substituted, there is no noticeable difference between the two types of S—C distances (Ealick, van der Helm, Ramalingam, Thyvelikakath & Berlin, 1977; Ealick, van der Helm, Baker & Berlin, 1979; Andreetti, Bocelli & Sgarabotto, 1978). The endocyclic C—S—C angle of 102·0 (2)° is close in value to that observed in the other compounds and seems to be independent of the substitution pattern in the heterocyclic ring.

The vinyl chain is twisted out of the plane of the thiochroman ring system by more than 30° [torsion angle, C(7)—C(6)—C(11)—C(13) = 33·1 (7)°] and has nearly ideal *anti* conformation for its double bond, C(11)—C(13) [torsion angle, C(6)—C(11)—C(13)—C(14) = −174·1 (4)°]. This places the methyl carbon, C(12), in a *syn* position with respect to C(14). The resulting steric strain is reflected in the angles C(12)—C(11)—C(13) of 123·6 (3)° and C(11)—C(13)—C(14) of 128·5 (2)°, both of which are significantly larger than 120°. The conformation about the C(13)—C(14) bond is skewed [torsion angle C(11)—C(13)—C(14)—P(15) = 92·6 (4)°] and as a result the triphenylphosphonium system is folded back towards the thiochroman ring system. The geometry around the P atom is nearly tetrahedral with an average P—C(sp^2) distance of 1·797 (4) Å and a slightly longer P—C(sp^3) distance of 1·809 (4) Å.

The Br^- anion has a number of short contacts with C atoms [$Br^-\cdots$C(20) = 3·618, $Br^-\cdots$C(14) = 3·643, $Br^-\cdots$C(14′) = 3·684, $Br^-\cdots$C(19) = 3·689, $Br^-\cdots$C(33) = 3·721, $Br^-\cdots$C(21) = 3·800, $Br^-\cdots$C(23) = 3·937, $Br^-\cdots$C(12) = 3·989 Å]. Of these, the contacts with C(14) and C(14′)(1−*x*, 1−*y*, −*z*) with H$\cdots Br^-$ distances of 2·75 and 2·76 Å and C—H$\cdots Br^-$ angles of 157 and 177°, indicate possible C—H$\cdots Br^-$ hydrogen bonding. Similar C—H$\cdots Br^-$ hydrogen bonds have been observed in a related structure {[$(C_6H_5)_3PCH(COC_6H_5)_2$]Br.$C_2H_5OH$; Antipin, Kalinin, Struchkov, Aladzheva, Mastryukova & Kabachnik (1980)} and in 2-amino-4-phenylthiazole hydrobromide (Form, Raper & Downie, 1974).

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Structure of the 1:1 Complex of 3-Pyridinecarbonitrile with Pentachlorophenol

By Z. Malarski, I. Majerz and T. Lis

Institute of Chemistry, University of Wrocław, 50383 *Wrocław, Poland*

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Abstract. $C_6HCl_5O.C_6H_4N_2$, $M_r = 370·45$, triclinic, $P\bar{1}$, $a = 7·713$ (5), $b = 8·679$ (5), $c = 13·914$ (9) Å, $\alpha = 117·08$ (5), $\beta = 94·13$ (6), $\gamma = 115·07$ (5)°, $V = 709·5$ (8) Å^3, $Z = 2$, $D_m = 1·72$, $D_x = 1·73$ Mg m^{-3}, μ(Mo $K\alpha$, $\lambda = 0·71069$ Å) = 1·01 mm^{-1}, $F(000) = 368$, $T = 301$ (1) K, final $R = 0·032$ for 1736 non-zero

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reflexions. The phenol OH and the pyridine-ring N atom are engaged in a molecular O—H···N-type hydrogen bond with a length of 2·703 (5) Å. The plane of the pyridine ring is twisted from the plane of the phenol ring by an angle of 56 (1)°.

Introduction. The complexes of phenols with tertiary amines are model compounds for investigations of the dynamic proton transfer equilibrium in the hydrogen bridges $O—H\cdots N \rightleftarrows O^{-}\cdots H—N^{+}$ (Ratajczak & Sobczyk, 1969; Grech, Kalenik, Malarski & Sobczyk, 1983; Zundel & Fritsch, 1984; Dega-Szafran & Dulewicz, 1981; Huyskens & Zeegers-Huyskens, 1964).

The scarce literature data refer to the structures of systems with the hydrogen bond $O^{-}\cdots H—N^{+}$ having a length over 2·70 Å (Talukdar & Chandchuri, 1976; Maartman-Moe, 1969; Van Bellingen, Germain, Piret & Van Meerssche, 1971). Structural information about the crystal and molecular structure of complexes with a weak O—H···N or very strong O···H···N bond with a formal proton transfer about 50% are not available, however.

We have decided to undertake systematic crystal-structure investigations of the complexes of phenols with aliphatic and pyridine amines, with the aim of finding a correlation between the structural data and spectroscopic (IR, UV) behaviour in these systems in order to explain the evolution of the potential-energy curve for the proton motion in OHN hydrogen bridges.

Previously, we have determined crystal and molecular structures of two complexes in the proximity of the 'critical' inversion region ($\Delta pK_a = pK_{a\ \rm base} - pK_{a\ \rm acid} \simeq 1\cdot2$) for the solid state, namely the complexes of pentachlorophenol with 4-methylpyridine ($\Delta pK_a = 0\cdot87$) (Malarski, Majerz & Lis, 1987) and with *N*-methylmorpholine ($\Delta pK_a = 2\cdot12$) (Majerz, Malarski & Lis, 1987). The hydrogen-bridge lengths for these adducts are 2·552 (4), and 2·564 (4) Å respectively.

We have now studied the complex of 3-pyridine carbonitrile with pentachlorophenol ($\Delta pK_a = -3\cdot91$), which is a typical example of a weak O—H···N hydrogen bond (Grech, Kalenik & Sobczyk, 1979; Malarski, Rospenk, Sobczyk & Grech, 1982).

Table 1. *Final atomic parameters with e.s.d.'s in parentheses*

For non-H atoms, $B_{\rm eq} = \frac{1}{3}(B_{11} + B_{22} + B_{33})$.

	x	y	z	$B_{\rm eq}/B_{\rm iso}$(Å^2)
O	0·1642 (5)	0·2234 (5)	0·2994 (3)	4·6 (3)
N(1)	0·0733 (6)	−0·1165 (6)	0·1027 (3)	4·8 (4)
N(2)	−0·5120 (7)	−0·7701 (7)	−0·0825 (4)	5·9 (4)
Cl(2)	0·2195 (2)	0·6141 (2)	0·4361 (1)	4·39 (8)
Cl(3)	0·3454 (2)	0·7909 (2)	0·6974 (1)	4·33 (8)
Cl(4)	0·3919 (2)	0·5409 (2)	0·7888 (1)	4·62 (9)
Cl(5)	0·3119 (2)	0·1116 (2)	0·6180 (1)	4·92 (9)
Cl(6)	0·1876 (2)	−0·0626 (2)	0·3571 (1)	4·68 (8)
C(1)	0·2130 (5)	0·2859 (6)	0·4099 (3)	3·1 (3)
C(2)	0·2464 (5)	0·4761 (6)	0·4875 (3)	3·0 (3)
C(3)	0·3025 (5)	0·5547 (5)	0·6035 (3)	3·1 (3)
C(4)	0·3218 (5)	0·4424 (6)	0·6442 (3)	3·0 (3)
C(5)	0·2875 (5)	0·2521 (6)	0·5683 (3)	3·4 (3)
C(6)	0·2332 (6)	0·1755 (5)	0·4521 (3)	3·2 (3)
C(7)	0·1416 (8)	−0·1023 (8)	0·0217 (5)	5·6 (5)
C(8)	0·0496 (9)	−0·2504 (10)	−0·0923 (5)	7·3 (6)
C(9)	−0·1230 (9)	−0·4278 (9)	−0·1242 (4)	5·7 (5)
C(10)	−0·1947 (6)	−0·4481 (6)	−0·0409 (4)	4·2 (4)
C(11)	−0·0926 (7)	−0·2873 (7)	0·0713 (4)	4·5 (4)
C(12)	−0·3745 (8)	−0·6300 (8)	−0·0657 (4)	4·7 (4)
H(1)	0·152 (7)	0·132 (6)	0·263 (4)	4·2 (13)
H(7)	0·262 (7)	0·037 (7)	0·043 (4)	7·1 (13)
H(8)	0·100 (8)	−0·231 (8)	−0·149 (5)	8·2 (14)
H(9)	−0·202 (7)	−0·543 (7)	−0·198 (4)	6·3 (12)
H(11)	−0·141 (6)	−0·305 (7)	−0·130 (4)	5·9 (11)

Experimental. Single crystals of the title compound were obtained by slow evaporation from CH_3CN solution of 3-pyridinecarbonitrile and pentachlorophenol in equimolar ratio. The compound crystallized mainly as colourless plates. D_m measured by flotation in 1,2-dibromoethane/CCl_4. Preliminary examination by rotation and Weissenberg photographs indicated the triclinic system. A specimen of dimensions from 0·5 to 0·8 mm was cut from a large crystal and sealed in a capillary. Syntex $P2_1$ diffractometer and Mo $K\alpha$ radiation for lattice parameters (15 reflexions in range $20 < 2\theta < 29°$) and intensity measurements; 2θ–ω scan technique; 1868 reflexions measured below $2\theta = 45°$ (h 0→8, k −9→8, l −14→14); after each group of 50 reflexions two standards measured, variation 3%; most calculations performed on a Nova 1200 computer with locally modified *XTL/XTLE* programs (Syntex, 1976); neutral-atom scattering factors from *International Tables for X-ray Crystallography* (1974); real and imaginary components of anomalous dispersion included for all non-H atoms; structure solved by direct methods; H atoms from difference synthesis; refinement based on F values; Cl, O, N, C anisotropic, H isotropic; 1736 reflexions with $I > 1\cdot96\sigma(I)$, $w = 1/\sigma^2(F)$ gave $R = 0\cdot032$ and $wR = 0\cdot040$; max. Δ/σ 0·01 (the $20\bar{1}$ reflexion suffered from strong extinction and was omitted from the calculations). Residual electron density in final difference Fourier map within −0·45 and 0·24 e Å^{-3}.

The final atomic parameters are given in Table 1.*

Discussion. The bond lengths and angles are given in Table 2. The molecular structure of the complex and atom-numbering scheme are shown in Fig. 1 and a stereoview of the cell packing in Fig. 2.

A single intermolecular hydrogen bond is formed by the phenol OH group with the pyridine N atom. The O—H···N bridge of length 2·703 (5) Å is characterized by a strongly asymmetric proton density distribution.

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* Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 43939 (13 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

The C(1)—O bond length of 1·335 (5) Å is only slightly shortened as result of the hydrogen bonding compared with this bond length for phenol in the gas phase, 1·364 Å (Pedersen, Larsen & Nygaard, 1969). The mean C—O bond length for phenols in the solid state is 1·36 (1) Å (Sakurai, 1962; Perrin & Michel, 1973; Bacon & Jude, 1973; Higashi & Osaki, 1977) because of the very weak hydrogen bond formed $[\nu_s(\mathrm{O{-}H\cdots O}) \simeq 3370\ \mathrm{cm}^{-1}]$. The IR spectrum of the complex of 3-pyridinecarbonitrile with pentachlorophenol is typical for the molecular form of the complex. The bands of pyridinium and phenolate ion absorption and low-frequency protonic absorption are absent, only the doublet of the $\nu_s(\mathrm{O{-}H\cdots N})$ band with the components at 2650 and 2930 cm⁻¹ is observed.

Table 2. *Principal interatomic distances* (Å) *and angles* (°)

O···N(1)	2·703 (5)	O—H(1)	0·67 (5)
H(1)···N(1)	2·08 (5)	O—C(1)	1·335 (5)
C(1)—C(2)	1·391 (6)	C(1)—C(6)	1·383 (6)
C(2)—C(3)	1·380 (5)	C(3)—C(4)	1·381 (6)
C(4)—C(5)	1·384 (6)	C(5)—C(6)	1·385 (5)
C(2)—Cl(2)	1·720 (5)	C(3)—Cl(3)	1·716 (4)
C(4)—Cl(4)	1·720 (3)	C(5)—Cl(5)	1·716 (5)
C(6)—Cl(6)	1·724 (4)		
N(1)—C(7)	1·311 (7)	N(1)—C(11)	1·322 (7)
C(7)—C(8)	1·371 (7)	C(8)—C(9)	1·377 (11)
C(9)—C(10)	1·370 (8)	C(10)—C(11)	1·383 (6)
C(10)—C(12)	1·454 (8)	C(12)—N(2)	1·127 (8)
O—H(1)···N(1)	155 (6)	C(1)—O—H(1)	116 (5)
C(7)—N(1)···H(1)	123 (2)	C(11)—N(1)···H(1)	117 (2)
O—C(1)—C(2)	117·2 (4)	O—C(1)—C(6)	124·7 (4)
C(2)—C(1)—C(6)	118·1 (4)	C(1)—C(2)—C(3)	121·1 (4)
C(1)—C(2)—Cl(2)	118·6 (4)	C(3)—C(2)—Cl(2)	120·3 (4)
C(2)—C(3)—C(4)	120·1 (4)	C(2)—C(3)—Cl(3)	120·1 (4)
C(4)—C(3)—Cl(3)	119·8 (4)	C(3)—C(4)—C(5)	119·7 (4)
C(3)—C(4)—Cl(4)	120·0 (4)	C(5)—C(4)—Cl(4)	120·4 (4)
C(4)—C(5)—C(6)	119·8 (4)	C(4)—C(5)—Cl(5)	120·1 (4)
C(6)—C(5)—Cl(5)	120·2 (4)	C(1)—C(6)—C(5)	121·3 (4)
C(1)—C(6)—Cl(6)	118·7 (4)	C(5)—C(6)—Cl(6)	120·0 (4)
C(7)—N(1)—C(11)	117·1 (5)	N(1)—C(7)—C(8)	124·2 (6)
C(7)—C(8)—C(9)	118·3 (7)	C(8)—C(9)—C(10)	118·6 (6)
C(9)—C(10)—C(11)	118·3 (5)	C(10)—C(11)—N(1)	123·4 (5)
C(9)—C(10)—C(12)	122·8 (5)	C(11)—C(10)—C(12)	118·9 (5)
C(10)—C(12)—N(2)	178·4 (6)		

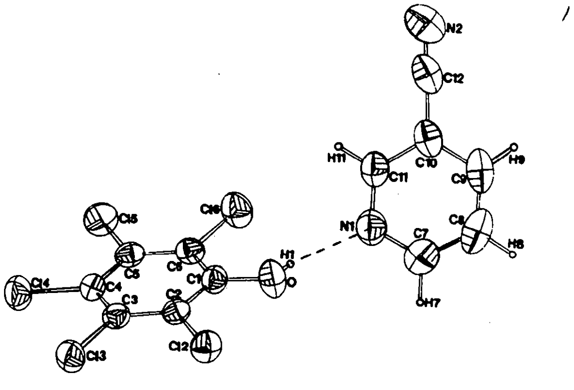

Fig. 1. The molecular structure and numbering scheme of the 1:1 complex of 3-pyridinecarbonitrile with pentachlorophenol. (Thermal ellipsoids for the non-H atoms have been scaled to include 50% probability.)

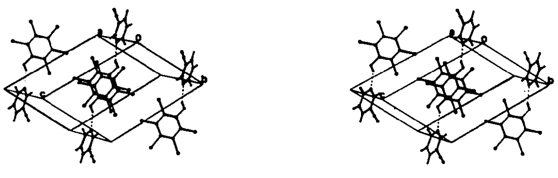

Fig. 2. A stereoview of the molecular packing in the unit cell.

Table 3. *Atom–plane distances* (Å) *and angle between normals to the planes* (°)

Plane *A*					
	N(1)	C(7)	C(8)	C(9)	C(10)
	0·003 (5)	−0·012 (7)	0·006 (8)	0·008 (8)	−0·007 (6)
	C(11)	C(12)	N(2)		
	0·005 (6)	−0·020 (7)*	−0·046 (6)*		
Plane *B*					
	C(1)	C(2)	C(3)	C(4)	C(5)
	−0·005 (5)	0·007 (5)	−0·005 (5)	0·001 (5)	0·001 (5)
	C(6)	O	Cl(2)	Cl(3)	Cl(4)
	0·001 (5)	−0·025 (5)*	0·017 (2)*	−0·008 (2)*	−0·016 (2)*
	Cl(5)	Cl(6)	H(1)		
	0·003 (2)*	0·018 (2)*	−0·05 (6)*		
A—*B*	56·0 (8)				

* Atoms not included in the definition of the planes.

The O and N bridge atoms are slightly bent out of the phenol-ring plane by −0·025 (5) and −0·021 (5) Å, respectively. The deviations of the Cl atoms from the ring plane range from −0·016 (1) to 0·018 (1) Å (Table 3). The length of the C—Cl bonds ranges from 1·720 (3) to 1·730 (3) Å and agrees within 0·01 Å with analogous bonds in other complexes of pentachlorophenol (Malarski *et al.*, 1987; Majerz *et al.*, 1987).

The average C—C bond length in the phenol ring is 1·384 (6) Å and these bonds are more equal than in the strongest complexes of pentachlorophenol (Malarski *et al.*, 1987; Majerz *et al.*, 1987). Similarly, all bond lengths in the 3-pyridinecarbonitrile molecule (Table 2) are in agreement with the data for 4-pyridinecarbonitrile (Laing, Sparrow & Sommerville, 1971). The C and N atoms of the cyano group deviate significantly from the plane of the pyridine ring (Table 3).

The pyridine and phenol rings are planar with an angle of 56 (1)° between the planes.

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Crystal Structure Determination by Neutron Diffraction of 2,2-Diphenyl-1-picrylhydrazyl (DPPH) Benzene Solvate (1/1)

BY J. X. BOUCHERLE

DRF/MDN, CENG, BP 85*X*, 38041 *Grenoble CEDEX, France*

B. GILLON*

ILL, 156*X*, 38042 *Grenoble CEDEX, France, and CMOA*, 23 *rue du Maroc*, 75019 *Paris, France*

J. MARUANI†

CMOA, 23 *rue du Maroc*, 75019 *Paris, France*

AND J. SCHWEIZER

DRF/MDN, CENG, BP 85*X*, 38041 *Grenoble CEDEX, France, and ILL*, 156*X*, 38042 *Grenoble CEDEX, France*

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Abstract. The crystal structure of 2,2-diphenyl-1-picrylhydrazyl (DPPH), syncrystallized with one benzene molecule per DPPH molecule (DPPH–C_6H_6), already determined at room temperature with X-rays [Williams (1967). *J. Am. Chem. Soc.* **89**, 4280–4287], was remeasured at low temperature (10 K) by neutron diffraction. Crystal data: $C_{18}H_{12}N_5O_6.C_6H_6$, $M_r = 472{\cdot}45$, monoclinic, Pc, $a = 7{\cdot}583\,(10)$, $b = 10{\cdot}593\,(15)$, $c = 14{\cdot}526\,(20)$ Å, $\beta = 108{\cdot}47\,(6)°$, $V = 1104{\cdot}57\,(2{\cdot}65)$ Å^3, $Z = 2$, $D_x = 1{\cdot}420$ g cm^{-3}, $\lambda = 1{\cdot}176$ Å, $R = 0{\cdot}032$ for 1264 unique observed reflections. The positions of the H atoms have been determined as well as the values of the temperature factors at 10 K. The shape of the molecule does not change significantly on cooling from room to low temperature.

Introduction. The stable aromatic free radical 2,2-diphenyl-1-picrylhydrazyl (DPPH) was one of the first paramagnetic species investigated by electron spin resonance. Its chemical stability and the narrowness of its resonance have led to the widespread use of its powder form as a standard in measuring unpaired spin concentrations and effective gyromagnetic ratios (Buchachenko, 1965). In addition, the hyperfine structure of the DPPH radical has been one of the most thoroughly investigated using magnetic resonance techniques (Maruani, 1980). Consequently DPPH appears as a very good candidate for a complete spin-density determination using polarized neutron diffraction, in order to compare the local values of the spin density given by the resonance techniques with the spin-density distribution, measured at each point of the molecule, by neutron diffraction (Boucherle, Gillon, Maruani & Schweizer, 1982).

DPPH is known in several forms (Weil & Anderson, 1965). Some of them are solvent free: DPPH-I (m.p. 379 K) is orthorhombic (Williams, 1965),

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[*] Present address: LLB, CENS, 91191 Gif sur Yvette CEDEX, France.

[†] Present address: LCP, 11 rue Pierre et Marie Curie, 75005 Paris, France.

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