O-AMINUMBENZOYLHYDRAZIDUM DI CHLORIDE

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ABSTRACT

The title compound was prepared and determined by x-ray diffraction analysis. The structure of the new compound crystalline in the triclinic system space group P_1^- with a=4.817 (5)Å, b=9.150 (5)Å, C=12.033 (5)Å and α=109.161 (5)°, β=92.209 (5)°and γ=101. 155 (5)°, Z=2.The final refinement of goodness of fit on F^2=1.052 The asymmetric unit of the compound contains two parts, one organic cation and two inorganic anion and the crystal structure was ensured by two types of interaction entered and intermolecular hydrogen bonds along the C axis N-H… Cl and N-H… O is resulting the formation of three dimensional Network and reinforcing the cohesion of the structure they have many area of application such us gas storage, catalyse...etc

Keywords: hybrid materials; hydrogen bond; X-ray diffraction.

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1. INTRODUCTION

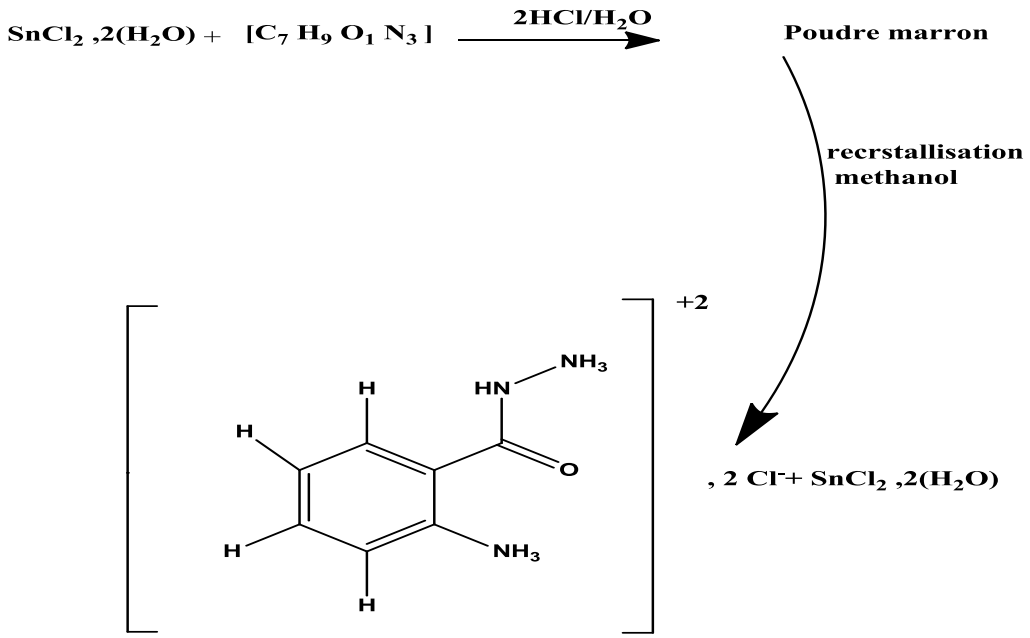
The O-amino benzoyle hydrazide has many biological activity[1] because of the acid hydrazide and amine function to exhibit anti-microbial, anti-fungal and Anti- tuberculostatic addition that, the coordination chemistry of this compound has been the subject of extensive investigation[2] because have donner atome.

In our investigation, we are interesting of research on hybrid material they are the object of an ever increasing the development of new materials containing multiple properties is a real challenge and is now attracting increasing interest from the scientific community the concept of "soft chemistry" at the end of the 1970s which has given substance to the notion of organic–inorganic hybrid material at the source of what represents the foundations of the concept of hybrid materials lies the word of life have a great potential application in many areas such as electrical, magnetic and optical properties[3,6]…etc in the other hand the important interaction between the organic and inorganic phases are the hydrogen bond when the relation ship between chemistry and biological study the transfer energy between organic cation and inorganic anion and they will give an important potential of the application. In this paper, we present a new organic-inorganic hybrid material o-aminumbenzoylhydrazidum Di-chloride and the hydrogen bond presented on this structure for modifying the crystal arrangement.

2. RESULTS AND DISCUSSION

EXPERIMENTAL

The title compound was crystallized by slow evaporation of an aqueous solution of o-amino benzoylhydrazide (C_7 H_9 O_1 N_3) , tin (II) chloride dihydrate and hydrochloric acid in a molar ratio of 1:1:1 under agitation during one hour at 60°C. the precipitate Formed and recrystallized from methanol.
Colourless prismatic crystals were obtained after one week.



Reaction scheme of the synthesis of the compound $[C_7 H_{11} O_1 N_3]^{+2}$, $2Cl^-$

REFINEMENT

All H atoms were located in difference Fourier maps but were placed in calculated positions and treated as riding on their parent C, N and O atoms, with C—H = 0.98 (Csp3) and 0.93 A ˚ (aromatic C), N—H = 0.89 A ˚ and Uiso(H) = 1.5Ueq(NH3 or Csp3) or 1.2Ueq(aromatic C).

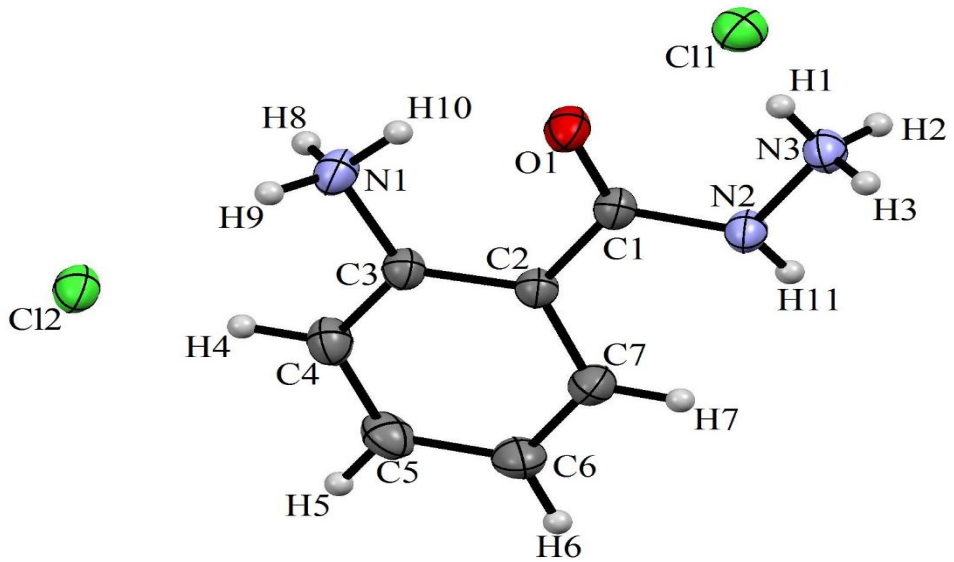


Figure1: The asymmetric unit of the title structure with the atom numbering schema. Displacement ellipsoids are drawn at the 50% probability level.

The asymmetric unit of title compound $(C_7H_{11}O\ N_3)^{2+}2Cl^-$ is formed of two entities an anionic part formed by the free chloride ions and a cationic part represented by the organic matrix protonated in the two amine functions all atoms are in a general position. The distances C-H is set at 0.93A° the distances N-H vary between 0.76A° and 0.94A° and the distances C-C vary between 1.379 (2) A° and 1.460 (2) A°and the angles C-C-C vary between 117.36° and 121.90° the angle H-N-H between 100 (2) °and 116 (2) °

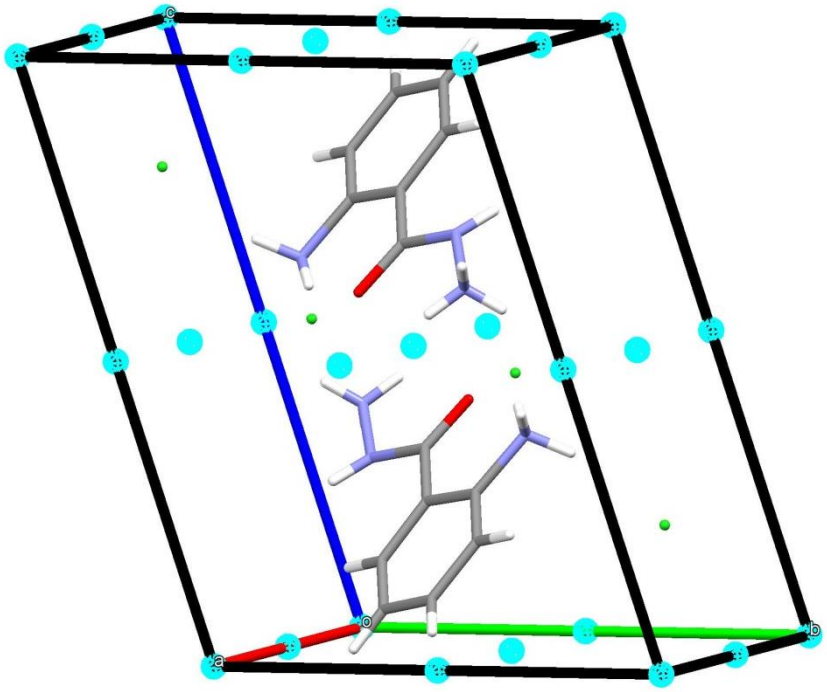


Figure2: perspective view of the $[C_7\ H_{11}\ O_1\ N_3]^{+2}$, $2Cl^-$

The cell contains two molecules of $[C_7\ H_{11}\ O_1\ N_3]^{+2}$, $2Cl^-$ which are deduced from each other by an inversion Centre relative to the space group P-1 the perspective view of the two molecules in the unit cell is shown in the figure2.

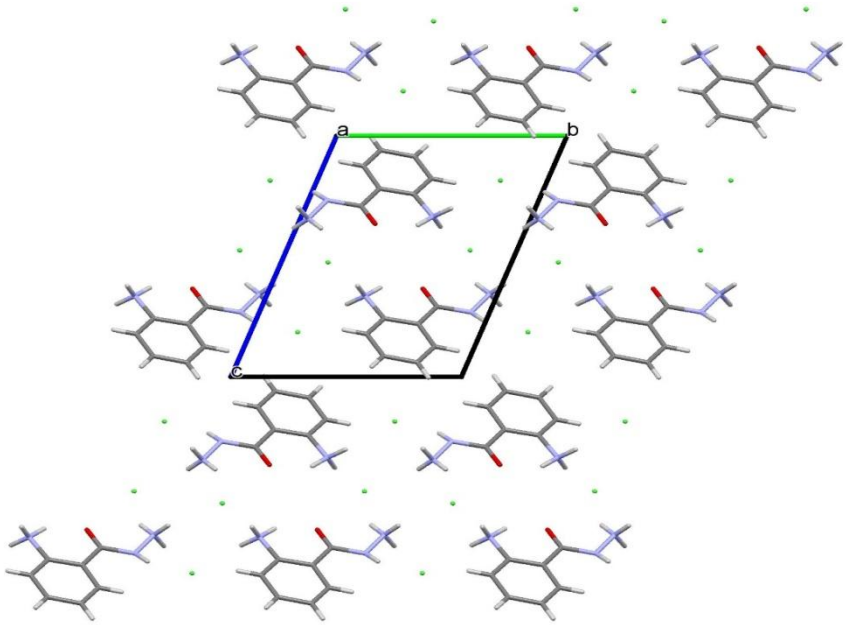


Figure3: The projection of the structure of the plane (bc) shows an alternation

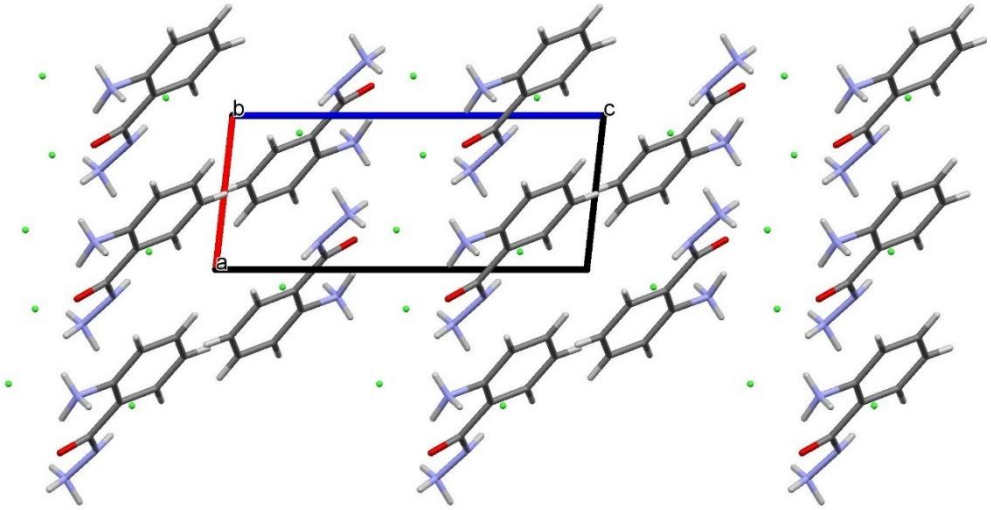


Figure4: The projection of the structure of the plane (ac) shows an alternation

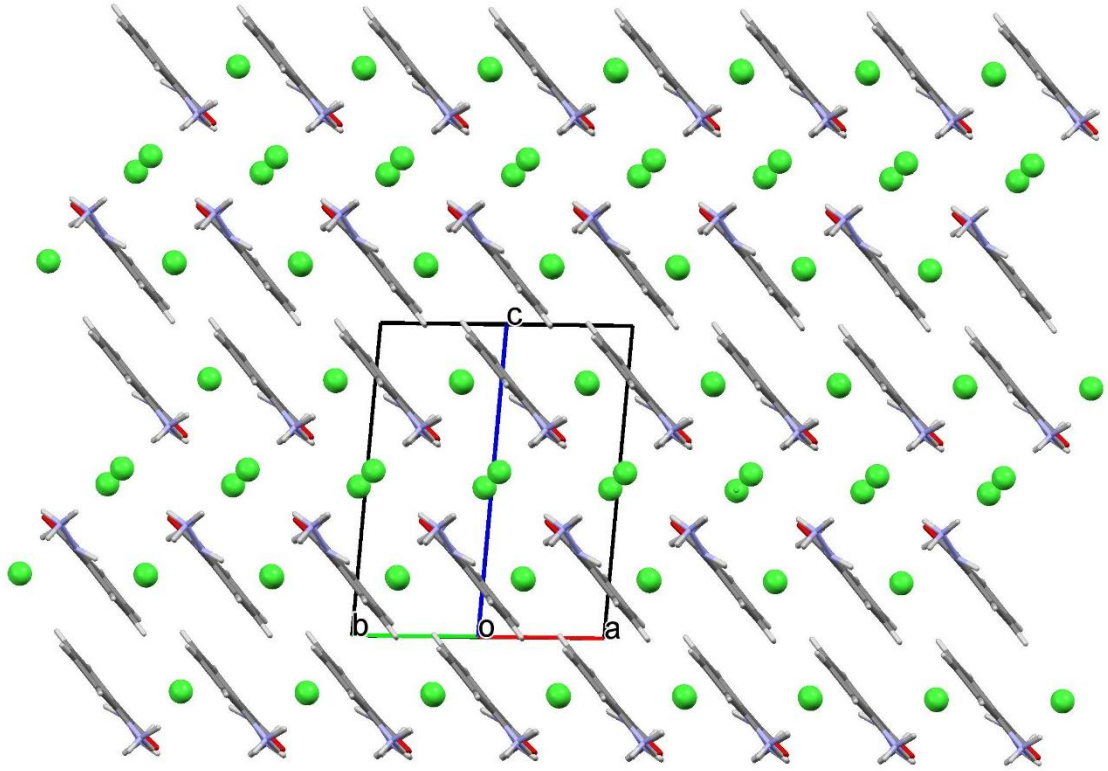


Figure5: The projection of the structure of the plane (110) shows an alternation

The projection of the structure on the plane (bc) shows an alternation of continuous and onions along the axis b, the forming layers at C=1/4 and C=3/4

alternating anions and cation-anion layers with C=0 and C=1. The layers of anions Cl^- are interposed between these double anion-cation mixed layers these layers are separated by a layer of chloride ions at C=1/2 . The view of the structure in the plane (110) allows us to confirm the arrangement of cations and anions in the network, the onion layers separated two double anion-cation layers at C=0.

The projection of the structure of the (ac), (bc) and (110) plans shows that there are not strong bonds of covalent bonding types which ensure the cohesion in the crystal between the cationic part $[C_7H_{11}ON_3]^{2+}$ and the anionic part Cl^- This brings us to think that this cohesion will be answered by the hydrogen bonding.

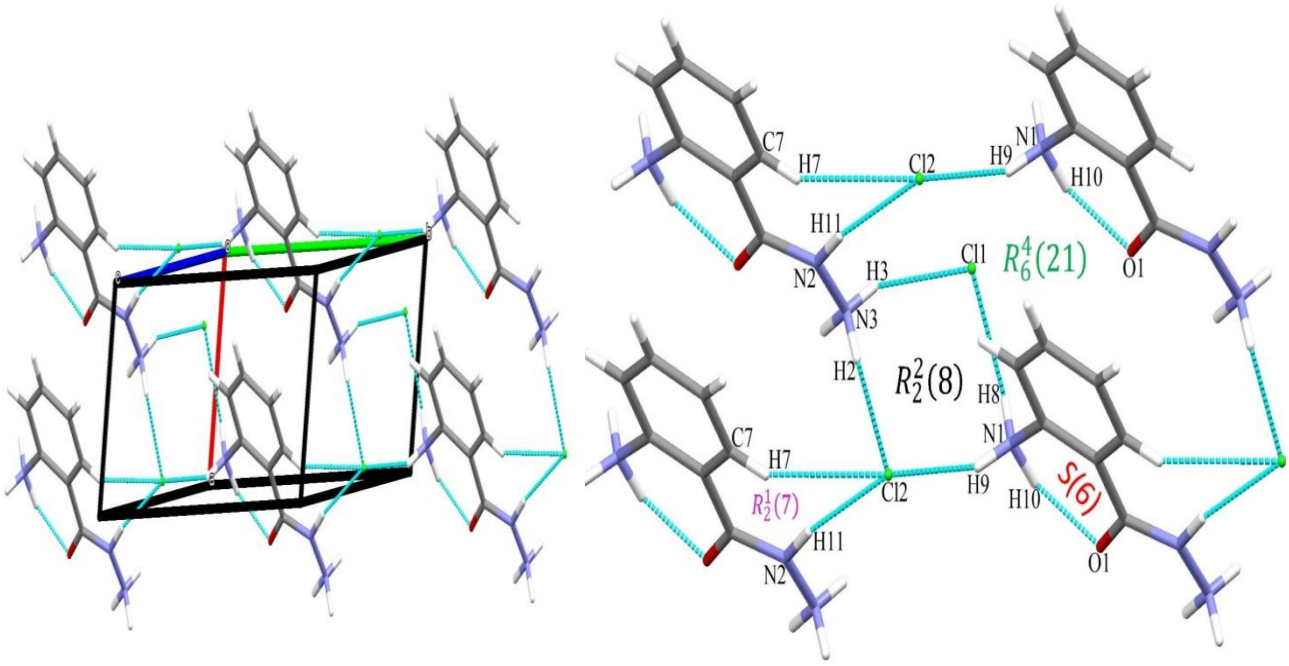


Figure6: The hydrogen bonds (dashed lines) in (I), viewed down the b axis.

Along the b the nitrogen and carbon atoms the structure links the molecule chains along the b axis by establishing intermolecular hydrogen bonds of the C-H… Cl and N-H… Cl type and form by combining a succession of cycles $\mathbf{R_2^1(7)}$,$\mathbf{R_2^2(8)}$ and $\mathbf{R_6^4(21)}$ and the interamolecular hydrogen bonds connect the nitrogen of the NH_3 group with the oxygen atom of the C=O group forming an **S (6)** rings.

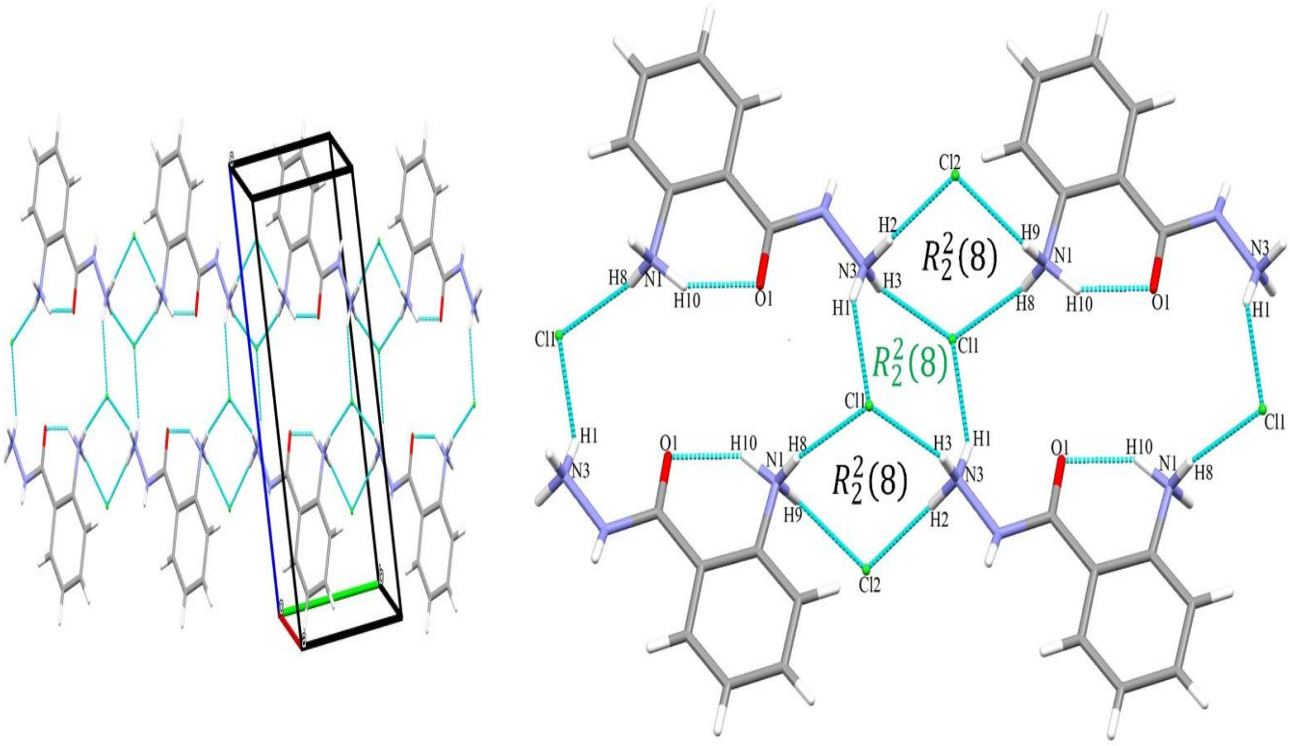


Figure6: The hydrogen bonds (dashed lines) in (I), viewed down the C axis.

The sequence of the structure along the C axis is built on the hydrogen bonds of type N-H… Cl form a succession of motifs Cycle $\mathbf{R_2^2(8)}$

EXPERIMENTAL:

Crystal Data

$C_7H_{11}N_3OCl_2$

Mr= 224.09 V=488.5(6) $Å^3$

Triclinic P-1 Z=2

a = 4.817(5)Å α=109.161(5)°

b= 9.150(5)Å β=92.209(5)°

c= 12.033(5)Å γ=101.155(5)°

Mo Kα radiation μ=0.628

T=293K 0.20×0.10×0.08

Data collection

Bruker Apex diffractometre

Measured reflections 13892 3157 reflections with I>2σ(I)

Independent reflections 3954 R_{int}=0.0215

Refinement

$R[F^2>2\sigma(F^2)]$= 0.0329 162 parameters

wR = 0.0867 H-atom parametres constrained

S= 1.052 $\Delta\rho_{min}$(e Å^{-3})=-0.26 , $\Delta\rho_{max}$(eÅ^{-3})=0.47

Table1. Hydrogen-bond geometrie (Å,°)

D—H···*A*	*D*—H (Å)	H···*A* (Å)	*D*···*A* (Å)	*D*—H···*A* (°)
N3—H1···Cl1^{i}	0.83 (2)	2.38 (2)	3.129 (4)	150 (2)
N3—H2···Cl2^{ii}	0.881 (19)	2.22 (2)	3.101 (4)	176.6 (17)
N3—H3···Cl1^{iii}	0.801 (19)	2.409 (19)	3.167 (4)	158.2 (19)
N1—H9··· Cl2^{ii}	0.90 (3)	2.35 (3)	3.190 (4)	156 (2)
N1—H8···Cl1^{iv}	0.76 (2)	2.36 (2)	3.116 (4)	169 (2)
N1—H10···O1^{i}	0.94 (3)	1.84 (3)	2.663 (3)	146 (2)
C7—H7··· Cl2^{ii}	0.93 (3)	2.90 (2)	3.703 (3)	144.95 (2)
N2—H11··Cl2 ^{v}	0.845 (19)	2.27 (2)	3.087 (3)	163.5 (18)

Codes de symétrie : **(i)** *x*−1, *y*, *z* ; **(ii)** *x*−1, *y*−1, *z* ; **(iii)** −*x*, −*y*, −*z*+1 ; **(iv)** −*x*+1, −*y*+1, −*z*+1 ; **(v)** *x*, *y*−1, *z*.

Data collection: APEX2 [7]; cell refinement : SAINT [7] ; data reduction :SAINT ; program (s) used to solve structure : SIR 2002 [7];program(s) used to refline structure : SIR2002 [8]; program(s) used to refine structure :SHELXL97 [9];molecular graphics:ORTEP-3 for Windows [10]; Software used to perapare material for

publication:WinGx[10],Mercury [11].

Tableau 1 :

Coordonnées atomiques et facteurs d'agitation thermiques isotropes

Atomes	X	Y	z	U(eq) [Å^2]
O1	−0.1922 (2)	0.33423 (11)	0.36517 (9)	0.0327 (3)
N1	0.1769 (3)	0.57221 (13)	0.33543 (11)	0.0295 (3)
N2	−0.1731 (2)	0.08128 (12)	0.26771 (10)	0.0267 (3)
N3	−0.3574 (3)	0.03423 (14)	0.34403 (11)	0.0284 (3)
C1	−0.0845 (3)	−0.0845 (3)	0.29148 (10)	0.0233 (3)
C2	0.1371 (2)	0.28668 (13)	0.21914 (10)	0.0229 (3)
C3	0.2625 (3)	0.44610 (14)	0.24218 (11)	0.0239 (3)
C4	0.4732 (3)	0.49181 (16)	0.17723 (13)	0.0304 (3)
C5	0.5602 (3)	0.37803 (18)	0.08633 (13)	0.0339 (4)
C6	0.4340 (3)	0.21873 (17)	0.05984 (13)	0.0343 (4)
C7	0.2275 (3)	0.17424 (15)	0.12581 (12)	0.0302 (3)
Cl1	0.25668 (7)	0.20293 (4)	0.52482 (3)	0.0320 (1)
Cl2	0.11469 (7)	0.79593 (4)	0.18517 (3)	0.0325 (1)

Tableau 2 :

Coordonnées des atomes d'hydrogène et facteurs d'agitation thermiques isotropes

Atomes	X	Y	z	U(iso) [Å^2]
H1	−0.430 (4)	0.107 (3)	0.3833 (18)	0.045 (5)*
H2	−0.510 (4)	−0.030 (2)	0.2985 (17)	0.035 (5)*
H3	−0.288 (4)	−0.009 (2)	0.3825 (17)	0.034 (5)*
H4	0.55565	0.59860	0.19480	0.0364*
H5	0.70254	0.40797	0.04306	0.0407*
H6	0.48895	0.14212	−0.00241	0.0411*
H7	0.14643	0.06722	0.10793	0.0363*
H8	0.310 (5)	0.623 (3)	0.3767 (19)	0.044 (6)*

H9	0.107 (5)	0.634 (3)	0.3017 (19)	0.049 (6)*
H10	0.033 (5)	0.523 (3)	0.370 (2)	0.055 (6)*
H11	−0.081 (4)	0.012 (2)	0.2347 (17)	0.042 (5)*

Tableau 3 :

Paramètres de déplacement atomique ($Å^2$)

Atomes	**$U_{(1,1)}$ou U**	**$U_{(2,2)}$**	**$U_{(3,3)}$**	**$U_{(1,2)}$**	**$U_{(1,3)}$**	**$U_{(2,3)}$**
O1	0.0365 (5)	0.0224 (4)	0.0412 (5)	0.0093 (4)	0.0168 (4)	0.0106 (4)
N1	0.0291 (5)	0.0194 (4)	0.0373 (6)	0.0040 (4)	0.0064 (4)	0.0067 (4)
N2	0.0289 (5)	0.0196 (4)	0.0328 (5)	0.0053 (4)	0.0099 (4)	0.0097 (4)
N3	0.0307 (5)	0.0228 (5)	0.0333 (5)	0.0045 (4)	0.0092 (4)	0.0120 (4)
C1	0.0228 (5)	0.0195 (4)	0.0276 (5)	0.0047 (4)	0.0027 (4)	0.0082 (4)
C2	0.0228 (5)	0.0202 (4)	0.0264 (5)	0.0048 (4)	0.0038 (4)	0.0087 (4)
C3	0.0232 (5)	0.0210 (5)	0.0288 (5)	0.0057 (4)	0.0045 (4)	0.0094 (4)
C4	0.0286 (6)	0.0265 (5)	0.0391 (7)	0.0050 (5)	0.0080 (5)	0.0155 (5)
C5	0.0326 (7)	0.0379 (7)	0.0390 (7)	0.0111 (6)	0.0145 (5)	0.0203 (6)
C6	0.0414 (7)	0.0321 (6)	0.0325 (6)	0.0130 (6)	0.0143 (5)	0.0114 (5)
C7	0.0365 (7)	0.0229 (5)	0.0305 (6)	0.0067 (5)	0.0089 (5)	0.0075 (4)
Cl1	0.0299 (2)	0.0316 (2)	0.0333 (2)	0.0064 (1)	0.0052 (1)	0.0095 (1)
Cl2	0.0353 (2)	0.0231 (2)	0.0405 (2)	0.0085 (1)	0.0097 (1)	0.0108 (1)

Tableau 4:

Paramètres géométriques :

1-Distances interatomiques **(Å)**

Atomes	**Distances (Å)**	**Atomes**	**Distances (Å)**
O1—C1	1.228 (2)	C1—C2	1.490 (2)
N1—C3	1.460(2)	C2—C3	1.397 (2)
N2—N3	1.411 (2)	C2—C7	1.401 (2)
N2—C1	1.360(2)	C3—C4	1.388 (2)

N1—H8	0.76 (2)	C4—C5	1.383 (3)
N1—H9	0.90 (3)	C5—C6	1.390 (3)
N1—H10	0.94 (3)	C6—C7	1.379 (3)
N2—H11	0.845(19)	C4—H4	0.9300
N3—H1	0.83 (2)	C5—H5	0.9300
N3—H2	0.881(19)	C6—H6	0.9300
N3—H3	0.801(19)	C7—H7	0.9300
Cl1···N3^{i}	3.129(4)	N3···O1	2.629 (3)
Cl1···C1	3.344(4)	N3···Cl1	3.376 (4)
Cl1···N2	3.384(4)	N3···Cl1^{iv}	3.167 (4)
Cl1···N3	3.376(4)	N3···Cl1^{vii}	3.129 (4)
Cl1···O1	3.465(4)	N2···H7	2.4900
Cl1···N1^{ii}	3.329(4)	C1···Cl1	3.344 (4)
Cl1···N1^{iii}	3.116 (4)	C1···C6^{vii}	3.487 (4)
Cl1···N3^{iv}	3.167(4)	C2···C5^{vii}	3.523 (4)
Cl2···N2^{v}	3.087(3)	C4···Cl2	3.526 (4)
Cl2···N3^{vi}	3.101(4)	C5···C2^{i}	3.523 (4)
Cl2···C4	3.526(4)	C6···C1^{i}	3.487 (4)
Cl2···N1	3.190(4)	C1···H10	2.39(3)
Cl1···H1^{i}	2.38 (2)	C7···H11	2.59(2)
Cl1···H10^{ii}	3.04 (3)	H1···Cl1^{vii}	2.38(2)
Cl1···H3^{iv}	2.409(19)	H1···O1	2.25(3)
Cl1···H8^{iii}	2.36 (3)	H2···Cl2^{x}	2.22(2)
Cl1···H3	3.01 (2)	H3···Cl1	3.01(2)
Cl1···H9^{ii}	2.98 (2)	H3···Cl1^{iv}	2.409 (19)
Cl2···H9	2.35 (3)	H4···Cl2	3.0600
Cl2···H11^{v}	2.27 (2)	H4···Cl2^{i}	2.9700
Cl2···H2^{vi}	2.22 (2)	H4···H8	2.4900
Cl2···H4^{vii}	2.9700	H4···H9	2.5800

Cl2···H4	3.0600	H5···Cl2^{viii}	3.0400
Cl2···H7^{v}	2.9000	H6···Cl2^{viii}	3.0900
Cl2···H6^{viii}	3.0900	H7···N2	2.4900
Cl2···H5^{viii}	3.0400	H7···Cl2^{ix}	2.9000
O1···N1	2.663(3)	H7···H11	2.0500
O1···Cl1	3.465(4)	H8···Cl1^{iii}	2.36(2)
O1···N3	2.629(3)	H8···H4	2.4900
O1···H10	1.84 (3)	H9···Cl1^{ii}	2.98(2)
O1···H1	2.25 (3)	H9···Cl2	2.35(3)
N1···Cl2	3.190(4)	H9···H4	2.5800
N1···Cl1^{ii}	3.329(4)	H10···Cl1^{ii}	3.04(3)
N1···Cl1^{iii}	3.116 (4)	H10···C1	2.39(3)
N1···O1	2.663(3)	H10···O1	1.84(3)
N2···Cl2^{ix}	3.087(3)	H11···C7	2.59(2)
N2···Cl1	3.384(4)	H11···H7	2.0500
N3···Cl2^{x}	3.101(4)	H11···Cl2^{ix}	2.27(2)

Les codes de symétries :

- (i) : x+1, y, z
- (ii) : −x, −y+1, −z+1
- (iii) : −x+1, −y+1, −z+1
- (iv) : −x, −y, −z+1
- (v) : x, y+1, z
- (vi) : x+1, y+1, z
- (vii) : x−1, y, z
- (viii) : −x+1, −y+1, −z
- (ix) : x, y−1, z
- (x) : x−1, y−1, z

2- Les angles des liaisons **(º)**

Atomes	**Angles (º)**	**Atomes**	**Angles (º)**
N3—N2—C1	116.58 (11)	C1—C2—C7	121.90 (11)
C3—N1—H10	106.8(16)	C1—C2—C3	120.74 (11)
H8—N1—H9	108 (3)	C3—C2—C7	117.36 (11)
H8—N1—H10	116 (2)	N1—C3—C4	117.01 (12)
H9—N1—H10	110 (2)	N1—C3—C2	121.43 (12)
C3—N1—H9	108.2(14)	C2—C3—C4	121.56 (12)

C3—N1—H8	108.0(18)	C3—C4—C5	119.79 (14)
C1—N2—H11	125.5(13)	C4—C5—C6	119.76 (14)
N3—N2—H11	112.3 (13)	C5—C6—C7	120.06 (14)
N2—N3—H3	113.3 (14)	C2—C7—C6	121.45 (13)
H1—N3—H2	100 (2)	C3—C4—H4	120.00
H1—N3—H3	114 (2)	C5—C4—H4	120.00
H2—N3—H3	110.5 (19)	C4—C5—H5	120.00
N2—N3—H1	112.0 (17)	C6—C5—H5	120.00
N2—N3—H2	106.2(13)	C5—C6—H6	120.00
O1—C1—N2	120.51 (12)	C7—C6—H6	120.00
O1—C1—C2	124.01 (12)	C2—C7—H7	119.00
N2—C1—C2	115.44 (11)	C6—C7—H7	119.00

3- les angles de torsion **(º)**

Atomes	**Angles (º)**
N3—N2—C1—O1	−9.75 (18)
N3—N2—C1—C2	172.76 (11)
O1—C1—C2—C3	8.11 (19)
O1—C1—C2—C7	−171.70 (13)
N2—C1—C2—C3	−174.50 (11)
N2—C1—C2—C7	5.70 (17)
C1—C2—C3—N1	−1.51 (19)
C1—C2—C3—C4	178.73 (12)
C7—C2—C3—N1	178.30 (12)
C7—C2—C3—C4	−1.46 (19)
C1—C2—C7—C6	−179.60 (13)
C3—C2—C7—C6	0.59(19)
N1—C3—C4—C5	−178.91 (13)
C2—C3—C4—C5	0.9 (2)

C3—C4—C5—C6	0.7 (2)
C4—C5—C6—C7	−1.5 (2)
C5—C6—C7—C2	0.9 (2)

4. ACKNOWLEDGEMENTS

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