1-Acetyl-4-({\(p\)}-chlorobenzylideneamino)-3-methyl-4,5-dihydro-1\(H\)-1,2,4-triazol-5-one

The molecules of the title compound, C\(_{12}\)H\(_{11}\)ClN\(_4\)O\(_2\), are arranged as layers, stacking approximately along the \(b\) axis through C—H⋯O intermolecular hydrogen bonds. Some of these hydrogen-bond interactions which link two centrosymmetrically related molecules generate \(\pi\)–\(\pi\)-stacking interactions between triazole rings.

Comment

The 1,2,4-triazole ring systems are typically planar 6\(\pi\)-electron partially aromatic systems, possessing an extensive chemistry (Temple, 1981; Benson, 1967). 1,2,4-Triazole and its derivatives are starting materials for the synthesis of many heterocycles (Desenko, 1995). In addition to its extensive chemical significance, the 1,2,4-triazole nucleus is also found to be associated with diverse pharmacological properties, such as analgesic, anti-asthmatic, diuretic, anti-inflammatory, fungicidal, bactericidal and pesticidal activities (Mohamed et al., 1993; Sharma & Bahel, 1982; Heubach et al., 1980; Bennur et al., 1976; Webb & Parsons, 1977). Knowledge of the molecular structure of these compounds is important for understanding their reactivity under condensation reaction conditions. Therefore, the crystal structure analysis of the title compound, (I), has been carried out.

The structure of (I) (Fig. 1) consists of one 1,2,4-triazole ring (ring A: N2/C8/N3/N4/C11) with an acetyl group substituted at N3, and a methyl group and O atom substituted at C11 and C8, respectively. It also has a benzene ring (ring B: C1–C6). N=C bond lengths [N4=C11 = 1.278 (3) \(\text{\AA}\) and N1=C7 = 1.269 (3) \(\text{\AA}\)] agree with literature values (Puvirasan et al., 1999; Liu et al., 1999). The triazole ring is planar and the maximum deviation is −0.0026 (3) \(\text{\AA}\) for atom C11. Atom O1 is also located in the plane. The bond lengths and angles in the acetyl group are comparable with reported values (Singh & Izydore, 1996). The dihedral angle between rings A and B is 7.29 (1)\(^\circ\), indicating that the whole molecule is nearly planar.

Atom H12 of the methyl group (C12) forms an intermolecular hydrogen bond with the acetyl group O atom (O2) of a symmetry-related molecule [C12⋯O2\(^\text{ii}\) = 3.41 \(\text{\AA}\); symmetry
**Experimental**

3-Methyl-4-(p-chlorobenzylideneamino)-4,5-dihydro-1H-1,2,4-triazol-5-one (0.01 mol) was treated with 10 ml of acetic anhydride and the mixture was refluxed for 30 min. After addition of 30 ml of absolute ethanol to the solution, the mixture was refluxed for one hour. The resulting product was filtered and dried in vacuo. Several recrystallizations of this product from ethanol gave the pure compound. Yield: 82%, m.p: 454–455 K. IR data (KBr/cm⁻¹): νC=O: 1769, 1697; νC=N: 1623, 1593; νbenzenoid ring: 820. ¹H NMR (δ/ppm. in DMSO-d₆): 2.40 (s, 3H), 2.50 (s, acetyl 3H), 7.36 (d, 2H, Ar—H), 7.60 (d, 2H, Ar—H), 9.36 (s, CH). ¹³C NMR (in DMSO-d₆): 166.24 (acetyl C—O), 155.79 (N—CH), 151.18 (triazole C—O), 148.14, 133.68, 132.95, 132.13 (2 C), 131.80 (2 C), 23.61, 12.18.

**Crystal data**

C₁₃H₁₁ClN₄O₂  
M₀ = 278.70  
Triclinic, P¹  
α = 6.910 (3) Å  
b = 7.5682 (10) Å  
c = 12.380 (3) Å  
α = 93.13 (2)°  
β = 98.20 (2)°  
γ = 95.86 (2)°  
V = 635.9 (3) Å³  
Dx = 1.456 Mg m⁻³  
Mo Kα radiation  
θ = 8.2–13.4°  
µ = 0.30 mm⁻¹  
T = 293 (2) K  
Plate, colorless  
0.25 × 0.15 × 0.10 mm

**Data collection**

Enraf-Nonius CAD-4 MACH3 diffractometer  
ω-2θ scans  
Absorption correction: none  
2723 measured reflections  
2503 independent reflections  
1527 reflections with I > 2σ(I)  
Rint = 0.015  
θmax = 26.0°  
h = 0 → 8  
k = 0 → 9  
l = −15 → 15  
3 standard reflections  
Frequency: 60 min  
Intensity decay: 0.1%

**Refinement**

Refinement on F²  
R(F²) > 2σ(F²) = 0.041  
wR(F²) = 0.121  
S = 1.02  
2593 reflections  
174 parameters  
H-atom parameters constrained

**Table 1**

Selected geometric parameters (Å, °).

<p>| | | | |</p>
<table>
<thead>
<tr>
<th></th>
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<tbody>
<tr>
<td>C1—C1</td>
<td>1.750 (3)</td>
<td>C8—N3</td>
<td>1.387 (3)</td>
</tr>
<tr>
<td>C4—C7</td>
<td>1.447 (3)</td>
<td>N3—N4</td>
<td>1.399 (2)</td>
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<tr>
<td>C7—N1</td>
<td>1.271 (3)</td>
<td>N3—C9</td>
<td>1.403 (3)</td>
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<tr>
<td>N1—N2</td>
<td>1.378 (2)</td>
<td>N4—C11</td>
<td>1.279 (3)</td>
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<tr>
<td>N2—C11</td>
<td>1.381 (3)</td>
<td>C9—O2</td>
<td>1.197 (3)</td>
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<tr>
<td>N2—C8</td>
<td>1.387 (3)</td>
<td>C9—C10</td>
<td>1.479 (3)</td>
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<tr>
<td>C8—O1</td>
<td>1.209 (2)</td>
<td>C11—C12</td>
<td>1.481 (3)</td>
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<tr>
<td>O2—C9—N3</td>
<td>119.8 (2)</td>
<td>N4—C11—N2</td>
<td>112.30 (18)</td>
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<tr>
<td>O2—C9—C10</td>
<td>124.5 (2)</td>
<td>N4—C11—C12</td>
<td>125.2 (2)</td>
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<tr>
<td>N3—C9—C10</td>
<td>115.73 (19)</td>
<td>N2—C11—C12</td>
<td>122.5 (2)</td>
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**Table 2**

Hydrogen-bonding geometry (Å, °).

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<tr>
<th>D—H···A</th>
<th>D—H</th>
<th>H···A</th>
<th>D···A</th>
<th>D—H···A</th>
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<tbody>
<tr>
<td>C7—H7···O1</td>
<td>0.93</td>
<td>2.28</td>
<td>2.938 (3)</td>
<td>127</td>
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<tr>
<td>C2—H2···O2²</td>
<td>0.93</td>
<td>2.52</td>
<td>3.162 (3)</td>
<td>126</td>
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<tr>
<td>C12—H12B···O2²</td>
<td>0.96</td>
<td>2.52</td>
<td>3.414 (3)</td>
<td>154</td>
</tr>
<tr>
<td>C10—H10C···O1iii</td>
<td>0.96</td>
<td>2.61</td>
<td>3.499 (3)</td>
<td>154</td>
</tr>
</tbody>
</table>

Symmetry codes: (i) 2 − x, 1 − y, 2 − z; (ii) 1 − x, 1 − y, 2 − z; (iii) x − 1, y, z.

The H atoms were positioned geometrically and refined using a riding model, fixing the aromatic C—H distance at 0.93 Å and the
methyl C—H distance at 0.96 Å, with \( U_{iso}(H) = 1.2U_{eq}(C) \) or 1.5\( U_{eq}(C) \) for the methyl group.

Data collection: CAD-4-PC Software (Enraf–Nonius, 1992); cell refinement: CAD-4-PC Software; data reduction: XCAD4 (Harms, 1997); program(s) used to solve structure: SHELXS97 (Sheldrick, 1997); program(s) used to refine structure: SHELXL97 (Sheldrick, 1997); molecular graphics: ORTEP-3 for Windows (Farrugia, 1997); software used to prepare material for publication: SHELXL97.

References


