even distribution throughout reciprocal space. The ratio between the scale factors for the two blocks of data was determined as 1.03. This value was kept fixed in the refinements. The diffractometer data were reduced to relative structure factors using an ALGOL program, DRAM,[9] which calculates intensities, Lorentz factors, and standard deviations. Averaging over symmetry related reflections gave 170 symmetry independent reflections. From these 11 were excluded because the intensities were found to be less than their standard deviation estimated from counting statistics, and 15 reflections with $\sin\theta/\lambda > 0.83$ were excluded because the reflections at high $\sin\theta/\lambda$ had broad and illshaped peaks, which made the assessment of the background difficult. The reflections were not corrected for absorption; μ (calc.) = 3.79 cm^{-1}.

STRUCTURE DETERMINATION

X-Ray results. The X-ray photographs indicated an orthorhombic symmetry. $h0l$ reflections were systematically absent for $h+l=2n+1$. Hence the space groups $Pmn2_1$ (No. 31) or the alternative setting $P2_1nm$ or $Pmnm$ (No. 59) were indicated, rather than the previously reported space group $Pnnm$ (No. 58). The former structure investigation was based upon 56 reflections recorded on film and measured by photometry. The results reported here were obtained from 748 reflections measured by X-ray counter methods and from 144 reflections obtained by neutron diffractometry. The X-ray results will be discussed first as they were obtained before the neutron results were available.

The density 7 g/cm^3 indicates the presence of 2 indium, 4 oxygen, and 2 hydrogen atoms per unit cell.[1] The indium atoms must occupy twofold positions and whatever the choice of space group only one parameter, y, is left to be determined from the Patterson function. Only one Patterson peak at $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ is big enough to be interpreted as an indium-indium vector and the coordinates of indium are chosen as $(0,\frac{1}{4},0)$. The arrangement of the indium atoms is centrosymmetric in all the possible space groups. The y-coordinate of the indium atom and an isotropic temperature factor were refined by a method suggested by Bhuiya and Stanley,[10] where the residual R' ($R' = \sum(|F_o| - k|F_c|)^2 / \sum|F_o|^2$, F_o and F_c are observed and calculated structure factors, k the scale factor) is minimized by finding for one atom at a time the position giving the lowest value of R', when the atomic position is varied within a given range. A program, D445, written by Danielsen [11] was used for this refinement. A minimum was found at $y = 0.2648$ giving a conventional R-value of 10.4 %. A Fourier synthesis was calculated phased on the location of the indium position. Thus the Fourier map exhibited $Pmnm$ symmetry. Peaks high enough to indicate oxygen positions were found only on planes near $z = 0$ and $z = 0.5$. Peaks at distances smaller than 2 Å from the indium were discarded. The four oxygen atoms in a unit cell would not be expected to be strictly equivalent. One would rather assume that they were paired in such a way that two oxygen atoms were belonging to hydroxyl groups. Two fold positions with $z = 0$ are not consistent with observed peaks in the Fourier when the space group $Pmnm$ is assumed. The structure could be described in the space group $P2_1nm$ without difficulty assuming that all atoms occupy positions of two-fold multiplicity.

Refinements were carried out in the space group $P2_1nm$ on the basis of a structure which corresponds to the earlier structure determination, the main difference being a shift in origin of approximately $-b/4$. In the refinement

the y parameter of indium was allowed to vary as were the x and y parameters of each of the oxygen atoms. Anisotropic temperature factor parameters and a scale factor were included too.

Two least squares programs were used. One is the fullmatrix least squares program ORFLS [12] as found in the X-ray 63 system. Another G403 [13] uses the block-diagonal approximation. This program also gives a weighting analysis scheme. The weights used were $w = 1/(\mu F)^2$ where $\mu F = \sqrt{\sigma F^2_{\text{count}} + kF_o^2} - |F_o|$, and $\sigma F^2_{\text{count}}$ were estimated from the counting statistics only.

The parameter k was adjusted to give an average of $w|F_o - F_c|^2$ which is nearly independent of the magnitude of F. A best fit was obtained for $k = 1.06$. A total of 16 parameters were refined using 748 reflections. The conventional R-value was found to be 5.1 % at the convergence of the refinement. A difference Fourier synthesis was calculated at the end of refinement but it gave no clear-cut evidence about the location of the hydrogen atom.

Neutron results. The hydrogen atoms are expected to lie in special positions $z = 0$ and $z = 1/2$. To test the suggestion of Christensen,[14] atoms with scattering lengths half the value of that for hydrogen were inserted in two positions in between the two oxygens with the shortest interatomic distance so that each of these atoms had a distance of 1 Å to the nearest oxygen atom. The H positions were refined using the step minimization program D445.[11]

With all other atoms fixed on the positions obtained by the X-ray investigation, the two inserted half hydrogen atoms moved to the same position at a distance of about 1.1 Å from one of the oxygen atoms.

Further refinement on all coordinates and anisotropic temperature factors were carried out using the least squares program G403 [13] and ORFLS.[12] In all cases the refinements were based on F_o. The weights used in the refinements were μF, with $k = 1.1$. A convergence point was reached at an R-value of 6.2 %.

Some reflections appeared to be affected by extinction, and a secondary extinction correction was applied using the Zachariasen formula [15] as modified by Larson:[16]

$$F_c^* = F_c(1 + g\beta(2\theta)F_c^2)^{-\frac{1}{4}}$$

where g and β are defined as by Zachariasen, $\beta(2\theta)$ in this case having the simple form $\beta(2\theta) = 1/\sin(2\theta)$. Two sets of refinements were carried out. In the first case 25 parameters were refined, 7 coordinates, 16 temperature factors, a scale factor, and the extinction factor g. In the second case indium and oxygen atoms were kept fixed on the positions obtained from the X-ray investigation, giving 20 parameters to refine. A final convergence was reached at R values of 4.2 % and 4.3 %, respectively. The corresponding weighted R values ($R_w^2 = \sum(|F_o| - k|F_c|)^2 w / \sum |F_o|^2 w$) were 5.3 % and 5.4 %. The values of g were 2.78×10^{-4} and 2.98×10^{-4}, respectively.

As the positions of indium and oxygen obtained from the neutron and the X-ray data differed somewhat, the Hamilton test [17] on the R_w factor ratio was performed.

We wish to test the hypothesis (at a 5 % significance level), that the coordinates of indium and oxygen atoms from the X-ray investigation fit the neutron data.

The ratio between the R_w values from the two refinements, $\mathcal{R} = R_w(\text{X-ray})/R_w(\text{neutron}) = 0.054/0.053 = 1.023$, has to be tested against $\mathcal{R}_{5,119,0.05}$. As $\mathcal{R}_{5,119,0.05} = 1.047$, we cannot reject the hypothesis.

Table 2. Scattering lengths and form factors.

	Neutron scattering lengths 10^{-12} cm	X-Ray form factors at $\sin\theta/\lambda = 0.5$
In	0.39	25.97
O	0.577	2.338
H	−0.372	0.071

Table 3. Positional and temperature parameters.

		X-Ray data		Neutron data. Refinement of all structural and temperature parameters		Neutron data. Refinement of temperature parameters and all hydrogen parameters	
In	x	0		0		0	
	y	0.26438	(7)	0.2673	(7)	0.26438	
	z	0		0		0	
	u_{11}	0.0042	(1)	0.0068	(10)	0.0070	(10)
	u_{22}	0.0052	(1)	0.0058	(8)	0.0075	(8)
	u_{33}	0.0060	(1)	0.0051	(8)	0.0050	(8)
	u_{12}	0		−0.0014	(7)	−0.0008	(7)
O_1	x	0.362	(1)	0.3608	(15)	0.362	
	y	0.478	(1)	0.4770	(4)	0.478	
	z	0		0		0	
	u_{11}	0.0020	(12)	0.0103	(9)	0.0101	(8)
	u_{22}	0.0078	(14)	0.0071	(6)	0.0070	(6)
	u_{33}	0.0064	(12)	0.0069	(8)	0.0064	(8)
	u_{12}	−0.0013	(10)	0.0000	(7)	−0.0004	(6)
O_2	x	0.636	(1)	0.6343	(5)	0.636	
	y	0.018	(1)	0.0186	(5)	0.018	
	z	0		0		0	
	u_{11}	0.0054	(15)	0.0076	(7)	0.0079	(7)
	u_{22}	0.0054	(13)	0.0096	(6)	0.0093	(6)
	u_{33}	0.0069	(13)	0.0080	(8)	0.0084	(9)
	u_{12}	−0.0005	(11)	0.0007	(7)	0.0001	(6)
H	x			0.520	(1)	0.520	(1)
	y			0.215	(2)	0.213	(1)
	z			0		0	
	u_{11}			0.0112	(26)	0.0123	(26)
	u_{22}			0.0409	(34)	0.0332	(30)
	u_{33}			0.0157	(16)	0.0158	(16)
	u_{12}			−0.0026	(26)	−0.0010	(25)

It is reassuring to notice that the final hydrogen atom coordinates for the two refinements differ less than two standard deviations.

CRYSTAL DATA

Crystal System: orthorhombic

Unit cell:[1] $a = 5.26 \pm 0.01$ Å
$b = 4.56 \pm 0.01$ Å
$c = 3.27 \pm 0.01$ Å

Space group: $P2_1nm$

$Dm = 7 \pm 0.7$ g/cm^3,[1] $Dx = 6.25$ g/cm^3

The X-ray scattering factors were taken from Vol. III of *International Tables for Crystallography*, and the neutron scattering lengths were the ones given in a list by the Neutron Diffraction Commission.[18] (Table 2).

Table 4. Interatomic distances and angles in Å and degrees.

Within octahedron	X-Ray data	Neutron data
In–O_1	2.140 (6)	2.125 (3)
In–O_1	2.140 (4)	2.138 (2)
In–O_2	2.220 (6)	2.233 (3)
In–O_2	2.201 (4)	2.207 (3)
In hydrogen bond		
O_1–O_2	2.543 (8)	2.537 (3)
O_1–H		1.458 (7)
O_2–H		1.079 (7)
$\angle O_1$–H–O_2		179.1 (6)

The parameters obtained are given in Table 3. Table 4 summarizes bond lengths and angles and Tables 5 and 6 give observed and calculated structure factors for X-ray and neutron data respectively. Numbers in parenthesis are standard deviations in unit of the last digit.

DISCUSSION

Whereas the geometric parameters agree well between the two diffraction techniques the thermal parameters differ considerably although they indicate quite small vibrations in accordance with the unusually high diffracted intensity at large $\sin\theta/\lambda$ values. In the X-ray case the temperature factor parameters are in error owing to the neglected absorption corrections and may be other systematic errors.

Table 5. Observed and calculated structure factors included in the refinements. X-Ray data.

h	k	l	Fo	|Fc|
2	0	0	71.6	77.5
4	0	0	47.2	46.7
6	0	0	46.3	46.1
8	0	0	36.6	36.3
10	0	0	21.2	20.5
12	0	0	15.0	15.2
0	1	0	7.4	7.4
2	1	0	24.8	21.5
3	1	0	65.8	66.0
4	1	0	6.5	5.9
5	1	0	49.1	48.9
6	1	0	9.1	7.7
7	1	0	36.1	35.4
8	1	0	5.5	4.3
9	1	0	28.9	26.8
11	1	0	20.1	20.4
13	1	0	12.6	14.7
0	2	0	47.7	55.1
1	2	0	13.6	13.8
2	2	0	64.0	68.8
3	2	0	11.3	11.4
4	2	0	59.4	61.2
5	2	0	8.9	8.6
6	2	0	36.3	36.5
7	2	0	7.1	6.1
8	2	0	27.5	25.5
10	2	0	24.9	25.0
12	2	0	17.9	18.5
0	3	0	16.2	16.7
1	3	0	56.2	61.7
2	3	0	18.8	18.8
3	3	0	47.0	47.3
4	3	0	12.2	12.7
5	3	0	39.3	39.1
6	3	0	11.1	10.9
7	3	0	33.2	33.1
8	3	0	9.3	7.9
9	3	0	24.5	24.0
10	3	0	6.4	6.8
11	3	0	16.6	16.4
12	3	0	5.9	5.0
13	3	0	11.5	13.4
0	4	0	55.2	57.3
1	4	0	18.1	18.7
2	4	0	41.7	44.5
3	4	0	15.3	16.0
4	4	0	31.2	33.1
5	4	0	13.7	13.4
6	4	0	33.2	33.4
7	4	0	11.0	10.8
8	4	0	26.8	27.5
9	4	0	9.2	8.3
10	4	0	17.1	16.3
11	4	0	6.1	5.7
12	4	0	11.6	12.5
0	5	0	17.9	18.4
1	5	0	33.8	35.6
2	5	0	17.8	18.7
3	5	0	37.3	37.9
4	5	0	16.5	16.0
5	5	0	30.6	30.8
6	5	0	13.5	13.5
7	5	0	21.2	21.2
8	5	0	10.3	10.3
9	5	0	18.7	18.2
10	5	0	7.6	7.9
11	5	0	15.5	16.2
12	5	0	6.2	6.4
0	6	0	27.5	26.7
1	6	0	17.8	18.6
2	6	0	30.0	30.2
3	6	0	17.2	17.3
4	6	0	29.2	29.7
5	6	0	15.3	14.9
6	6	0	20.3	20.6
7	6	0	12.1	11.8
8	6	0	15.6	15.7
9	6	0	10.4	9.9
10	6	0	14.5	15.7
11	6	0	7.8	7.9
0	7	0	19.1	18.2
1	7	0	26.9	26.3
2	7	0	17.5	17.3
3	7	0	20.0	19.4
4	7	0	13.9	15.0
5	7	0	18.4	18.1
6	7	0	12.6	13.3
7	7	0	19.3	18.9
8	7	0	11.0	11.1
9	7	0	13.7	13.6
10	7	0	8.2	8.8
11	7	0	7.1	8.5
0	8	0	22.7	20.8
1	8	0	16.7	16.5
2	8	0	19.0	17.7
3	8	0	15.8	15.4
4	8	0	14.6	14.6
5	8	0	14.6	13.9
6	8	0	14.4	15.2
7	8	0	12.4	11.7
8	8	0	13.1	13.0
9	8	0	9.0	9.7
10	8	0	7.4	8.6
0	9	0	14.4	14.5
1	9	0	12.5	12.0
2	9	0	15.3	14.6
3	9	0	17.5	15.7
4	9	0	13.9	13.8
5	9	0	14.3	13.0
6	9	0	11.6	11.7
7	9	0	7.3	7.9
8	9	0	9.5	9.6
0	10	0	10.8	9.6
1	10	0	13.7	13.5
2	10	0	11.6	10.4
3	10	0	12.3	12.5
4	10	0	10.9	10.2
5	10	0	12.5	11.5
6	10	0	7.0	7.9
7	10	0	9.3	9.9
0	11	0	12.3	12.1
1	11	0	9.7	9.0
2	11	0	10.4	11.4
3	11	0	6.8	5.4
4	11	0	10.4	10.4
1	0	1	66.7	62.6
3	0	1	81.8	75.0
5	0	1	54.8	51.0
7	0	1	31.4	29.9
9	0	1	27.6	26.1
11	0	1	24.7	23.7
13	0	1	12.1	14.1
1	1	1	17.7	17.7
2	1	1	65.2	69.8
3	1	1	10.1	9.4
4	1	1	53.7	52.5
5	1	1	9.3	8.6
6	1	1	41.6	40.9
8	1	1	32.0	31.0
9	1	1	6.7	5.3
10	1	1	23.1	22.3
12	1	1	16.1	16.5
0	2	1	11.8	12.4
1	2	1	71.0	75.9
2	2	1	11.5	11.5
3	2	1	47.0	45.7
4	2	1	9.2	8.7
5	2	1	41.1	40.4
6	2	1	6.9	7.2
7	2	1	39.8	38.5
8	2	1	6.7	5.5
9	2	1	26.0	25.5
11	2	1	15.8	15.1
13	2	1	13.3	14.5
0	3	1	47.3	52.4
1	3	1	16.9	17.6
2	3	1	51.3	53.3
3	3	1	14.3	14.3
4	3	1	47.2	46.7
5	3	1	12.5	12.5
6	3	1	33.9	33.7
7	3	1	9.2	8.8
8	3	1	26.0	25.0
9	3	1	7.4	7.4
10	3	1	22.1	21.0
12	3	1	15.0	16.0
0	4	1	16.0	17.2
1	4	1	40.6	40.6
2	4	1	16.8	17.0
3	4	1	47.1	46.3
4	4	1	14.9	14.4
5	4	1	36.0	35.9
6	4	1	12.0	11.6
7	4	1	23.0	22.8
8	4	1	9.3	8.8
9	4	1	20.9	20.5
10	4	1	8.2	7.3
11	4	1	17.5	18.8
12	4	1	6.1	5.6
0	5	1	39.6	41.6
1	5	1	18.5	18.8
2	5	1	33.7	33.3
3	5	1	15.5	16.4
4	5	1	28.7	28.1
5	5	1	14.8	14.2
6	5	1	26.5	27.0
7	5	1	11.1	11.3
8	5	1	23.6	22.7
9	5	1	9.6	9.6
10	5	1	14.6	14.8
11	5	1	7.6	7.0
12	5	1	10.0	11.2
0	6	1	17.6	18.2
1	6	1	32.4	32.2
2	6	1	16.8	17.6
3	6	1	23.5	24.2
4	6	1	14.9	15.3
5	6	1	22.8	22.4
6	6	1	14.0	13.4
7	6	1	22.5	22.3
8	6	1	11.1	10.8
9	6	1	15.4	15.6
10	6	1	7.0	8.2
11	6	1	8.9	10.2
0	7	1	19.8	20.0
1	7	1	16.7	16.9
2	7	1	23.8	23.1
3	7	1	16.4	16.3
4	7	1	23.9	23.6
5	7	1	14.2	14.5
6	7	1	16.4	16.4
7	7	1	12.7	11.6
8	7	1	12.4	12.2
9	7	1	9.8	9.5
10	7	1	11.7	12.9
0	8	1	17.2	16.1
1	8	1	17.8	16.7
2	8	1	16.4	15.9
3	8	1	19.0	18.7
4	8	1	14.8	14.2
5	8	1	16.1	15.6
6	8	1	12.9	12.5
7	8	1	11.4	11.2
8	8	1	10.1	10.3
9	8	1	10.0	10.6
10	8	1	8.3	8.7
0	9	1	18.1	16.8
1	9	1	16.3	14.9
2	9	1	14.9	13.0
3	9	1	13.4	13.4
4	9	1	9.8	9.7
5	9	1	11.9	12.1
6	9	1	11.4	11.9
7	9	1	10.5	10.9
8	9	1	11.2	11.0
0	10	1	13.1	12.9
1	10	1	11.8	10.7
2	10	1	13.2	13.0
3	10	1	8.5	8.9
4	10	1	11.7	12.0
5	10	1	7.8	8.6
6	10	1	10.9	10.7
7	10	1	8.2	8.3
0	11	1	6.9	5.2
1	11	1	10.7	11.2
2	11	1	8.4	7.6
3	11	1	10.6	11.2
4	11	1	9.8	8.8
2	0	2	57.5	57.7
4	0	2	41.1	39.9
6	0	2	40.9	40.2
8	0	2	32.4	32.7
10	0	2	20.5	18.7
12	0	2	13.0	14.1
0	1	2	5.8	5.5
1	1	2	55.0	59.1
2	1	2	16.0	12.8
3	1	2	53.7	53.1
4	1	2	5.1	4.8
5	1	2	42.3	42.0
6	1	2	6.5	6.6
7	1	2	32.8	31.5
9	1	2	25.8	24.3
11	1	2	18.8	18.8
13	1	2	11.4	13.7
0	2	2	40.8	45.2
1	2	2	10.3	10.3
2	2	2	52.4	54.2
3	2	2	8.6	9.2
4	2	2	49.6	51.0
5	2	2	7.6	7.4
6	2	2	32.4	32.2
7	2	2	6.4	5.4
8	2	2	24.2	23.0
9	2	2	5.7	4.4
10	2	2	22.6	22.9
12	2	2	15.4	17.2
0	3	2	12.9	13.7
1	3	2	46.8	50.2
2	3	2	16.0	15.0
3	3	2	38.5	40.4
4	3	2	11.8	11.0
5	3	2	34.2	34.3
6	3	2	10.1	9.7
7	3	2	31.3	29.7
8	3	2	7.8	7.2
9	3	2	22.6	21.9
10	3	2	5.9	6.2
11	3	2	14.2	15.2
0	4	2	45.0	47.7
1	4	2	15.4	15.8
2	4	2	37.8	38.2
3	4	2	13.1	13.8
4	4	2	29.2	29.1
5	4	2	11.6	11.9
6	4	2	30.1	29.9
7	4	2	9.4	9.8
8	4	2	25.2	25.1
9	4	2	7.0	7.6
10	4	2	15.1	15.0
12	4	2	10.3	11.7
0	5	2	15.7	16.1
1	5	2	29.5	31.2
2	5	2	15.6	16.4
3	5	2	31.8	33.4
4	5	2	14.1	14.2
5	5	2	27.5	27.6
6	5	2	12.8	12.2
7	5	2	19.8	19.2
8	5	2	9.5	9.4
9	5	2	16.8	16.7
11	5	2	12.9	15.0
12	5	2	6.0	6.0
0	6	2	23.3	23.7
1	6	2	16.1	16.5
2	6	2	26.6	26.9
3	6	2	15.1	15.5
4	6	2	27.1	26.7
5	6	2	14.0	13.5
6	6	2	18.9	18.7
7	6	2	11.1	10.7
8	6	2	15.1	14.4
9	6	2	8.5	9.1
10	6	2	13.3	14.5
11	6	2	7.3	7.3
0	7	2	15.7	16.4
1	7	2	23.7	23.7
2	7	2	16.4	15.6
3	7	2	17.9	17.5
4	7	2	14.8	13.6
5	7	2	16.4	16.5
6	7	2	12.6	12.2
7	7	2	17.6	17.4
8	7	2	11.3	10.3
9	7	2	12.1	12.6
10	7	2	7.3	8.2
0	8	2	20.0	18.9
1	8	2	15.0	15.0
2	8	2	17.0	16.1
3	8	2	14.9	14.1
4	8	2	14.2	13.3
5	8	2	13.3	12.8
6	8	2	14.5	14.0
7	8	2	10.0	10.8
8	8	2	11.3	12.0
9	8	2	8.9	9.0
0	9	2	14.1	13.3
1	9	2	11.7	11.0
2	9	2	14.3	13.4
3	9	2	14.8	14.4
4	9	2	12.7	12.7
5	9	2	12.4	12.0
6	9	2	9.9	10.8
7	9	2	7.4	7.4
8	9	2	7.6	9.0
0	10	2	7.8	8.8
1	10	2	12.5	12.5
2	10	2	10.4	9.6
3	10	2	12.0	11.6
4	10	2	9.2	9.4
5	10	2	9.5	10.7
6	10	2	7.3	7.4
0	11	2	9.7	11.2
1	11	2	8.9	8.4
2	11	2	10.4	10.6
3	11	2	5.5	5.0
4	11	2	8.2	9.7
1	0	3	44.2	42.7
3	0	3	52.0	49.7
5	0	3	39.8	38.4
7	0	3	24.3	23.8
9	0	3	23.1	21.6
11	0	3	19.3	20.2
0	1	3	43.8	49.7
1	1	3	7.7	7.8
2	1	3	46.0	45.6
3	1	3	5.8	5.6
4	1	3	39.2	38.4
5	1	3	5.8	6.2
6	1	3	35.0	31.9
8	1	3	26.2	25.3
9	1	3	5.0	4.5
10	1	3	19.1	18.8
12	1	3	13.1	14.2
0	2	3	7.5	8.3
1	2	3	44.6	49.0
2	2	3	8.0	7.9
3	2	3	33.4	34.3
4	2	3	6.2	6.5
5	2	3	31.9	31.2
7	2	3	32.2	31.1
8	2	3	5.5	4.5
9	2	3	21.6	21.2
11	2	3	13.0	12.8
0	3	3	34.7	37.5
1	3	3	10.9	11.9
2	3	3	35.6	38.5
3	3	3	10.3	10.5
4	3	3	35.2	35.4
5	3	3	8.9	9.7
6	3	3	27.6	26.9
7	3	3	7.3	7.2
8	3	3	21.7	20.6
9	3	3	5.7	6.2
10	3	3	18.0	17.8
11	3	3	5.5	3.8
12	3	3	12.7	13.8
0	4	3	11.9	12.8
1	4	3	28.7	30.8
2	4	3	11.8	12.8
3	4	3	34.8	35.4
4	4	3	11.1	11.3
5	4	3	28.6	28.6
6	4	3	10.7	9.4
7	4	3	19.0	18.6
8	4	3	7.7	7.3
9	4	3	17.4	17.3
10	4	3	6.4	6.2
11	4	3	14.8	16.1
0	5	3	30.6	32.1
1	5	3	13.3	14.6
2	5	3	26.5	27.6
3	5	3	12.2	13.0
4	5	3	22.8	22.5
5	5	3	12.2	11.5
6	5	3	22.7	22.1
7	5	3	9.5	9.4
8	5	3	19.5	19.1
9	5	3	7.1	8.1
10	5	3	11.7	12.6
11	5	3	5.8	6.1
0	6	3	12.7	14.5
1	6	3	26.2	25.7
2	6	3	14.0	14.2
3	6	3	20.2	19.5
4	6	3	12.4	12.5
5	6	3	19.2	18.4
6	6	3	12.0	11.2
7	6	3	18.8	18.7
8	6	3	9.7	9.2
9	6	3	12.9	13.3
10	6	3	7.1	7.0
0	7	3	15.9	16.2
1	7	3	13.8	13.8
2	7	3	18.2	19.0
3	7	3	12.6	13.5
4	7	3	19.8	19.6
5	7	3	11.8	12.1
6	7	3	13.2	13.8
7	7	3	10.6	9.8
8	7	3	9.2	10.4
9	7	3	6.7	8.1
0	8	3	13.4	13.4
1	8	3	14.4	13.9
2	8	3	13.8	13.3
3	8	3	16.0	15.7
4	8	3	12.7	12.0
5	8	3	13.6	13.2
6	8	3	11.0	10.6
7	8	3	9.5	9.6
8	8	3	7.8	8.8
0	9	3	14.9	14.2
1	9	3	12.7	12.6
2	9	3	11.5	11.0
3	9	3	10.4	11.4
4	9	3	8.0	8.2
5	9	3	10.9	10.4
6	9	3	9.8	10.2
7	9	3	9.1	9.4
0	10	3	10.7	11.0
1	10	3	8.9	9.2
2	10	3	10.8	11.1
3	10	3	6.2	7.7
4	10	3	9.7	10.3
5	10	3	6.0	7.5
1	11	3	8.4	9.7
2	0	4	37.4	35.6
4	0	4	29.8	27.1
6	0	4	31.2	28.7
8	0	4	25.9	24.7
10	0	4	13.9	14.5
12	0	4	9.4	11.3
1	1	4	32.4	36.2
2	1	4	7.8	6.5
3	1	4	35.4	34.3
5	1	4	31.5	29.2
6	1	4	5.0	4.8
7	1	4	24.5	23.1
9	1	4	19.7	18.6
11	1	4	13.8	14.9
0	2	4	26.4	29.6
1	2	4	6.0	6.4
2	2	4	33.3	34.5
3	2	4	6.5	6.0
4	2	4	35.7	34.5
5	2	4	6.3	5.2
6	2	4	24.8	23.2
7	2	4	4.5	4.0
8	2	4	18.0	17.2
9	2	4	5.1	3.4
10	2	4	17.8	18.0
0	3	4	8.6	8.9
1	3	4	29.8	32.8
2	3	4	10.2	9.8
3	3	4	27.6	27.7
4	3	4	8.2	7.7
5	3	4	26.7	24.6
6	3	4	7.2	7.1
7	3	4	23.6	22.3
8	3	4	5.0	5.5
9	3	4	17.6	17.0
10	3	4	5.7	4.9
11	3	4	10.8	12.1
0	4	4	31.2	32.3
1	4	4	10.4	10.8
2	4	4	26.0	26.4
3	4	4	10.0	9.7
4	4	4	22.0	20.8
5	4	4	10.3	8.7
6	4	4	24.0	22.3
7	4	4	8.2	7.4
8	4	4	20.0	19.4
9	4	4	7.5	6.0
10	4	4	10.2	11.8
0	5	4	11.0	11.4
1	5	4	21.6	22.1
2	5	4	10.8	11.7
3	5	4	25.4	24.3
4	5	4	10.3	10.4
5	5	4	21.8	20.6
6	5	4	10.6	9.2
7	5	4	15.0	14.7
8	5	4	7.8	7.4
9	5	4	12.5	13.1
0	6	4	17.5	17.3
1	6	4	12.4	12.1
2	6	4	20.0	19.9
3	6	4	11.5	11.5
4	6	4	21.1	20.2
5	6	4	10.6	10.3
6	6	4	15.7	14.3
7	6	4	9.4	8.3
8	6	4	10.6	11.3
9	6	4	6.4	7.2
0	7	4	12.6	12.3
1	7	4	17.6	17.9
2	7	4	12.4	11.8
3	7	4	13.9	13.2
4	7	4	11.0	10.4
5	7	4	13.7	12.7
6	7	4	9.5	9.5
7	7	4	12.9	13.7
8	7	4	7.4	8.2
9	7	4	8.8	10.1
0	8	4	14.9	14.6
1	8	4	12.7	11.6
2	8	4	12.8	12.4
3	8	4	11.8	10.9
4	8	4	11.6	10.4
5	8	4	10.7	10.0
6	8	4	10.5	11.1
7	8	4	8.5	8.7
0	9	4	11.1	10.4
1	9	4	8.8	8.6
2	9	4	11.2	10.5
3	9	4	11.3	11.4
4	9	4	9.4	10.1
5	9	4	9.8	9.6
6	9	4	8.3	8.7
0	10	4	6.6	7.1
1	10	4	9.3	10.0
2	10	4	7.7	7.7
3	10	4	8.7	9.3
3	0	5	32.8	30.5
5	0	5	28.4	25.0
7	0	5	17.4	16.1
9	0	5	15.9	15.5
2	1	5	28.6	27.5
4	1	5	27.3	24.3
5	1	5	4.7	4.1
6	1	5	23.4	21.4
8	1	5	19.4	17.8
10	1	5	13.8	13.7
0	2	5	4.5	5.0
1	2	5	29.5	29.6
2	2	5	4.5	4.9
3	2	5	22.6	21.6
4	2	5	4.9	4.2
5	2	5	23.4	20.5
7	2	5	23.7	21.7
9	2	5	15.1	15.3
0	3	5	22.5	23.4
1	3	5	6.8	7.4
2	3	5	24.3	24.3
3	3	5	6.6	6.7
4	3	5	25.0	23.3
5	3	5	7.5	6.6
6	3	5	19.0	18.4
7	3	5	4.7	5.0
8	3	5	14.5	14.6
9	3	5	4.7	4.6
10	3	5	11.0	13.1
0	4	5	7.6	8.3
1	4	5	19.3	19.8
2	4	5	8.3	8.4
3	4	5	24.2	23.5
4	4	5	8.4	7.6
5	4	5	21.0	19.5
6	4	5	6.5	6.5
7	4	5	14.0	13.0
8	4	5	4.9	5.2
9	4	5	13.1	12.6
0	5	5	21.7	21.4
1	5	5	10.1	9.7
2	5	5	19.1	18.5
3	5	5	8.8	8.8
4	5	5	16.5	15.3
5	5	5	8.9	8.1
6	5	5	16.7	15.7
7	5	5	7.5	6.7
8	5	5	13.6	13.9
9	5	5	5.6	6.0
0	6	5	9.9	9.9
1	6	5	17.8	17.7
2	6	5	9.9	9.8
3	6	5	14.1	13.4
4	6	5	9.4	8.7
5	6	5	13.5	13.0
6	6	5	8.2	8.0
7	6	5	13.4	13.7
8	6	5	6.3	6.8
0	7	5	12.4	11.3
1	7	5	8.8	9.7
2	7	5	13.2	13.4
3	7	5	10.1	9.6
4	7	5	15.2	14.1
5	7	5	9.4	8.7
6	7	5	11.1	10.1
7	7	5	5.7	7.3
0	8	5	10.1	9.6
1	8	5	10.0	10.0
2	8	5	10.1	9.6
3	8	5	11.3	11.4
4	8	5	9.4	8.8
5	8	5	9.0	9.8
6	8	5	6.5	7.9
0	9	5	8.4	10.5
1	9	5	8.1	9.3
2	9	5	6.7	8.1
3	9	5	8.3	8.4
2	0	6	21.1	20.8
4	0	6	18.6	16.3
6	0	6	19.3	18.3
8	0	6	15.1	16.4
2	1	6	4.2	4.0
3	1	6	22.3	20.6
5	1	6	19.9	18.2
6	1	6	4.1	3.2
7	1	6	16.2	15.0
9	1	6	11.2	12.6
0	2	6	15.8	17.3
1	2	6	3.5	3.8
2	2	6	20.2	20.6
3	2	6	3.7	3.7
4	2	6	22.2	21.4
5	2	6	3.7	3.3
6	2	6	16.6	14.8
8	2	6	11.0	11.5
0	3	6	4.3	5.4
1	3	6	19.0	19.8
2	3	6	5.4	6.0
3	3	6	18.0	17.0
4	3	6	4.8	4.8
5	3	6	16.6	15.6
6	3	6	4.8	4.7
7	3	6	15.4	14.8
0	4	6	15.7	20.1
1	4	6	6.2	6.7
2	4	6	16.9	16.4
3	4	6	6.5	6.1
4	4	6	12.9	13.1
5	4	6	5.9	5.6
6	4	6	15.7	14.7
7	4	6	5.0	5.0
8	4	6	12.4	13.2
0	5	6	7.0	7.2
1	5	6	13.0	13.9
2	5	6	7.1	7.5
3	5	6	15.1	15.7
4	5	6	7.1	6.8
5	5	6	13.1	13.6
6	5	6	6.2	6.2
7	5	6	9.2	10.0
0	6	6	10.3	11.1
1	6	6	7.4	7.9
2	6	6	11.5	13.0
3	6	6	7.9	7.6
4	6	6	14.0	13.4
5	6	6	6.6	6.9
6	6	6	8.8	9.7
0	7	6	7.9	8.2
1	7	6	10.2	12.0
2	7	6	7.1	7.9
3	7	6	8.4	8.9
4	7	6	7.7	7.1
5	7	6	8.6	8.7
0	8	6	8.3	9.9
1	8	6	7.5	7.9
2	8	6	8.6	8.5
3	0	7	20.0	17.9
5	0	7	15.8	15.1
7	0	7	10.9	10.1
2	1	7	16.8	15.8
4	1	7	15.7	14.4
6	1	7	11.6	13.1
0	2	7	3.1	2.9
1	2	7	15.4	17.3
3	2	7	13.8	12.5
5	2	7	12.6	12.4
7	2	7	13.3	13.6
0	3	7	13.7	13.6
1	3	7	4.0	4.4
2	3	7	15.6	14.3
4	3	7	13.7	14.1
6	3	7	11.4	11.4
1	4	7	5.8	11.8
2	4	7	4.2	5.1
3	4	7	14.4	14.3
4	4	7	4.9	4.7
5	4	7	11.8	12.2
6	4	7	4.3	4.1
0	5	7	9.2	13.1
1	5	7	4.4	5.9
2	5	7	7.4	11.3
3	5	7	4.1	5.4
4	5	7	7.7	9.5
5	5	7	4.5	5.1
1	6	7	6.5	11.1
2	6	7	5.6	6.1
3	6	7	7.8	8.5
2	0	8	13.4	11.7
4	0	8	10.5	9.4
1	1	8	9.8	11.9
3	1	8	13.7	11.8
0	2	8	8.6	9.7
2	2	8	11.2	11.7
1	3	8	11.3	11.4
2	3	8	3.2	3.5

Table 6. Observed and calculated structure factors included in the refinements. Neutron data.

h	k	l	Fo	|Fc|
2	0	0	0.32	0.31
4	0	0	1.71	1.74
6	0	0	1.10	1.12
8	0	0	1.49	1.60
0	1	0	0.25	0.24
1	1	0	1.14	1.18
2	1	0	1.87	1.89
3	1	0	1.39	1.51
4	1	0	1.64	1.66
5	1	0	0.78	0.75
0	2	0	1.63	1.78
1	2	0	0.42	0.41
2	2	0	0.53	0.52
4	2	0	1.90	1.98
5	2	0	0.54	0.60
8	2	0	0.82	0.86
0	3	0	0.59	0.57
1	3	0	1.56	1.56
2	3	0	1.83	1.92
4	3	0	0.50	0.55
6	3	0	1.21	1.26
7	3	0	1.23	1.30
0	4	0	1.98	2.00
1	4	0	0.84	0.80
2	4	0	0.10	0.14
3	4	0	0.70	0.65
4	4	0	1.12	1.10
6	4	0	0.97	1.00
0	5	0	0.58	0.56
1	5	0	0.13	0.14
2	5	0	1.47	1.45
3	5	0	1.65	1.55
5	5	0	1.01	1.01
6	5	0	1.29	1.24
0	6	0	0.79	0.75
1	6	0	0.90	0.89
2	6	0	0.62	0.63
4	6	0	1.45	1.44
0	7	0	0.57	0.58
1	7	0	1.07	1.22
2	7	0	1.11	1.04
3	7	0	0.64	0.57
1	0	1	0.15	0.11
3	0	1	2.90	2.65
5	0	1	2.01	1.87
7	0	1	0.84	0.86
1	1	1	1.59	1.53
2	1	1	0.25	0.20
3	1	1	1.11	1.04
5	1	1	1.86	1.75
1	2	1	2.19	2.29
2	2	1	0.59	0.61
3	2	1	0.48	0.57
7	2	1	2.18	2.18
1	3	1	1.39	1.46
2	3	1	0.42	0.41
3	3	1	1.03	1.10
4	3	1	1.10	1.09
5	3	1	1.39	1.31
2	4	1	1.18	1.18
3	4	1	2.02	2.07
5	4	1	1.25	1.31
6	4	1	0.64	0.65
0	5	1	1.57	1.54
1	5	1	1.14	1.07
3	5	1	0.53	0.64
5	5	1	1.32	1.28
6	5	1	0.75	0.77
1	6	1	1.31	1.29
2	6	1	1.35	1.21
3	6	1	0.43	0.45
0	7	1	0.71	0.80
1	7	1	0.73	0.76
0	0	2	2.02	2.04
4	0	2	1.69	1.64
6	0	2	1.11	1.08
8	0	2	1.48	1.54
1	1	2	1.15	1.14
2	1	2	2.00	1.89
3	1	2	1.45	1.44
4	1	2	0.66	0.66
6	1	2	1.63	1.57
0	2	2	1.77	1.75
1	2	2	0.44	0.39
2	2	2	0.49	0.53
4	2	2	2.04	1.95
1	3	2	1.53	1.49
2	3	2	1.88	1.84
4	3	2	0.48	0.55
6	3	2	1.35	1.21
7	3	2	1.25	1.25
0	4	2	1.86	1.94
1	4	2	0.80	0.76
3	4	2	0.65	0.61
4	4	2	1.03	1.03
5	4	2	0.58	0.58
2	5	2	1.42	1.38
3	5	2	1.51	1.46
4	5	2	0.61	0.71
5	5	2	0.97	0.96
0	6	2	0.68	0.70
1	6	2	0.85	0.85
4	6	2	1.50	1.37
3	0	3	2.66	2.53
5	0	3	1.70	1.67
0	1	3	0.49	0.43
1	1	3	1.48	1.44
3	1	3	0.99	0.95
5	1	3	1.61	1.58
1	2	3	2.17	2.18
2	2	3	0.51	0.54
3	2	3	0.52	0.52
1	3	3	1.32	1.32
2	3	3	0.35	0.43
3	3	3	1.01	0.98
4	3	3	0.99	0.98
5	3	3	0.24	0.22
2	4	3	1.00	1.05
3	4	3	1.85	1.89
0	5	3	1.44	1.42
1	5	3	0.96	0.97
3	5	3	0.66	0.59
1	6	3	1.19	1.17
2	6	3	1.00	1.09
0	0	4	1.90	1.94
4	0	4	1.27	1.31
1	1	4	1.00	0.93
2	1	4	1.65	1.67
3	1	4	1.16	1.19
4	1	4	0.62	0.54
5	1	4	1.01	0.97
0	2	4	1.44	1.41
1	2	4	0.41	0.33
2	2	4	0.58	0.54
3	2	4	0.14	0.17
4	2	4	1.81	1.74
1	3	4	1.27	1.26
2	3	4	1.58	1.58
0	4	4	1.72	1.75
1	4	4	0.62	0.65
3	0	5	2.02	2.07
1	1	5	1.15	1.18
2	1	5	0.20	0.27
1	2	5	1.61	1.77

The structure can be described as a packing of indium-oxygen octahedra. Chains of octahedra sharing edges are stacked along the [001] direction. The parallel chains are interlinked by sharing the corners of the octahedra. Fig. 1 shows the systems of octahedra and the hydrogen bonds joining them together. The exceptionally strong hydrogen bond seems to be correlated with a hardness not often found in metal oxide hydroxides.

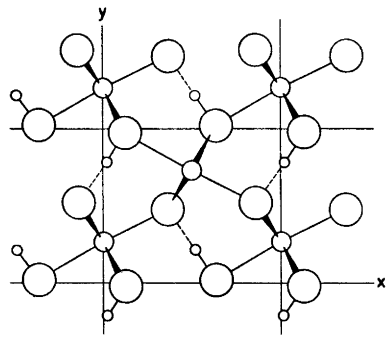

Fig. 1. Projection in the [001] direction of five octahedra in the indium oxide hydroxide structure. Hydrogen atoms are indicated by small circles, indium atoms by medium sized circles and oxygen atoms by large circles.

The octahedra lie with faces almost parallel with the $x-z$ plane, so that the oxygen atoms have the y coordinates approximately 0 and 0.5.

The layer of oxygen atoms with $y \simeq 0$ consists of OH^- ions and the layer of oxygen atoms with $y \simeq 0.5$ consists of O^{2-} ions.

The hydrogen bond is linear (Table 4). The four next nearest oxygen atoms (having z coordinates $\pm\frac{1}{2}$) to the hydrogen atom form a rectangle with the hydrogen atom positioned on the normal to the plane through the center, and with hydrogen-oxygen distances 2.80 Å and 2.81 Å. The distance of the hydrogen from the rectangle plane is 0.2 Å.

The nearest indium atoms are at distances 2.54 Å and 2.74 Å.

The vibration of the hydrogen atom is greatest in a direction nearly parallel to the y direction. This can be explained as a movement of the atom in a potential minimum elongated along the y direction and governed by the repulsion from the indium atoms.

The shortest oxygen-hydrogen distance is in good agreement with other oxygen-hydrogen distances found in short hydrogen bonds, when it is compared with the oxygen-oxygen distance (Hamilton and Ibers).[19]

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