

The Crystal and Molecular Structure of 4,5-Dicyano-2-imidazolyl(phenyl)bromonium Ylide, $C_{11}H_5N_4Br$

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Crystals of $C_{11}H_5N_4Br$ belong to the monoclinic space group $P2_1/c$, with $a=10.742$ (6), $b=5.963$ (4), $c=18.100$ (7) Å, $\beta=99.42$ (5)°, and $Z=4$. The structure has been refined to an R index of 0.059 on the basis of 1389 independent reflections measured on an X-ray diffractometer. This study has confirmed the ylide nature of the compound; the dicyanoimidazolyl and phenyl groups show almost equal attachment to the bromine atom with Br–C bond lengths of 1.890 (6) and 1.929 (8) Å, respectively. The C–Br–C bond angle is 99.3 (3)°, and the angle of twist of the ring systems about the Br–C bonds is 58°. Within the imidazolyl ring there is crystallographic evidence for the localization of the π -electron density.

Introduction

The existence of stable iodonium ylides is now well established (Hayasi, Okada & Kawanisi, 1970; Karele & Neiland, 1968). However, prior to the preparation of the title compound (Sheppard & Webster, 1973), the only mention of a chloronium or bromonium ylide was the proposed bromonium ylide intermediate in the reaction of 3,5-di-*t*-butylbenzene-1,4-diazooxide with 2,6-diisopropyl-4-bromophenol (Pirkle & Koser, 1968). Since no X-ray structural information exists on any halonium ylide, we thought it important to verify the bonding mode of the bromine atom and to obtain the detailed structural parameters of the 4,5-dicyano-2-imidazolyl(phenyl)bromonium ylide.

Experimental

Clear, colorless, needle-shaped crystals of the title compound were grown by slow evaporation of a chloroform solution of the substance. One broad needle, $0.50 \times 0.25 \times 0.05$ mm in size, was mounted along the needle axis in a thin-walled glass capillary. Crystallographic parameters were determined from preliminary data collected on an Enraf–Nonius CAD-4 diffractometer with graphite crystal monochromated copper radiation. Cell dimensions were calculated from a least-squares refinement of the angular settings of 12 reflections, and are given in Table 1.

Intensity data for the hkl – $\bar{h}kl$ quadrant were collected by the ω - 2θ scan technique with a take-off angle of 3.5°. The scan rate was variable and was determined by a fast (20° min⁻¹) prescan. Calculated speeds based on

Table 1. *Crystal data*

$C_{11}H_5N_4Br$	F.W. 272.9
Monoclinic	Space group $P2_1/c$
$a=10.742$ (6) Å	$Z=4$
$b=5.963$ (4)	$F(000)=536$
$c=18.100$ (7)	$V=1143$ Å ³
$\beta=99.42$ (5)°	$D_c=1.59$ g cm ⁻³
$\lambda(Cu K\alpha)=1.5418$ Å	$\mu=85.7$ cm ⁻¹

the net intensity gathered in the prescan ranged from 20 to 0.5° min⁻¹. Moving background counts were collected for 25% of the total scan range at each end of the scan. For each intensity the scan width was determined by the equation: scan range = $A + B \tan \theta$, where $A=0.9^\circ$ and $B=0.45^\circ$. Aperture settings were determined in a similar manner with $A=4$ mm and $B=4$ mm. Two standard reflections, $\bar{3}10$ and 106, were measured periodically and showed no significant change in intensity during data collection.

The standard deviations of the intensities, σ_I , were estimated from the formula

$$\sigma = \{ [C_N + (T_c/2T_B)^2 \cdot (B_1 + B_2)] + (0.02)^2 [C_N + (T_c/2T_B)^2 \cdot (B_1 + B_2)]^2 \}^{1/2}$$

where C_N is the counts collected during scan time T_c and B_1 and B_2 are background intensities, each collected during the background time T_B .

All reflections with 2θ less than 140° were measured. A slow scan was performed on a total of 1389 unique reflections. Since these data were scanned at a speed which would yield a net count of 4000, the calculated standard deviations were all very nearly equal. No reflection was subjected to a slow scan unless a net count of 20 was obtained in the prescan. Based on these considerations, the data set of 1389 reflections

* Contribution No. 2251.

(used in the subsequent structure determination and refinement) was considered observed, and consisted in the main of those for which $I > 2\sigma(I)$.

The intensities were corrected for Lorentz, polarization, and absorption effects (Webe, Busing & Levy, 1962). The linear absorption coefficient was 85.7 cm^{-1} , and the transmission factors varied from 0.36 to 0.60.

Structure solution and refinement

The position of the bromine atom was deduced from a three-dimensional Patterson map, and two structure-factor Fourier-map cycles revealed the positions of all non-hydrogen atoms.

The refinement was based on the least-squares minimization of the quantity $\sum[w(|F_o| - |F_c|)^2]$ with a weighting scheme based on essentially unit weights. Scattering factors for Br, N, and C were taken from the compilations of Cromer & Waber (1965); those for H were from *International Tables for X-ray Crystallography* (1962). The scattering by bromine was corrected for the real and imaginary components of anomalous dispersion using Cromer's (1965) table. Extinction was not deemed to be significant, and no correction was made. Computations were carried out using local modifications of existing programs on either an IBM 360/50 or a UNIVAC 1110 computer.

Isotropic refinement led to an R value of 0.095; a difference Fourier map gave good indications of the positions of the five hydrogen atoms in the asymmetric unit. Several cycles of anisotropic refinement of the non-hydrogen atoms (positional parameters of the hydrogen atoms were not refined) led to the final agreement values shown in Table 2. The largest parameter shifts in the last cycle of refinement were less than 0.2 of their estimated standard deviations. A final difference Fourier map indicated no unaccounted electron density. No systematic variation of $w(|F_o| -$

Table 2. *Agreement indices*

$$R_1 = \frac{\sum||F_o| - |F_c||}{\sum|F_o|} = 0.059$$

$$R_2 = \left\{ \frac{\sum w|F_o|^2 - |F_c|^2}{\sum w|F_o|^2} \right\}^{1/2} = 0.063$$

Goodness-of-fit

$$\left\{ \frac{\sum w|F_o|^2 - |F_c|^2}{(M - S)} \right\}^{1/2} = 2.37$$

$M = 1389$ observations
 $S = 145$ parameters

$|F_c|)^2$ vs $|F_o|$ or $(\sin \theta)\lambda$ was observed. The final parameters are listed in Table 3.*

Discussion

The molecular structure, illustrated in Fig. 1, clearly establishes the ylide nature of the compound, and shows the almost equal attachment of the dicyanoimidazolyl and phenyl groups to the bromine atom. Both Br-C bond distances (Table 4) fall within the normal range bounded by 1.86 Å in bromobenzene (Sutton, 1965) and 1.977(3) Å in 4-bromo-2,3-dicarbomethoxy-2-cyclohepten-1-one (Atwood, Williams, Garner & Cone, 1974). However, the shorter Br-C(imidazolyl) length of 1.890(6) Å vs the Br-C(phenyl) distance of 1.929(8) Å may be readily explained in terms of the

* The table of $|F_o|$ and $|F_c|$ has been deposited with the British Library Lending Division as Supplementary Publication No. SUP 31073 (14 pp., 1 microfiche). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 13 White Friars, Chester CH1 1NZ, England.

Table 3. *Final fractional coordinates and thermal parameters* ($\times 10^4$) for 4,5-dicyano-2-imidazolyl(phenyl)bromonium ylide

Standard deviations in parentheses to last digit quoted. Anisotropic thermal parameters defined by $\exp[-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + 2\beta_{12}hk + 2\beta_{13}hl + 2\beta_{23}kl)]$.

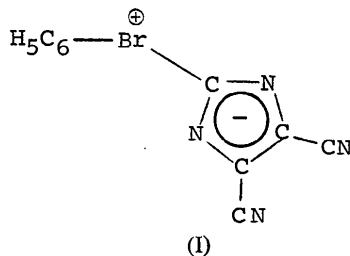
	x/a	y/b	z/c	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
Br	0.39613 (8)	0.37335 (16)	0.32462 (4)	98.6 (9)	340.5 (31)	14.3 (2)	-19.1 (5)	17.8 (3)	1.0 (7)
N(1)	0.4262 (6)	0.2814 (11)	0.1760 (3)	98 (7)	249 (22)	17 (2)	10 (10)	16 (3)	0 (5)
N(2)	0.3033 (6)	0.5896 (11)	0.1900 (3)	90 (7)	271 (25)	20 (2)	6 (10)	20 (3)	-5 (5)
N(3)	0.4409 (8)	0.2339 (13)	-0.0163 (4)	152 (10)	382 (29)	18 (2)	0 (14)	26 (4)	-11 (6)
N(4)	0.2004 (9)	0.8284 (16)	0.0142 (5)	39 (11)	514 (42)	48 (4)	50 (17)	19 (5)	77 (10)
C(1)	0.3705 (7)	0.4193 (12)	0.2199 (4)	83 (7)	240 (28)	16 (2)	17 (11)	16 (3)	0 (6)
C(2)	0.3849 (7)	0.3764 (14)	0.1077 (4)	86 (8)	270 (25)	16 (2)	-3 (12)	17 (3)	-4 (6)
C(3)	0.3102 (7)	0.5622 (13)	0.1162 (4)	79 (8)	305 (30)	18 (2)	1 (12)	14 (3)	11 (6)
C(4)	0.4188 (8)	0.2927 (14)	0.0397 (4)	120 (10)	259 (28)	18 (2)	-14 (12)	17 (4)	-1 (6)
C(5)	0.2511 (8)	0.7155 (16)	0.0607 (5)	90 (9)	412 (37)	30 (3)	17 (14)	21 (4)	19 (8)
C(6)	0.2465 (8)	0.2018 (13)	0.3321 (4)	95 (9)	242 (26)	18 (2)	2 (11)	20 (3)	5 (6)
C(7)	0.1681 (9)	0.2854 (15)	0.3785 (5)	119 (11)	342 (34)	30 (3)	20 (15)	30 (4)	-15 (8)
C(8)	0.0615 (9)	0.1522 (21)	0.3851 (6)	98 (10)	626 (54)	51 (5)	-6 (20)	43 (5)	-5 (13)
C(9)	0.0404 (10)	-0.0437 (19)	0.3474 (7)	105 (12)	470 (47)	62 (5)	-50 (18)	19 (6)	8 (12)
C(10)	0.1207 (11)	-0.1187 (18)	0.3011 (6)	143 (11)	352 (43)	52 (4)	-43 (16)	15 (5)	-13 (12)
C(11)	0.2272 (9)	0.0047 (15)	0.2933 (5)	138 (12)	264 (30)	34 (3)	-14 (15)	28 (5)	-27 (8)

Table 3 (cont.)

Calculated positions 0.96 Å from the bonded carbon atom.
Isotropic thermal parameter set at 5.0 Å².

	x	y	z
H(1)	0.1861	0.4246	0.4055
H(2)	0.0053	0.1988	0.4200
H(3)	-0.1270	-0.1270	0.3519
H(4)	-0.2630	-0.2630	0.2740
H(5)	-0.0385	-0.0385	0.2605

added electrostatic attraction implied by the ylide formulation (I).



The C-Br-C bond angle of 99.3(3)° is indicative of the use of bromine atom *p*-orbitals in the Br-C bonds; the angle is greater than the theoretical 90° value primarily because of non-bonded repulsions between the two organic substituents. Another feature of this portion of the structure is the 'angle of twist' of the ring systems about the Br-C bonds. The normal to the plane formed by Br, C(1), C(6) and C(9) makes an angle of 90° with the normal of the imidazolyl ring, while it makes an angle of 58° with that of the phenyl group (Fig. 2).

Table 4. Bond angles (°)

C(1)-Br-C(6)	99.3 (3)	C(2)-C(3)-C(5)	129.1 (7)
Br-C(1)-N(1)	119.8 (5)	C(2)-C(4)-N(3)	176.3 (9)
Br-C(1)-N(2)	120.4 (5)	C(3)-C(5)-N(4)	176.2 (10)
N(1)-C(1)-N(2)	119.8 (6)	Br-C(6)-C(7)	116.8 (6)
C(1)-N(1)-C(2)	99.8 (6)	Br-C(6)-C(11)	118.6 (6)
C(1)-N(2)-C(3)	101.4 (6)	C(7)-C(6)-C(11)	124.6 (8)
N(1)-C(2)-C(3)	109.8 (6)	C(6)-C(7)-C(8)	115.6 (8)
N(2)-C(3)-C(2)	109.2 (6)	C(7)-C(8)-C(9)	120.8 (9)
N(1)-C(2)-C(4)	122.9 (7)	C(8)-C(9)-C(10)	121.3 (10)
N(2)-C(3)-C(5)	121.7 (7)	C(9)-C(10)-C(11)	120.0 (10)
C(3)-C(2)-C(4)	127.3 (7)	C(10)-C(11)-C(6)	117.6 (9)

Least-squares plane results, given in Table 5, also show the phenyl group itself to be planar to within 0.006 Å; the maximum deviation for any atom of the dicyanoimidazolyl group from its plane is 0.02 Å.

Quite a large number of recent crystallographic studies have dealt with imidazole or the imidazolyl group bonded to various substrates. A compilation of the important parameters of several well-determined structures is presented in Table 6. The imidazole molecule (II) is a good ligand for bonding to metal ions because of its high Lewis basicity imparted by the lone pair of electrons on the nitrogen atom:

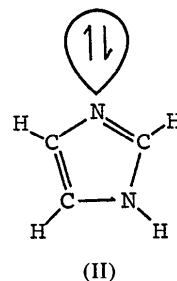


Table 5. Least-squares planes and perpendicular distances from them (Å)

The equations of the planes are expressed in orthogonalized space as $AX + BY + CZ - P = 0$. Atoms marked with an asterisk were not included in the least-squares plane calculation.

Plane 1 (dicyanoimidazolyl moiety)

$$-0.8030X - 0.5834Y - 0.1217Z + 4.6166 = 0$$

N(1)	-0.003	C(2)	0.007
N(2)	-0.012	C(3)	0.008
N(3)	-0.002	C(4)	-0.002
N(4)	0.014	C(5)	-0.019
C(1)	0.010	Br*	-0.032

Plane 2 (phenyl group)

$$-0.4046X + 0.4911Y - 0.7715Z + 4.6612 = 0$$

C(6)	-0.000	C(10)	0.006
C(7)	0.003	C(11)	-0.004
C(8)	-0.001	Br*	-0.050
C(9)	-0.003		

Plane 3

$$0.5277X - 0.8421Y - 0.1118Z + 0.7896 = 0$$

Br	0.005	C(2)*	-0.691
C(1)	0.000	C(3)*	0.696
C(6)	-0.009	C(7)*	-1.045
C(9)	0.004	C(8)*	-1.013
N(1)*	1.168	C(10)*	1.009
N(2)*	-1.127	C(11)*	1.023

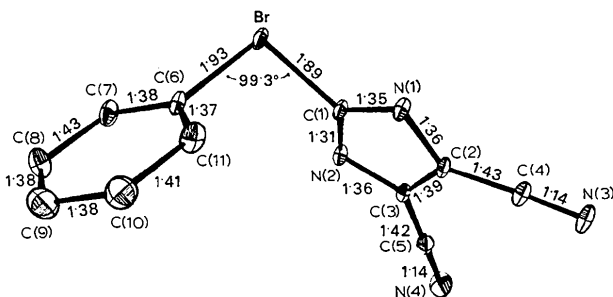


Fig. 1. Molecular structure with the atoms represented as their 40% ellipsoids for thermal motion. The standard deviations of the bond distances are all less than 0.01 Å.

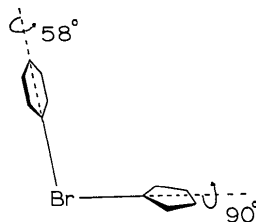
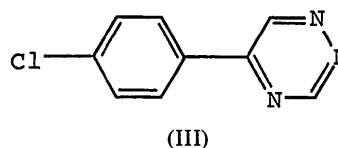


Fig. 2. Orientation of the phenyl and imidazolyl rings relative to the plane of Br, C(1), C(6) and C(9).

However, the coordination of the molecule causes no large or systematic changes of the structural parameters of the ligand itself: those of the four metal ion adducts are quite similar to those of *N*-(β -D-ribofuranosyl)imidazole and of imidazole (Table 6).

In the 4,5-dicyano-2-imidazolyl(phenyl)bromonium ylide, there is crystallographic evidence for the localization of the π -electron density of the imidazolyl ring. In particular, the C(1)–N(2) bond length of 1.310(9) Å may be compared with the C–N(avg) of 1.316(3) Å in 5-(*p*-chlorophenyl)-1,2,4-triazine (Atwood, Krass & Paudler, 1974), a molecule whose electronic structure is thought to be dominated by the canonical form (III):



In the present study the other three C–N lengths are significantly greater (1.350–1.365 Å), than in the triazine, and the C(2)–C(3) bond length, although longer than the 1.335 Å C=C standard (Bartell & Bonham, 1960), is certainly contracted from the 1.43 Å delocalized length quoted for cyclopentadienyl systems (Churchill & Kalra, 1973). Thus there appears to be a dominance of the resonance form (IV):

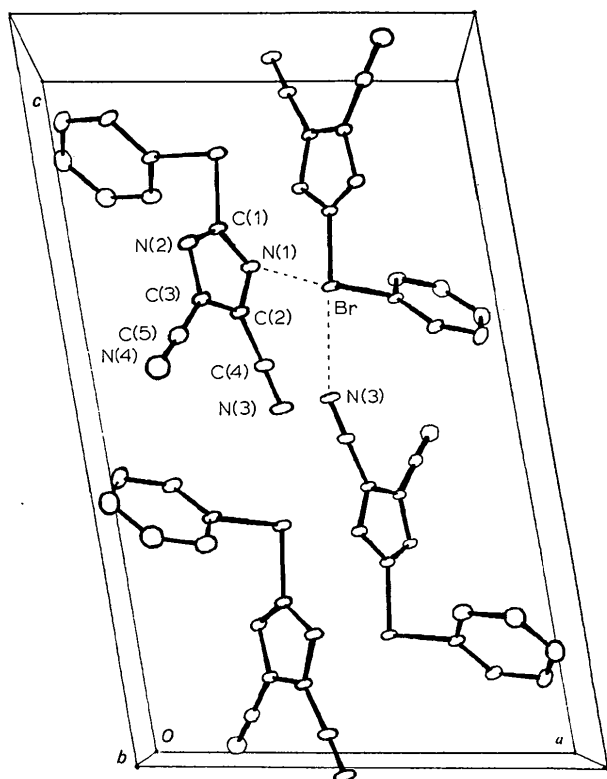
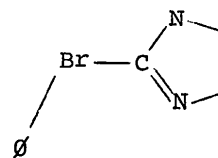


Fig. 3. Packing of four molecules in the unit cell.



A pattern of π -bond localization may be seen for the remaining molecules given in Table 6, but, again, there is no general pattern for the behavior.

The 4,5-dicyano-2-imidazolyl(phenyl)bromonium ylide does differ quite markedly from the others shown in Table 6 in one respect: the N–C–N bond angle of 119.8(6)° is 8° larger than the largest of the remaining molecules. This effect appears as a clear-cut example of the concept of isovalent hybridization (Bent, 1961). In the other molecules, the carbon atom in question is bound to either a carbon or a hydrogen atom. The more electronegative bromine atom demands more *p*-character in the Br–C bond, and as a result the carbon atom must place more *s*-character in the bonds to the nitrogen atoms: hence a larger N–C–N angle.

The parameters associated with the cyano groups are normal in every respect, as are those of the phenyl group. It should be noted that the larger thermal motion of the phenyl-carbon atoms causes greater bond length and angle uncertainties than for the imidazolyl group.

Table 6. Comparison of bond lengths (Å) and angles (°) in selected imidazole complexes

	ImH ^a	Cd(2MImH) ₆ - (BF ₄) ₂ ^b	Cd(ImH) ₆ - (NO ₃) ₂ ^c	Cd(ImH) ₂ Cl ₂ ^d	Ag(ImH) ₂ NO ₃ ^e	ImR ^f	2,4ClImBrC ₆ H ₅
C(1)–N(1)	1.326 (5)	1.341 (3)	1.316 (4)	1.351 (4)	1.323 (10)	1.351 (3)	1.350 (9)
C(1)–N(2)	1.349 (5)	1.350 (5)	1.325 (5)	1.337 (4)	1.336 (11)	1.316 (3)	1.310 (9)
N(1)–C(2)	1.378 (5)	1.337 (5)	1.366 (5)	1.375 (4)	1.364 (10)	1.376 (3)	1.366 (9)
N(2)–C(3)	1.369 (5)	1.347 (7)	1.357 (6)	1.362 (4)	1.404 (11)	1.378 (3)	1.361 (9)
C(2)–C(3)	1.358 (5)	1.352 (5)	1.340 (6)	1.348 (4)	1.360 (11)	1.360 (3)	1.391 (9)
N(1)–C(1)–N(2)	111.3 (3)	107.6 (3)	110.6 (3)	111.2 (2)	110.9	111.9 (2)	119.8 (6)
C(1)–N(1)–C(2)	105.4 (3)	106.2 (3)	106.1 (3)	105.4 (2)	107.1	107.0 (2)	99.8 (6)
C(1)–N(2)–C(3)	107.2 (3)	110.6 (3)	107.7 (3)	107.7 (2)	107.1	105.0 (2)	101.4 (6)
N(1)–C(2)–C(3)	109.8 (3)	112.0 (4)	108.9 (4)	109.8 (2)	109.4	105.7 (2)	109.8 (6)
N(2)–C(3)–C(2)	106.3 (3)	103.6 (5)	106.6 (4)	106.0 (2)	105.5	110.3 (2)	109.2 (6)

References: (a) Martinez-Carrera (1966); (b) Reedijk & Verschoor (1973); (c) Mighell & Santoro (1971); (d) Flook, Freeman, Hug & Rosalky (1973); (e) Antti & Lundberg (1971); (f) James & Matsushima (1973).

The unit-cell packing, given in Fig. 3, is dominated by the tendency for the Br^+ ion to associate itself with both the negatively charged imidazolyl ring and the cyano nitrogen atom on neighboring molecules. The $\text{Br}-\text{N}(3)$ ($x, \frac{1}{2}-y, \frac{1}{2}+z$) separation is $2.912(6)$ Å, and the $\text{Br}-\text{N}(1)$ ($1-x, \frac{1}{2}+y, \frac{1}{2}-z$) distance is $3.094(6)$ Å; the next closest approach to Br is $\text{C}(3)$ ($1-x, -\frac{1}{2}+y, \frac{1}{2}-z$) at $3.665(8)$ Å. There are many intermediate non-bonding lengths not involving the bromine atom, the shortest of which is $\text{N}(3)-\text{N}(3)$ ($1-x, \bar{y}, \bar{z}$) at $3.08(2)$ Å.

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The Crystal and Molecular Structure of Dichlorobis-(*N,N*-diisopropylacetamidinato)methyltantalum(V)–1·0 Benzene

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Crystals of the title compound are monoclinic, space group $P2_1/n$, with $a=11.772$ (12), $b=16.250$ (13), $c=15.632$ (13) Å, $\beta=109.98$ (12)°, $Z=4$. The intensities of 1748 independent reflexions above background were collected by counter methods and refined to R 0.079. The structure of the molecule is disordered but it is clear that the seven-coordinate monomer has a distorted pentagonal bipyramidal geometry with the two bidentate ligands occupying the pentagonal girdle. While the two chlorine atoms occupy the axial positions and the methyl group the fifth equatorial position in most unit cells, there is evidence for interchange of position between the methyl group and the chlorine atoms in some unit cells. There is also evidence for disorder in the fifth equatorial position.

During our studies of the structure of seven-coordinate complexes, we have prepared the four compounds of formula $\text{Me}_n\text{TaCl}_{3-n}[\text{R}-\text{N}-\text{C}(\text{Me})-\text{N}-\text{R}]_2$ with $n=0, 1$ and $\text{R}=\text{C}_3\text{H}_7$, C_6H_{11} and have carried out crystal structure analyses of three; namely with $n=0$, $\text{R}=\text{C}_3\text{H}_7$ [monoclinic form, Drew & Wilkins (1974a),

orthorhombic form, Drew & Wilkins (1975a)], with $n=0$, $\text{R}=\text{C}_6\text{H}_{11}$ (Drew & Wilkins, 1975b) and with $n=1$, $\text{R}=\text{C}_6\text{H}_{11}$ (Drew & Wilkins, 1974b). In this paper we report the crystal structure of the fourth compound $\text{MeTaCl}_2[\text{C}_3\text{H}_7-\text{N}-\text{C}(\text{Me})-\text{N}-\text{C}_3\text{H}_7]_2$, (I)