Electronic Supplementary Information

$CO(NH_2)_2 \cdot NH_4Cl$: an ultraviolet birefringent material with the conjugated C=O groups

Ya Wang,[a, b, #] Jian Han,[a, #] AbudukadiTudi, [a, b] Zhizhong Zhang, [a, b] Zhihua Yang, [a] Shilie Pan [a, *]

[a] CAS Key Laboratory of Functional Materials and Devices for Special Environments; Xinjiang Technical Institute of Physics & Chemistry, CAS; Xinjiang Key Laboratory of Electronic Information Materials and Devices, 40-1 South Beijing Road, Urumqi 830011, China.

[b] Center of Materials Science and Optoelectronics Engineering,University of Chinese Academy of Sciences, Beijing 100049, China.

[#] These authors contributed equally to this work.

* Corresponding authors: slpan@ms.xjb.ac.cn.

CONTENTS

Table S1. The molar ratios of starting compositions, experimental temperatures and the determined crystalline phases.

No.	Molar ratios		Experimental temperature	Product
	$CO(NH_2)_2$	NH_4Cl		
1	1	1	room temperature	$CO(NH_2)_2 \cdot NH_4Cl$, $CO(NH_2)_2$
2	1	2	room temperature	no assignment
3	1	3	room temperature	no assignment
4	2	1	room temperature	no assignment
5	3	1	room temperature	$CO(NH_2)_2 \cdot NH_4Cl$, NH_4Cl
6	3	1	40 ℃	$CO(NH_2)_2 \cdot NH_4Cl$, NH_4Cl

Table S2. Crystal data and structure refinement for $CO(NH_2)_2 \cdot NH_4Cl$.

Empirical formula	$CO(NH_2)_2 \cdot NH_4Cl$
Formula weight	113.55
Temperature	293(2) K
Wavelength	0.71073 Å
Crystal system, space group	Orthorhombic, *Pmna*
Unit cell dimensions	a = 7.909(3) Å
	b = 17.113(6) Å
	c = 8.049(3) Å
Volume	1089.4(7) $Å^3$
Z, Calculated density	8, 1.385 mg/cm^3
Absorption coefficient	0.577 mm^{-1}
F(000)	480
Crystal size	0.24 × 0.20 × 0.12 mm^3
Limiting indices	$-8 \le h \le 10$, $-22 \le k \le 18$, $-10 \le l \le 8$
Reflections collected / unique	6080 / 1349 [R(int) = 0.0519]
Completeness to theta = 27.46	99.60%
Refinement method	Full-matrix least-squares on F^2
Goodness-of-fit on F^2	1.042

Final *R* indices [$I > 2\text{sigma}(I)$][a]	$R_1 = 0.0448$, $wR_2 = 0.1212$
R indices (all data)[a]	$R_1 = 0.0682$, $wR_2 = 0.1395$
Largest diff. peak and hole	0.240 and -0.392 e.A^{-3}

[a] $R_1 = \Sigma||F_o| - |F_c||/\Sigma|F_o|$ and $wR_2 = [\Sigma w(F_o^2 - F_c^2)^2 / \Sigma wF_o^4]^{1/2}$ for $F_o^2 > 2\sigma(F_o^2)$.

Table S3. Atomic coordinates ($\times 10^4$), equivalent isotropic displacement parameters (Å$^2 \times 10^3$) of each atom for $CO(NH_2)_2 \cdot NH_4Cl$. U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

Atom	x	y	z	U_{eq}
C(1)	5000	1587(2)	518(4)	39(1)
C(2)	5000	3404(2)	5439(4)	42(1)
N(1)	2500	5041(2)	2500	48(1)
N(2)	2500	4(2)	7500	46(1)
N(3)	3585(3)	1996(1)	508(4)	61(1)
N(4)	5000	3034(2)	3997(4)	54(1)
N(5)	5000	2958(2)	6784(4)	80(1)
Cl(1)	5000	1098(1)	5233(1)	45(1)
Cl(2)	0	3900(1)	4669(1)	44(1)
O(1)	5000	858(1)	582(3)	48(1)
O(2)	5000	4135(1)	5527(3)	53(1)

Table S4. Bond lengths (Å) and angles (°) for $CO(NH_2)_2 \cdot NH_4Cl$.

C(1)-O(1)	1.249(4)	O(1)-C(1)-N(3)	121.97(15)
C(1)-N(3)	1.321(3)	O(1)-C(1)-N(3)#1	121.97(15)
C(1)-N(3)#1	1.321(3)	N(3)-C(1)-N(3)#1	116.0(3)
C(2)-O(2)	1.254(4)	O(2)-C(2)-N(5)	121.9(3)
C(2)-N(5)	1.325(4)	O(2)-C(2)-N(4)	121.9(3)
C(2)-N(4)	1.322(4)	N(5)-C(2)-N(4)	116.2(3)
N(1)-H(1)	0.861(16)	H(1)-N(1)-H(2)	114(3)
N(1)-H(2)	0.855(16)	H(3)-N(2)-H(4)	115(3)
N(2)-H(3)	0.867(15)	C(1)-N(3)-H(5)	118(2)
N(2)-H(4)	0.859(16)	C(1)-N(3)-H(6)	121(2)
N(3)-H(5)	0.836(16)	H(5)-N(3)-H(6)	120(3)

N(3)-H(6)	0.852(16)	C(2)-N(4)-H(7)	119(2)
N(4)-H(7)	0.845(17)	C(2)-N(4)-H(8)	121(3)
N(4)-H(8)	0.847(16)	H(7)-N(4)-H(8)	120(3)
N(5)-H(9)	0.861(17)	C(2)-N(5)-H(9)	121(3)
N(5)-H(10)	0.866(17)	C(2)-N(5)-H(10)	126(3)
		H(9)-N(5)-H(10)	113(3)

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

Table S5. Hydrogen bond lengths (Å) and bond angles (°) in $CO(NH_2)_2 \cdot NH_4Cl$.

D-H	H...A	D...A	<(DHA)	
0.840(16)	2.65(2)	3.419(4)	152(3)	N5-H10...Cl1
0.837(16)	2.448(16)	3.280(3)	173(3)	N5-H9...Cl2_#1
0.826(16)	2.690(19)	3.458(3)	155(3)	N4-H8...Cl1
0.825(16)	2.482(17)	3.302(3)	173(3)	N4-H7...Cl2_#2
0.825(15)	2.672(17)	3.448(3)	157(2)	N3-H6...Cl2_#2
0.813(15)	2.482(16)	3.279(2)	167(2)	N3-H5...Cl1_#3
0.835(15)	2.083(17)	2.902(2)	167(3)	N1-H2...O2_#6
0.841(15)	2.447(16)	3.282(2)	172(3)	N1-H1...Cl2

[a]Symmetry codes: #1 x+1/2, y, -z+3/2; #2 x+1/2, y, -z+1/2; #3 x-1/2, y, -z+1/2; #4 x-1/2, y, -z+3/2;#5 -x+1, -y, -z+1;#6 -x+1, -y+1, -z+1

Table S6. Calculated refractive indices and birefringence at different wavelength of $CO(NH_2)_2 \cdot NH_4Cl$.

Wavelength (nm)	n_x	n_y	n_z	Δn
533.7	1.59457	1.60917	1.68849	0.09391
635.5	1.58697	1.60127	1.67938	0.09241
735.7	1.58252	1.59664	1.67403	0.09151
842.0	1.57950	1.59350	1.67040	0.09090
944.4	1.57752	1.59145	1.66803	0.09050
1075.0	1.57580	1.58966	1.66595	0.09015

Table S7. Calculated values of $\Delta\alpha$ of different chromospheres for $CO(NH_2)_2 \cdot NH_4Cl$.

Chromosphere	α_{xx}	α_{xy}	α_{yy}	α_{xz}	α_{yz}	α_{zz}	$\Delta\alpha$

s							
C=O	6.86	9.06	7.93	0.37	0.40	-1.66	9.10
$CO(NH_2)_2$(1)	30.16	0.00	33.11	0.00	0.30	22.26	9.72
$CO(NH_2)_2$(2)	22.31	0.00	33.42	0.00	0.02	30.60	10.00
NH_4(1)	5.58	0.00	5.35	0.01	0.00	5.29	0.43
NH_4(2)	5.19	0.00	5.61	0.02	0.00	5.03	0.52

Table S8. Birefringence and bond population of the C=O bonds.

Compounds	Δn (1064 nm)	Δn of C=O (1064 nm)	Bond population of the C=O bonds
COS	0.574	0.221	1.2
$(Mg(CO(NH_2)_2)_4(H_2O)_2)Br_2$	0.150	0.021	0.94-0.95
$CO(NH_2)_2$	0.116	0.077	0.93
$Li_2(SO_4)(CO(NH_2)_2)_3$	0.096	0.026	0.94-0.96
$CO(NH_2)_2 \cdot NH_4Cl$	0.090	0.086	0.91-0.92
$((NH_2)_2CO)H_3PO_4$	0.037	0.035	0.85

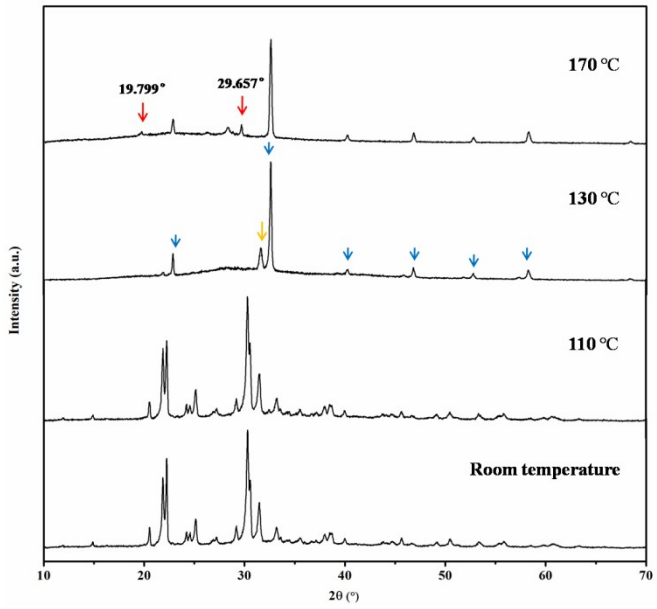

Figure S1. Variable-temperature powder XRD, XRD peaks of $C_3H_3N_3O_3$, $CO(NH_2)_2$ and NH_4Cl are indicated by light red, orange and blue arrows, respectively.

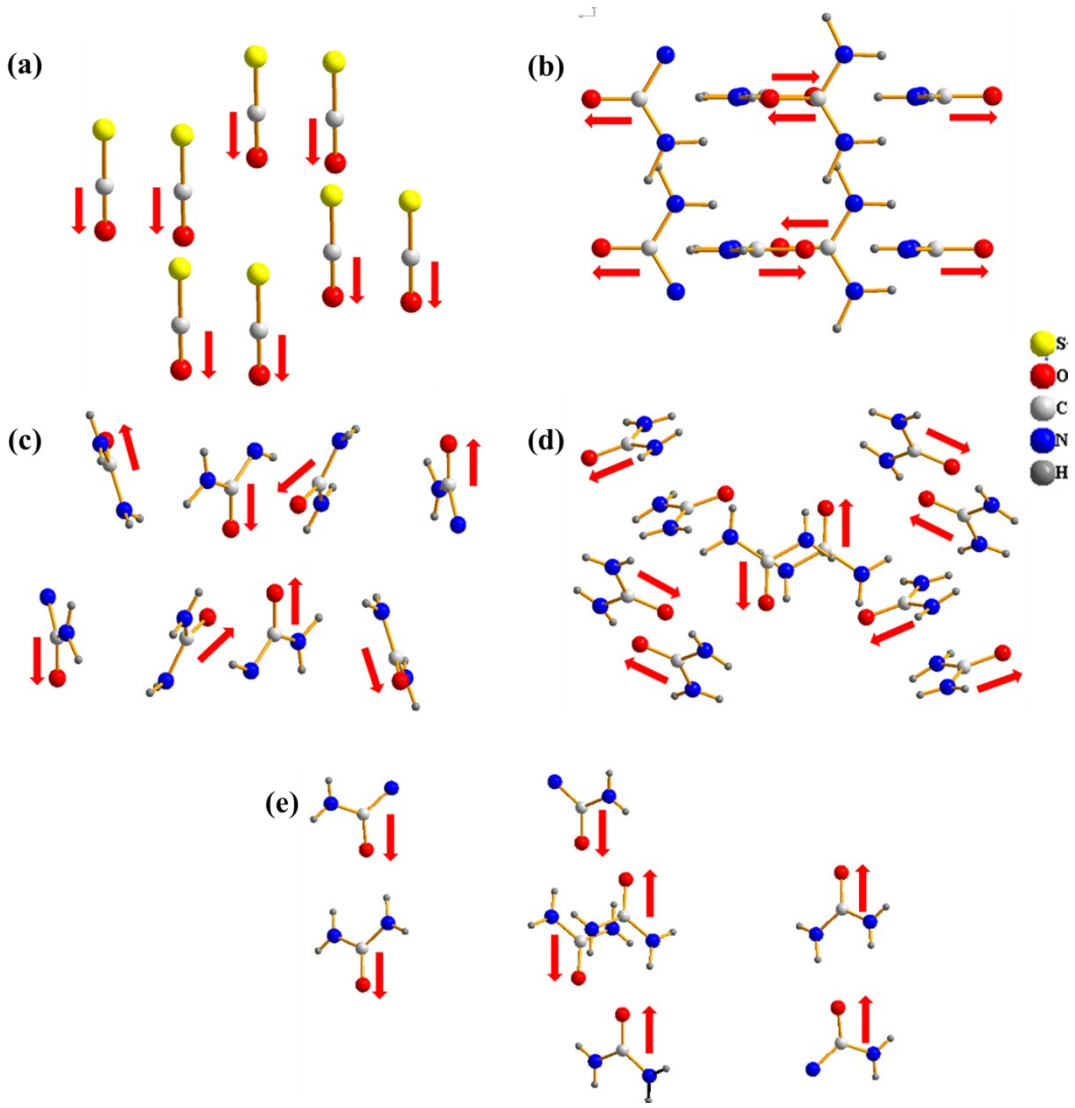

Figure S2. The arrangements of the C=O groups in (a) COS; (b) $CO(NH_2)_2$; (c) $(Mg(CO(NH_2)_2)_4(H_2O)_2)Br_2$; (d) $Li_2(SO_4)(CO(NH_2)_2)_3$; (e) $((NH_2)_2CO)H_3PO_4$.