Supplementary Materials

Table S1. Crystal data and structure refinement for 100 K.

Identification code	pyox99
Empirical formula	C5 H6 N2 O4
Formula weight	158.12
Temperature	100 K
Wavelength	0.71073 A
Crystal system, space group	TRICLINIC, P -1
Unit cell dimensions	a = 3.6202(1) A alpha = 117.336(3) deg. b = 9.8258(3) A beta = 95.831(2) deg. c = 10.4146(3) A gamma = 94.177(2) deg.
Volume	324.39(2) A^3
Z, Calculated density	2, 1.619 Mg/m^3
Absorption coefficient	0.142 mm^-1
F(000)	164
Crystal size	0.38 x 0.29 x 0.23 mm
Theta range for data collection	3.93 to 29.52 deg.
Limiting indices	-4<=h<=4, -12<=k<=13, -14<=l<=14
Reflections collected / unique	15797 / 1662 [R(int) = 0.0336]
Completeness to theta = 26.00	99.7 %
Absorption correction	Empirical
Max. and min. transmission	1.1672 and 0.9567
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	1662 / 0 / 103
Goodness-of-fit on F^2	1.240
Final R indices [I>2sigma(I)]	R1 = 0.0401, wR2 = 0.0827
R indices (all data)	R1 = 0.0560, wR2 = 0.0892
Largest diff. peak and hole	0.358 and -0.319 e.A^-3

Table S2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 100 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
O(001)	4262(3)	8407(1)	8639(1)	22(1)
O(002)	1002(3)	6993(1)	6393(1)	21(1)
O(003)	1708(3)	11000(1)	8833(1)	23(1)
O(004)	-1410(3)	9651(1)	6583(1)	23(1)
N(005)	3438(3)	3353(1)	6014(1)	19(1)
N(006)	4130(3)	4874(1)	6952(1)	20(1)
C(007)	6639(4)	3638(2)	8061(2)	21(1)
C(008)	616(4)	9700(2)	7605(2)	19(1)
C(009)	4935(4)	2578(2)	6660(2)	21(1)
C(00A)	2112(4)	8239(2)	7556(2)	18(1)
C(00B)	6064(4)	5078(2)	8206(2)	21(1)

Table S3. Bond lengths [A] and angles [deg] for 100 K.

O(001)-C(00A)	1.2393(16)
O(002)-C(00A)	1.2632(16)
O(003)-C(008)	1.3217(16)
O(003)-H(003)	0.85(2)
O(004)-C(008)	1.2096(16)
N(005)-C(009)	1.3361(18)
N(005)-N(006)	1.3433(16)
N(005)-H(005)	0.8600
N(006)-C(00B)	1.3375(18)
N(006)-H(006)	0.8600
C(007)-C(009)	1.386(2)
C(007)-C(00B)	1.387(2)
C(007)-H(007)	0.9300
C(008)-C(00A)	1.5514(19)
C(009)-H(009)	0.9300
C(00B)-H(00B)	0.9300
C(008)-O(003)-H(003)	112.7(13)
C(009)-N(005)-N(006)	108.92(11)
C(009)-N(005)-H(005)	125.5
N(006)-N(005)-H(005)	125.5
C(00B)-N(006)-N(005)	108.89(11)
C(00B)-N(006)-H(006)	125.6
N(005)-N(006)-H(006)	125.6
C(009)-C(007)-C(00B)	105.66(12)
C(009)-C(007)-H(007)	127.2
C(00B)-C(007)-H(007)	127.2
O(004)-C(008)-O(003)	122.22(12)
O(004)-C(008)-C(00A)	122.08(12)
O(003)-C(008)-C(00A)	115.69(12)
N(005)-C(009)-C(007)	108.30(12)
N(005)-C(009)-H(009)	125.9
C(007)-C(009)-H(009)	125.9
O(001)-C(00A)-O(002)	126.84(12)
O(001)-C(00A)-C(008)	117.60(12)
O(002)-C(00A)-C(008)	115.55(12)
N(006)-C(00B)-C(007)	108.23(12)
N(006)-C(00B)-H(00B)	125.9
C(007)-C(00B)-H(00B)	125.9

Symmetry transformations used to generate equivalent atoms:

Table S4. Anisotropic displacement parameters (A^2 x 10^3) for 100 K. The anisotropic displacement factor exponent takes the form: 2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
O(001)	28(1)	17(1)	17(1)	5(1)	-3(1)	4(1)
O(002)	26(1)	14(1)	16(1)	3(1)	-2(1)	2(1)
O(003)	31(1)	15(1)	17(1)	4(1)	-3(1)	4(1)
O(004)	29(1)	19(1)	19(1)	7(1)	-3(1)	4(1)
N(005)	23(1)	15(1)	13(1)	3(1)	0(1)	1(1)
N(006)	24(1)	15(1)	19(1)	6(1)	1(1)	3(1)
C(007)	22(1)	23(1)	18(1)	10(1)	0(1)	4(1)
C(008)	21(1)	14(1)	17(1)	5(1)	2(1)	2(1)
C(009)	24(1)	18(1)	21(1)	9(1)	4(1)	5(1)
C(00A)	20(1)	16(1)	16(1)	6(1)	2(1)	3(1)
C(00B)	22(1)	21(1)	15(1)	5(1)	1(1)	2(1)

Table S5. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 100 K.

	x	y	z	U(eq)
H(003)	2990(50)	10860(20)	9480(20)	35
H(005)	2229	2946	5140	23
H(006)	3441	5598	6775	24
H(007)	7905	3427	8758	26
H(009)	4847	1509	6242	25
H(00B)	6883	6023	9032	25

Table S6. Torsion angles [deg] for 100 K.

C(009)-N(005)-N(006)-C(00B)	-0.18(15)
N(006)-N(005)-C(009)-C(007)	0.08(16)
C(00B)-C(007)-C(009)-N(005)	0.05(16)
O(004)-C(008)-C(00A)-O(001)	177.94(13)
O(003)-C(008)-C(00A)-O(001)	-1.87(19)
O(004)-C(008)-C(00A)-O(002)	-1.6(2)
O(003)-C(008)-C(00A)-O(002)	178.58(12)
N(005)-N(006)-C(00B)-C(007)	0.22(15)
C(009)-C(007)-C(00B)-N(006)	-0.17(16)

Symmetry transformations used to generate equivalent atoms:

Table S7. Hydrogen bonds for 100 K [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
O(003)-H(003)...O(001)	0.85(2)	2.259(19)	2.7005(14)	112.5(15)
O(003)-H(003)...O(001)#1	0.85(2)	1.88(2)	2.6622(14)	152.1(18)
N(005)-H(005)...O(002)#2	0.86	1.91	2.7025(15)	153.0
N(005)-H(005)...O(004)#2	0.86	2.32	2.9181(15)	126.6
N(006)-H(006)...O(001)	0.86	2.51	3.0821(15)	124.4
N(006)-H(006)...O(002)	0.86	1.85	2.6975(15)	168.5
C(007)-H(007)...O(003)#3	0.93	2.87	3.6108(17)	138.0
C(009)-H(009)...O(004)#3	0.93	2.48	3.2235(17)	136.9
C(00B)-H(00B)...O(001)	0.93	2.77	3.2164(17)	110.4
C(00B)-H(00B)...O(003)#1	0.93	2.71	3.6248(17)	168.4

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,-y+2,-z+2 #2 -x,-y+1,-z+1 #3 x+1,y-1,z

Table S8. Crystal data of 1H-Pyrazol-2-ium hydrogen oxalate.

	Yu et al.[14]	**Karthiga Devi et al.**[25]	**Our work**
a [Å]	3.7286 (7)	3.775	3.6202 (1)
b [Å]	9.836 (2)	9.93	9.8258 (3)
c [Å]	10.487 (2)	10.55	10.4146 (3)
α (°)	117.35 (3)	117	117.336 (3)
β (°)	97.01 (3)	97.13	95.831 (2)
γ (°)	93.65 (3)	93.79	94.177 (2)
V (Å^3)	335.92 (14)	345	324.39 (2)
R	0.0965		0.0403
Crystal system	Triclinic	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$	$P\bar{1}$

Figure S1. Raman spectra of 1H-Pyrazol-2-ium hydrogen oxalate recorded at 300 K (upper panel) and the sample deposited on a quartz plate (see text) (lower panel).