Inorganica Chimica Acta, *81* (1984) 127–132

On the Interaction Between Mercury(II) Cyanide, or Chloride, and Diazoles

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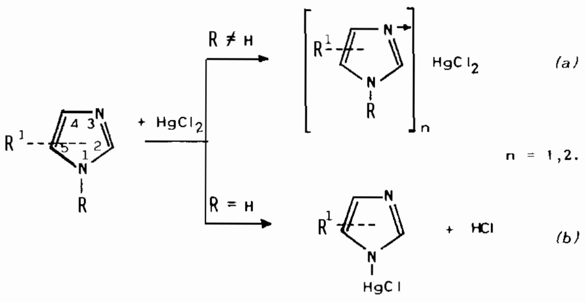
Received June 4, 1983

Adducts such as $L_2Hg(CN)_2$ (L = imidazole, substituted imidazoles, or 3,5-dimethylpyrazole), and (1-vinylimidazole)$_4Hg_3(CN)_6$ are obtained and characterized through infrared and nmr data, while the preparation of $HgCl_2$(3,5-dimethylpyrazole)$_2\cdot 2H_2O$ cannot be repeated. The adducts are dissociated in acetone solution. The different behaviour of mercury(II) chloride and cyanide is discussed.

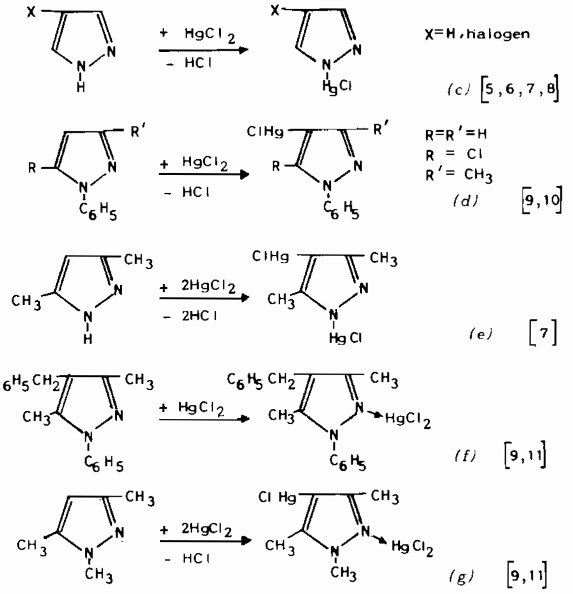
Introduction

While the coordination chemistry of mercury(II) halides is well established and a recent review on the subject is available [1], the same cannot be said about the behaviour of mercury(II) halides or pseudohalides towards diazoles, in spite of the fact that imidazole [2] or, to a minor extent, pyrazole [3] rings are constituents of several important natural products.

In a preceding paper of ours [4], imidazole (ImH) and various substituted imidazoles (imH) were found to react with mercury(II) chloride giving either N-adducts or N-mercurated derivatives, according to one of the two following patterns:



Path (a) is followed with nitrogen substituted imidazoles, path (b) with 1-unsubstituted ligands, and no C-mercurated derivative has been found. While the reactions of this class of diazoles with mercury chloride are now easily classified, the same cannot be said of others of the diazole family. Indeed the action of $HgCl_2$ on a pyrazole may yield different types of products, namely N-mercurated derivatives (c), C-mercurated compounds (d), C- and N-mercurated species (e), adducts (f) when neither C- nor N-ring mercuration is possible, or even adducts of already mercurated products as in (g):



In addition, there is a report [12] on the isolation of adducts such as $HgCl_2L_2\cdot 2H_2O$ and $Hg(NO_3)_2L_2\cdot 4H_2O$ where L is 3,5-dimethylpyrazole: the existence of such species does not fit into the reaction patterns listed above.

In spite of such a varied reactivity of diazoles toward mercury(II) chloride, we assumed that another HgX_2 compound could be found which might be able to give only adducts and might be unable to undergo either C- or N-mercuration. Such a compound will allow the study of the interaction

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0020-1693/84/$3.00

TABLE I.

Compound and Formula	M.P. °C	Yield %	Elemental Analysis %[a]			Additional Data[b]
			C	H	N	
1) $(ImH)_2 \cdot Hg(CN)_2$	110–	64	24.51	2.21	21.83	
$C_8H_8HgN_6$	113		24.71	2.07	21.62	
2) $(2\text{-}MeImH)_2 \cdot Hg(CN)_2$	145–	72	28.69	2.90	20.17	$\Lambda = 3.87$ ohm^{-1} cm^2 mol^{-1}
$C_{10}H_{12}HgN_6$	146		28.65	2.86	20.05	Mw = 220 (calcd: 418.83)
3) $(4(5)\text{-}MeImH)_2 \cdot Hg(CN)_2$	138	72	28.82	3.09	20.42	
$C_{10}H_{12}HgN_6$			28.80	2.88	20.16	
4) $(benzoImH)_2 \cdot Hg(CN)_2$	205	89	39.42	2.69	17.47	
$C_{16}H_{12}HgN_6$			39.27	2.45	17.18	
5) $(1\text{-}MeIm)_2 \cdot Hg(CN)_2$	115	71	28.36	2.69	20.44	$\Lambda = 3.22$ ohm^{-1} cm^2 mol^{-1}
$C_{10}H_{12}HgN_6$			28.79	2.88	20.15	Mw = 201 (calcd: 416.83)
6) $(1,2\text{-}Me_2Im)_2 \cdot Hg(CN)_2$	140	67	32.02	3.64	19.17	
$C_{12}H_{16}HgN_6$			32.37	3.60	18.88	
7) $(1\text{-}vinylIm)_4 \cdot 3Hg(CN)_2$	65	93	27.97	2.36	17.64	
$C_{26}H_{24}Hg_3N_{14}$			27.50	2.11	17.27	
8) $(1\text{-}PhCH_2Im)_2 \cdot Hg(CN)_2$[c]	114	61	46.75	3.59	14.77	
$C_{22}H_{20}HgN_6$			46.45	3.52	14.80	
9) $(3,5\text{-}Me_2PzH)_2 \cdot Hg(CN)_2$	82	80	32.55	3.19	18.37	
$C_{12}H_{16}HgN_6$			32.38	3.59	18.89	

[a] Found/calcd. [b] Conductivity was measured in a 10^{-3} molar acetone solution at 20 °C; molecular weight was determined by osmometry in acetone solution (see Text). [c] Ref. 4.

TABLE II. Selected Infrared Data.[a]

Compound	ν(N–H)	ν(C–H) azole	ν(C≡N)	1500–1600	low frequency region
1) $(ImH)_2 \cdot Hg(CN)_2$	3210m	3123m	2180m	1530m, 1510w	420m, 320w, 273w
2) $(2\text{-}MeImH)_2 \cdot Hg(CN)_2$	3200m	3122m, 3110m	2180m[b]	1565m	400m, 382m, 370m, 310w, 270w
3) $(4(5)\text{-}MeImH)_2 \cdot Hg(CN)_2$	3200m	3120m	2180m	1578m, 1540vw, 1520w	408w, 360m, 318w, 280m
4) $(benzoImH)_2 \cdot Hg(CN)_2$	3178m	3120m	2182w	1595m	432m, 422m, 388vw, 320w, 270w
5) $(1\text{-}MeIm)_2 \cdot Hg(CN)_2$		3130m, 3120m	2180w[b]	1530m, 1514m	448w, 413m, 320vw
6) $(1,2\text{-}Me_2Im)_2 \cdot Hg(CN)_2$		3140w, 3120m	2178w	1570w, 1538m, 1510m	440m, 400m, 312w, 278m
7) $(1\text{-}vinylIm)_4 \cdot 3Hg(CN)_2$[c]		3160m, 3120m	2180m	1590w, 1505m	445m, 429m, 350vw, 339vw
8) $(1\text{-}PhCH_2Im)_2 \cdot Hg(CN)_2$[d]		3120m	2180w	1590w, 1520s	482m, 428m, 318w, 278w
9) $(3,5\text{-}Me_2PzH)_2 \cdot Hg(CN)_2$	3240m	3150m, 3120m	2180m	1580m	450w, 420w, 380m, 310vw, 280w

[a] Nujol mull. [b] Acetone solution: 2180 broad. [c] ν(C=C) at 1648 cm^{-1}. [d] Ref. 4.

between diazoles and mercury(II) without the complications due to any further reaction, such as mercuration. Therefore we tried mercury(II) cyanide for which the following equilibria are completely shifted to the left [13], while for the chloride they are somewhat shifted to the right [13] indeed a mercury(II) derivative produced by such equilibria is likely to be responsible for the reported ring mercurations.

$$HgX_2 + H_2O \rightleftharpoons Hg(OH)X + H^+ + X^-$$

$$HgX_2 \rightleftharpoons HgX^+ + X^- ;$$

TABLE III. Proton Magnetic Resonance Data.

Compound	Solvent	δ values[a] for azole substituents in position		
		1	2	4 + 5
ImH	$(CD_3)_2CO$	12.97s, NH	7.87s	7.18s
$(ImH)_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	10.73s, broad, NH	7.90s	7.22s
2-MeImH	$(CD_3)_2CO$	9.50s, broad, NH	2.37s, Me	6.97s
$(2\text{-MeImH})_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	10.60s, broad, NH	2.42s, Me	7.05s
4(5)-MeImH	$(CD_3)_2CO$	11.77s, NH	7.68s	2.23s, Me; 6.84s
$(4(5)\text{-MeImH})_2 \cdot Hg(CN)_2$[b]	$(CD_3)_2CO$	9.80s, broad, NH	7.74s	2.23s, Me; 6.90s
benzoImH	$(CD_3)_2CO$	11.67s, v. broad, NH	8.30s	7.12–7.38m; 7.58–7.82m, C_6H_4
$(benzoImH)_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	[c]	8.37s	7.18–7.45m; 7.68–7.88m, C_6H_4
1-MeIm	$(CD_3)_2CO$	3.63s, Me	7.51s	7.03s; 6.97s
$(1\text{-MeIm})_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	3.80s, Me	7.70s	7.02s; 7.20s
$1,2\text{-Me}_2Im$	$(CD_3)_2CO$	3.47s, Me	2.22s, Me	6.80s, 6.92s
$(1,2\text{-Me}_2Im)_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	3.64s, Me	2.37s, Me	6.90s; 7.09s
1-vinylIm	$(CD_3)_2CO$	4.78d; 4.89d; 5.31d; 5.52d; 7.03s; 7.23s; 7.32s	7.48s	7.09d; 7.93s
$(1\text{-vinylIm})_4 \cdot 3Hg(CN)_2$	$(CD_3)_2CO$	5.00d; 5.14d; 5.53d; 5.72d; 7.28s; 7.35s; 7.44s	7.67s	7.19s; 8.09s
1-PhCH_2Im	$CDCl_3$	5.00s, CH_2	7.47s	6.84d; 6.95–7.40m
$(1\text{-PhCH}_2Im)_2 \cdot Hg(CN)_2$[f]	$CDCl_3$	5.07s, CH_2; 7.00–7.50m, Ph	7.63s	6.92c
$3,5\text{-Me}_2PzH$	$(CD_3)_2CO$	11.98s, NH	5.81s[d]	2.21s, Me[e]
$(3,5\text{-Me}_2PzH)_2 \cdot Hg(CN)_2$	$(CD_3)_2CO$	11.33s, v. broad, NH	5.93s[d]	2.27s, Me[e]

[a] s = singlet; d = doublet; m = multiplet; c = complex. [b] Unchanged down to −60 °C. [c] Not observed, but ν(NH) was found in the infrared spectrum. [d] 4-position. [e] 3- and 5-position. [f] Ref. 4.

Here we report the results obtained by reaction of various imidazoles and of two pyrazoles with mercury cyanide; in addition were investigated the products claimed to be mercury chloride or nitrate adducts of 3,5-dimethylpyrazole [12].

Results and Discussion

The interaction of mercury(II) cyanide with a diazole gave the corresponding adducts, white, crystalline, and air-stable according to the reaction

$$Hg(CN)_2 + nL \rightleftharpoons L_2Hg(CN)_2$$

1–9

From analytical and spectral data (Tables I–III) all the compounds but 7 were identified as 2:1 adducts; they are not electrolyte in acetone solution, thus ruling out ionic structures such as $[L_4Hg]$-$[Hg(CN)_4]$, *i.e.* a derivative of a very stable complex anion. The diazoles employed include both 1-unsubstituted and 1-substituted imidazoles, as well as those having a methyl group on the 2-, or 4(5)-position, or a 1-benzyl group or benzoderivative. No product was isolated with pyrazole ($pK_a = 2.52$), while the more basic 3,5-dimethylpyrazole ($pK_a = 4.37$) [14] gave smoothly the compound 9 although such a ligand undergoes both N- and C-mercuration with mercury chloride according to the reaction (e). Similarly no compound was isolated from 2-phenylimidazole, 2-PhimH, less basic than imidazole or the three methylimidazoles; the singular reactivity of 2-PhimH was already ascertained in the chemistry of gold(I), where $(2\text{-PhimAu})_n$ rather than an adduct of the type $(imH)_nAuCl$ was isolated [15]. The reaction was carried out in water (compounds 1, 2, 3, 5 and 6), in water plus a methanol solution of the ligand (compound 4), in methanol (compound 6), or even in diethylether (compound 9); the yields were generally good.

The infrared spectra (Table II) showed the presence of the cyanide and of the organic ligands.

For the latter a ν(CH) due to the heterocycle was found at above 3100 cm^{-1}; ν(NH) was observed at *ca.* 3200 cm^{-1} for the derivatives of 1-unsubstituted azoles, while at least one band in the 1500–1600 cm^{-1} region is typical of the 'ring breathing' vibration. In the low-frequency region vibrations due to ν(Hg–C) or to δ(HgCN) (at 442 or 341 cm^{-1}, resp. in $Hg(CN)_2$ [16]) cannot be assigned owing to the presence of the organic ligand. The ν(C≡N) band was observed at 2180 ± 2 cm^{-1} in all the complexes, *i.e.* at a value lower than in the starting mercury(II) cyanide (2194 cm^{-1}). The lowering of the vibration can be explained as the result of two possible canonical forms

$$-\text{Hg}-\text{C}{\equiv}\text{N}\text{: (A); (B) }\overset{+}{\text{Hg}}{=}\text{C}{=}\overset{-}{\underline{\text{N}}}$$

Indeed, an increase in the coordination number results in an increase in the negative charge on the Hg atom; as a result the contribution of (B) is now bigger, thus decreasing the CN frequency. However, the lowering of ν(CN), $\Delta\nu$, is only 15 cm^{-1}, much less than the $\Delta\nu$ observed upon coordination of a very good donor; such as CN^-: for example ν(CN) was found at 2161 or 2148 cm^{-1} in $Hg(CN)_3^-$ or in $Hg(CN)_4^{--}$, resp. [17]. A big value of $\Delta\nu$ is paralleled by a sharp increase in the molar extinction coefficient along the series $Hg(CN)_2$, $Hg(CN)_3^-$, and $Hg(CN)_4^{--}$, while qualitative assessment of the intensity of ν(CN) in the series $(diazole)_2Hg(CN)_2$ reveals only modest changes in comparison with mercury cyanide. Since the infrared evidence suggests that the complexes here reported do not contain a strong N → Hg donor bond it is not surprising that molecular weight determinations by osmometry in acetone solution give a result which is approximately half the calculated value. Clearly, solvation takes place according to the scheme

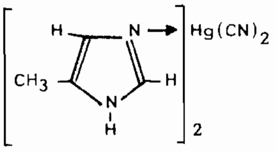
$$L_2Hg(CN)_2 + n\cdot\text{solvent} \rightleftharpoons L_{2-n}(\text{solvent})_nHg(CN)_2 + nL$$

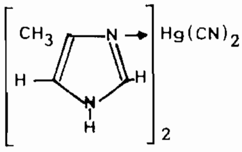
$$n = 1, 2$$

It should be noted that, accordingly, the complexes obtained are soluble in donor solvents, such as acetone, methanol, ethanol, acetonitrile or nitromethane, but are insoluble in benzene, carbon tetrachloride, chloroform or dichloromethane.

The proton nmr spectra of the compounds reveal that the organic ligands do not undergo any alteration when the complexes are formed. In the complex the CH or CH_3 signals are generally displaced to lower field by comparison with the ligand. This observation proves that the dissociation established through molecular weight determinations is only partial and that much of the complex is still present. However, such a dissociation and/or fast exchange of the N–H proton, between the nitrogen atoms or with the solvent, explains why the 4- and 5-CH protons of imidazole in 1, of 2-methylimidazole in 2, or the 3- and 5-methyl protons of 3,5-dimethylpyrazole in 9 are magnetically equivalent.

With 4(5)-methylimidazole the complex 3 is formed which may exist either as a 5-methyl- or as a 4-methylimidazole adduct:





In the proton nmr spectra, unchanged down to –60 °C, the methyl signal was found at the same value in the complex as in the parent diazole, while the CH signals are displaced to lower field upon coordination. Therefore the complex should preferably be formulated as the 5-methyl isomer, the one having a bigger distance between the CH_3 group and the donor nitrogen atom. In addition, a C-13 nmr spectrum was recorded for 3 and for $Hg(CN)_2$. The spectrum of the acceptor showed only one signal at 144.12 ppm from Me_4Si; the satellites expected for the presence of ^{199}Hg nuclei having spin ½ could not be observed owing to the limited solubility of the compound, to the low abundance of the isotope (16.86%) and, finally, to the fact that satellites due to $^{199}Hg-^{13}C$ scalar coupling are significantly broader than the uncoupled ^{13}C resonance [18]. The signal due to the cyano group was observed in the adduct at 143.43 ppm; in addition signals at 11.83, 133.13, 119.80, and 136.79 were observed and assigned through an off-resonance spectrum to the methyl group, to the 5-, 4-, and 2-C respectively, because they are quartet, singlet, doublet and doublet respectively. Of the doublets the higher was assigned to the 2-C by analogy with the spectrum of imidazole, for which values of 135.7 (2-C) and 121.8 (4- and 5-C) were found [19].

Therefore the available evidence suggests that all the complexes but 7 may be considered as additional examples of four-coordination around the metal atom. Although no structural assignment can be proposed for the 1-vinylimidazole derivative, 7, the presence of ν(C=C) at 1648 cm^{-1} proves that the double bond of the olefin is not involved in coordination.

Finally, the reaction between mercury(II) chloride and 3,5-dimethylpyrazole (pzH) was repeated in the experimental conditions mentioned in the literature [12] and detailed in the experimental part. In no case was an adduct obtained: neither was $HgCl_2(pzH)_2\cdot 2H_2O$ nor anything approaching this formulation.

From the reaction a white powder was indeed obtained which could not be purified because of its insolubility. Since the infrared spectrum did not show any absorption above 3000 cm^{-1}, neither water nor an NH-function is present, thus disproving the structure proposed; in addition, a complete analysis suggests a formula such as $Hg_6Cl_7(pz)_4$. This powder is likely to be a mixture of various mercury pyrazolates; if the metal has the usual +2 oxidation state, then the presence is required of three monoanionic pz^- anions, $C_5H_7N_2^-$, and of one dianionic pz^{--} dianion, $C_5H_6N_2^{--}$; at the moment two metal compounds are known where a twice deprotonated pyrazole is present, namely a mercury(II) (see reaction (e), above [7]) and an alkylplatinum(II) derivative [20].

In conclusion, mercury(II) cyanide gives adducts, $L_2Hg(CN)_2$ (L = imidazole, or substituted imidazoles, or 3,5-dimethylpyrazoles), or (1-vinylimidazole)$_4Hg_3(CN)_6$, thus revealing a different behaviour in comparison with mercury(II) chloride. Usually this last reacts with 1-unsubstituted diazoles giving N-mercurated products, or with 1-substituted diazoles giving either C-mercurated compounds (pyrazoles) or N-mercurated derivatives (imidazoles). It is not surprising, then, that the preparation of the adduct $HgCl_2$(3,5-dimethylpyrazole)$_2 \cdot 2H_2O$ [12] cannot be repeated, while an adduct can be obtained when the cyanide replaces the chloride.

The results reported here also make evident the fact that the reactivity of mercury(II) depends very much on the counter-anion involved. As a consequence, any generalization should be looked at with caution, especially if it concerns the behaviour of mercury(II) in solution towards natural products containing diazole rings, such as nucleic acids or purines.

Experimental

The samples were pumped to constant weight (20 °C, *ca.* 0.1 Torr). Carbon, hydrogen, and nitrogen analyses were carried out by Mr. A. Canu (University of Sassari). Infrared spectra from 4000 to 250 cm^{-1} were recorded on a Perkin-Elmer 457 instrument; ^{1}H-nmr spectra were recorded on a Varian EM-390 instrument operating at room temperature and 90 MHz. Molecular weight determination by osmometry were carried out by Pascher's Mikroanalytisches Laboratorium, Bonn (FRG). Analytical, infrared and nmr data are given in the Table I, II and III respectively.

Bis(benzoimidazole)dicyanomercury(II), (4)

To an aqueous solution (6 ml) of mercury(II) cyanide (250 mg; 0.99 mmol) an ethanol solution (5 ml) of benzoimidazole (500 mg; 4.23 mmol) was added under stirring. The white precipitate (0.43 g) was filtered and washed with water and with diethyl ether. The compound is soluble in acetone, methanol, acetonitrile, and nitromethane, but insoluble in benzene, ether, nitrobenzene and chlorinated methanes.

The compound 1 was prepared similarly, while ethanol was omitted in the preparation of 2, 3, 5, 6, and 8.

Bis(3,5-dimethylpyrazole)dicyanomercury(II), (9)

The compound 9 was obtained by stirring a suspension of mercury cyanide (257 mg; 1.02 ml) in diethyl ether (15 ml) with an ether solution (15 ml) of 3,5-dimethylpyrazole (310 mg; 3.23 mmol); the precipitate was washed with water and with diethyl ether.

Tetrakis(1-vinylimidazole)hexacyanotrimercury(II), (7)

After 3 days a methanol solution (20 ml) of mercury cyanide (280 mg; 1.11 mmol) and 1-vinylimidazole (420 mg; 4.46 mmol) was concentrated under reduced pressure to small volume (*ca.* 3 ml); eight drops of water were added and the precipitate (390 mg) was filtered. The compound is insoluble in benzene, ether, chlorinated solvents, and nitrobenzene, and is soluble in acetone, methanol, acetonitrile, nitromethane and dimethylsulphoxide.

Interaction of mercury(II) chloride with 3,5-dimethylpyrazole

The attempt to prepare the adduct $HgCl_2(C_5H_8N_2)_2 \cdot 2H_2O$ claimed in the literature [12] was carried out as follows. Mercury(II) chloride (462 mg; 1.70 mmol) and 3,5-dimethylpyrazole (358 mg; 3.73 mmol) in 50% ethanol (30 ml) gave a white precipitate, which after a few hours was filtered and washed with ethanol (120 mg). Found: C, 13.13; H, 1.66; Cl, 13.5; N, 6.10; Hg, 65.4%. The compound is insoluble in common organic solvents; upon heating it blackens gradually without melting up to 280 °C. Similarly negative results were obtained for the preparation of $Hg(NO_3)_2(C_5H_8N_2)_2 \cdot 4H_2O$ [12].

Acknowledgements

We thank Mrs. Maria Bonfà (University of Milan) for ^{13}C nmr spectra, the CNR for financial support, and Biblioteca Chimica della Facoltà di Scienze MFN (University of Milan).

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