Table 1. *Comparison of selected geometric parameters (Å,°) for (I) and (II)*

	(I)	(II)
C1—C8a	1.413 (3)	1.403 (6)
C1—C15	1.414 (4)	–
C2—C9	1.464 (3)	1.485 (6)
C1—C2	1.416 (3)	1.402 (6)
C2—C3	1.396 (3)	1.403 (6)
C3—C3a	1.394 (4)	1.396 (6)
C4—C3a	1.392 (4)	1.383 (6)
C4—C5	1.381 (5)	1.401 (7)
C5—C6	1.377 (5)	1.382 (8)
C6—C7	1.376 (5)	1.392 (7)
C7—C8	1.394 (4)	1.369 (7)
C8—C8a	1.381 (4)	1.396 (6)
C8a—C3a	1.471 (3)	1.478 (6)
C2—C1—C8a	109.2 (2)	108.8 (4)
C1—C2—C3	107.8 (2)	108.9 (4)
C2—C3—C3a	109.7 (2)	108.9 (4)
C3—C3a—C8a	107.4 (2)	106.9 (4)
C4—C3a—C8a	126.5 (3)	127.3 (4)
C3a—C4—C5	129.4 (3)	128.7 (5)
C4—C5—C6	128.6 (3)	128.7 (5)
C5—C6—C7	130.1 (3)	129.8 (5)
C6—C7—C8	128.8 (3)	128.7 (5)
C7—C8—C8a	128.2 (3)	129.4 (5)
C8—C8a—C3a	128.3 (2)	127.3 (4)
C1—C8a—C3a	105.8 (2)	106.5 (4)

For both compounds, data collection: *MSC/AFC Data Reduction and Refinement Software* (Rigaku Corporation, 1988); cell refinement: *MSC/AFC Data Reduction and Refinement Software*; program(s) used to solve structures: *SHELXS*86 (Sheldrick, 1990); program(s) used to refine structures: *SHELXL*93 (Sheldrick, 1993); molecular graphics: *ORTEP*II (Johnson, 1976) and *TEXSAN* (Molecular Structure Corporation, 1992); software used to prepare material for publication: *WORD*6.0.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: BS1032). Services for accessing these data are described at the back of the journal.

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Tris(1-phenacyl-2-pyridone) Hydroxonium Tetrafluoroborate, a Hydrogen-Bonded Complex

S. G. Zhukov,[a] V. B. Rybakov,[a] E. V. Babaev,[a]
K. A. Paseshnichenko[a] and H. Schenk[b]

[a]*Department of Chemistry, Moscow State University, 119899 Moscow, Russian Federation, and* [b]*Laboratory for Crystallography, Amsterdam University, Nieuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands. E-mail: ksenia@biocryst.phys.msu.su*

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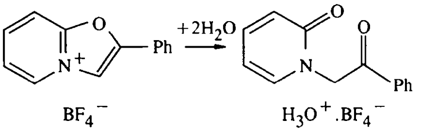
Abstract

The hydrolytic cleavage of 2-phenyloxazolo[3,2-*a*]pyridinium tetrafluoroborate results in the formation of the title tris(1-phenacyl-2-pyridone) hydroxonium tetrafluoroborate complex, $3C_{13}H_{11}NO_2.H_3O^+.BF_4^-$. The structure is built up from hydrogen-bonded cations and disordered BF_4^- anions. The strong hydrogen bonding causes considerable redistribution of electron density in the pyridone moiety.

Comment

Recently, it was found that mesoionic 2-oxo-3-benzoyloxazolo[3,2-*a*]pyridine can be transformed to oxazolo[3,2-*a*]pyridinium tetrafluoroborate by the action of H_2SO_4 and HBF_4 (Babaev & Orlova, 1997). Having attempted to confirm the structure of this compound, we discovered that prolonged standing of the reaction mixture (one week at room temperature) resulted in the unexpected formation of *N*-phenacyl-2-pyridone, (I). Although the ring opening of the former compound is known to occur by the action of alkali (Pauls

& Kroehnke, 1976), no data on the extremely mild hydrolysis by the action of water have been reported. The formation of pyridone is the result of hydrolytic cleavage of the oxazolium fragment by two molecules of water (see scheme below).



(I)

The first water molecule causes ring opening and formation of pyridone, while the second serves as the acceptor of a proton. The *N*-phenacyl-2-pyridone molecule forms a strong hydrogen bond *via* the pyridone O atom to the H_3O^+ cation positioned on the 3 axis [O2···H1 1.50 (2), O2···O1 2.524 (2) Å and O2···H1—O1 171 (2)°]. As a result of hydrogen bonding, the C2—O2 bond in the present structure is 0.02 Å longer than the longest in known structures containing the *N*-alkylated pyridone moiety (Nawata, Matsuura, Ando & Iitaka, 1990; Hsu & Craven, 1974; Schwalbe & Saenger, 1973). The other bond lengths and angles are ordinary for compounds of this type. In the difference Fourier maps, no sign of proton transfer from H_3O^+ to the pyridone O atom was found. The BF_4^- anion is orientationally disordered over two sets of sites and does not form any considerably short contacts with the hydrogen-bonded complex cation; the shortest F···H(pyridone) distances are 2.40 Å, *i.e.* 0.2 Å shorter than the sum of the van der Waals radii. We have not found any examples of such compounds with the H_3O^+ cation coordinated by three neutral organic molecules in a search of the Cambridge Structural Database (Allen & Kennard, 1993).

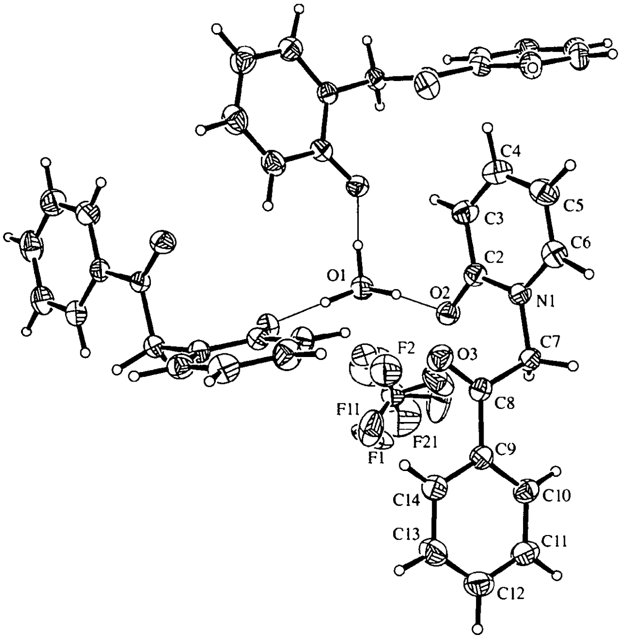


Fig. 1. The formula unit of tris(1-phenacyl-2-pyridone) hydroxonium tetrafluoroborate showing 25% probability displacement ellipsoids.

Experimental

Single crystals of the title compound were obtained by evaporation of a water solution of 2-phenyloxazolo[3,2-*a*]pyridinium tetrafluoroborate. Chemical analysis: found C 62.75, H 4.40%; $C_{39}H_{36}BF_4N_3O_7$ requires C 62.83, H 4.46%.

Crystal data

$3C_{13}H_{11}NO_2.H_3O^+.BF_4^-$
$M_r = 745.52$
Trigonal
$R\bar{3}$
$a = 14.020 (2)$ Å
$c = 33.005 (5)$ Å
$V = 5618.3 (14)$ Å^3
$Z = 6$
$D_x = 1.322$ Mg m^{-3}
D_m not measured

Mo $K\alpha$ radiation
$\lambda = 0.7107$ Å
Cell parameters from 22 reflections
$\theta = 15.4–17.0°$
$\mu = 0.104$ mm^{-1}
$T = 293 (2)$ K
Irregular
0.55 × 0.40 × 0.33 mm
Colourless

Data collection

Enraf–Nonius CAD-4 diffractometer
ω scans
Absorption correction: none
2724 measured reflections
2724 independent reflections
1355 reflections with $I > 2\sigma(I)$

$\theta_{max} = 26.9°$
$h = 0 \rightarrow 15$
$k = 0 \rightarrow 15$
$l = -41 \rightarrow 42$
2 standard reflections
frequency: 150 min
intensity decay: none

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.053$
$wR(F^2) = 0.130$
$S = 1.027$
2724 reflections
225 parameters
All H atoms refined
$w = 1/[\sigma^2(F_o^2) + (0.051P)^2]$
where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} = 0.012$

$\Delta\rho_{max} = 0.151$ e Å^{-3}
$\Delta\rho_{min} = -0.154$ e Å^{-3}
Extinction correction: *SHELXL93*
Extinction coefficient: 0.0011 (2)
Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. *Fractional atomic coordinates and equivalent isotropic displacement parameters (Å^2)*

$$U_{eq} = (1/3)\Sigma_i\Sigma_j U^{ij} a_i^* a_j^* \mathbf{a}_i.\mathbf{a}_j.$$

	x	y	z	U_{eq}
O1	1/3	2/3	0.02200 (9)	0.0633 (8)
N1	0.55916 (14)	0.55407 (14)	0.08376 (5)	0.0502 (5)
O2	0.44653 (13)	0.57930 (12)	0.04134 (5)	0.0603 (5)
O3	0.36388 (13)	0.39143 (13)	0.10647 (5)	0.0657 (5)
C2	0.5147 (2)	0.6164 (2)	0.07057 (7)	0.0534 (6)
C3	0.5497 (2)	0.7171 (2)	0.09097 (9)	0.0659 (7)
C4	0.6222 (2)	0.7485 (2)	0.12176 (8)	0.0747 (8)
C5	0.6657 (3)	0.6831 (3)	0.13395 (9)	0.0742 (8)
C6	0.6330 (2)	0.5866 (2)	0.11477 (7)	0.0612 (7)
C7	0.5208 (2)	0.4469 (2)	0.06510 (8)	0.0512 (6)
C8	0.4073 (2)	0.3640 (2)	0.08081 (7)	0.0493 (6)
C9	0.3530 (2)	0.2508 (2)	0.06390 (7)	0.0518 (6)

C10	0.4024 (2)	0.2193 (2)	0.03446 (7)	0.0590 (7)
C11	0.3484 (3)	0.1142 (2)	0.01876 (9)	0.0704 (8)
C12	0.2450 (3)	0.0404 (3)	0.03236 (10)	0.0810 (9)
C13	0.1952 (3)	0.0699 (3)	0.06187 (11)	0.0825 (9)
C14	0.2491 (2)	0.1753 (2)	0.07741 (9)	0.0681 (7)
F1†	0.5770 (6)	0.3274 (8)	0.1594 (3)	0.236 (6)
F11‡	0.5684 (6)	0.3173 (5)	0.1372 (3)	0.145 (5)
F2†	2/3	1/3	0.1069 (3)	0.145 (4)
F21‡	2/3	1/3	0.1905 (4)	0.228 (9)
B1	2/3	1/3	0.1498 (2)	0.083 (2)

† Site occupancy = 0.580 (12). ‡ Site occupancy = 0.420 (12).

Table 2. *Selected geometric parameters (Å, °)*

N1—C6	1.362 (3)	C2—C3	1.412 (3)
N1—C2	1.374 (3)	C3—C4	1.345 (4)
N1—C7	1.455 (3)	C4—C5	1.392 (4)
O2—C2	1.272 (2)	C5—C6	1.349 (4)
C6—N1—C2	122.4 (2)	N1—C2—C3	116.3 (2)
C6—N1—C7	119.4 (2)	C4—C3—C2	121.0 (3)
C2—N1—C7	118.1 (2)	C3—C4—C5	120.7 (3)
O2—C2—N1	117.8 (2)	C6—C5—C4	118.9 (3)
O2—C2—C3	125.8 (2)	C5—C6—N1	120.6 (3)
C6—N1—C7—C8	101.1 (2)	N1—C7—C8—C9	177.7 (2)
C2—N1—C7—C8	−75.6 (2)	O3—C8—C9—C14	1.1 (3)
N1—C7—C8—O3	−2.2 (3)	O3—C8—C9—C10	179.8 (2)

Data collection: *CAD*-4 *Software* (Enraf–Nonius, 1989). Cell refinement: *CAD*-4 *Software*. Data reduction: *SDP-Plus* (Frenz, 1985). Program(s) used to solve structure: *SHELXS*96 (Sheldrick, 1996). Program(s) used to refine structure: *SHELXL*93 (Sheldrick, 1993). Molecular graphics: *ORTEX* (McArdle, 1995). Software used to prepare material for publication: *SHELXL*93.

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Supplementary data for this paper are available from the IUCr electronic archives (Reference: NA1311). Services for accessing these data are described at the back of the journal. A packing diagram has also been deposited.

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(−)-2-Azabicyclo[2.2.1]hept-5-en-3-one (Lactam)

Bernadette Suchod,[a] Janine Lajzerowicz[a] and André Collet[b]

[a]*Laboratoire de Spectrométrie Physique, Unité mixte de Recherche (C5588), Université Joseph Fourier – Grenoble 1 – CNRS, BP 87 – 38402 Saint Martin d'Hères CEDEX, France, and* [b]*Ecole normale supérieure de Lyon, Stéréochimie et Interactions Moléculaires (UMR CNRS 117), 46 allée d'Italie, 69364 Lyon CEDEX 07, France. E-mail: suchod@coucou.ujf-grenoble.fr*

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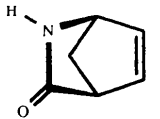
Abstract

In the lactam structure, C_6H_7NO, molecules are linked by hydrogen bonds between N—H and C═O [N···O 2.885 (2) Å] to form ribbons parallel to the [100] 2_1 axis. The shortest intermolecular distances outside the ribbons are relatively large [C···O 3.535 (2) and C···C 3.600 (2) Å].

Comment

The (+) and (−) enantiomers of the title bicyclic lactam are key synthons for the preparation of carbocyclic nucleosides used as chemotherapeutic agents. It was discovered recently (Potter *et al.*, 1996) that the (±)-lactam crystallizes as a conglomerate; this finding induced us to focus on the possibility of resolving this compound by the entrainment method (Jacques, Collet & Wilen, 1981). An amazing observation was made during this entrainment process: under certain conditions, the crystallization of supersaturated solutions proceeds *via* a series of oscillations of their enantiomeric composition from (+) to (−), and conversely, until the final racemic equilibrium composition is achieved. To understand this behaviour, it seemed useful to elucidate the crystallographic structure of the enantiomer, (I).



(I)

The structure of (I) is organized in ribbons of molecules linked by hydrogen bonds between C═O and N—H [N···O 2.885 (2) Å]. These ribbons are parallel to [100] and the molecules are related by a 2_1 axis. In the plane perpendicular to the ribbons, their section has a herring-bone disposition. Outside the ribbons, the structure is relatively loose and the shortest intermolecular distances are large [C4···C5$(-x, \frac{1}{2}+y, \frac{1}{2}-z)$ 3.600 (2) and C7···O8$(-x, y-\frac{1}{2}, \frac{1}{2}-z)$ 3.535 (2) Å].

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