HAMILTON, W. C. (1965). *Acta Cryst.* **18**, 502–510.
International Tables for X-ray Crystallography (1974). Vol. IV. Birmingham: Kynoch Press.
KURAHASHI, M. (1976). *Bull. Chem. Soc. Jpn*, **49**, 2927–2933.
MAIN, P., FISKE, S. J., HULL, S. E., LESSINGER, L., GERMAIN, G., DECLERCQ, J. P. & WOOLFSON, M. M. (1980). *MULTAN80. A System of Computer Programs for the Automatic Solution of Crystal Structures from X-ray Diffraction Data.* Univs. of York, England, and Louvain-la-Neuve, Belgium.
MOTHERWELL, W. D. S. (1974). *EENY.* Potential-energy calculation program version of June 1974. Univ. Chemical Laboratory, Lensfield Road, Cambridge, England.
MOTHERWELL, W. D. S. (1978). *PLUTO78.* A program for plotting molecular and crystal structures. Univ. of Cambridge, England.
NORTH, A. C. T., PHILLIPS, D. C. & MATHEWS, F. (1968). *Acta Cryst.* A**24**, 351–359.
STEWART, J. M., MACHIN, P. A., DICKINSON, C. W., AMMON, H. L., HECK, H. & FLACK, H. (1976). The *XRAY76* system. Tech. Rep. TR-446. Computer Science Center, Univ. of Maryland, College Park, Maryland.
STEWART, R. F., DAVIDSON, E. R. & SIMPSON, W. T. (1965). *J. Chem. Phys.* **42**, 3175–3187.

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Structure of the (1/1) Complex *cis*-3-(2,5-Dihydroxyphenyl)-5-hydroxy-3-methyl-2-(*N*-methylpropionamido)-2,3-dihydrobenzo[*b*]furan–($^2H_6$)Acetone, $C_{19}H_{21}NO_5.C_3D_6O$*

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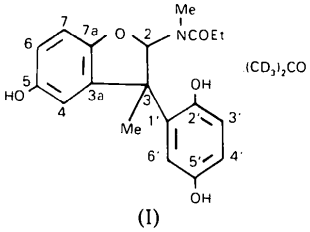
Abstract. $M_r = 407{\cdot}49$, triclinic, $P\bar{1}$, $a = 7{\cdot}263$ (3), $b = 15{\cdot}272$ (4), $c = 10{\cdot}051$ (3) Å, $\alpha = 103{\cdot}90$ (1), $\beta = 81{\cdot}23$ (1), $\gamma = 88{\cdot}53$ (1)°, $V = 1067{\cdot}3$ (6) Å^3, $Z = 2$, $D_x = 1{\cdot}268$ (1) g cm^{-3}, $\lambda(\mathrm{Cu}\,K\alpha) = 1{\cdot}54178$ Å, $\mu = 7{\cdot}11$ cm^{-1}, $F(000) = 428$, $T = 293$ K, final $R = 0{\cdot}045$ for 3039 independent observed reflections. The *N*-methylpropionamido and hydroquinone substituents situated at adjacent C atoms of the dihydrofuryl ring are almost eclipsed. The system of intermolecular hydrogen bonds in the crystal lattice consists of three types, two of which bond pairs of enantiomeric molecules into aggregates. The third type involves the acetone molecule. The aggregates form chains parallel to [011] in the crystal.

Introduction. Several dihydrobenzofuran as well as hydroquinone derivatives (MacNicol, McKendrick & Wilson, 1978; Helgeson, Lauer & Cram, 1983) are reported to have complexing properties for some organic molecules in the solid state. Similar properties were found recently (Kozerski, 1981) for some derivatives of 3-(2,5-dihydroxyphenyl)benzo[*b*]furan. They are chemically close to the dianine derivatives which in the solid state form hexahost cavities able to trap one or two simple guest molecules (MacNicol *et al.*, 1978). The title compound (I) is one of a series for which the complexing ratio of 1:1 was postulated on the basis of ^{1}H NMR spectra (Kozerski, 1981). The present X-ray structural investigation has been performed in order to elucidate the complexing mode of the solvent molecules which seems to be different from that of the dianine derivatives. A verification of NMR conclusions concerning the configuration of amide residues is also of interest.



(I)

Experimental. Crystal (0·2 × 0·2 × 0·3 mm) obtained from $(CD_3)_2CO$ solution from a ^{1}H NMR measurement. D_m not determined. Siemens AED automatic diffractometer, Cu $K\alpha$ radiation, $\omega/2\theta$ scan, $2\theta_{\max} = 108°$, $hkl_{\max} = 8$, 18, 12. Stability controlled with one reflection (430) at 50 reflection intervals (±4% intensity variation). 4018 unique reflections collected, 3049 with $I > 2\sigma_I$ regarded as observed. Cell constants refined against 28 reflections. Lorentz–polarization correction, no absorption correction. Space group $P\bar{1}$. Structure solved by direct methods (*SHELX76*, Sheldrick, 1976) followed by ρ maps. H and D atoms found from $\Delta\rho$ maps. Structure refined (F_{hkl}) by the full-matrix least-squares procedure (*XRAY70*, Stewart, Kundell &

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* Molecular Structures and Inclusion Properties of Some Benzofuran Derivatives. III. Part II: Urbańczyk-Lipkowska, Krajewski, Gluziński, Kozerski & Czugler (1982).

Table 1. *Fractional atomic coordinates* ($\times 10^4$) *and equivalent isotropic thermal parameters with e.s.d.'s in parentheses*

$B_{eq} = 8\pi^2(U_1U_2U_3)^{1/3}$, where U_i are the eigenvalues of the U_{ij} matrix.

	x	y	z	B_{eq}(Å^2)
C(1)	11008 (3)	7166 (2)	1209 (2)	3·0 (1)
C(2)	9719 (4)	7560 (2)	607 (2)	3·3 (1)
C(3)	8477 (4)	8261 (2)	1377 (3)	3·4 (1)
C(4)	8558 (3)	8566 (1)	2779 (2)	3·1 (1)
C(5)	9879 (3)	8193 (1)	3383 (2)	2·9 (1)
C(6)	11127 (3)	7489 (1)	2617 (2)	2·8 (1)
O(7)	12197 (2)	6457 (1)	440 (2)	3·7 (1)
O(8)	7286 (3)	9257 (1)	3540 (2)	4·1 (1)
C(9)	12620 (3)	7091 (2)	3276 (2)	3·0 (1)
C(10)	14582 (4)	7230 (2)	2536 (3)	4·0 (1)
C(11)	12603 (3)	7478 (2)	4815 (2)	2·9 (1)
C(12)	12751 (3)	8358 (2)	5555 (2)	3·1 (1)
C(13)	12893 (3)	8510 (2)	6958 (2)	3·4 (1)
C(14)	12896 (4)	7798 (2)	7595 (2)	3·7 (1)
C(15)	12802 (4)	6920 (2)	6854 (3)	3·8 (1)
C(16)	12677 (3)	6784 (2)	5458 (3)	3·3 (1)
O(17)	12685 (3)	5940 (1)	4582 (2)	4·0 (1)
C(18)	12442 (3)	6060 (2)	3228 (3)	3·4 (1)
N(19)	10708 (3)	5679 (1)	2909 (2)	3·1 (1)
C(20)	9005 (4)	6024 (2)	3879 (3)	4·0 (1)
C(21)	10608 (4)	4954 (1)	1853 (2)	3·2 (1)
O(22)	9073 (2)	4673 (1)	1642 (2)	3·9 (1)
C(23)	12372 (4)	4456 (2)	988 (3)	3·9 (1)
C(24)	13211 (5)	3827 (2)	1754 (3)	5·3 (1)
O(25)	13050 (3)	9360 (1)	7761 (2)	4·3 (1)
C1(Ac)	7562 (4)	9567 (2)	7334 (3)	5·0 (2)
C2(Ac)	7663 (6)	8612 (3)	7366 (5)	7·6 (1)
C3(Ac)	7401 (8)	10279 (4)	8629 (4)	8·1 (5)
O(Ac)	7551 (4)	9793 (2)	6265 (2)	6·0 (1)

Table 2. *Bond lengths* (Å) *and angles* (°) *with e.s.d.'s in parentheses*

C(1)–C(2)	1·387 (4)	C(13)–C(14)	1·389 (4)
C(1)–C(6)	1·399 (3)	C(13)–O(25)	1·371 (3)
C(1)–O(7)	1·365 (3)	C(14)–C(15)	1·378 (4)
C(2)–C(3)	1·378 (3)	C(15)–C(16)	1·386 (4)
C(3)–C(4)	1·384 (3)	C(16)–O(17)	1·374 (3)
C(4)–C(5)	1·388 (4)	O(17)–C(18)	1·451 (3)
C(4)–O(8)	1·378 (3)	C(18)–N(19)	1·449 (3)
C(5)–C(6)	1·383 (3)	N(19)–C(20)	1·453 (3)
C(6)–C(9)	1·537 (4)	N(19)–C(21)	1·349 (3)
C(9)–C(10)	1·549 (3)	C(21)–O(22)	1·235 (3)
C(9)–C(11)	1·516 (3)	C(21)–C(23)	1·506 (3)
C(9)–C(18)	1·573 (3)	C(23)–C(24)	1·534 (5)
C(11)–C(12)	1·386 (3)	C1(Ac)–O(Ac)	1·206 (4)
C(11)–C(16)	1·372 (4)	C1(Ac)–C2(Ac)	1·466 (6)
C(12)–C(13)	1·394 (4)	C1(Ac)–C3(Ac)	1·468 (5)
C(1)–C(2)–C(3)	121·4 (2)	C(11)–C(12)–C(13)	118·2 (2)
C(2)–C(1)–C(6)	120·8 (2)	C(12)–C(11)–C(16)	119·7 (2)
C(2)–C(1)–O(7)	120·8 (2)	C(11)–C(16)–C(15)	123·0 (2)
C(1)–C(6)–C(5)	117·4 (2)	C(11)–C(16)–O(17)	113·9 (2)
C(1)–C(6)–C(9)	121·2 (2)	C(12)–C(13)–C(14)	120·8 (2)
C(6)–C(1)–O(7)	118·4 (2)	C(12)–C(13)–O(25)	121·8 (2)
C(2)–C(3)–C(4)	117·9 (3)	C(13)–C(14)–C(15)	121·1 (2)
C(3)–C(4)–C(5)	121·1 (2)	C(14)–C(13)–O(25)	117·4 (2)
C(3)–C(4)–O(8)	117·2 (2)	C(14)–C(15)–C(16)	117·1 (2)
C(4)–C(5)–C(6)	121·3 (2)	C(15)–C(16)–O(17)	123·0 (2)
C(5)–C(4)–O(8)	121·7 (2)	C(16)–O(17)–C(18)	107·8 (2)
C(5)–C(6)–C(9)	121·4 (2)	O(17)–C(18)–N(19)	107·8 (2)
C(6)–C(9)–C(10)	109·3 (3)	C(18)–N(19)–C(20)	118·1 (2)
C(6)–C(9)–C(11)	116·9 (2)	C(18)–N(19)–C(21)	123·8 (2)
C(6)–C(9)–C(18)	113·6 (2)	C(20)–N(19)–C(21)	117·7 (2)
C(10)–C(9)–C(11)	106·2 (2)	N(19)–C(21)–O(22)	119·9 (2)
C(10)–C(9)–C(18)	109·6 (2)	N(19)–C(21)–C(23)	119·9 (2)
C(11)–C(9)–C(18)	100·8 (2)	O(22)–C(21)–C(23)	120·1 (2)
C(9)–C(11)–C(12)	130·5 (2)	C(21)–C(23)–C(24)	110·5 (2)
C(9)–C(11)–C(16)	109·2 (2)	C2(Ac)–C1(Ac)–O(Ac)	121·6 (3)
C(9)–C(18)–O(17)	107·2 (2)	C2(Ac)–C1(Ac)–C3(Ac)	120·3 (4)
C(9)–C(18)–N(19)	117·0 (2)	C3(Ac)–C1(Ac)–O(Ac)	118·0 (4)

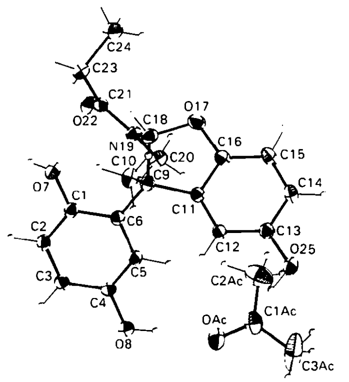

Fig. 1. *ORTEP* (Johnson, 1965) diagram of the title compound with the crystallographic labelling of the atoms. Projection on the *bc* plane. Thermal-motion ellipsoids set out at 40% probability level.

Baldwin, 1970) with anisotropic temperature factors for non-H atoms [isotropic temperature factors for H(D) invariant, set as B_{eq} of the adjacent atom + 1 Å^2]. Scattering factors from *International Tables for X-ray Crystallography* (1974). 10 reflections deleted in the last refinement step. Final $R = 0{\cdot}045$ (unit weights), $S = 1{\cdot}5844$, average $\Delta/\sigma = 0{\cdot}053$. Max. electron density amplitude on the $\Delta\rho$ maps $< 0{\cdot}6\ \text{e Å}^{-3}$. Computations performed on CYBER-76 and CYBER-73 computers.

^{13}C NMR chemical shifts for N substituents determined at frequency 50·3 MHz (Varian XL-100) in ($^2H_4$)methanol against internal TMS under conditions of slow rotation around the amide bond for observation of sharp lines in both rotamers (acquisition time 1·5 s, pulse angle 45°, digital resolution 0·7 Hz). ^1H NMR spectrum taken at 100 MHz (JEOL-JNM-4H) from a solution in ($^2H_8$)dioxane.

Discussion. The refined atomic coordinates and the B_{eq} values are presented in Table 1.* Table 2 gives the bond lengths and valence angles. Fig. 1 shows the *ORTEP* (Johnson, 1965) diagram for the title molecule with the crystallographic labelling of the atoms.

The bond lengths and angles in the benzofuran and hydroquinone rings are not unusual. As in other benzofuran derivatives (Ponnuswamy & Parthasarathy,

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* Lists of structure factors, anisotropic thermal parameters, refined H- and D-atom coordinates and some NMR data have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 39535 (31 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

1981; Urbańczyk-Lipkowska, Krajewski, Gluziński, Kozerski & Czugler, 1982; Urbańczyk-Lipkowska, Krajewski, Gluziński, Andreetti & Bocelli, 1982) the C(9)—C(18) bond is somewhat elongated. The torsion angle N(19)—C(18)—C(9)—C(6) is −14·7 (3)°; this causes many short intramolecular non-bonding distances in the environment of C(9) [*e.g.* C(6)···N(19) 2·868 (3) Å]. Thus, the space around C(9) may be regarded as particularly overcrowded which may result in elongation of some bond lengths involving C(9).

The amide bond in the title compound has the *trans* conformation, similar to that found in the di-*O*-acetylated derivative (Urbańczyk-Lipkowska, Krajewski, Gluziński, Kozerski & Czugler, 1982). However, in the case of the tri-*O*-acetylated derivative (Urbańczyk-Lipkowska, Krajewski, Gluziński, Andreetti & Bocelli, 1982) the *syn* conformation for the amide bond was observed. The literature data for some NMR investigations (Stewart & Siddall, 1970; La Planche & Rogers, 1963) lead to the conclusion that in unsymmetrical tertiary amides the predominant conformation along the amide bond is that in which a larger substituent at N is situated in the position *cis* to the carbonyl group. The present ^1H and ^{13}C NMR studies on the title compound in solution indicate that the *trans* conformation prevails (60%). This conclusion is based on the signals from both rotamers, observed in a condition of hindered rotation around the amide bond, which is slow on the ^1H and ^{13}C NMR time scale (data deposited). On the other hand it is unlikely that the observed anisochrony of the chemical shifts in ^1H and ^{13}C NMR were induced by other conformational changes in the molecule such as hindered rotation around the C(18)—N(19) or C(9)—C(6) bonds since these changes usually have no effect on NMR spectra at ambient temperatures. The proximity of the phenyl ring and the carbonyl moiety in the *cis* conformation along the C(9)—C(18) bond prevents such a rotation. It is generally acknowledged, although not documented quantitatively, that the anisochrony of the NMR signals of the substituents on N originates from the magnetic anisotropy of the carbonyl group and the electric field effect borne out in each site by an electric dipole (Bothner-By & Pople, 1965; ApSimon, Demarco, Mathieson, Craig, Karim, Saunders & Whalley, 1970; Piccini-Leopardi & Reisse, 1981). These studies indicate that in ^1H resonance the substituents on N situated *cis* with respect to the O atom usually give resonances at lower frequencies than the *trans* ones. This was confirmed also by nuclear Overhauser enhancement (NOE) studies (Anet & Bourn, 1965; Lewin & Frucht, 1975). The same conclusion with respect to the direction of the anisochrony is derived from ^{13}C NMR studies on simple amides (Piccini-Leopardi & Reisse, 1981; McFarlane, 1970; Levy & Nelson, 1972; Dorman & Bovey, 1973*a*) and amino acids (Dorman & Bovey, 1973*b*; Voelter & Oster, 1973). According to this general consensus the prevailing (60%) *trans* rotamer in solution was assigned based on the fact that the C(9) and C(18) atoms give high-frequency shifts in this form while the *N*-methyl group absorbs at lower frequencies as shown by the present ^1H and ^{13}C NMR results.

The arrangement of the hydroquinone hydroxyl H atoms resembles that of the *β* modification of hydroquinone (Lindeman, Shklover & Struchkov, 1981). The hydroxyl H atoms are diverted in opposite directions (Fig. 1) and twisted away from the hydroquinone plane. However, the twisting is not symmetrical with respect to the hydroquinone ring centre. Both H atoms [H(O7) and H(O8)] are positioned on the same side of the hydroquinone plane at unequal distances from it [0·5 (1) and 0·1 (1) Å, respectively].

The H(O25) hydroxyl H atom is found to be coplanar with the aromatic ring of the dihydrobenzofuran moiety.

All hydroxyl H atoms of the title molecule are involved in relatively strong intermolecular hydrogen bonds (Fig. 2). Three types of hydrogen bonds can be identified in the crystal structure [bond, acceptor symmetry, donor···acceptor distance (Å), H···acceptor distance (Å), angle at H (°): O(7)—H(O7)···O(22), 2−*x*, 1−*y*, −*z*, 2·672 (2), 1·79 (3), 168 (3); O(8)—H(O8)···O(Ac), *x*, *y*, *z*, 2·702 (3), 1·82 (3), 175 (3); O(25)—H(O25)···O(8), 2−*x*, 2−*y*, 1−*z*, 2·763 (3), 1·83 (3), 173 (3)]. The molecules of the title compound associated with acetone form chains in the crystal parallel to [011] with no H bonds between them. The adjacent molecules in a chain are mutually bonded by pairs of H bonds centred at ($0\frac{1}{2}0$) and at ($00\frac{1}{2}$).

The structure of the title compound seems to be approximately isomorphous to that of the uncomplexed diacetylated derivative (Urbańczyk-Lipkowska, Krajewski, Gluziński, Kozerski & Czugler, 1982). The space volume occupied by the disordered acetyl group in the latter is comparable with that accommodating the acetone molecule in the present structure.

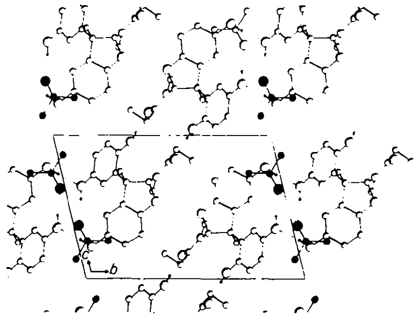

Fig. 2. A projection on the *bc* plane of the crystal showing the intermolecular H-bond system. C-attached H atoms omitted for clarity. Atoms of acetone molecules shaded for distinction.

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References

Anet, F. A. L. & Bourn, A. J. R. (1965). *J. Am. Chem. Soc.* **87**, 5250–5251.

ApSimon, J. W., Demarco, P. V., Mathieson, D. W., Craig, W. G., Karim, A., Saunders, L. & Whalley, W. B. (1970). *Tetrahedron*, **26**, 119–146.

Bothner-By, A. A. & Pople, J. A. (1965). *Annu. Rev. Phys. Chem.* **16**, 43–64.

Dorman, D. E. & Bovey, F. A. (1973*a*). *J. Org. Chem.* **38**, 1719–1722.

Dorman, D. E. & Bovey, F. A. (1973*b*). *J. Org. Chem.* **38**, 2379–2383.

Helgeson, R. C., Lauer, M. & Cram, D. J. (1983). *J. Chem. Soc. Chem. Commun.* pp. 101–103.

International Tables for X-ray Crystallography (1974). Vol. IV. Birmingham: Kynoch Press.

Johnson, C. K. (1965). *ORTEP*. Report ORNL-3794. Oak Ridge National Laboratory, Tennessee.

Kozerski, L. (1981). *J. Mol. Struct.* **75**, 95–99.

La Planche, L. A. & Rogers, M. T. (1963). *J. Am. Chem. Soc.* **85**, 3728–3730.

Levy, G. C. & Nelson, G. L. (1972). *J. Am. Chem. Soc.* **94**, 4897–4901.

Lewin, A. H. & Frucht, M. (1975). *Org. Magn. Reson.* **7**, 206–225.

Lindeman, S. V., Shklover, V. E. & Struchkov, Yu. T. (1981). *Cryst. Struct. Commun.* **10**, 1173–1179.

McFarlane, W. (1970). *J. Chem. Soc. Chem. Commun.* pp. 418–419.

MacNicol, D. D., McKendrick, J. J. & Wilson, D. R. (1978). *Chem. Soc. Rev.* **7**, 65–87.

Piccini-Leopardi, C. & Reisse, J. (1981). *J. Magn. Reson.* **42**, 60–72.

Ponnuswamy, M. N. & Parthasarathy, S. (1981). *Cryst. Struct. Commun.* **10**, 1203–1209.

Sheldrick, G. M. (1976). *SHELX76*. Program for crystal structure determination. Univ. of Cambridge, England.

Stewart, J. M., Kundell, F. A. & Baldwin, J. C. (1970). The *XRAY70* system. Computer Science Center, Univ. of Maryland, College Park, Maryland.

Stewart, W. E. & Siddall, T. H. III (1970). *Chem. Rev.* **70**, 517–551.

Urbańczyk-Lipkowska, Z., Krajewski, J. W., Gluziński, P., Andreetti, G. D. & Bocelli, G. (1982). *Cryst. Struct. Commun.* **11**, 1537–1542.

Urbańczyk-Lipkowska, Z., Krajewski, J. W., Gluziński, P., Kozerski, L. & Czugler, M. (1982). *Cryst. Struct. Commun.* **11**, 1531–1536.

Voelter, W. & Oster, O. (1973). *Org. Magn. Reson.* **5**, 547–548.

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10-(2-Methoxyphenyl)pyrido[3,2-*b*][1,4]benzothiazine, $C_{18}H_{14}N_2OS$

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Abstract. $M_r = 306{\cdot}38$, monoclinic, $P2_1/c$, $a = 11{\cdot}859\,(1)$, $b = 8{\cdot}121\,(1)$, $c = 16{\cdot}016\,(1)$ Å, $\beta = 101{\cdot}59\,(1)°$, $V = 1511{\cdot}0\,(3)$ Å^3, $Z = 4$, $D_x = 1{\cdot}347$ g cm^{-3}, λ(Cu $K\alpha$) = 1·5418 Å, $\mu = 18{\cdot}07$ cm^{-1}, $F(000) = 640$, $T = 295$ K. Final $R = 0{\cdot}050$ for 1708 observed reflections. The central ring of the tricyclic ring system is in a boat conformation. The folding angle between the planes of the pyrido and benzo rings is 158·7 (1)°. The plane of the 10-phenyl ring, almost perpendicular to the plane of the central ring [dihedral angle 95·4 (1)°], nearly bisects the tricyclic ring system.

Introduction. During the continuous search for biologically active phenothiazines, a series of substituted phenothiazines (Chiou, Reeves & Biehl, 1976; Biehl, Patrizi, Lapis & Reeves, 1974), heteroarylphenothiazines (Jovanovic & Biehl, 1983; Jovanovic, Biehl, Rosenstein & Chu, 1984), and their pyridobenzothiazine analogs (Jovanovic & Biehl, 1983) have been prepared. One of the interesting findings of these studies was the significant electronic interactions between the phenothiazine tricyclic ring and electron-withdrawing 10-aryl substituents. The examination of the interaction in the solid state by X-ray analysis can be used to compare the study of the same effects in solution by NMR spectroscopy (Jovanovic & Biehl, 1983, 1984).

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