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Metal Complexes with Mixed Ligands

2. The Molecular and Crystal Structure of Catena-di-μ-perchlorato-[di-μ-imidazolato-diperchlorato-octa-imidazolo-tricopper(II)]; $Cu_3(C_3H_3N_2)_2(C_3H_4N_2)_8(ClO_4)_4$

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The detailed crystal structure of catena-di-μ-perchlorato[di-μ-imidazolato-diperchlorato-octa-imidazolo-tricopper(II)],

$$Cu_3(C_3H_3N_2)_2(C_3H_4N_2)_8(ClO_4)_4$$

has been determined using three-dimensional X-ray diffraction data collected with a linear diffractometer. The crystals are monoclinic, space group $P2_1/a$, with cell dimensions and the corresponding standard deviations: $a = 16.644 \pm 0.005$ Å, $b = 15.524 \pm 0.005$ Å, $c = 9.226 \pm 0.001$ Å and $\beta = 94.68 \pm 0.05°$ at 25°C. There are two formula units per unit cell.

The structure is built up of tri-nuclear units, consisting of three octahedrally (distorted) coordinated Cu-atoms connected to each other through two imidazolato bridges. Each unit is centrosymmetric around the central Cu-atom. The tri-nuclear units are linked together through two perchlorate groups forming a chain running along the c-direction. The chains are held together by hydrogen and van der Waals-bonds. No hydrogen atoms could be located directly, but a reasonable hydrogen-bonding scheme was inferred from short O – N distances. The centrosymmetric copper atom (Cu_1) is coordinated to two imidazolate nitrogens (Cu – N; 1.975 Å), two imidazole nitrogens (Cu – N; 2.009 Å) and two perchlorate oxygens (Cu – O; 2.755 Å), and each of the two symmetry related copper atoms (Cu_2) to three imidazole nitrogens (Cu – N; 1.986, 1.987, 1.991 Å), one imidazolate nitrogen (Cu – N; 1.976 Å) and two perchlorate oxygens (Cu – O; 2.581, 2.930 Å).

At this institute crystal structure studies on metal imidazole complexes were started some years ago,[1–3] and during the same time aqueous equilibria $Cu^{2+} - OH^- - C_3H_4N_2$ (imidazole) in 3.0 M $NaClO_4$-medium have been extensively studied by Sjöberg.[4] The main part of his investigation was carried out as a series of emf-titrations. In a typical set of these titrations, acid (Cu^{2+}, $C_3H_5N_2^+$)-perchlorate solutions were titrated with OH^-, and the titrations were mostly continued until precipitation occurred. The compositions of these precipitates were found to be different depending on the ratio C/B in the solution (B = the total copper concentration and C = the total imidazole concentration). Sjöberg found the following three phases:

Phase 1, formed by OH^--addition to solutions where $C/B < 2$, consists of a light blue coloured microcrystalline precipitate. Attempts to obtain sharp X-ray powder photographs of this phase failed, but the composition could be determined to be $Cu(OH)_{1.60}(ClO_4)_{0.40}$. No imidazole could be found in the precipitate. Attempts to obtain crystals suitable for X-ray structure determinations are in progress (G. Ivarsson). The region of existence for this phase is determined by the equilibrium constant:[4] $[Cu^{2+}]^5[H^+]^{8-}[ClO_4^-]^2 = 10^{33.64 \pm 0.35}$ M^{-1}.

Phase 2 (blue-violet coloured crystals) is always formed from solutions with $C/B > 2$. In pure form phase 2 is best obtained when $4 < C/B < 8$. At higher ratios a mixture of phases 2 and 3 is formed. The formula of phase 2 was determined to be $Cu(C_3H_4N_2)_4(ClO_4)_2$, and the crystals were found to be monoclinic with $a = 8.198$ Å, $b = 16.293$ Å, $c = 9.353$ Å and $\beta = 125.81°$. The unit cell contains two formula units and the space group is $P2_1/n$. At present we are collecting three-dimensional X-ray data for the crystal. The region of existence for this phase is determined by the equilibrium constant:[4] $[Cu^{2+}][C_3H_5N_2^+]^4[H^+]^{4-}[ClO_4^-]^2 = 10^{14.65 \pm 0.09}$ M^3.

Phase 3 (violet-blue crystals) is obtained from solutions with $C/B > 8$ and $B < 1$ mM. No solubility data for this phase could be obtained.

The aim of the present work is to present analysis results, crystal data and a complete structure determination of phase 3.

EXPERIMENTAL

Crystal preparation and analyses. The crystals used have been chosen from precipitations obtained by Sjöberg.[4] In a typical preparation 1.40 ml of a solution with the composition: $[Na^+] = 3.495$ M, $[OH^-] = 0.495$ M, $[ClO_4^-] = 3.000$ M were added to 50.0 ml of a solution with the composition: $[Na^+] = 2.984$ M, $[ClO_4^-] = 3.000$ M, $[Cu^{2+}] = 0.80$ mM, $[H^+] = 1.123$ mM, $[C_3H_5N_2^+] = 12.89$ mM. The solution was then allowed to stand in a thermostated room at 25°C. Within a few days, well defined dark violet prismatic crystals were formed.

The copper content of the crystals was determined electrolytically according to Kolthoff-Sandell.[5] Confirmatory copper analyses were also made using atomic absorption methods. The nitrogen content was determined using the Kjeldahl[6] method. IR-spectra indicated that the crystals contained neither H_2O nor OH^-. The Cu and N analyses gave the following results (in weight %): $Cu = 14.96 \pm 0.08$ and $N = 21.9 \pm 0.2$. This corresponds to 3.33 imidazoles ($C_3H_4N_2$) and (or) imidazolates ($C_3H_3N_2$)$^-$ per copper atom. In order to obtain the number of imidazolates per copper atom, we determined the uptake of protons when the crystals were dissolved in an acidic solution of known H^+-concentration. The H^+-concentration was chosen so that the reaction occurred according to the formula:

$$(Cu(C_3H_3N_2)_x(C_3H_4N_2)_{3.33-x}(ClO_4)_{2-x})_n + n(3.33+x)H^+ \rightarrow$$
$$\rightarrow 3.33nC_3H_5N_2^+ + nCu^{2+} + n(2-x)ClO_4^-$$

This determination gave a H^+-uptake of 4 $(= 3.33 + x)H^+$ per Cu which gives $x = 0.67$. Thus the formula $Cu_3(C_3H_3N_2)_2(C_3H_4N_2)_8(ClO_4)_4$ could be established.

Crystal data and space group. From rotation photographs around the c- and b-axes and the corresponding Weissenberg photographs (zero, first and second layer lines) taken with Cu$K\alpha$-radiation it was concluded that the crystals are monoclinic. The cell dimensions and the angle were refined from a Guinier photograph and the following parameters and their corresponding standard deviations were obtained:

$a = 16.644 \pm 0.005$ Å, $b = 15.524 \pm 0.005$ Å, $c = 9.226 \pm 0.001$ Å, $\beta = 94.68° \pm 0.05°$, $V = 2375.89$ Å^3.

By the flotation method (using bromoform and acetone) the density of the crystals was determined to be 1.77 g/cm^3. With two of the above mentioned formula units in the cell the calculated density is 1.771 g/cm^3.

Systematic extinctions were found for $h0l$, $h = 2n+1$ and $0k0$, $k = 2n+1$. This is characteristic for the space group $P2_1/a$ (No. 14).

Collection and reduction of intensity data. Single-crystal intensity data were collected and measured using an automatic linear diffractometer (PAILRED). The radiation was Mo$K\alpha$ with a LiF-monochromator. The crystal used was an approximately rectangular prism with dimensions of $0.024 \times 0.018 \times 0.017$ cm. It was rotated around the c-axis (parallel to the 0.024-edge of the crystal) and intensities for $hk0 - hk8$ were measured ($\sin \theta \leq 0.58$). The half scan-intervals were: for all $hk0 - hk3 = 1.0°$ and for $hk4 - hk8$ with $\theta > 22° = 1.0°$ and with $\theta \leq 22° = 2.0°$. Scan speed used was 0.5 degrees/min. Each reflection was measured only once. Background intensities, B_1 and B_2, were measured during 20 sec (t_b) before and after each scan. The intensity, I_{obs}, for a reflection was calculated from the measured total intensity T (peak + background) by substracting for the background counts B_1 and B_2 according to the relation:

$$I_{obs} = T - \frac{t_T}{2t_b}(B_1 + B_2)$$

where t_T stands for the total scanning time. Relative counting statistical error of each reflection, $\Delta I/I$, was calculated using the formula:

$$\frac{\Delta I}{I} = \frac{[T + t^2(B_1 + B_2)]^{1/2}}{T - t(B_1 + B_2)}$$

where $t = t_T/2t_b$.

A total of 5597 independent reflections were measured. Of these all with $\Delta I/I > 0.5$ were omitted. This reduction resulted in a final data set of 2674 observed reflections used in the refinement. When the refinement was completed the structure factors for the "unobserved reflections" were calculated as a check which confirmed that they all had low values. The intensity data were corrected for Lorentz and polarization factors and for absorption, the linear absorption coefficient was calculated to be 16.965 cm^{-1}. The absorption correction gave differences in the transmission factors of up to 6 %.

Computer programs used. The diffractometer data correction was made by a program written by the authors. The computer programs for Lorentz and polarization corrections, Fourier summations, and for the calculation of distances and angles were originally written by A. Zalkin. A modified version of a program written by Gantzel, Sparks and Trueblood was used for structure factor calculation and refinement of the structural parameters. Correction for absorption was made by a program originally written by P. Coppens, L. Leiserowitz and D. Rabinovich revised by Olle Olofsson and Mats Elfström. The computer program ORTEP [7] was used to produce the stereoscopic figures. The computers used were CD 3600 and CD 3200.

STRUCTURE DETERMINATION AND REFINEMENT

The copper and chlorine atoms were located from a three-dimensional Patterson synthesis, and the other atoms were found by standard Fourier methods. The structure was refined by full-matrix least-squares techniques. Throughout the refinement the observed reflections were weighted with $\omega = 1$. The weighting schemes of Hughes and of Cruickshank [8] were also tested but without improvements. The atomic scattering factors for Cu^{2+}, Cl$^-$, O, N, and C were used.[18] With individual isotropic temperature factors the refinement converged to a conventional R-value of 0.121. The refinement was then completed with anisotropic temperature factors for all atoms. This reduced R to a final value of 0.050. All parameter shifts in the final cycle were less than 10 % of the estimated standard deviation. A final difference Fourier synthesis was calculated in which no abnormalities could be detected. No hydrogen atoms could be located with certainty. The final atomic coordinates and vibrational parameters are given in Tables 1 and 2. A comparison between observed and calculated structure factors is reported in Table 3.

Table 1. The atomic positional fractional coordinates and their estimated standard deviations ($10^4 \times \sigma$ in parenthesis).

	x	y	z
Cu_1	0.0000(0)	0.0000(0)	0.0000(0)
Cu_2	0.8530(0)	0.2591(0)	0.3793(1)
Cl_1	0.8170(1)	0.1386(1)	0.7772(2)
Cl_2	0.9034(1)	0.4038(1)	0.0631(2)
O_1	0.8743(4)	0.0789(4)	0.8385(8)
O_2	0.8279(4)	0.2205(4)	0.8451(7)
O_3	0.7389(4)	0.1050(6)	0.7944(13)
O_4	0.8250(6)	0.1459(4)	0.6242(8)
O_5	0.8566(5)	0.3444(5)	0.1397(8)
O_6	0.8579(5)	0.4197(5)	0.9275(8)
O_7	0.9747(5)	0.3609(7)	0.0242(12)
O_8	0.9154(6)	0.4811(6)	0.1364(11)
N_1	0.6897(4)	0.3387(4)	0.9180(8)
N_2	0.5766(3)	0.4093(4)	0.9397(7)
N_3	0.9268(5)	0.1721(5)	0.3073(9)
N_4	0.9768(4)	0.0718(4)	0.1682(6)
N_5	0.9412(3)	0.3164(4)	0.5024(7)
N_6	0.0042(5)	0.3911(5)	0.6780(9)
N_7	0.7783(4)	0.3471(4)	0.4484(9)
N_8	0.7113(5)	0.4678(5)	0.4693(9)
N_9	0.7596(4)	0.1971(4)	0.2805(7)
N_{10}	0.6820(7)	0.0982(8)	0.1724(13)
C_1	0.6580(5)	0.4142(5)	0.9625(9)
C_2	0.5569(5)	0.3300(5)	0.8766(10)
C_3	0.6285(5)	0.2858(6)	0.8618(11)
C_4	0.9340(5)	0.1452(5)	0.1747(9)
C_5	0.9990(6)	0.0527(7)	0.3080(11)
C_6	0.9693(6)	0.1123(7)	0.3995(10)
C_7	0.9355(5)	0.3508(5)	0.6297(10)
C_8	0.0179(5)	0.3327(8)	0.4589(12)
C_9	0.0556(6)	0.3816(8)	0.5720(14)
C_{10}	0.7681(5)	0.4276(5)	0.3997(10)
C_{11}	0.7206(5)	0.3336(5)	0.5437(10)
C_{12}	0.6791(5)	0.4101(6)	0.5607(11)
C_{13}	0.7579(7)	0.1126(7)	0.2463(14)
C_{14}	0.6856(6)	0.2343(8)	0.2318(11)
C_{15}	0.6371(6)	0.1704(10)	0.1673(12)

Table 2. The final vibrational parameters and their estimated standard deviations ($10^4 \times \sigma$ in parenthesis). Anisotropic temperature factors have been calculated according to the formula
$\exp[-(h^2B_{11}+k^2B_{22}+l^2B_{33}+hkB_{12}+hlB_{13}+klB_{23})]$.

	B_{11}	B_{22}	B_{33}	B_{12}	B_{13}	B_{23}
Cu_1	0.0023(0)	0.0024(0)	0.0063(3)	−0.0005(1)	0.0026(1)	−0.0020(1)
Cu_2	0.0026(0)	0.0029(0)	0.0060(3)	0.0000(0)	0.0003(1)	−0.0031(1)
Cl_1	0.0028(0)	0.0026(1)	0.0065(3)	0.0009(3)	−0.0003(2)	−0.0012(2)
Cl_2	0.0035(1)	0.0044(1)	0.0076(3)	−0.0012(1)	0.0013(2)	−0.0014(2)
O_1	0.0066(4)	0.0050(3)	0.0203(14)	0.0064(6)	−0.0074(10)	0.0004(10)
O_2	0.0055(3)	0.0032(3)	0.0166(11)	0.0002(5)	−0.0013(8)	−0.0058(8)
O_3	0.0038(3)	0.0096(6)	0.0480(26)	−0.0049(7)	0.0136(14)	−0.0188(19)
O_4	0.0158(7)	0.0050(3)	0.0066(11)	−0.0013(8)	0.0059(12)	−0.0007(9)
O_5	0.0085(4)	0.0073(4)	0.0143(13)	−0.0036(7)	0.0088(11)	0.0038(11)
O_6	0.0087(4)	0.0070(4)	0.0124(12)	−0.0007(7)	−0.0031(11)	0.0039(10)
O_7	0.0049(4)	0.0152(8)	0.0320(22)	0.0037(9)	0.0075(13)	−0.0071(21)
O_8	0.0124(6)	0.0086(6)	0.0273(19)	−0.0108(10)	0.0046(17)	−0.0193(16)
N_1	0.0035(3)	0.0048(4)	0.0131(12)	0.0026(5)	0.0038(9)	−0.0016(9)
N_2	0.0031(2)	0.0029(3)	0.0094(10)	0.0010(4)	0.0021(7)	0.0011(7)
N_3	0.0038(3)	0.0030(3)	0.0090(12)	0.0024(5)	0.0001(9)	−0.0035(10)
N_4	0.0033(2)	0.0037(3)	0.0042(9)	−0.0002(4)	0.0015(7)	−0.0012(7)
N_5	0.0028(2)	0.0039(3)	0.0080(10)	−0.0013(4)	0.0014(7)	−0.0052(8)
N_6	0.0052(4)	0.0058(4)	0.0133(12)	−0.0015(6)	−0.0030(10)	−0.0065(11)
N_7	0.0030(3)	0.0029(3)	0.0087(12)	0.0000(5)	0.0021(9)	−0.0011(9)
N_8	0.0064(4)	0.0047(4)	0.0152(14)	0.0051(6)	0.0067(12)	0.0047(11)
N_9	0.0040(3)	0.0043(3)	0.0066(10)	−0.0026(5)	0.0017(7)	−0.0032(8)
N_{10}	0.0095(6)	0.0136(9)	0.0234(21)	−0.0135(12)	0.0122(18)	−0.0184(22)
C_1	0.0031(3)	0.0048(4)	0.0115(14)	0.0011(6)	0.0043(10)	0.0018(11)
C_2	0.0041(4)	0.0039(4)	0.0122(14)	0.0005(6)	0.0045(11)	−0.0016(11)
C_3	0.0040(4)	0.0048(4)	0.0145(16)	0.0019(6)	0.0039(11)	−0.0022(12)
C_4	0.0046(4)	0.0035(4)	0.0061(12)	0.0016(6)	−0.0002(9)	−0.0040(9)
C_5	0.0068(5)	0.0067(6)	0.0088(14)	0.0075(9)	0.0010(13)	−0.0020(13)
C_6	0.0071(6)	0.0070(6)	0.0071(14)	0.0069(9)	−0.0011(12)	−0.0037(13)
C_7	0.0046(4)	0.0044(4)	0.0106(14)	0.0002(6)	0.0005(11)	−0.0057(11)
C_8	0.0037(4)	0.0092(7)	0.0179(20)	−0.0041(9)	0.0027(13)	−0.0048(18)
C_9	0.0049(5)	0.0083(7)	0.0244(24)	−0.0060(10)	−0.0036(16)	−0.0034(20)
C_{10}	0.0047(4)	0.0039(4)	0.0122(14)	0.0011(6)	0.0043(11)	0.0005(11)
C_{11}	0.0039(4)	0.0048(4)	0.0109(14)	0.0019(6)	0.0047(10)	0.0028(11)
C_{12}	0.0048(4)	0.0058(5)	0.0150(17)	0.0035(8)	0.0058(13)	0.0038(14)
C_{13}	0.0067(6)	0.0072(7)	0.0208(22)	−0.0059(10)	0.0003(17)	−0.0082(18)
C_{14}	0.0041(4)	0.0107(8)	0.0125(16)	−0.0052(10)	−0.0015(12)	0.0016(17)
C_{15}	0.0047(5)	0.0161(11)	0.0127(18)	−0.0113(12)	0.0052(13)	−0.0136(22)

DESCRIPTION AND DISCUSSION OF THE STRUCTURE

The structure is built up from coupled tri-nuclear Cu-units (complexes) which are connected through perchlorate bridges forming symmetry related chains directed along the *c*-axis. They are parallel per half unit lengths in *a*- and *b*-directions related with a mirror plane with glide *a*/2. Schematically

Table 3. Observed and calculated structure factors (×10). Values marked with an asterisk were not included in the final refinement.

H	K		
L=	0		
9	18	179	150
9	17	216	241
8	17	154	148
3	17	433	404
2	17	160	189
-2	17	168	189
-3	17	404	404
-4	17	173	175
-9	17	226	241
-5	16	269	253
-6	16	329	306
-7	16	208	162
-8	16	160	202
-1	16	313	285
-1	15	445	414
-3	15	185	126
-5	15	182	173
-7	15	438	402
-11	14	150	113
-10	14	326	288
-6	14	300	286
-5	14	171	156
-4	14	311	289
-1	14	522	509
0	14	140	151
-1	13	286	240
-2	13	169	169
-3	13	426	408
-4	13	265	295
-5	13	261	229
-6	13	454	465
-7	13	133	10*
-10	13	212	176
-11	13	390	371
-12	13	175	193
-15	12	175	151
-14	12	308	304
-13	12	184	205
-9	12	481	497
-8	12	337	320
-7	12	158	160
-6	12	290	209
-4	12	254	247
-3	12	176	168
-2	12	280	273
-1	11	264	232
-3	11	287	302
-4	11	264	276
-6	11	263	261
-7	11	408	385
-8	11	333	318
-10	11	154	155
-11	11	197	253
-12	11	357	367
-13	11	397	421
-15	11	177	176
-18	10	153	143
-15	10	219	239
-13	10	193	193
-12	10	193	221
-10	10	480	462
-9	10	349	373
-8	10	392	385
-7	10	233	250
-6	10	478	482
-5	10	217	200
-4	10	441	492
-3	10	262	244
-2	10	637	649
-1	10	268	275
0	10	179	196
-1	9	147	140
-3	9	452	451
-5	9	328	321
-7	9	179	199
-8	9	603	618
-9	9	205	218
-10	9	251	285
-11	9	135	98
-12	9	214	168
-16	9	162	182
-17	9	174	193
-15	8	175	169
-14	8	231	230
-13	8	171	163
-12	8	349	365
-11	8	169	175
-8	8	198	235
-6	8	848	878
-5	8	711	693
-4	8	153	169
-3	8	712	725
0	8	951	992
-1	7	236	262
-2	7	707	707
-3	7	255	286
-4	7	504	529
-7	7	119	145
-8	7	278	284
-10	7	262	281
-11	7	411	392
-15	7	175	154
-18	7	268	243
-18	6	211	253
-16	6	397	413
-15	6	150	102
-11	6	150	158
-10	6	528	567
-8	6	481	457
-6	6	240	206
-5	6	144	207
-2	6	578	561
-1	6	658	651
0	6	365	393
-1	5	448	431
-2	5	234	246
-3	5	514	475
-4	5	125	61
-6	5	863	899
-5	5	463	422
-9	5	743	720
-8	5	612	610
-10	5	477	486
-11	5	399	375
-12	5	471	486
-13	5	264	265
-14	5	231	221
-16	5	204	237
-17	5	194	194
-18	5	192	247
-20	4	218	245
-14	4	611	599
-13	4	345	360
-11	4	136	128
-10	4	260	294
-9	4	573	560
-8	4	881	911
-7	4	164	187
-6	4	956	900
-5	4	188	175
-4	4	527	502
-3	4	96	34
-1	4	481	477
-1	3	778	768
-3	3	1350	1361
-4	3	919	922
-5	3	351	352
-6	3	249	155
-7	3	992	990
-8	3	749	747
-9	3	672	700
-10	3	261	244
-11	3	179	162
-12	3	832	807
-13	3	406	436
-14	3	136	100
-15	3	159	189
-16	3	241	218
-18	3	148	163
-19	3	204	230
-18	2	195	231
-17	2	234	255
-16	2	153	161
-15	2	305	309
-14	2	134	126
-13	2	231	233
-12	2	257	273
-10	2	1173	1149
-9	2	231	189
-8	2	402	444
-7	2	518	514
-6	2	353	411
-5	2	129	77
-4	2	1408	1352
-3	2	859	918
-3	1	81	50
-5	1	1435	1386
-6	1	369	374
-7	1	353	345
-8	1	626	639
-11	1	458	463
-12	1	410	412
-14	1	246	257
-15	1	302	303
-17	1	204	227
-14	0	407	444
-12	0	643	634
-10	0	247	244
-8	0	287	275
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the chains may be written in the following way ($Im^- = C_3H_3N_2^-$, $ImH = C_3H_4N_2$):

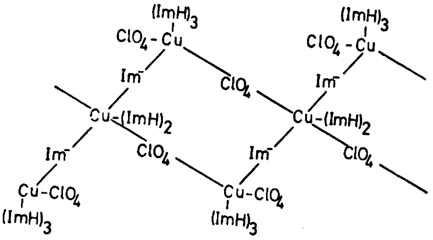


Fig. 1.

The symmetry related strings are held together by hydrogen and van der Waals' bonds. The molecular packing is illustrated in Fig. 2. In one formula unit one copper atom (Cu_1) is placed in the center of symmetry. The other two copper atoms (Cu_2 and $\overline{\mathrm{Cu}}_2$) are related to one another by this center of symmetry and each one is connected to Cu_1 through an imidazolate bridge. The other ligands, imidazole nitrogens, and perchlorate oxygens, give a distorted octahedral coordination around the copper atoms. The arrangements around Cu_1 and Cu_2 are shown in Figs. 3a and 3b, respectively.

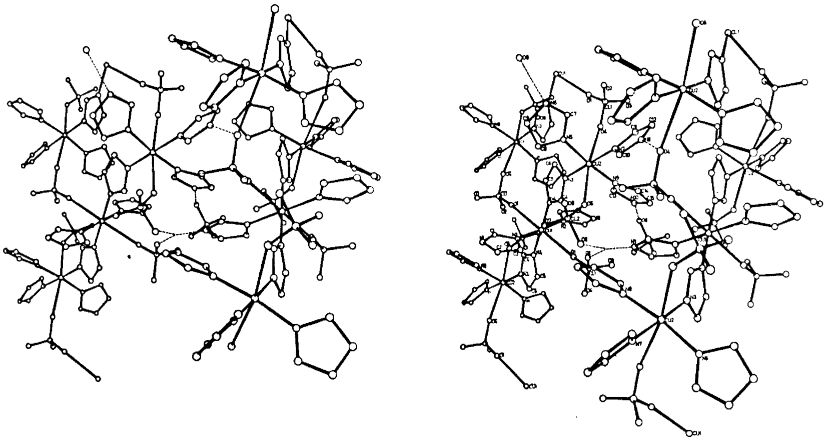


Fig. 2. Stereoscopic illustration of the molecular packing of $\mathrm{Cu_3(C_3H_3N_2)_2(C_3H_4N_2)_8(ClO_4)_4}$ viewed along the *b*-axis. The labels C2 and C3 should be interchanged in the figure.

The coordination around Cu_1. The Cu_1-atoms are surrounded by two imidazole nitrogens (N_2 and $\overline{\mathrm{N}}_2$) and two imidazolate nitrogens (N_4 and $\overline{\mathrm{N}}_4$) in an angular distorted square with sides 2.804(9) and 2.830(9) Å. Considering the symmetry the copper atom must be lying in the plane formed by these four nitrogen atoms; and the calculated copper-nitrogen distances are 2.009(6) Å ($\mathrm{Cu}_1-\mathrm{N}_2$) and 1.975(6) Å ($\mathrm{Cu}_1-\mathrm{N}_4$). The angles between coordinating directions are 89.48(25)° ($\mathrm{N}_2-\mathrm{Cu}_1-\mathrm{N}_4$) and 90.52(25)° ($\mathrm{N}_4-\mathrm{Cu}_1-\overline{\mathrm{N}}_2$). At a distance of 2.755(6) Å below and above this plane oxygen atoms (O_1 and $\overline{\mathrm{O}}_1$)

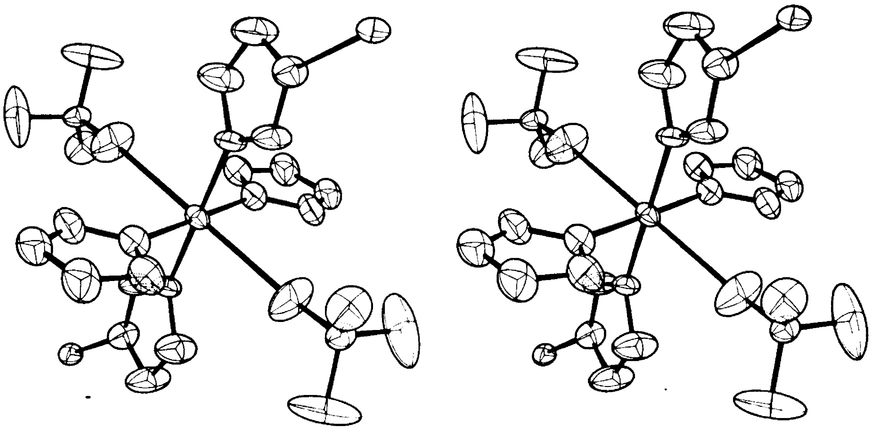


Fig. 3a. Stereoscopic diagram (viewed along the *b*-axis) of the Cu_1-coordination with thermal ellipsoids scaled to enclose 50 % probability.

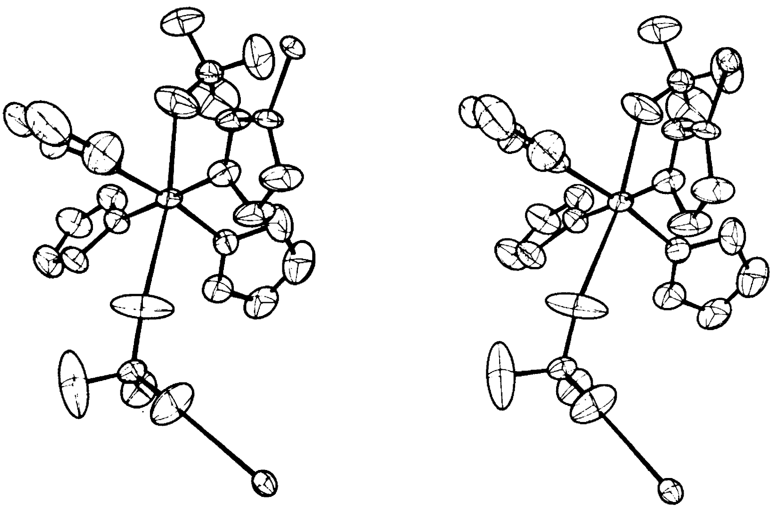


Fig. 3b. Stereoscopic diagram (viewed along the *b*-axis) of the Cu_2-coordination with thermal elipsoids scaled to enclose 50 % probability.

from perchlorate groups are situated completing the octahedral coordination. The angles are: $O_1-Cu_1-\bar{O}_1=180°$, $O_1-Cu_1-N_2=90.57° \pm 0.23$ and $O_1-Cu_1-N_4=89.17° \pm 0.24$, which means that the coordination directions from the copper atom are very close to those in a regular octahedron (Fig. 3a).

The coordination around Cu_2. The Cu_2-atoms are surrounded by three different imidazole nitrogens (N_5, N_7, N_9) and one imidazolate nitrogen (N_3) at distances 1.991(6) Å, 1.987(7) Å, 1.986(7) Å, and 1.976(8) Å, respectively. When the positions of these four nitrogen atoms are used to calculate a plane of best fit (least squares) it is found that N_5 and N_9 are below and N_3 and N_7 are above the plane, all at a distance of 0.067 Å from the plane. The copper atom which is 0.053 Å above this plane is furthermore coordinated to two oxygen atoms O_4 and O_5 from perchlorate groups, the distance Cu_2-O_5 (O_5 above the plane) = 2.581(8) Å being much shorter than the distance Cu_2-O_4 (O_4 below the plane) which is 2.930(7) Å. The calculated bond angles (Table 4b) show considerable distortion from a regular octahedral coordination figure around Cu_2. For example, the angles $O_4-Cu_2-O_5 = 169.83°$, $N_9-Cu_2-O_4 = 84.00°$, and $N_9-Cu_2-O_5 = 85.84°$ which means that both the Cu_2-O_4 and Cu_2-O_5 bonds are bent towards the Cu_2-N_9 bond direction (Fig. 3b).

Cu – N bond length discussion. If we compare the distances from copper to imidazole nitrogens in the two types of octahedrons we find that the three distances Cu_2-N are all within less than one standard deviation from the mean value 1.988 Å and this mean value differs from the Cu_1-N distance of 2.009(6) Å by almost three standard deviations. Comparing the distances from copper to imidazolate nitrogens we find that the distances in the two octahedrons are almost equal namely 1.976(8) Å and 1.975(6) Å. The imidazolate nitrogens seem to be more strongly attached to the Cu atoms and the Cu – N distance is significantly different from at least the Cu_1-imidazole nitrogen distance of 2.009(6) Å. This is also shown in catena-μ-imidazolato-chloro-diimidazolo copper(II), $Cu(C_3H_3N_2)(C_3H_4N_2)_2Cl$ [3] where the imidazolate nitrogens bind to copper forming an infinite chain with $Cu-N(Im^-) = 1.963(5)$ Å and 1.986(5) Å and the distance $Cu-N(ImH) = 2.058(4)$ Å. If Cu – N(ImH) bond lengths are compared with Cu – N(amino) and Cu – N-(peptide) bond lengths previously reported,[16] the shortest distances are found for Cu – N(peptide) with an average of 1.92 Å. The average for the Cu – N-(amino) bond distance is 2.00 Å while the Cu – N(imidazole) bond lengths range from 1.95 Å to 2.01 Å.

These differences may reflect the differences in coordination brought about in some complexes when the ligands are multidentate and in others when there are mixed ligands. In bisimidazolato-copper(II) [10] the reported Cu – N distances range from 1.96 Å to 2.00 Å. In (glycyl-L-histidinato)copper(II) sesquihydrate [11] the distance Cu – N is 1.98 Å where N is the imidazole nitrogen in the dipeptide molecule which is chelating the copper atom.The distance Cu – N is 2.011(7) Å in (β-alanyl-L-histidinato)copper(II) dihydrate [12] where N belongs to the imidazole-ring of the peptide, which is chelating another copper atom. In dihistamino-copper(II) perchlorate [13] the Cu – N-(imidazole) distance is 1.985(9) Å and the histamine molecule forms a chelate with copper. Cu – N(imidazole) distances reported in three complexes of copper(II) with mixed imidazole and glycine peptide ligands [14] range from 1.95(1) Å to 1.96(1) Å which are somewhat shorter than distances in the present work and the other comparisons (Table 4a). In the complexes given above it can be seen that the Cu – N(imidazole) distances can vary from about 1.95 Å to about 2.01 Å. It is reasonable to suggest that these variations depend on the differences in the surroundings of a copper – N(imidazole) bond and corresponding imidazole ring from one coordination situation to another, and general rules for the variations are difficult to find.

Table 4. Bond lengths and bond angles around the copper atoms.

(a) Bond lengths

Present work		Comparison values			
Bond-type	$l(\sigma(l) \times 10^3)$, Å	Bond-type	l, Å	Bond-type	l, Å
$Cu_1-N_4(Im^-)$	1.975(6)	$Cu-N(Im^-)$ [10]	1.96 – 2.00		
$Cu_2-N_3(Im^-)$	1.976(8)	$Cu-N(Im^-)$ [3]	1.986, 1.963		
$Cu_1-N_2(ImH)$	2.009(6)	$Cu-N(ImH)$ [3]	2.058		
$Cu_2-N_5(ImH)$	1.991(6)	$Cu-N(ImH)$ [11]	1.977		
$Cu_2-N_7(ImH)$	1.987(7)	$Cu-N(ImH)$ [12]	2.011		
$Cu_2-N_9(ImH)$	1.986(7)	$Cu-N(ImH)$ [13]	1.985		
		$Cu-N(ImH)$ [14]	1.95, 1.96		
		$Cu-N$ [13]	1.95 – 1.98		
$Cu_1-O_1(ClO_4^-)$	2.755(6)	$Cu-O(ClO_4^-)$[13]	2.617	$Cu-OH_2$ [11]	2.458
$Cu_2-O_4(ClO_4^-)$	2.930(7)	$Cu-O(ClO_4^-)$[14]	2.97	$Cu-OH_2$ [12]	2.483
$Cu_2-O_5(ClO_4^-)$	2.581(8)	$Cu-O=$ [11]	2.943	$Cu-OH_2$ [14]	2.36 – 2.40
		$Cu-O=$ [14]	2.36	$Cu-OH_2$ [15]	2.461, 2.778

(b) Bond angles

Angle	$\theta(10\sigma(\theta))°$	Angle	$\theta(10\sigma(\theta))°$	Angle	$\theta(10\sigma(\theta))°$
$N_2-Cu_1-N_4$	89.5(3)	$\bar{O}_1-Cu_1-N_4$	89.2(3)	$O_4-Cu_2-O_5$	169.9(3)
$N_4-Cu_1-\bar{N}_2$	90.6(3)	$O_1-Cu_1-N_2$	90.6(3)	$O_5-Cu_2-N_3$	90.3(3)
$N_3-Cu_2-N_5$	92.7(3)	$O_4-Cu_2-N_3$	89.5(3)	$O_5-Cu_2-N_5$	101.1(3)
$N_5-Cu_2-N_7$	87.9(3)	$O_4-Cu_2-N_5$	89.1(3)	$O_5-Cu_2-N_7$	88.9(3)
$N_7-Cu_2-N_9$	89.7(3)	$O_4-Cu_2-N_7$	91.3(3)	$O_5-Cu_2-N_9$	85.9(3)
$N_9-Cu_2-N_3$	89.9(3)	$O_4-Cu_2-N_9$	84.0(3)		
$Cu_1-N_2-(C_1-N)$	124.5(6)	$Cu_2-N_3-C_4-$	129.8(7)	$Cu_2-N_7-C_{10}-$	127.0(7)
$Cu_1-N_2-(C_2=C)$	127.1(6)	$Cu_2-N_3-C_6=$	122.9(7)	$Cu_2-N_7-C_{11}=$	126.3(6)
$Cu_1-N_4-C_4-$	130.6(6)	$Cu_2-N_5-C_7-$	126.4(6)	$Cu_2-N_9-C_{13}-$	125.5(7)
$Cu_1-N_4-C_5=$	125.0(6)	$Cu_2-N_5-C_8=$	124.7(7)	$Cu_2-N_9-C_{14}=$	125.7(7)

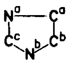
Comparisons

Angle	Range	Mean value	No. in sample
$Cu-N-(C=C)$ (present work)	122.8 – 127.0	125	6
$Cu-N-(C=C)$[18]	121 – 129	125	14
$Cu-N-(C-N)$ (present work)	124.5 – 130.5	127	6
$Cu-N-(C-N)$[18]	121 – 131	126	14

Symmetry and bond lengths in the ligands imidazolate and imidazole. The dimensions of the imidazole rings are about the same as those found in other publications (see Table 5). In the imidazole ring N^a (symbols given in Table 5) should be chemically equivalent to N^b and C^a to C^b which gives a symmetry where we should have equal distances between C^c-N^a and C^c-N^b as well as between N^a-C^a and N^b-C^b. This is not found in the present work where the distances are significantly different: $C_4-N_3=1.308(11)$ Å, $C_4-N_4=1.348(10)$ Å and $N_3-C_6=1.410(13)$ Å, $N_4-C_5=1.346(12)$ Å.

Table 5. Dimensions of the imidazolate (Im^-) and imidazole (ImH) rings.

Designation for the atoms in the ring		N^a	N^b	C^a	C^b	C^c
Corresponding atoms (see Fig. 2) in ring	1	N_2	N_1	C_2	C_3	C_1
	2	N_3	N_4	C_6	C_5	C_4
	3	N_5	N_6	C_8	C_9	C_7
	4	N_7	N_8	C_{11}	C_{12}	C_{10}
	5	N_9	N_{10}	C_{14}	C_{15}	C_{13}



(a) Bond lengths

Bond-type:	C^a-N^a	N^a-C^c	C^c-N^b	N^b-C^b	C^b-C^a
Present work, ring	$l(\sigma(l)\times 10^3)$, Å	$l(\sigma(l)\times 10^3)$, Å	$l(\sigma(l)\times 10^3)$, Å	$l(\sigma(l)\times 10^3)$, Å	$l(\sigma(l)\times 10^3)$, Å
1(ImH)	1.389(11)	1.356(11)	1.362(11)	1.378(10)	1.392(12)
2(Im^-)	1.410(13)	1.308(11)	1.348(10)	1.346(12)	1.372(14)
3(ImH)	1.391(11)	1.301(11)	1.347(12)	1.359(15)	1.397(17)
4(ImH)	1.370(12)	1.334(11)	1.339(12)	1.369(13)	1.389(13)
5(ImH)	1.401(12)	1.349(14)	1.403(17)	1.346(19)	1.381(18)
Mean values	1.39	1.33	1.36	1.36	1.39
Comparison values					
Range in 15 samples [16]	1.32 – 1.44	1.29 – 1.39	1.32 – 1.40	1.33 – 1.41	1.36 – 1.49
Mean values [16]	1.37	1.33	1.35	1.37	1.39
$Cu(Im)_2$ [10]	1.370, 1.375	1.33	1.33	1.370, 1.375	1.39, 1.36
$Cu(Im)(ImH)_2Cl$;[3] (ImH)	1.401(7)	1.325(8)	1.375(9)	1.364(10)	1.385(9)
(Im^-)	1.402(12)	1.329(9)	1.333(9)	1.360(10)	1.365(11)
Free imidazole [17]	1.38	1.33	1.35	1.37	1.36

(b) Angles

Angle type:	$C^a-N^a-C^c$	$N^a-C^c-N^b$	$C^c-N^b-C^b$	$N^b-C^b-C^a$	$C^b-C^a-N^a$
Present work, ring	$\theta(10\sigma(\theta))^\circ$	$\theta(10\sigma(\theta))^\circ$	$\theta(10\sigma(\theta))^\circ$	$\theta(10\sigma(\theta))^\circ$	$\theta(10\sigma(\theta))^\circ$
1(ImH)	108.4(7)	108.2(7)	109.5(7)	106.5(8)	107.7(8)
2(Im^-)	105.9(8)	113.7(7)	104.5(7)	110.9(9)	105.2(9)
3(ImH)	108.8(8)	111.5(8)	106.3(9)	109.0(10)	104.6(10)
4(ImH)	106.5(8)	110.6(8)	108.6(7)	105.4(9)	108.9(8)
5(ImH)	109.0(9)	105.6(10)	111.1(12)	106.3(10)	108.1(10)
Mean values	107.6	110	108	107.5	107
Comparison values					
Range in 15 samples [16]	105 – 113	107 – 114	105 – 109	104 – 110	106 – 111
Mean values [16]	108	111	108	106	108
$Cu(Im)_2$ [10]	105, 106	114, 112	105, 106	108, 108	108, 108
$Cu(Im)(ImH)_2Cl$;[3] (ImH)	107.1	109.6	108.7	106.3	108.3
(Im^-)	104.9	112.9	106.3	108.4	107.6
Free imidazole [17]	105	111	107	106	110

A reasonable explanation is that the perchlorate oxygens are bond to Cu_1 and Cu_2 at different distances and impose a certain distortion to the imidazolate rings especially to the carbon atoms (steric hindrance). The planarity of the imidazole rings is shown in Table 6 where the planes of best fit are given. The distances of the copper atoms from the planes show that the

Table 6 (a). Planes of best fit (least squares).

Plane	Description	
1	Four donor atoms (N_3, N_5, N_7, and N_9)	$0.1994x+0.5706y-0.7966z+2.2371=0$
2	Imidazole ring 1	$0.1299x+0.3963y-0.9089z-4.1761=0$
3	3	$-0.3076x+0.8444y-0.4386z-2.5857=0$
4	4	$-0.6131x-0.2861y-0.7364z-12.2853=0$
5	5	$0.4294x+0.2193y-0.8761z+3.7478=0$
6	Imidazolate ring 2	$0.8403x+0.5406y-0.3972z+14.1028=0$

(b). Deviations in Å from planes (max. standard deviation of the atomic positional fractional coordinates $xyz \times 10^4$ in parentheses).

Plane 1		2		3		4		5		6	
$Cu_2(1)$	0.053	$Cu_1(0)$	−0.122	$Cu_2(1)$	0.174	$Cu_2(1)$	0.037	$Cu_2(1)$	0.053	$Cu_1(0)$	−0.117
$N_3(9)$	0.067	$N_1(8)$	−0.010	$N_5(9)$	0.004	$N_7(9)$	−0.027	$N_9(7)$	0.002	$Cu_2(1)$	−0.376
$N_5(7)$	−0.067	$N_2(7)$	−0.005	$N_6(9)$	−0.007	$N_8(9)$	−0.013	$N_{10}(13)$	0.017	$N_3(9)$	−0.002
$N_7(9)$	0.067	$C_1(9)$	0.009	$C_7(10)$	0.001	$C_{11}(10)$	0.025	$C_{13}(14)$	−0.012	$N_4(6)$	−0.006
$N_9(7)$	−0.067	$C_2(11)$	0.007	$C_8(12)$	−0.008	$C_{11}(10)$	0.019	$C_{14}(11)$	0.008	$C_4(9)$	0.005
		$C_3(10)$	−0.001	$C_5(14)$	0.009	$C_{12}(11)$	−0.004	$C_{15}(12)$	−0.015	$C_5(11)$	0.005
										$C_6(10)$	−0.001

copper atoms do not lie in any of the planes calculated. The way the planes are packed in the space is best shown in the stereoscopic drawings (Figs. 3a and 3b).

The surroundings of the nitrogens (hydrogen bonds). There are six different Cu-bonded nitrogens in one formula unit. These nitrogens are all surrounded by three neighbours, one copper and two carbons which form a nearly planar triangular arrangement (sp^2-hybrid). The sum of the three coordination angles are for all nitrogens close to 360°.

N_2	N_4	N_3	N_5	N_7	N_9
359.90	359.92	358.45	359.87	359.62	359.98

The nitrogens at the other end of the imidazole rings are within hydrogen bond distances from perchlorate oxygens in symmetry related chains [15,18] (Table 8, Fig. 2). A bifurcated hydrogen bond from N_1 to O_2 and O_6 seems very possible. The oxygen atoms which are in hydrogen bond contact to imidazole nitrogens are lying close to the plane of the corresponding imidazole rings. There are some short distances from copper-coordinating nitrogens to the

Table 7. Angles in the perchlorate groups.

Angles	$\theta(10\sigma(\theta))°$	Angles	$\theta(10\sigma(\theta))°$
$O_1-Cl_1-O_2$	110.7(4)	$O_5-Cl_2-O_6$	105.9(5)
$O_1-Cl_1-O_3$	108.0(5)	$O_5-Cl_2-O_7$	108.8(6)
$O_1-Cl_1-O_4$	109.5(5)	$O_5-Cl_2-O_8$	112.4(6)
$O_2-Cl_1-O_3$	111.3(5)	$O_6-Cl_2-O_7$	104.7(6)
$O_2-Cl_1-O_4$	110.2(4)	$O_6-Cl_2-O_8$	108.8(6)
$O_3-Cl_1-O_4$	107.4(7)	$O_7-Cl_2-O_8$	115.9(7)

Table 8. Hydrogen bonds.

Bond	$l(\sigma(l) \times 10^3)$, Å	Bond	$l(\sigma(l) \times 10^3)$, Å
$N_6-H\cdots O_8$	2.877(13)	$\cdot O_2$	3.059(9)
$N_8-H\cdots O_4$	2.944(11)	$N_1-H:$	
$N_{10}-H\cdots O_6$	2.978(15)	$\cdot O_6$	3.062(11)

Table 9. Short distances from copper bonded nitrogens to the carbon atoms of adjacent imidazole rings.

	l, Å		l, Å		l, Å
N_3,C_{13}	2.969	N_7,C_{14}	2.990	N_2,C_4	3.448
N_3,C_8	3.182	N_7,C_7	2.989	N_2,C_5	3.362
N_5,C_6	3.350	N_9,C_{11}	3.327	N_4,C_2	3.452
N_5,C_{10}	3.425			N_4,C_1	3.469

carbon atoms of adjacent imidazole rings (Table 9). These contacts may be formed through limited rotation of the imidazole ligands around a Cu – N bond to give a configuration of lowest energy.

The perchlorate groups and Cu – O bond discussion. There are two different perchlorate groups. One (I) is connected to both Cu_1 and Cu_2 and the other (II) only to Cu_2. The only significant difference between them is that one O – Cl bond distance in group II is 1.385(10) Å which is shorter than the other three 1.432(9) Å, 1.431(8) Å, 1.430(8) Å. This oxygen has the shortest of all hydrogen bond (O . . . H – N) distances to a nitrogen, 2.877 Å. In the perchlorate group I the O – Cl distances are 1.433(7) Å, 1.413(7) Å, 1.422(8) Å, and 1.423(6) Å (International tables[18] 1.41 – 1.48 Å; Bonnet et Jeannin[13] 1.40 – 1.44 Å). The angles are given in Table 7. Three of the four oxygens in both groups are involved in bonds stronger than van der Waals forces with other atoms but O_3 and O_7 are not.

There are three different Cu – O distances. The shortest distance 2.581(8) Å corresponds to a situation where the oxygen belongs to an unshared perchlorate group (II) and the distances 2.755(7) Å and 2.930(7) Å to situations where the oxygens belong to shared perchlorate groups (I). The difference between the last two values is due to the fact that the value 2.93 Å refers to the Cu_2-octahedron, where one perchlorate group is shared and one unshared, while 2.755 Å refers to the Cu_1-octahedron where both the perchlorate groups are shared. If the distances between the oxygens coordinating on either side of a copper atom are compared we find that $(Cu_1-O_1)+(Cu_1-\bar{O}_1)$ and $(Cu_2-O_4)+(Cu_2-O_5)$ are both approximately equal to 5.510 Å.

The distorted octahedral configuration around a copper atom (mostly 4 short bonds and 2 long bonds) is in agreement with what is found in many other determinations and this behaviour is theoretically discussed by Orgel.[9]

Differences between the two longer distances within the same octahedron have also been reported [11,12,14] (Table 4). The tendency towards a five-coordinated Cu(II) atom can be noticed for Cu_2 but the distance 2.93 Å is not long enough to omit a $Cu_2 - O_4$ interaction.

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