

# Crystal Structure of Tetrapropylammonium Dichlorocuprate(I); Comparison of Anionic Configurations in Halocuprates(I) Crystallizing with Symmetrical Tetraalkylammonium and Related Cations

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Andersson, Staffan and Jagner, Susan, 1986. Crystal Structure of Tetrapropylammonium Dichlorocuprate(I); Comparison of Anionic Configurations in Halocuprates(I) Crystallizing with Symmetrical Tetraalkylammonium and Related Cations. – Acta Chem. Scand. A 40: 52–57.

The crystal structure of tetrapropylammonium dichlorocuprate(I) has been determined from single-crystal X-ray diffraction data.  $[\text{N}(\text{C}_3\text{H}_7)_4][\text{CuCl}_2]$  crystallizes in space group  $Pn/2n$  with  $a = 12.628(3)$ ,  $b = 7.587(2)$ ,  $c = 8.869(2)$  Å,  $\beta = 91.39(2)^\circ$  and  $Z = 2$ . Full-matrix least-squares refinement of 116 structural parameters gave  $R = 0.046$  for 839 observed [ $I > 3.0 \sigma(I)$ ] reflections. The dichlorocuprate(I) ion is a nearly linear monomer with  $\text{Cu}-\text{Cl} = 2.071(2)$  Å and  $\text{Cl}-\text{Cu}-\text{Cl} = 178.5(1)^\circ$ . The anionic configurations of tetraphenylarsonium, tetraphenylphosphonium and symmetrical tetraalkylammonium halocuprates(I) characterized hitherto by crystal-structure determination are compared. There would appear to be a tendency towards increased coordination number of copper(I) with decreasing size of the cation. Trends in the concentrations of the ligands in the crystalline phases indicate that the geometry of the anion is determined by the degree of dilution imposed on the ligands by the cations.

Crystal structure studies on chlorocuprates(I),<sup>1–3</sup> bromocuprates(I)<sup>1,2,4–7</sup> and iodocuprates(I)<sup>8–16</sup> crystallizing with tetraphenylarsonium, tetraphenylphosphonium and symmetrical alkylammonium cations indicate that the configuration of the anion in the solid state is strongly dependent on the size of the cation. Suppression of X-Cu-X catenation and prevalence of species with lower copper(I) coordination numbers would seem to be favoured the larger the cation. In the chlorocuprate(I) series, discrete digonal entities have been obtained with tetrabutylammonium,<sup>1</sup> tetraphenylarsonium<sup>2</sup> and tetraphenylphosphonium,<sup>2</sup> whereas the anion in tris(tetraethylammonium) catena- $\mu$ -chloro-ennea- $\mu$ -chloro-heptacuprate(I) is an infinite  $[\text{Cu}_7\text{Cl}_{10}]^{3-}$  chain containing both two- and three-coordinated copper(I).<sup>3</sup> It therefore seemed conceivable that the tetrapropylammonium compound might contain a discrete

anion with trigonal-planar coordinated copper(I). Although discrete bromocuprates(I) containing three-coordinated copper(I), viz.  $[\text{Cu}_4\text{Br}_6]^{2-}$ ,  $[\text{Cu}_2\text{Br}_4]^{2-}$  and  $[\text{Cu}_2\text{Br}_5]^{3-}$  have been obtained with tetrapropylammonium,<sup>5</sup> tetraethylammonium<sup>6</sup> and tetramethylammonium,<sup>7</sup> respectively, and discrete  $[\text{Cu}_2\text{I}_4]^{2-}$  ions with tetrabutylammonium,<sup>8</sup> tetraphenylarsonium,<sup>9</sup> tetraphenylphosphonium,<sup>10</sup> tetrapropylammonium<sup>11</sup> and tetraethylammonium,<sup>10</sup> no such chlorocuprate(I) analogues have yet been reported. In an investigation of the tetrapropylammonium-chlorocuprate(I) system, crystals of tetrapropylammonium dichlorocuprate(I) were obtained and a structural determination undertaken.

## Experimental

Tetrapropylammonium dichlorocuprate(I) was prepared by dissolving tetrapropylammonium chloride and copper(I) chloride (molar ratio 1 : 1)

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in ethanol under nitrogen and allowing the solution to evaporate slowly under nitrogen at room temperature. Colourless rectangular plates, m.p. 99–100°, were deposited from the concentrated solution after approximately a week. Crystals of  $[N(C_3H_7)_4][CuCl_2]$  are monoclinic, space group  $P2/n$  (No. 13,<sup>17a</sup> non-standard setting), with  $a = 12.628(3)$ ,  $b = 7.587(2)$ ,  $c = 8.869(2)$ ,  $\beta = 91.39(2)^\circ$ ,  $Z = 2$ ,  $D_c = 1.25 \text{ g cm}^{-3}$  and  $\mu(\text{MoK}\alpha) = 1.62 \text{ mm}^{-1}$ .

Diffracted intensities from a crystal,  $0.18 \times 0.27 \times 0.25 \text{ mm}$ , were measured at approximately 290 K for  $2\theta < 60^\circ$  with a Syntex  $P2_1$  diffractometer, using graphite-monochromated  $\text{MoK}\alpha$  radiation and the  $\omega$ - $2\theta$  scan mode with a variable  $2\theta$  scan rate of  $3\text{--}29^\circ \text{ min}^{-1}$ . A 96-step profile was recorded for each reflection and the Lehmann and Larsen profile-analysis method<sup>18</sup> was used to calculate the intensities.<sup>19</sup> Of the 2532 independent reflections measured, excluding those systematically absent, 839 had  $I > 3.0 \sigma(I)$  and were considered observed. Correction was made for Lorentz and polarisation effects; an empirical correction<sup>20</sup> for the effects of absorption was made after solution of the structure. The unit-cell dimensions were determined from diffractometer setting angles for 15 reflections.

### Structure determination and refinement

The positions of the copper and chlorine atoms were determined by direct methods (MITHRIL)<sup>21</sup> and those of the nitrogen and carbon atoms from subsequent electron-density maps.<sup>22</sup> Full-matrix least-squares refinement<sup>22</sup> of positional and isotropic thermal parameters gave  $R =$

0.12; after an empirical correction<sup>20</sup> for the effects of absorption  $R = 0.088$ . Allowance for anisotropic thermal parameters gave  $R = 0.066$  for 74 parameters and 839 reflections. Comparable refinement based on the data uncorrected for absorption yielded  $R = 0.069$ . Inclusion of positional parameters for the hydrogen atoms, located from a difference map, gave  $R = 0.046$  (116 parameters; 839 reflections). The isotropic temperature factors of the hydrogen atoms were set equal to the equivalent isotropic values of the carrying carbon atoms (Table 1) and not refined. Atomic scattering factors were taken from the *International Tables for X-Ray Crystallography*<sup>17b</sup> and weights were calculated according to  $w = [\sigma^2(F_o) + 0.0007 F_o^2]^{-1}$ . A final difference map showed a maximum residual electron density of  $0.46 \text{ e } \text{Å}^{-3}$ . Atomic coordinates and equivalent isotropic thermal parameters for the non-hydrogen atoms are given in Table 1. Structure factors, anisotropic thermal parameters, hydrogen-atom coordinates and carbon-hydrogen distances may be obtained from the authors.

### Discussion

In tetrapropylammonium dichlorocuprate(I) the anion is a nearly linear monomer (Fig. 1) with a Cu-Cl distance and a Cl-Cu-Cl angle similar to those determined for e.g.  $[\text{As}(\text{C}_6\text{H}_5)_4][\text{CuCl}_2]$  and  $[\text{P}(\text{C}_6\text{H}_5)_4][\text{CuCl}_2]$ .<sup>2</sup> Bond distances and angles within the cation (Fig. 2) are given in Table 2 and the structure of the compound is illustrated in Fig. 3. The closest distances between copper(I) and carbon and chlorine and carbon are

Table 1. Fractional coordinates and equivalent isotropic thermal parameters ( $\text{Å}^2$ ) for the non-hydrogen atoms in  $[N(C_3H_7)_4][CuCl_2]$ .  $B_{eq}$  is defined as  $\frac{8\pi^2}{3} \sum \sum U_{ij} a_i^* a_j^* a_i \cdot a_j$ . Estimated standard deviations are given in parentheses.

Atom	x	y	z	$B_{eq}$
Cu	0.2500	0.1536(1)	0.7500	5.04(3)
Cl	0.0869(1)	0.1501(3)	0.7689(2)	8.38(6)
N	0.2500	0.0054(7)	0.2500	3.7(1)
C(1)	0.1696(4)	0.1192(7)	0.3325(6)	4.6(2)
C(2)	0.1119(6)	0.2563(9)	0.2363(7)	6.1(2)
C(3)	0.0384(6)	0.3609(10)	0.3386(8)	6.9(2)
C(4)	0.3097(4)	-0.1075(7)	0.3672(6)	4.6(1)
C(5)	0.2431(6)	-0.2423(10)	0.4482(8)	6.5(2)
C(6)	0.3123(7)	-0.3496(10)	0.5533(7)	7.7(2)

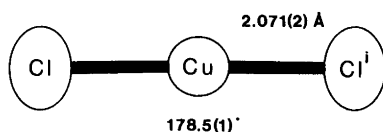


Fig. 1. The dichlorocuprate(I) ion in  $[N(C_2H_5)_4][CuCl_2]$ . Estimated standard deviations in the distance and angle are given in parentheses. The thermal ellipsoids enclose 50% probability.<sup>23</sup> Symmetry code:  $i: \frac{1}{2}-x, y, 1\frac{1}{2}-z$ .

$Cu \cdots C(1) = 3.824(5) \text{ \AA}$  and  $Cl \cdots C(4^i) = 3.642(6) \text{ \AA}$  [symmetry code:  $(i): x - \frac{1}{2}, -y, \frac{1}{2} + z$ ].

Halocuprate(I) ions characterized hitherto by crystal-structure determination of compounds containing symmetrical tetraalkylammonium and related cations are summarized in Table 3. There would appear to be a trend towards increased X-Cu-X catenation and a higher coordination number of copper(I) with decreasing size of the cation. Whereas the coordination of copper(I) in the chlorocuprates(I) and bromocuprates(I) changes from digonal through trigonal to tetrahedral, no crystalline iodocuprate(I) containing two-coordinated copper(I) has yet been isolated. It would also seem that the bromocuprates(I) show less tendency towards formation of infinite chains than do the chlorocuprates(I) and iodocuprates(I). The chlorocuprates(I) differ, moreover, from the bromocuprates(I) and the iodocuprates(I) in that no discrete chlorocuprate(I) containing three-coordinated copper(I) has yet been isolated.

In order to examine the effect of the size of the cation on the anionic configuration, the concen-

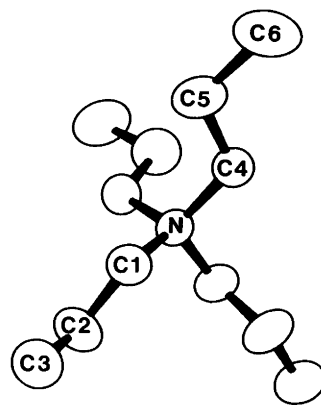


Fig. 2. The tetrapropylammonium ion showing the atomic numbering. The orientation of the cation is approximately the same as that in Fig. 3 in which N is situated at  $\frac{2}{3}, 1-y, \frac{2}{3}$ . The thermal ellipsoids enclose 50% probability.<sup>23</sup> The positions of the hydrogen atoms are not shown.

trations of copper(I) and halide in crystalline halocuprates(I) containing symmetrical tetraalkylammonium and related cations have been calculated (Table 4). Only those compounds for which structural data are available have been included. The tetraethylammonium compound containing the  $[Cu_6I_{11}]^{5-}$  ion has also been omitted since this compound crystallizes as  $[N(C_2H_5)_4]_6[Cu_6I_{11}]I$ .<sup>15</sup> In all three series (Table 4) the ligand concentration is seen to increase with decreasing size of the cation. Although tetraphenylarsonium and tetraphenylphosphonium are formally slightly smaller than tetrabutylammonium, the ligand concentrations in the former halocuprates(I) are slightly

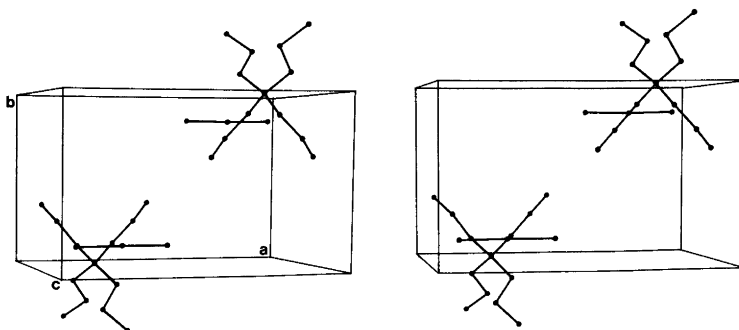


Fig. 3. Stereoscopic view<sup>23</sup> of the unit cell. All atoms are represented as spheres of radius 0.05 Å. Hydrogen atoms have been omitted.

Table 2. Bond lengths (Å) and angles (°) within the tetrapropylammonium ion. Estimated standard deviations are given in parentheses. Symmetry code: (i):  $\frac{1}{2}-x, y, \frac{1}{2}-z$ .

N-C(1)	1.533(6)	N-C(4)	1.532(6)
C(1)-C(2)	1.520(8)	C(4)-C(5)	1.516(9)
C(2)-C(3)	1.535(10)	C(5)-C(6)	1.503(10)
C(1)-N-C(1')	111.4(5)	C(4)-N-C(4')	112.0(5)
C(1)-N-C(4)	108.2(3)	C(1)-N-C(4')	108.6(3)
N-C(1)-C(2)	115.5(4)	N-C(4)-C(5)	115.4(4)
C(1)-C(2)-C(3)	108.1(5)	C(4)-C(5)-C(6)	109.7(6)

lower than in the corresponding tetrabutylammonium compounds. This can undoubtedly be ascribed to the greater rigidity of the phenyl rings as compared with the butyl chains, leading to larger effective volumes for the tetraphenylarsonium and tetraphenylphosphonium cations. The copper(I) concentration also increases with decreasing size of the cation but there are discontinuities in the bromocuprate(I) series: Whereas it is difficult to rationalize the degree of polymerization in the sequence  $[\text{Cu}_2\text{Br}_5]^{3-}$ ,  $[\text{Cu}_2\text{Br}_4]^{2-}$  and  $[\text{Cu}_4\text{Br}_6]^{2-}$  in terms of dilution of copper(I), the experimental configurations are readily understandable in terms of the ligand concentrations.

It would thus appear that the anionic configurations of these halocuprates(I) in the solid state are determined by the degree of dilution imposed on the ligands by the cations. Large cations hin-

der a high overall ligand concentration and thus tend to favour formation of small, discrete entities with a low copper(I) coordination number. Conversely, small cations permit a high ligand concentration resulting in polynuclear species in which copper(I) attains a high coordination number. That several different anions have been obtained with the same cation in the iodocuprate(I) series<sup>10-15</sup> suggests that a given cation may be associated with a fairly broad ligand concentration range within which the formation of two or three species is favourable. It is, however, evident that crystallization of *e.g.* an infinite chlorocuprate(I) chain, containing tetrahedrally coordinated copper(I), with tetrabutylammonium as cation, or of a discrete monomeric diiodocuprate(I) ion with tetramethylammonium as cation, would be extremely unlikely to occur. Several attempts have

Table 3. Halocuprate(I) ions characterized by crystal-structure determination of compounds containing symmetrical tetraalkylammonium and related cations. Infinite chains are denoted as such; the remaining species are discrete entities. C.N. denotes the coordination number of copper(I).

Cation	Chlorocuprate(I)			Bromocuprate(I)			Iodocuprate(I)		
	Species	C.N.	Ref.	Species	C.N.	Ref.	Species	C.N.	Ref.
$\text{N}(\text{C}_4\text{H}_9)_4^+$	$[\text{CuCl}_2]^-$	2	1	$[\text{CuBr}_2]^-$ $[\text{CuBrCl}]^-$	2 2	1 4	$[\text{Cu}_2\text{I}_4]^{2-}$	3	8
$\text{As}(\text{C}_6\text{H}_5)_4^+$	$[\text{CuCl}_2]^-$	2	2				$[\text{Cu}_2\text{I}_4]^{2-}$	3	9
$\text{P}(\text{C}_6\text{H}_5)_4^+$	$[\text{CuCl}_2]^-$	2	2	$[\text{CuBr}_2]^-$	2	2	$[\text{Cu}_2\text{I}_4]^{2-}$	3	10
$\text{N}(\text{C}_3\text{H}_7)_4^+$	$[\text{CuCl}_2]^-$	2	this work	$[\text{Cu}_4\text{Br}_6]^{2-}$	3	5	$[\text{Cu}_2\text{I}_4]^{2-}$ $(\text{Cu}_3\text{I}_4)_\infty$ $[\text{Cu}_5\text{I}_7]^{2-}$	3 3,4 4	11 12 13
$\text{N}(\text{C}_2\text{H}_5)_4^+$	$(\text{Cu}_7\text{Cl}_{10}^{3-})_\infty$	2,3	3	$[\text{Cu}_2\text{Br}_4]^{2-}$	3	6	$[\text{Cu}_2\text{I}_4]^{2-}$ $(\text{Cu}_2\text{I}_3)_\infty$ $[\text{Cu}_6\text{I}_{11}]^{5-}$ $(\text{Cu}_2\text{I}_5)_\infty$	3 4 4 4	10 14 15 16
$\text{N}(\text{CH}_3)_4^+$				$[\text{Cu}_2\text{Br}_5]^{3-}$	3	7			
$\text{NH}_4^+$	$(\text{CuCl}_3^{2-})_\infty$	4	24	$(\text{CuBr}_3^{2-})_\infty$	4	24			

Table 4. Concentrations (mol dm<sup>-3</sup>) of copper(I) and halide, X, in crystalline halocuprates(I) containing symmetrical tetraalkylammonium and related cations.

Compound	[Cu]	[X]	Ref.
<i>Chlorocuprates(I)</i>			
[N(C <sub>4</sub> H <sub>9</sub> ) <sub>4</sub> ][CuCl <sub>2</sub> ]	3.28	6.56	1
[As(C <sub>6</sub> H <sub>5</sub> ) <sub>4</sub> ][CuCl <sub>2</sub> ]	3.00	5.99	2
[P(C <sub>6</sub> H <sub>5</sub> ) <sub>4</sub> ][CuCl <sub>2</sub> ]	3.02	6.05	2
[N(C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> ][CuCl <sub>2</sub> ]	3.91	7.82	present work
[N(C <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> ] <sub>3</sub> [Cu <sub>7</sub> Cl <sub>10</sub> ]	11.0	15.7	3
(NH <sub>4</sub> ) <sub>2</sub> [CuCl <sub>3</sub> ]	10.0	30.1	24
<i>Bromocuprates(I)</i>			
[N(C <sub>4</sub> H <sub>9</sub> ) <sub>4</sub> ][CuBr <sub>2</sub> ]	3.18	6.36	1
[P(C <sub>6</sub> H <sub>5</sub> ) <sub>4</sub> ][CuBr <sub>2</sub> ]	2.93	5.87	2
[N(C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>4</sub> Br <sub>6</sub> ]	6.85	10.3	5
[N(C <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>2</sub> Br <sub>4</sub> ]	5.22	10.4	6
[N(CH <sub>3</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>2</sub> Br <sub>5</sub> O]	5.54	11.1	7
(NH <sub>4</sub> ) <sub>2</sub> [CuBr <sub>3</sub> ]	8.23	24.7	24
<i>Iodocuprates(I)</i>			
[N(C <sub>4</sub> H <sub>9</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>2</sub> I <sub>4</sub> ]	2.95	5.90	8
[As(C <sub>6</sub> H <sub>5</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>2</sub> I <sub>4</sub> ]	2.76	5.53	9
[N(C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>2</sub> I <sub>4</sub> ]	3.60	7.21	11
[N(C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> ] <sub>2</sub> [Cu <sub>5</sub> I <sub>7</sub> ]	7.23	10.1	13
[N(C <sub>3</sub> H <sub>7</sub> ) <sub>4</sub> ][Cu <sub>3</sub> I <sub>4</sub> ]	8.81	11.8	12
[N(C <sub>2</sub> H <sub>5</sub> ) <sub>4</sub> ][Cu <sub>2</sub> I <sub>3</sub> ]	8.28	12.4	14
[N(CH <sub>3</sub> ) <sub>4</sub> ][Cu <sub>2</sub> I <sub>3</sub> ]	10.6	16.0	16

been made to prepare a crystalline diiodocuprate(I) containing a monomeric [CuI<sub>2</sub>]<sup>-</sup> ion using large cations,<sup>25</sup> but, as yet, no such entity has been reported. It is, perhaps, not unconceivable that the degree of ligand dilution necessary for the attainment of this anionic configuration is such that the concentration would be insufficient to permit the formation of a solid phase.

In order to examine the effect of cation size further, as well as lack of symmetry with respect to exposure of the positive charge, attempts are being made to prepare crystalline halocuprates(I) containing unsymmetrical quaternary alkylammonium and related cations. With, for example, phenyltrimethylammonium as cation a discrete [Cu<sub>2</sub>Br<sub>4</sub>]<sup>2-</sup> dimer containing trigonal-planar coordinated copper(I) has been obtained;<sup>26</sup> the ligand concentration in this crystalline phase is 10.8 mol dm<sup>-3</sup>, which is also in agreement with the values obtained for the bromocuprate(I) series in Table 4. Similarly, trimethylsulfonium, which would be

expected to have an exposed unipositive charge, crystallizes with an infinite double [Cu<sub>2</sub>I<sub>3</sub>]<sup>-</sup> chain,<sup>27</sup> the ligand concentration in the crystalline phase being 16.9 mol dm<sup>-3</sup> (cf. Table 4). It seems possible that dilution of the ligands by the cations may prove to be a determinative factor also for the anionic configurations of halocuprates(I) crystallizing with unsymmetrical cations of low positive charge.

*Acknowledgement.* Financial support from the Swedish Natural Science Research Council (NFR) is gratefully acknowledged.

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Received June 18, 1985.