

Structure of α -Acetyl- β -(3-chloroanilino)-*N*-methylacrylamide

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(Received 14 June 1989; accepted 18 May 1990)

Abstract. $C_{12}H_{13}ClN_2O_2$, $M_r = 252.70$, triclinic, $P\bar{1}$, $a = 9.193$ (1), $b = 9.243$ (2), $c = 9.706$ (2) Å, $\alpha = 123.15$ (2), $\beta = 115.19$ (2), $\gamma = 66.67$ (2)°, $V = 610.15$ Å³, $Z = 2$, $D_m = 1.398$, $D_x = 1.377$ Mg m⁻³, $\lambda(\text{Mo } K\alpha) = 0.71069$ Å, $\mu = 0.257$ mm⁻¹, $F(000) = 264$, room temperature, $R = 0.0395$, $wR = 0.0318$ for 994 observed reflections. Molecules in the crystal are discrete but contain intramolecular N—H...O bonds whose dimensions are normal for this type of compound [N(8)...O(14) = 2.583 (6), H(N8)...O(14) = 1.86, N(15)...O(13) = 2.665 (6), H(N15)...O(13) = 2.01 Å, N(8)—H(N8)...O(14) = 137, N(15)—H(N15)...O(13) = 146°]. One bond in the acetoacetamide group [C(12)—N(15), length 1.319 (6) Å] has partial double-bond character, suggesting some electron delocalization. A similar effect has also been observed in 2-(4-chlorophenylhydrazono)-*N*-methylacetoacetamide [Mehdi, Sadanandam, Shetty & Rama Rao (1986). *Acta Cryst.* C42, 347–349].

Introduction. The title compound exhibits broad spectrum fungicidal activity against *Penicillium tardum*, *Helminthosporium halodes* and *Fusarium oxysporicum*, and the X-ray investigation was undertaken to study the conformation of the molecule.

Experimental. Pale-yellow crystals, obtained on recrystallization from ethanol, were prepared by mixing *N*-methylacetoacetamide with ethyl formate and 3-chloroaniline and heating under reflux in an oil bath. A suitable crystal of dimensions 0.38 × 0.33 × 0.15 mm was mounted on an Enraf-Nonius CAD-4 automatic diffractometer. D_m by flotation in zinc bromide and thallous formate solution. Cell parameters obtained by least-squares refinement from 25 reflections with $14 < 2\theta < 34^\circ$. Intensity data collected using monochromated Mo $K\alpha$ radiation and ω - θ scan technique for $4 < 2\theta < 50^\circ$, two standard reflections ($\bar{6}11$, $5\bar{1}1$) measured after every 80 reflections showed no significant variation in intensity; Lorentz and polarization corrections applied but not absorption. A total of 1996 reflections (hkl ranges 0–10, 0–10, 0–11) were measured. Of these, 1878 were unique and 1189 were classified as observed [$F > 4\sigma(F)$]; $R_{\text{int}} = 0.0117$. Non-H atoms were located

using *SHELXS86* (Sheldrick, 1985) and refined by full-matrix methods, with anisotropic thermal parameters, using the 994 reflections with $F > 5\sigma(F)$ to $R = 0.081$. The phenyl ring was then constrained to be a regular planar hexagon (C—C = 1.395 Å), and H atoms riding on corresponding C atoms at 1.08 Å, and all $U(\text{H})$ refined. The constraints were then removed and full-matrix weighted $\{w = [\sigma^2(F) + g(F^2)]^{-1}$, $g = 9.21 \times 10^{-4}$ (refined)} least-squares refinement on F yielded $R = 0.0395$ and $wR = 0.0318$ at convergence [$\Delta/\sigma = 0.033$ (max.) and 0.0037 (mean) over the last three cycles] with largest peak in final difference Fourier synthesis +0.311 and minimum of -0.283 e Å⁻³. Neutral-atom scattering factors from *International Tables for X-ray Crystallography* (1974, Vol. IV) and refinements performed with *SHELX76* (Sheldrick, 1976).

Discussion. The molecular structure, numbering scheme and bond lengths are depicted in Fig. 1. Final atomic coordinates and equivalent isotropic thermal parameters for all non-H atoms are given in Table 1, while valence angles and hydrogen-bond data are listed in Table 2.* The structure consists of independent molecules which are almost planar. Except for the C(12)—N(15) bond [1.319 (6) Å], all other molecular and intramolecular bonds are normal for this type of compound. A comparison with the bonds in 2-(4-chlorophenylhydrazono)-*N*-methylacetoacetamide (Mehdi, Sadanandam, Shetty & Rama Rao, 1986) is shown in Table 3. The partial double-bond character of C(12)—N(15) suggests electron delocalization and this effect is also observed in the acetoacetamide [(II), Table 3]. The intramolecular distances N(8)...O(14) and N(15)...O(13) here also compare well with the distances N...O(1) and N(3)—O(2) in the acetoacetamide (Table 3). In both cases the molecule tends to remain planar because rotation is inhibited by the hydrogen

* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 52725 (8 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

