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NOTE

Supramolecular Structure of 1-Phenyl-3-methyl-4-benzoylpyrazolon-5-one

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The compound 1-phenyl-3-methyl-4-benzoylpyrazolon-5-one with the molecular formula $C_{17}H_{14}N_2O_2$, exists in a keto-enamine tautomeric form. The pyrazolone ring makes dihedral angles of 28.36(3) and 59.31(5)° with the two phenyl rings. The molecules are linked into two-dimensional supramolecular structure by a combination of N-H···O and C-H···O hydrogen-bonding interactions.

Key Words: 1-Phenyl-3-methyl-4-benzoylpyrazolon-5-one, Supramolecular structure.

1-Phenyl-3-methyl-4-benzoylpyrazolon-5-one, an effective β-diketonate, is widely used and well known for its extractive ability[1-3]. In recent years, its metal complexes have also been found to have good antibacterial and biological properties. Its metal complexes have analgesic activity[4-6]. For our interest in super supramolecular chemistry, herein we've mainly studied the supramolecular structure of the this compound.

1-Phenyl-3-methyl-5-pyrazolone was purchased from Alfa Aesar and used without any purification. The reagents and solvents were analytical grade reagents from Tianjin Chemical Reagent Factory. C, H and N analyses were carried out with a GmbH VariuoEL V3.00 automatic elemental analyzer. Melting points were measured by the use of a microscopic melting point apparatus made in Beijing Taike Instrument Limited Company and the thermometer was uncorrected. X-Ray single crystal structure was determined on a Bruker Smart APEX CCD area detector.

General procedure: 1-Phenyl-3-methyl-5-pyrazolone (3.75 g) was dissolved in 80 mL of 1,4-dioxane by heating. Calcium hydroxide (6 g) was added followed by addition of benzoyl chloride (3 mL) dropwise within 1 min. The temperature increased during the first few minutes and the reaction mixture became a thick paste. The reaction mixture was refluxed for 0.5 h. Calcium complex of the desired ligand formed in the flask was decomposed by pouring the mixture into dilute hydrochloric acid (50 mL). An orange precipitate was formed slowly. It was filtered under suction, washed with a little water and 1,4-dioxane and recrystallised from a slightly acidified methanol-water mixture to destroy any undecomposed calcium complex. Yield 79.8 %. m.p. 89-91 ºC. Anal. calcd. (%) for $C_{17}H_{14}N_2O_2$: C, 73.25; H, 5.04; N, 10.01. Found (%): C, 73.37; H, 5.07; N, 10.07.

Colourless needle-like single crystals suitable for X-ray diffraction studies were obtained by slow evaporation from a solution of dichloromethane at room temperature over a period of approximately one month.

TABLE-1
CRYSTAL DATA AND REFINEMENT PARAMETERS FOR THE TITLE COMPOUND

Empirical formula	$C_{17}H_{14}N_2O_2$
Formula weight	278.30
Temperature	298(2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2(1)/c
Cell dimensions	a = 13.4824(16) Å, b = 9.1016(11) Å, c =11.4348(12) Å, β = 90.8170(10)
Volume	871.56(18) Å^3
Z	4
Density (calculated)	1.318 mg/m^3
Absorption coefficient	0.088 mm^{-1}
$F_{(000)}$	584
Index ranges	$-16 \le h \le 14$, $-10 \le k \le 9$, $-13 \le l \le 13$
Reflections collected/unique	6863 / 2472 [R_{int} = 0.0548]
Data/restraints/parameters	2472 / 0 / 192
Goodness of fit indicator	1.030
Final R indices [I > 2σ(I)]	R_1=0.0468, wR_2=0.1080
R indices (all data)	R_1=0.0858, wR_2=0.1371
Largest diff. peak and hole	0.299 and -0.229 e Å^{-3}

X-Ray structure determination: The single crystal of the title compound with approximate dimensions of 0.45 mm

TABLE-2
SELECTED BOND LENGTHS (Å) AND ANGLES (°) FOR THE TITLE COMPOUND

Bond	Lengths	Bond	Lengths	Bond	Lengths
N(1)-N(2)	1.377(3)	O(1)-C(1)	1.247(3)	C(2)-C(11)	1.466(3)
N(1)-C(1)	1.391(3)	O(2)-C(11)	1.224(3)	C(3)-C(4)	1.484(3)
N(1)-C(5)	1.418(3)	C(1)-C(2)	1.433(3)	C(5)-C(10)	1.371(4)
N(2)-C(3)	1.330(3)	C(2)-C(3)	1.381(3)	C(5)-C(6)	1.372(4)
C(6)-C(7)	1.375(4)	C(11)-C(12)	1.490(3)	C(14)-C(15)	1.372(4)
C(7)-C(8)	1.349(5)	C(12)-C(17)	1.384(3)	C(15)-C(16)	1.380(4)
C(8)-C(9)	1.370(5)	C(12)-C(13)	1.385(3)	C(16)-C(17)	1.382(4)
C(9)-C(10)	1.390(4)	C(13)-C(14)	1.380(3)	–	–
Bond	Angles	Bond	Angles	Bond	Angles
N(2)-N(1)-C(1)	108.78(18)	N(2)-N(1)-C(1)	108.78(18)	C(2)-C(3)-C(4)	132.4(2)
C(3)-N(2)-N(1)	109.26(19)	C(3)-N(2)-N(1)	109.26(19)	C(10)-C(5)-C(6)	120.1(2)
O(1)-C(1)-N(1)	122.4(2)	O(1)-C(1)-N(1)	122.4(2)	C(5)-C(6)-C(7)	119.4(3)
C(3)-C(2)-C(1)	107.3(2)	C(3)-C(2)-C(1)	107.3(2)	C(8)-C(7)-C(6)	121.6(3)
C(7)-C(8)-C(9)	119.2(3)	O(2)-C(11)-C(12)	119.9(2)	C(13)-C(12)-C(11)	119.3(2)
C(8)-C(9)-C(10)	120.6(3)	C(2)-C(11)-C(12)	119.6(2)	C(14)-C(15)-C(16)	119.9(3)
C(5)-C(10)-C(9)	119.2(3)	C(17)-C(12)-C(13)	118.6(2)	C(15)-C(16)-C(17)	119.9(3)
O(2)-C(11)-C(2)	120.5(2)	C(17)-C(12)-C(11)	122.0(2)	C(16)-C(17)-C(12)	120.7(2)

× 0.17 mm × 0.14 mm was placed on a Bruker Smart 1000 diffractmeter equipped with Apex CCD area detector. The diffraction data were collected using a graphite monochromated MoK$_\alpha$ radition (λ = 0.71073 Å) at 298(2) K. The structure was solved by using the program SHELXS-97[7] and Fourier difference techniques and refined by full-matrix least-squares method on F^2 using SHELXL-97[8]. Details of the data collection and refinements of the title compound are given in Table-1. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were added theoretically. CCDC: 894881.

The structure of the title compound is shown in Fig. 1. The dihedral angles formed by the pyrazolone ring with the two phenyl rings C5-C10 and C12-C17 are 28.36(3) and 59.31(5)º, respectively. Two carbonyl oxygen atoms are available for coordination with metals. The pyrazolone ring is planar and atoms O1, N1, N2, C1, C2, C3, C4 and C11 are almost coplanar, the largest deviation being 0.035 (3) Å for atom O1. But the O2 atom is not on the mean plane with a deviation of 0.652(2) Å. The dihedral angle between this mean plane and the pyrazolone ring of 1-phenyl-3-methyl-4-benzoylpyrazolon-5-one is 0.39(1)°. A strong intermolecular N2-H2···O1 hydrogen bond (Table-2) stabilized the molecules into a 1D infinite chain parallel to the c-axis (Fig. 2)[9,10]. This linkage is further stabilized by a pair of weak intermolecular C13-H13···O2 hydrogen-bonding interactions to form an infinite 2D supramolecular structure (Fig. 2) (Table-3)[11,12].

TABLE-3
HYDROGEN-BONDING DATA (Å, °)

D-H···A	d(D-H)	d(H···A)	d(D···A)	∠D-H···A
N2-H2···O1	0.86	1.88	2.668(3)	151
C13-H13···O2	0.93	2.55	3.288(3)	137

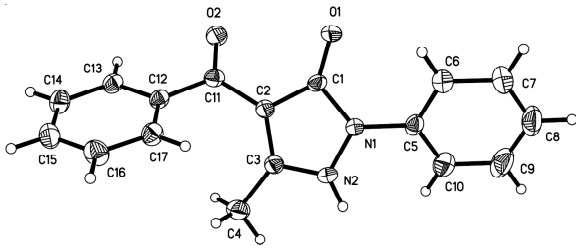

Fig. 1. Molecule structure of the title compound

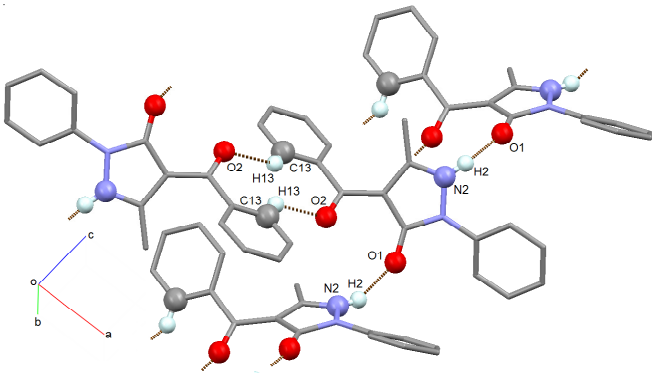

Fig. 2. View of the supramolecular structure within the title compound (hydrogen atoms, except those forming hydrogen bonds, are omitted for clarity)

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