

Preparation, Spectroscopy and X-Ray Structure Analysis of *N, N'*-Bis(3-nitrophenylmethylene)-cyclohexane-1,2-diamine

KODJO CHARLES GUILLAUME^{*1,2}, KANGAH NIAMEKE JEAN-BAPTISTE²,
KABLAN AHMONT LANDRY CLAUDE³, BOSSON ANTOINE KOUAME¹, KABLAN
RICHMOND JEAN FRANÇOIS⁴, ZIAO NAHOSSE² and RETAILLEAU PASCAL⁵

¹Laboratoire de Chimie Bioorganique et de Substances Naturelles, Université Nangui Abrogoua, UFR-SFA, 02 B.P. 801 Abidjan 02, Côte d'Ivoire

²Laboratoire de thermodynamique et physicochimie du milieu, Université Nangui Abrogoua, Côte d'Ivoire

³UFR des Sciences Biologiques, Université Péléforo Gon Coulibaly de Korhogo, BP 1328 Korhogo, Côte d'Ivoire

⁴Laboratoire de Cristallographie et Physique Moléculaire, UFR SSMT (Sciences des Structure et de la Matière Technologique), Université Félix Houphouët Boigny, 01 BP V34 Abidjan 01, Côte d'Ivoire

⁵Institut de Chimie des Substances Naturelles, 1 av. de la Terrasse, 91198, Gif sur Yvette
charles.kodjo@yahoo.fr

Received 19 November 2016 / Accepted 9 December 2016

Abstract: The compound *N, N'*-Bis(3-nitrophenylmethylene)cyclohexane-1,2-diamine (α -diimine Schiff base) was synthesized and characterized by MS, IR, NMR ¹H and X-ray single crystal analysis. While the cyclohexane-1,2-diamine used for the synthesis of this composite is a racemic *cis* and *trans* mixture, we observed by x-ray diffraction for the product *N, N'*-bis(3-nitrophenylmethylene) cyclohexane-1,2-diamine only the (1*R*,2*R*)*trans* enantiopure. The crystalline network cohesion of this compound is assured by a combination of C-H \cdots O and C-H \cdots N hydrogen bonds. It crystallizes in the monoclinic non-standard I2/a space group.

Keywords: Schiff base, α -Diimine, Crystal structure, Enantiopure, Hydrogen bonds, I2/a space group

Introduction

Schiff bases are formed when any primary amine reacts with an aldehyde or a ketone¹⁻⁴. Schiff bases have considerable flexible synthetic procedure; consequently a wide variety of them can be prepared. Structurally they are compounds in which the carbonyl group (C=O) has been replaced by an imine or azomethine group (C=N). The imine or azomethine groups are present in various natural derived or artificial compounds such as pigment, dyes,

catalysts *etc.*⁵⁻⁶. They are also used as intermediates in organic synthesis as well as polymers stabilizers⁷. Schiff bases are known to exhibit a broad range of biological activities including antifungal, antibacterial, antimalaria, antiproliferative, antiinflammatory, antiviral and antipyretic properties⁸⁻¹¹. The imine group present in such compounds has been shown to be critical to their biological activities^{12,13}. The present paper describes the preparation and characterization of α -diimine Schiff base.

Experimental

3-Nitrobenzaldehyde and cyclohexane-1,2-diamine were procured from Aldrich and used without further purification. All the solvents were dried before use. Melting points were determined in capillary tube using an MPD Mitamura Riken Kogyo (Japan) electrothermal melting point apparatus and are uncorrected. IR spectra in the range 4000-400 cm^{-1} were obtained on a Bruker-Vector FTIR spectrophotometer, with samples investigated as thin film from CHCl_3 solution. The ^1H NMR spectra were recorded on a Bruker-Avance-300 spectrometer, operating at 300 MHz. The mass spectra were recorded on a TOF LCT Premier (WATERS) Spectrometer coupled to an HPLC Alliance 2695 chain.

Synthesis

3-Nitrobenzaldehyde (0.4 mmol) and cyclohexane-1,2-diamine (0.2 mmol) were dissolved in ether (30 mL). At room temperature, the mixture was stirred for four days to give a pale yellow precipitate. The precipitate obtained was filtered and recrystallized in ethanol (R_f : 0.71 in hexane/acetone (50:50), yield: 80%, mp 76-77 $^\circ\text{C}$). A good quality of colorless single crystals was obtained by slow evaporation of acetone solution after 3 days.

Results and Discussion

Spectroscopy Analysis

MS study

The high resolution mass spectrum (HR-ESI-MS) of the title compound shows a peak corresponding to the molecular ion at m/z 381.1560 $[\text{M}+\text{H}]^+$, what allows to propose $\text{C}_{20}\text{H}_{20}\text{N}_4\text{O}_4$ empirical formula.

IR study

The IR spectrum shows a characteristic band at 1642 cm^{-1} and another at 1580 cm^{-1} indicating the presence of two imine functions ($\text{C}=\text{N}$) in this structure. The bands located between 2939 cm^{-1} and 2845 cm^{-1} correspond to $\nu_{\text{C-H}}$ elongation vibrations of cyclohexane fragment.

NMR study

The ^1H NMR spectrum shows one singlet peak at 8.45 ppm corresponding to the highly deblinded H_7 and H_{12} imine protons. The singlet peak at 8.28 ppm is for H_{11} and H_{16} protons isolated by nitro groups in meta position. The signals of consecutive aromatic protons H_9 , H_{10} , H_{11} on the one hand and H_{18} , H_{19} and H_{20} on the other hand, appear as an AMX type spectrum but with very small coupling constants. So that, instead of presenting doubled doublets signals, we observe rather two doublets at 8.28 ppm and 8.15 ppm and a triplet at 7.91 ppm. The multiplet at 3.49 ppm is associated to H_1 and H_2 protons. The multiplet signal at 2.15 ppm corresponds to the two protons in equatorial position fixed on C_3 and C_6 carbon atoms. The integration curve of the multiplet at 1.90 ppm indicates that it contains four protons: H_3 and H_6 in axial position and H_4 and H_5 in equatorial position. The last multiplet at 1.51 ppm signals H_4 and H_5 protons in axial position. For a given carbon atom, proton in equatorial position is more deblinded than in axial position one¹⁴.

X-ray analysis

The title compound crystallizes in the non-standard I2/a space group of monoclinic system. Indeed, structure has been solved and refined in C2/c centrosymmetric space group. However, since refining of the structure in non-standard space group I2/a leads to the same values for R but to GooF value (0.92) much closer to ideal value 1, we preferred for this compound the non-standard space group I2/a.

While the cyclohexane α -diamine used for synthesis is a *cis* and *trans* racemic mixture, asymmetric unit of the title compound is constituted by an enantiopure monomer represented in Figure 1. Equatorial orientations of the two substituents in position 1 and 2 on cyclohexane indicate that it is *trans* isomer. According to conventional stereo descriptors¹⁵, the absolute configurations of the asymmetric carbons C₁* and C₂* are 1R and 2R. The crystallization of this compound is therefore carried out according to a spontaneous resolution and produces a mechanical mixture of crystals. We subsequently selected randomly and resolved the *trans* (1R, 2R) isomer structure. The *trans* (1S, 2S) enantiomer as well as *cis* isomers are probably present in the mixture obtained after crystallization.

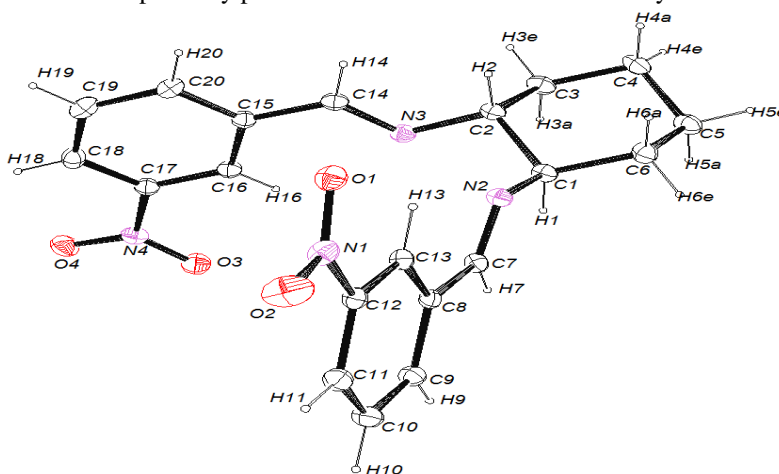


Figure 1. Molecular structure of *N,N'*-bis(3-nitrophenylmethylene)cyclohexane-1,2-diamine with atomic numbering scheme. (Displacement ellipsoids are drawn at 50% probability level)

The data given in the Table 1 in which selected geometrical parameters are grouped, shows similarity of carbon-carbon bonds of aromatic rings of this molecule. With an average value of about 1.381 Å, these links conform to the values proposed by Allen *et al.*,¹⁶. The valence angles associated to these aromatic rings do not undergo major deformations and have values around 120° characteristic of benzene ring. The lengths of N₁-O₁ bonds [1.217 (4) Å] and N₁-O₂ [1.202 (4) Å] on the one hand and N₄-O₄ [1.222 (4) Å] and N₄-O₃ [1.226 (4) Å] on the other hand, describe N=O double bonds and show the similarity of all these bonds. This result illustrates the delocalization of the free pair of electron along the two O-N-O bonds.

Like many Schiff bases¹⁷⁻²⁰, the two azomethine functions present in the structure of title compound are significantly shorter than value suggested by literature¹⁶. Indeed, with lengths of 1,259(4) Å for N₂ = C₇ and 1,272(4) Å for N₃ = C₁₄, azomethine functions of this compound are 6.74% and 5.77% shorter than value proposed by Allen *et al.*,¹⁶.

The Table 1 shows that the valence angles $C_1-N_2=C_7$ [$117.3(3)^\circ$], $N_2=C_7-C_8$ [$119.7(3)^\circ$], $C_2-N_3=C_{14}$ [$118.5(3)^\circ$] and $N_3=C_{14}-C_{15}$ [$123.5(4)^\circ$] in which atoms of imine functions are involved are congruent with their SP^2 hybridization characters.

The values of C-C-C valence angles between 109.55° and 112.6° observed for cyclohexane attest that the structure adopts non-plane conformation in which carbon atoms respect their SP^3 hybridizations. The fact that the sum of torsion angles $C_7-N_2-C_1-C_2$ and $C_7-N_2-C_1-C_6$ on the one hand and $C_{14}-N_3-C_2-C_1$ and $C_{14}-N_3-C_2-C_3$ on the other hand is not 0° but respectively about 13° and 8° is further proof of the non-planarity of cyclohexane ring.

Table 1. Selected bond lengths and angles (\AA , $^\circ$)

C_5-C_6	1.510 (5)	$C_6-C_5-C_4$	111.7 (4)
C_5-C_4	1.511 (6)	$C_5-C_4-C_3$	109.55(4)
C_4-C_3	1.523 (5)	$C_4-C_3-C_2$	112.6 (4)
C_2-C_3	1.522 (4)	$C_3-C_2-C_1$	110.7 (3)
C_2-C_1	1.530 (5)	$C_2-C_1-C_6$	110.0 (3)
C_1-C_6	1.518 (5)	$C_1-C_6-C_5$	112.6 (3)
C_1-N_2	1.452 (4)	$C_1-N_2=C_7$	117.3(3)
$N_2=C_7$	1.259 (4)	$N_2=C_7-C_8$	119.7 (3)
C_7-C_8	1.474 (4)	$C_8-C_{13}-C_{12}$	119.6 (3)
C_8-C_9	1.404 (4)	$C_{13}-C_{12}-C_{11}$	122.7 (4)
C_9-C_{10}	1.372 (5)	$C_{12}-C_{11}-C_{10}$	118.4 (4)
$C_{10}-C_{11}$	1.370 (5)	$C_{11}-C_{10}-C_9$	120.0 (4)
$C_{11}-C_{12}$	1.383 (5)	$C_9-C_8-C_{13}$	117.8 (4)
$C_{12}-C_{13}$	1.358 (4)	$O_2-N_1-O_1$	123.5 (4)
$C_{13}-C_8$	1.387 (4)	$C_2-N_3=C_{14}$	118.5 (3)
$C_{12}-N_1$	1.487 (4)	$N_3=C_{14}-C_{15}$	123.5 (4)
N_1-O_1	1.217 (4)	$C_{18}-C_{19}-C_{20}$	120.2 (4)
N_1-O_2	1.202 (4)	$C_{19}-C_{20}-C_{15}$	120.6 (4)
C_2-N_3	1.472 (4)	$C_{20}-C_{15}-C_{16}$	119.3 (4)
$N_3=C_{14}$	1.272 (4)	$C_{16}-C_{17}-C_{18}$	121.9 (4)
$C_{14}-C_{15}$	1.459 (5)	$O_4-N_4-O_3$	121.9 (4)
$C_{15}-C_{16}$	1.401 (4)	$C_7-N_2-C_1-C_2$	126.0(4)
$C_{17}-C_{18}$	1.386 (5)	$C_{14}-N_3-C_2-C_1$	123.4(3)
$C_{18}-C_{19}$	1.384 (5)	$C_7-N_2-C_1-C_6$	-112.8(3)
$C_{19}-C_{20}$	1.381 (5)	$C_{14}-N_3-C_2-C_3$	-115.3(3)
$C_{20}-C_{15}$	1.387 (5)	$N_2-C_7-C_8-C_9$	-174.7 (3)
$C_{17}-N_4$	1.459 (5)	$N_3-C_{14}-C_{15}-C_{20}$	170.7 (3)
N_4-O_4	1.222 (4)	$N_2-C_7-C_8-C_{13}$	4.5 (5)
N_4-O_3	1.226 (4)	$N_3-C_{14}-C_{15}-C_{16}$	-4.2 (6)

Conformational analysis of the structure obtained clearly shows that the hexanic cycle adopts chair conformation more stable than the boat form²¹⁻²³.

In order to analyze relative orientations of the different parts of the molecule, various mean planes are defined and recorded in Table 2.

And Table 3 shows the different dihedral angles between these mean planes thus defined.

Table 2. Mean planes

Plane	Atoms defining each plane
I	C ₂ , C ₃ , C ₅ , C ₆
II	C ₈ , C ₉ , C ₁₀ , C ₁₁ , C ₁₂ , C ₁₃
III	C ₁ , N ₂ , C ₇ , C ₈
IV	C ₂ , N ₃ , C ₁₄ , C ₁₅
V	C ₁₅ , C ₁₆ , C ₁₇ , C ₁₈ , C ₁₉ , C ₂₀

Table 3. Dihedral angles between these mean planes(°)

	II	III	IV	V
I	80	85	79	75
II	0	5	69	63
IV	69	65	0	4
V	63	59	4	0

With a dihedral angle of 85°, plane **III** is practically perpendicular to the plane **I**. With an angle of 75°, plane **V** is that which deviates most from the median plane of cyclohexane chair plate. While the two planes **III** and **IV** which connect the two aromatic rings of the structure to cyclohexane describe 65° angle, aromatic rings tend to approach and define between them a slightly smaller angle of 63°. It is probably in order to avoid an important steric hindrance caused by this slight approximation of the two aromatic rings that the two nitro groups substituted on these benzene rings position themselves anti-mutually to the farthest possible.

The smallness of the angles between planes **II** and **III** (5 °) on the one hand and planes **IV** and **V** (4°) on the other, illustrates the co-planarity of each aromatic nucleus with central plane containing azomethine function. The main crystal parameters of the structure are shown in Table 4.

Table 4. Crystal data collection and structure refinement parameters

Empirical formula: C ₂₀ H ₂₀ N ₄ O ₄	10212 observed reflexions
Formula weight: 380,40	3423 independante reflexions
Temperature : 293K	254 parameters, F(0 0 0) : 1600
X-ray: Cu K α ; λ = 1,54197Å	Index ranges (h, k, l) -17~18 ; -19~18 ; -19~11
Crystal system: Monoclinic	Theta range for data collection: 3.9° ≤ θ ≤ 68.2°
Space group : I2/a	Absorption coefficient (μ) : 0.760 (mm-1)
a = 15.498 (5)Å	Goodness of fit : 0.92
b = 15.844 (5)Å	Refinement on F ²
c = 16.598 (5)Å	Final R indices [I > 2 σ (I)]: R1 = 0.065 wR2 = 0.1450
β = 106.14(3)°	R indices (all data) : R1 = 0.227 wR2 = 0.1607
V = 3915(2)Å ³	Maximum; minimum $\Delta\rho$ (eÅ ⁻³) : 0.17; -0.19
D _{calc} = 1.291Mg m ⁻³	Enraf-Nonius CAD-4 diffractometer
Z = 8	Structure determination: SHELXS 97

The analysis of the crystalline stacking allowed us to know the environment of each molecule inside the crystal. All intermolecular interactions contributing to stabilization of crystal network are compiled in Table 5.

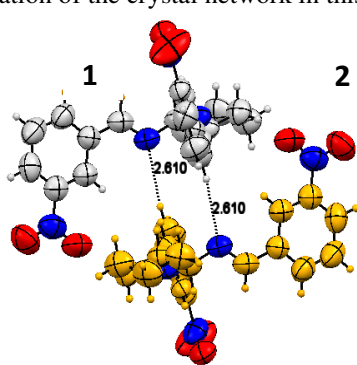
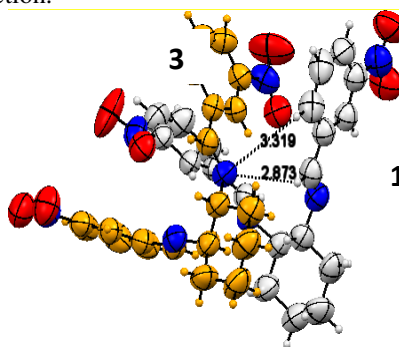
Table 5. Selected intermolecular interactions (Å, °)

$D-H\cdots A$	$D-H$	$H\cdots A$	$D\cdots A$	$D-H\cdots A$
$C_9-H_9\cdots N_3^i$	0.93	2.610	3.520	166.51
$C_{20}-H_{20}\cdots N_2^{ii}$	0.93	3.319	3.995	131.35
$C_{14}-H_{14}\cdots N_2^{ii}$	0.93	2.873	3.590	134.79
$C_7-H_7\cdots O_1^{iii}$	0.93	2.701	3.583	158.68
$C_1-H_1\cdots O_1^{iii}$	0.979	2.837	3.736	152.86
$C_5-H_5\cdots O_2^{iii}$	0.97	2.639	3.453	141.72
$C_1-H_1\cdots O_2^{iii}$	0.979	3.112	3.280	130.36
$C_5-H_5\cdots O_3^{iiii}$	0.97	2.906	3.682	137.87
$C_4-H_4\cdots O_3^{iiii}$	0.97	3.343	3.739	106.74

Codes de symétrie: (i) $\frac{1}{2}-x, \frac{1}{2}-y, \frac{1}{2}-z$; (ii) $-x, \frac{1}{2}-y, z$; (iii) $x, \frac{1}{2}+y, -z$; (iiii) $\frac{1}{2}+x, y, \frac{1}{2}-z$

In the centrosymmetric dimer consisting of the molecules **1** (x, y, z) and **2** ($\frac{1}{2}-x, \frac{1}{2}-y, \frac{1}{2}-z$), the hydrogen atom H_9 of one forms a bond with the nitrogen N_3 of the other (Figure 2). With a length of 2.61Å and linearity angle of 166.51° this interaction is conform to weak hydrogen bond²⁴.

The molecule **3** ($-x, \frac{1}{2}-y, z$) symmetric of molecule **1** with regard to the C2 rotation axis parallel to $[0, 0, 1]$ direction, establishes with its N_2 atom a three-center hydrogen bonds with H_{14} and H_{20} hydrogen atoms as illustrated in Figure 3. Participating thus in the stabilization of the crystal network in this direction.

**Figure 2.** Intermolecular interaction between 1 and 2**Figure 3.** Three-center interaction between 1 and 3

The Figure 4 shows that O_1 and O_2 oxygen atoms of molecule **4** ($x, \frac{1}{2} + y, -z$) symmetric of **1** with regard to the glide plane b perpendicular to the direction $[0\ 0\ 1]$ also establish three-center type hydrogen bonds with respectively hydrogen atoms H_1 and H_7 and then H_1 and H_5 of the molecule **1**. Since the hydrogen atom H_1 is moreover also engaged in a three-center interaction with the O_1 and O_2 atoms, this hydrogen bonding series describes W non-bonding interactions.

In view of all these intermolecular interactions described above, it is therefore easy to realize that the molecules inside the crystal are held in a cluster around C2 rotation axis parallel to c crystallographic axis in infinite chain as illustrated in Figure 5. Within each cluster, the cohesion is then ensured by $C-H\cdots O$ and $C-H\cdots N$ type hydrogen bonds.

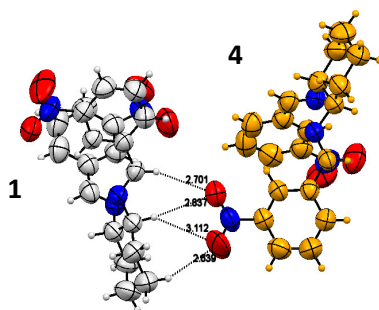


Figure 4. W intermolecular interaction between **1** and **4**

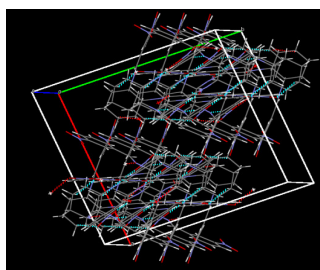


Figure 5. View of crystal network showing two clusters of molecules linking into a chain along [0 0 1]

The cohesion between the different clusters is essentially ensured by another C-H \cdots O type hydrogen bond as illustrated in Figure 6 in addition to classic Van der Waals type interactions. Indeed, with its H₄ and H₅ atoms, the molecule **5** ($\frac{1}{2} + x, \frac{1}{2} - z$) symmetric of the molecule **1** compared with the glide plane **a** perpendicular to [001] direction and positioned at [0 0 $\frac{1}{4}$], housed in the neighboring cluster, interacts with oxygen atom O₃ under tree-center type interaction.

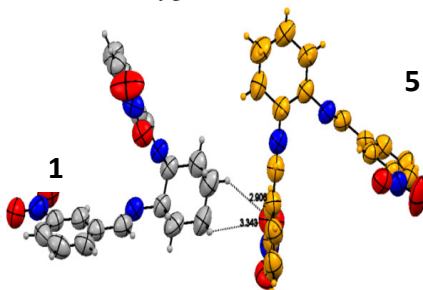


Figure 6. Three-center interaction between **1** and **5**

Conclusion

The title compound was easily synthesized by stirring the reaction mixture at room temperature and characterized by conventional spectrometry methods (NMR, IR HR-ESI-MS). The single crystal x-ray diffraction characterization showed that this compound, which crystallizes in non-standard I2/a monoclinic space group is a pure *trans*-(1R 2R) enantiomer, in contrast to racemic *cis* and *trans* cyclohexane α -diamine mixture used for the synthesis. The three-dimensional organization of the molecules inside the crystal takes place around each C2 rotational symmetry axis parallel to c crystallographic axis. Molecules are bound in cluster along these axes in infinite chain. In addition to classic Van der waals forces, crystalline network cohesion of this compound is assured by a combinaison of C-H \cdots O and C-H \cdots N hydrogen bounds.

Supplementary material

Crystallographic data for the structural analysis have been deposited at the Cambridge Crystallographic Data Centre, CCDC No. **1494923**. Copies of this information may be obtained free of charge from the Director, CCDC, 12 Union Road, Cambridge, CB21E Z, UK (Fax: +44-1223-336033; email: deposit@ccdc.cam.ac.uk or <http://www.ccdc.cam.ac.uk>).

Reference

1. Vogel A J, *Practical Organic Chemistry*, London, Longmans, 1966, 653
2. March J, *Advanced Organic Chemistry*, Third Edition, 1985, 796
3. Vollhardt K P C, *Organic Chemistry*, International Student Edition, 1987, 643
4. Da Silva C M, Da Silva D L, Modolo L V, Alves R B, De Resende M A, Martins C V B and De Fatima A, *J Adv Res.*, 2011, **2**(1), 1-8; DOI:10.1016/j.jare.2010.05.004
5. Bringmann G, Dreyer M, Faber J H, Dalsgaard P W, Staerk D, Jaroszewski J W, Henry N D, Frank M B, Reto B and Søren B C, *J Nat Prod.*, 2004, **67**(5), 743-748; DOI:10.1021/np0340549
6. De Souza A O, Galetti F C S, Silva C L, Bicalho B, Parma M M, Fonseca S F, Marsaioli A J, Trindade A C L B, Gil R P F and Bezerra F S, *Quim Nova.*, 2007, **30**(7), 1563-1566.
7. Dhar D N and Taploo C L, *J Sci Ind Res.*, 1982, **41**(8), 501-506.
8. Sawada H, Yanagida K, Inaba Y, Sugiyama M, Kawase T and Tomita T, *Eur Polym J.*, 2001, **37**(7), 1433-1439; DOI:10.1016/S0014-3057(01)00005-2
9. Ma D Y, Zhang L X, Rao X Y, Wu T L, Li D H and Xie X Q, *J Coord Chem.*, 2013, **66**(9), 1486-1496; DOI:10.1080/00958972.2013.783699
10. Siddiqui J I, Iqbal A, Ahmad S and Weaver G W, *Molecules*, 2006, **11**(2), 206-211; DOI:10.3390/11020206
11. Dao V T, Gaspard C, Mayer M, Werner G H, Nguyen S N and Michelot R J, *Eur J Med Chem.*, 2000, **35**(9), 805-813; DOI:10.1016/S0223-5234(00)00165-3
12. Guo Z, Xing R, Liu S, Zhong Z, Ji X, Wang L, et al. *Carbohydr Res.*, 2007, **342**(10), 1329-1332; DOI:10.1016/j.carres.2007.04.006
13. Kumar S, Dhar N D and Saxena N P, *J Sci Ind Res.*, 2009, **68**, 181-187.
14. Silverstein R M, Kiemle D J and Webster F X, *Identification Spectrométrie de composés Organiques*, De Boeck Supérieur, 3th Ed., (in)organic chemistry collection, 2016, 175.
15. Aloui F, El Abed R, Marinetti A, Ben Hassine B, *Com Rend Chimie*, 2009, **12**(1-2), 284-290; DOI:10.1016/j.crci.2008.01.009
16. Allen F H, Kennard O and Wattson D G, *J Chem Soc Perkin Trans.*, 1987, **2**, S1-S19; DOI:10.1039/P29870000051
17. Glidewell C, Low J N, Skakle J M S and Wardell J L, *Acta Cryst.*, 2005, **E61**, o1699-o1701; DOI:10.1107/S1600536805014248
18. Dutta S and Chakravorty A, *Polyhedron*, 1994, **13**, 1811-1816; DOI:10.1016/0277-5387(94)80002-2
19. Elbayoumi M A, El-Asser M and Halin F A, *J Ann Chem Soc.*, 1971, **93**(3), 586-590; DOI:10.1021/ja00732a004
20. Elmali A, Kabak M and Elerman Y, *J Mole Struct.*, 1999, **484**(1-3), 229-234; DOI:10.1016/S0022-2860(98)00897-7
21. Rebek J J, Marshall L, Wolak R, Parris K, Killoran M, Askew B, Nemeth D and Islam N, *J Am Chem Soc.*, 1985, **107**(25), 7476-7481; DOI:10.1021/ja00311a042
22. Thuery P, Nierlich M, Baldwin B, Aoki Y and Hirose T, *J Chem Soc Perkin Trans.*, 1999, **2**, 2077-2086; DOI:10.1039/A903816G
23. Remmert S and Parish C, *J Comput Chem.*, 2009, **30**(6), 992-998; DOI:10.1002/jcc.21125
24. Jeffrey A G, *An Introduction to Hydrogen Bond*, Oxford University Press, Inc., 1997.