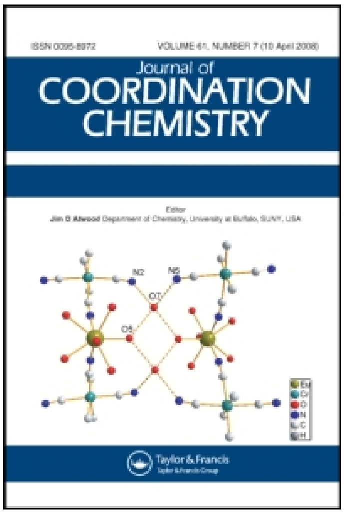

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Coordination Behavior of Sulfathiazole: Crystal Structure of $[Cu(en)_2(OH_2)_2][Sulfathiazole]_2 \cdot 2H_2O$ (en = ethylenediamine): Antibacterial activity

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COORDINATION BEHAVIOR OF SULFATHIAZOLE: CRYSTAL STRUCTURE OF $[Cu(en)_2(OH_2)_2][SULFATHIAZOLE]_2 \cdot 2H_2O$ (en = ETHYLENEDIAMINE): ANTIBACTERIAL ACTIVITY

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The preparation, spectroscopic, magnetic properties and crystal structure of $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ (en = ethylenediamine; stz stands for the deprotonated form of sulfathiazole, 4-amino-N-2-thiazolylbenzenosulfonamide) are reported. The complex crystallizes in the triclinic system, space group P1, with cell constants $a = 8.321(1)$ Å, $b = 7.441(1)$ Å, $c = 13.627(3)$ Å, $\alpha = 86.02(1)°$, $\beta = 101.81(1)°$, $\gamma = 97.88(1)°$ and Z = 1. The metal ion is six coordinate by four *N* atoms of two ethylenediamine ligands in the basal plane and two O atoms of two water molecules in the axial positions. The stereochemistry of the metal center is irregular and can be considered a distorted octahedron. Two deprotonated sulfathiazole molecules are present in the complex and a center of symmetry is located at the copper(II) ion. This is the first crystallographic evidence of sulfathiazole acting as a counter ion. The susceptibility of certain strains of bacterium towards the complex was measured, it shows good antibacterial activity against *Salmonella typhimurium* strain.

Keywords: Copper; Antibacterial; Ethylenediamine; Sulfathiazole

INTRODUCTION

Sulfanilamide derivatives are of considerable value in chemotherapy due to their antibacterial activity. The complex formation between metal ions and sulfa drugs have been extensively studied [1–5] with a view to establish the relationship and importance of metal-drug interactions. Bult [6] indicates that the literature on the chemistry of the metal sulfonamide derivatives, especially the structure determinations, is often incomplete and conflicting. In fact, the only crystal structures of the sulfathiazole complexes have been recently reported by Casanova [7–10]. The crystal structure of the $[Zn(stz)_2 \cdot H_2O]$ complex shows the drug acting as a bridging ligand that interacts through the N_{amino} and the $N_{thiazole}$ atoms [7], while in $[Cu(stz)py_3Cl]$ the drug behaves as a monodentate ligand binding to the metal ion via the $N_{thiazole}$ atom only [8]. These results seem

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to suggest that the different behavior of the ligand depends on its deprotonation and/or the metal ion, and it is difficult to give a generally valid account of the structures of other metal-sulfonamides. The preparation properties and crystal structure of $[Cu(Hstz)_2(EtOH)Cl_2]$ [9] and $[Cu(stz)_2(Him)_2 \cdot MeOH$ [10] were also published and the behavior of the sulfathiazole is as a mono or bidentate ligand respectively.

As a consequence, three types of behavior of the sulfathiazole have been observed; in neutral form, it is monodentate through the N_{amino} atom, when deprotonated, it can act as a monodentate or bidentate ligand. In the present work we report the synthesis and spectroscopic studies of $[Cu(en)_2(OH_2)_2][stz)]_2 \cdot 2H_2O$ where the sulfathiazole behaves as a deprotonated counter ion.

EXPERIMENTAL

Physical Methods

Fourier transform infrared (FTIR) spectra of the sulfathiazole ligand and its metal complex as KBr pellets were recorded in the spectral range 4000–400 cm^{-1} with a Perkin Elmer Series 2000 apparatus. FTIR spectra as polyethylene pellets were registered between 450–120 cm^{-1} by using a Brucker IFS 66V spectrophotometer. Spectra were scanned with resolution of 2 cm^{-1}. Two hundred scans were accumulated for both the mid and far FTIR measurements. EPR spectra were recorded on a Brucker ECS 106 spectrometer by the X-band method (9.76 GHz). α-α'-Diphenyl-β-picrylhydrazide free radical was used as the g marker. UV–Visible spectra were recorded using a Perkin Elmer recording spectrometer. Carbon, nitrogen and hydrogen microanalysis were done by the microlabs in the Venezuelan Institute of Scientific Research (IVIC). Magnetic susceptibility was measured on a Johnson Matthey Susceptibility Balance at room temperature using $HgCo(NCS)_4$ as calibrant.

Antibacterial Activity Test

In vitro antibacterial activity of the ligand and the complexes were tested using the paper disc diffusion method [11]. The chosen strains include G(+) *S. aureus* and *B. cereus* and G(−) *P. aeruginosa*, *S. typhimurium* and *E. coli*. The liquid medium containing the bacterial subcultures was autoclaved for 20 min at 15 lb pressure before inoculation. The bacteria were cultured for 24 h at 36°C in an incubator. Mueller Hinton broth was used for preparing basal media for the bioassay of the organisms. Nutrient agar was poured on a plate and allowed to solidify. The test compounds in DMSO solutions were added dropwise on a 10 mm diameter filter paper disc placed in the center of the agar plates. The plates were then kept at 5°C for 1 h and transferred to an incubator maintained at 36°C. The width of the growth inhibition zone around the disc was measured after 24 h of incubation. Four replicates were taken for each treatment.

MATERIALS AND METHODS

All reactants and solvents were analytical grade where possible, purchased from Aldrich and used without further purification. All solvents were distilled by standard techniques before use. Copper(II) dichloride (1 mmol) was added to a solution of sulfathiazole (2 mmol) in methanol (50 mL). To the resulting yellowish mixture, (10 mmol) of ethylenediamine was also added. A blue precipitate was obtained immediately which was filtered out and the filtrate was left standing at room temperature. After a few days, blue crystals were obtained from this filtrate. Anal. calcd. for $C_{22}H_{42}N_{10}O_8S_4Cu$(%): C, 34.54; H, 5.23; N, 18.32: S, 16.75. Found: C, 34.58; H, 5.13; N, 18.61; S, 17.12.

Crystal Structure Determination of $[Cu(en)_2(OH_2)_2]$ $[stz]_2 \cdot 2H_2O$

For structural determination a single crystal of $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ with approximate dimensions $0.1 \times 0.2 \times 0.4$ mm was selected under the microscope using polarized light and mounted in a sealed glass capillary containing a small amount of the mother liquor. Preliminary oscillation and Weissenberg photographs allowed determination of approximate cell parameters and possible space group. The intensities of 3719 reflections were collected at 293(2) K in the θ–2θ mode on a Nicolet P3/F diffractometer automated by Crystal Logic Inc., using monochromated Mo Kα radiation ($\lambda = 0.7107$ Å) Unit cell parameters were calculated from the least-squares fitting of the angular settings of 36 reflections with $10° \leq 2\theta \leq 20°$. Three check reflections (− 1 − 12), (003) and (− 114) were measured at intervals of 97 reflections in order to monitor crystal stability and alignment. No significant variation was detected in the intensities of the standards.

The structure was solved by direct methods with SHELXS-97 [12]. The positions for the hydrogen atoms were obtained in successive difference Fourier maps with SHELXL-97 [13]. The refinement of atomic coordinates and anisotropic thermal parameters was carried out by full matrix least squares techniques on F^2 with SHELXL-97. The hydrogen atoms of the ligand were included in geometrically calculated positions and refined using a riding model maintaining the C–H distances at 0.93 and 0.97 Å for the aromatic and secondary-NH_2 groups, respectively. They were assigned isotropic thermal parameters equal to 1.5 times the Ueq of the carbon atom to which they are attached. The details of the data collection procedure and the structure refinement results are summarized in Table I. Selected bond lengths and angles are summarized in Table II.

RESULTS AND DISCUSSION

The complex appears to be air stable, soluble in DMSO and slightly soluble in CH_3CN. The experimental conductivity value of a 10^{-3} M solution in DMSO at room temperature, falls in the range of a 2 : 1 electrolyte [14]. Thermal gravimetric analysis of the complex shows a mass loss equivalent to two water molecules at 150°C, which means that these water molecules are coordinated to the metal.

In the infrared spectrum of the sulfathiazole, changes in frequency and intensity are observed when complexed and show similarities with the infrared spectrum of the complex which has characteristic bands of the sulfathiazole ligand. The bands of 3396 and 3386 cm^{-1}, assigned to the ν_{asym} (N–H) and ν_{sym} (N–H) vibrations of the NH_2 group are shifted with respect to those of the ligand (3350 and 3320 cm^{-1}). As expected, the vibrational bands due to $\nu(SO_2)_{asym}$ (1328 cm^{-1}), $\nu_{sym}(SO_2)$ (1138 cm^{-1}), and scissors and wagging SO_2 (570 and 550 cm^{-1}) remain unchanged. There is no change of the

TABLE I Crystal data and structure refinement for $CuC_{22}H_{40}N_{10}O_8S_4$

Empirical formula	$CuC_{22}H_{40}N_{10}O_8S_4$
Formula weight	764.42
Temperature	293(2) K
Wavelength	0.71070 Å
Crystal system	Triclinic
Space group	PI
Unit cell dimensions	$A=8.321(1)$ Å, $\alpha=86.02(1)^\circ$
	$B=7.441(1)$ Å, $\beta=101.81(1)^\circ$
	$C=13.627(3)$ Å, $\gamma=97.887(1)^\circ$
Volume	817.3(2) Å^3
Z	1
Calculated density	1.533 mg/m^3
Absorption coefficient	0.984 mm^{-1}
F(000)	399
Crystal size	$0.1 \times 0.2 \times 0.4$ mm
Theta range for data collection	1.53°–25.00°
Index ranges	$-9 \le h \le 9$
	$-8 \le k \le 8$
	$-9 \le l \le 16$
Reflections collected/unique	3719/2871 [$R_{\text{int}}=0.0619$]
Completeness to $2\theta=25.000^\circ$	99.9%
Refinement method	Full-matrix least-squares on F^2
Data/restraints/parameters	2871/0/221
Goodness-of-fit on F^2	1.029
Final R indices [$I > 2\sigma(I)$]	$R_1=0.0525$, $wR_2=0.1328$
R indices (all data)	$R_1=0.0674$, $wR_2=0.1434$
Largest diff. Peak and hole	0.520 and -0.800 e. A^{-3}

TABLE II Selected bond lengths [Å] and angles [°] for $CuC_{22}H_{40}N_{10}O_8S_4$[a]

Cu–N(5)	2.033(3)	C(2)–S(1)#1	1.735(4)
Cu–N(5)#1	2.033(3)	C(3)–N(1)	1.377(6)
Cu–N(4)	2.042(3)	N(2)–S(2)#1	1.578(3)
Cu–N(4)#1	2.042(3)	S(2)–O(1)#1	1.451(3)
O(4)–HOA	0.84(9)	S(2)–O(2)#1	1.454(3)
O(4)–HOB	0.70(6)	S(2)–N(2)#1	1.578(3)
O(3)–HOC	0.87(10)	S(2)–C(4)#1	1.761(4)
O(3)–HOD	0.68(9)	O(1)–S(2)#1	1.451(3)
S(1)–C(2)#1	1.735(4)	O(2)–S(2)#1	1.454(3)
S(1)–C(1)#1	1.753(4)	C(4)–S(2)#1	1.761(4)
C(1)–N(1)	1.304(5)	C(7)–N(3)	1.369(5)
C(1)–N(2)	1.366(5)	N(4)–C(10)	1.456(6)
C(1)–S(1)#1	1.753(4)	C(11)–N(5)	1.480(6)
C(2)–C(3)	1.316(7)	C(10)–C(11)	1.441(8)
N(5)–Cu–N(5)#1	180.0	C(1)–N(1)–C(3)	111.3(4)
N(5)–Cu–N(4)	84.13(14)	C(1)–N(2)–S(2)#1	121.4(3)
N(5)#1–Cu–N(4)	95.87(14)	O(1)#1–S(2)–O(2)#1	114.47(19)
N(5)–Cu–N(4)#1	95.87(14)	O(1)#1–S(2)–N(2)#1	113.97(17)
N(5)#1–Cu–N(4)#1	84.13(14)	O(2)#1–S(2)–N(2)#1	105.46(18)
N(4)–Cu–N(4)#1	180.0	O(1)#1–S(2)–C(4)#1	106.32(18)
C(2)#1–S(1)–C(1)#1	89.3(2)	O(2)#1–S(2)–C(4)#1	108.09(18)
N(1)–C(1)–N(2)	121.4(3)	N(2)#1–S(2)–C(4)#1	108.29(17)
N(1)–C(1)–S(1)#1	112.7(3)	C(8)–C(9)–C(4)	120.6(4)
N(2)–C(1)–S(1)#1	125.9(3)	C(10)–N(4)–Cu	108.2(3)
C(3)–C(2)–S(1)#1	109.6(3)	C(11)–C(10)–N(4)	111.5(4)
C(2)–C(3)–N(1)	117.1(4)	C(11)–N(5)–Cu	108.7(3)

[a] Symmetry transformations used to generate equivalent atoms: #1: $-x+1, -y+1, -z$.

bands at 1531, 921 and 678–632 cm^{-1}, which have been assigned to the characteristic thiazole ring and ν(S–N) and ν(C–S) vibrations, respectively. The presence of a M–N stretching vibration at 445 cm^{-1} for the metal complex supports coordination by the ethylenediamine as a quadridentate N_4 chelating agent.

The UV–visible spectra of the sulfathiazole ligand and the complex in DMSO solution have major absorption maxima at 34130 and 33780 cm^{-1}, respectively, assigned to the $\pi \rightarrow \pi^*$ transitions within the organic molecule. The *d–d* electronic spectrum of the copper complex exhibits an asymmetric broad band centered at 15300 and 17850 cm^{-1} assignable to $^2B_{1g} \rightarrow {}^2B_{2g}$ and $^2B_{1g} \rightarrow {}^2E_{2g}$ transitions respectively characteristic of octahedral geometry [15]. A corrected magnetic moment has been calculated from the molar magnetic susceptibility using Pascal's constants [16] and the value of 1.85 BM indicates the presence of one unpaired electron. The EPR spectrum of the complex (Fig. 1) was obtained as a dimethylsulphoxide glass at 77 K and shows a typical pattern of magnetically dilute $S = 1/2$ centers with spectral characteristics resembling those of copper(II) in other six coordinate geometries ($g_{\parallel}, g_{\perp}$ and $A_{\parallel}$ values are 2.26, 2.06 and $170.10^{-4}\,cm^{-1}$, respectively and no $A_{\perp}$ hyperfine splittings were observed). According to Hathaway and Billing [17], this feature is consistent with an elongated tetragonal-octahedral symmetry (4 + 2 coordination). The existence of a $g_{\perp}$ value lower than its respective $g_{\parallel}$ value ($g_{\parallel} > g_{\perp} > 2.0023$) indicates [18] that the unpaired electron most likely resides in the $d_{x^2-y^2}$ orbital having $^2B_{1g}$ as a ground state term. The axial symmetry parameter, $G = 4.3$, obtained by the relation $(g_{\parallel} - 2)/(g_{\perp} - 2)$, for the complex, supports [19] the contention that there is no exchange interaction

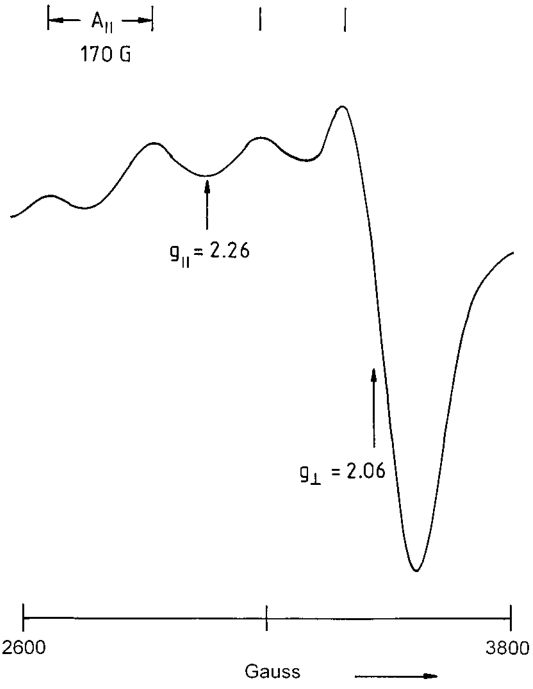

FIGURE 1 EPR spectrum of the $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ compound in dimethylsulfoxide glass at 77 K.

between the copper centers ($G > 4$) and that the local tetragonal axes are aligned in a parallel fashion or only slightly mis-aligned. The magnetic moment of the copper complex is in agreement with the EPR and the electronic spectra.

Description of the Structure

The molecular structure of $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ and the atom labelling scheme, are shown in Fig. 2 (ORTEP-III) [20]. The crystal is centrosymmetric. The center of symmetry is located at the copper(II). A structural search in the Cambridge Structural Database indicates that this is the first crystallographic determination of deprotonated sulfathiazole acting as a counter ion. The coordination environment of copper(II) is composed of four nitrogen atoms from the ethylenediamine and two oxygen atoms of water molecules. The coordination polyhedron around copper(II) can be approximated as a distorted octahedron. The C5, C6, C7, C8, and C9 atoms in the sulfonamide, lie in a plane, but N1, C1, S1, C2 and C3 are in a distorted plane. The S2-C4 (1.761(4) Å), S2-N2 (1.578(3) Å), C1-N2 (1.366(5) Å) and C1-N1 (1.304(5) Å) distances are close to the distances reported for sulfathiazole II [21].

The Cu–N4 and Cu–N5 distances are 2.042(3) Å and 2.033(3) Å, the values obtained are longer than Cu–N bond lengths in similar compounds. The planes of the ethylendiamine rings are distorted and related by a center of symmetry. There is an angle of 53.2(2)° between ethylendiamine and benzene rings.

The structure can be described in terms of sulfathiazole chains in zig-zags along the *b*-axis and between the coordination polyhedra centered at copper(II), coincident with the center of the face C and parallel to [220]. The oxygen atoms of coordinated water and lattice water and the nitrogen atoms link separate molecules by hydrogen bonds. The hydrogen bonds which form are of the N–H...N, N–H...O, O–H...N, and O–H...O types as indicated in Table III. The molecular packing in crystals is illustrated in Fig. 3.

Antibacterial Activity

Antibacterial activities of the complex were tested *in vitro* (50 and 20 μg per disc) against two Gram-positive (*S. aureus* 6538 and *B. cereus* 9634) and three Gram-negative bacteria (*Salmonella typhimurium* 14028, *P. aeruginosa* 9027 and *E. coli* 10536) by the disc diffusion method [11] using nutrient agar as medium. The complex was found to

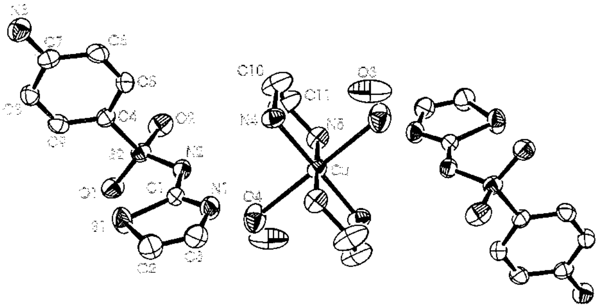

FIGURE 2 The molecular structure of $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ indicating the atom labelling scheme (ORTEP-III, ellipsoids drawn at the 50% level of probability). Hydrogen atoms are omitted for clarity.

TABLE III Hydrogen bonds with H...A < r(A) + 2.000 Å and ⟨DHA⟩ 110° for $CuC_{22}H_{40}N_{10}O_8S_4$

D–H	*d(D–H)*	*d(H...A)*	*⟨DHA⟩*	*d(D...A)*	*A*
O4–HOA	0.844	2.166	154.10	2.948	N2
O4–HOB	0.701	2.081	163.94	2.761	O3
O3–HOC	0.865	2.001	150.77	2.788	O1[$-x,-y,-z$]
O3–HOD	0.682	2.222	144.35	2.804	O2[$-x,-y+1,-z$]
N3–H3A	0.860	2.477	174.95	3.335	O3[$x,y,z+1$]
N4–H3B	0.860	2.311	168.18	3.158	N1[$-x+1,-y+1,-z+1$]
N4–H4A	0.860	2.455	159.59	3.274	O4[$-x+1,-y+1,-z$]
N4–H4A	0.860	2.517	118.63	3.025	N5[$-x+1,-y+1,-z$]
N4–H4B	0.860	2.449	160.61	3.273	N3[$x,y,z-1$]
N5–H5A	0.860	2.392	143.89	3.128	O4
N5–H5A	0.860	2.436	126.18	3.025	N4[$-x+1,-y+1,-z$]
N5–H5B	0.860	2.511	133.86	3.168	O2[$x+1,y,z$]

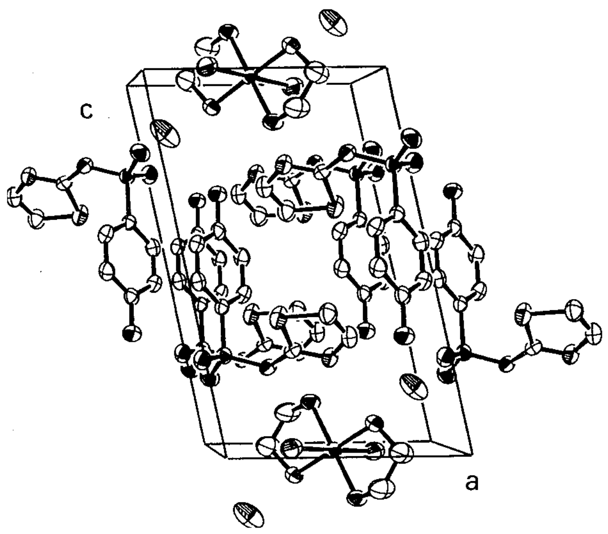

FIGURE 3 Packing arrangement for $[Cu(en)_2(OH_2)_2][stz]_2 \cdot 2H_2O$ (ORTEP-III, ellipsoids drawn at the 50% level of probability). Hydrogen atoms are omitted for clarity.

possess good bactericidal activity against *Salmonella typhimurium* 14028. However, it shows no bacterial activity against *S. aureus* 6538, *B. cereus* 9634, *P. aeruginosa* 9027 and *E. coli* 10536.

The average bactericidal diameters of the complex against *Salmonella typhimurium* was 25 and 20 mm at concentrations of 50 and 20 µg/disc respectively. The same values are obtained when the amoxicilline antibiotic is used as control at concentration of 200 µg/disc. Following the Shungu *et al.*, suggestions [22,23] the complex can be considered as highly sensitive.

Supplementary Data

Listings of atomic position and anisotropic thermal parameters for the crystal structure have been deposited at the Cambridge Crystallographic Data Center under the deposition number CCDC 136945.

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