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# SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL ACTIVITY OF Fe(II) MIXED LIGANDS COMPLEX OF SUCCINIC ACID AND HEXAMETHYLENETETRAMINE

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## ABSTRACT

*An Fe(II) complex has been synthesized from iron(II) sulphate, succinic acid and hexamethylenetetramine. The nature of bonding and the structural features of the complex have been deduced from elemental analysis, molar conductance, pH measurements, IR, UV-Vis spectral studies. Spectroscopic and analytical studies reveal octahedral geometry for this complex. The electronic spectrum of iron(II) complex in DMSO was recorded, and its salient features are reported. The activity studies data indicate that the complex is more potent antibacterial agent than the ligands and ciprofloxacin drug against the tested pathogenic bacteria strains. The synthesized compound can serve as potential photoactive materials as indicated from their characteristic absorption in the ultraviolet region.*

**Key Words:** Succinic acid, hexamethylenetetramine, antibacterial activity.

## INTRODUCTION

Many microorganisms, which cause damage to human health, exhibit drug resistance due to improper and abusive use of antimicrobial agents called antibiotics. Antimicrobial resistance has a significant public health and economic impact. Thus, there is a need for measures to reduce this problem. Emphasis on health care costs has provoked a renewed interest in the design and development of novel and cost effective antimicrobial agents with increased biological activity against these resistant strains [1-8]. Research has shown that design and synthesis of transition metal complexes derived from mixed base ligands might be a possible way to obtain materials endowed with antiviral, antibacterial and antifungal properties [3]. Broad empirical screening of chemical compounds for antimicrobial activity represents an alternative strategy for the development of new antimicrobials complexes with enhanced microbial activities, selective toxicity and chemical stability [4, 9]. The following bacteria *Staphylococcus aureus, Staphylococcus aubeus, Streptococcus pneumoniae, Enterococcus species, Pseudomonas species and Klebsiella* species are associated with a high incidence of infections that are resistant to treatment with antibiotics [2]. Our interest in synthesis of metal complex with antibacterial activities has led to the report herein the synthesis, spectroscopic studies and biological activities of mixed ligands complex of iron(II) with succinic acid and hexamethylenetetramine which have donor and acceptor property due to electron-withdrawing and releasing substituents (OH

and $NH_2$ groups). We are also interested to examine the optical properties of this compound.

## MATERIALS AND METHODS

All the chemicals and solvents used in this study viz, hexamethylenetetramine (HMTA, $C_6H_{12}N_4$), succinic acid ($C_4H_6O_4$), iron(II) sulphate heptahydrate salts ($FeSO_4.7H_2O$), ethanol, methanol, ammonia were all obtained from Sigma Aldrich and were of analytical grade. The solvents were used without further purification. Melting point determination was carried out using a Thomas Hoover capillary melting point apparatus. Infrared spectra of the ligands and complex were recorded using pressed KBr discs in the range (4000-450 $cm^{-1}$) on Perkin-Elmer FT-IR spectrometer 1.00 versions (Wattman) and the Electronic spectrum was measured in the range 200-700 nm at room temperature with Perkin Elmer spectrum Bk UV-Vis spectrophotometer.

### Synthesis of the Iron(II) Complex:

To a stirred solution of hexamethylenetetramine (5 mmol, 0.701 g) in 10 ml methanol, 5 ml aqueous solution of $FeSO_4.7H_2O$ (5 mmol, 1.390 g) was added dropwise with stirring at 20 $^oC$ for 10 minutes. Then a 5 ml methanol solution of succinic acid (5 mmol, 0.591 g) was added dropwise with stirring to the mixture. The resulting mixture was stirred for three hours. The reddishbrown complex formed was filtered, washed with methanol and dried under vacuum. On the fifth day, brown crystals were observed to form from the precipitate.

### Biological Activity

Antibacterial Screening: All the species used for this test were derived from stock cultures obtained from Environmental Science Laboratory, Federal University of Petroleum Resources, Effurun, Delta State, Nigeria. Antibacterial activity was tested against *Staphylococcus aureus, Staphylococcus aubeus* and *Escherichia coli*) using the paper disc plate method. The antibacterial activities of the ligands, and metal complex were evaluated using disc-diffusion method. Nutrient Agar was employed as bacterial growth medium. Antibacterial agents (100 µg) of the complex were loaded on the discs by soaking in test solution, and the discs were placed on the surface of the sterile nutrient agar medium with aid of the sterile forceps and the plates were incubated at 37 °C for 24 h. Antibacterial activity was evaluated by measuring the diameter of growth inhibition zone (IZ) in mm around the discs using Zone Reader. Compound was considered active when the IZ was greater than 6 mm. Antibacterial activity studies were performed in triplicate, and the average was taken as the final reading. Hexamethylenetetramine, succinic acid and ciprofloxacin were recorded under similar conditions [3].

## RESULTS

The physical properties of the complex and ligands listed in Table 1 shows that the Fe(II) complex was isolated as brown crystals. The iron(II) complex decomposed on melting within a temperature range of 160 - 163 °C. The variation in the color with increasing temperature could be due to changes in the crystal structure. The Fe(II) complex is air stable and insoluble in water and partially soluble in ethanol (Table 2). The complex and its ligands were generally soluble in methanol and ammonia.

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**Table 1: Physical properties of ligands and Fe (II) complex**

| Ligand/complex | Colour | Physical state | yield (%) | Melting point (°C) |
|---|---|---|---|---|
| $(CH_2)_6N_4$ | colorless | crystalline | - | 200 |
| $C_4H_6O_4$ | colorless | crystalline | | 186 |
| $FeSO_4.7H_2O$ | colorless | crystalline | - | 100 |
| Fe(II) complex | brown | crystalline | 68.5 | 160-163 |

**Table 2: Solubility of the ligands and Fe(II) complex in water and some solvents**

| Compound | Solvents | | | |
|---|---|---|---|---|
| | Water | Ethanol | Methanol | Ammonia |
| $(CH_2)_6N_4$ | Soluble | Insoluble | Insoluble | Soluble |
| $C_4H_6O_4$ | Insoluble | Insoluble | Insoluble | Soluble |
| $FeSO_4.7H_2O$ | Insoluble | Soluble | Soluble | Soluble |
| Fe(II) complex | Insoluble | Partially soluble | soluble | soluble |

The elemental composition was determined from energy dispersive X- ray analysis (EDX) spectrum to be calculated: C 15.42, H 15.38, N 10.22, O 16.26, Fe 42.72 %; found: C 15.98, H 15.56, N 9.28, O 16.34, Fe 42.84 %.

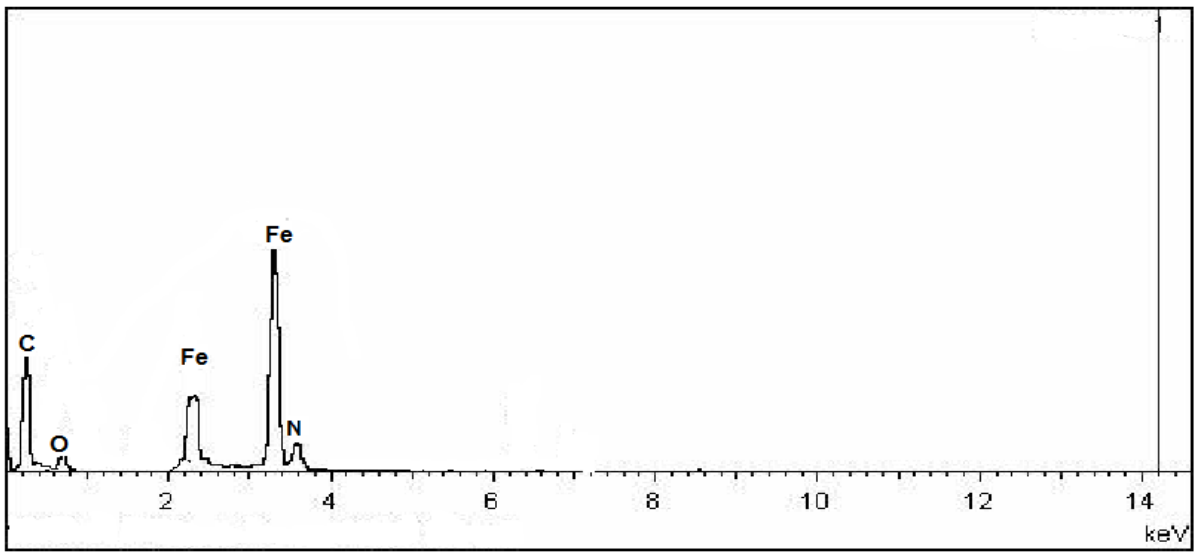


Figure I: EDX spectrum of Fe(II) complex

The infrared spectrum of the complex (Fig.2) was compared with that of the free ligands and qualitative differences between the spectra used in diagnosing the different vibrational frequencies of the metal and ligands in the complex. The Fe(II) vibrational bands were observed at 2991, 2053, 1445, 977, 697 cm-$^{1}$ assigned to $\upsilon_{as}(CH_2)$, $\upsilon_{s}(CH_2)$, $\upsilon$(C-N), $_{\delta}(CH_2)$, $\upsilon$(M-O) vibrational modes, respectively. Similar vibrational frequencies are recorded in the literature for similar compounds [11, 12].

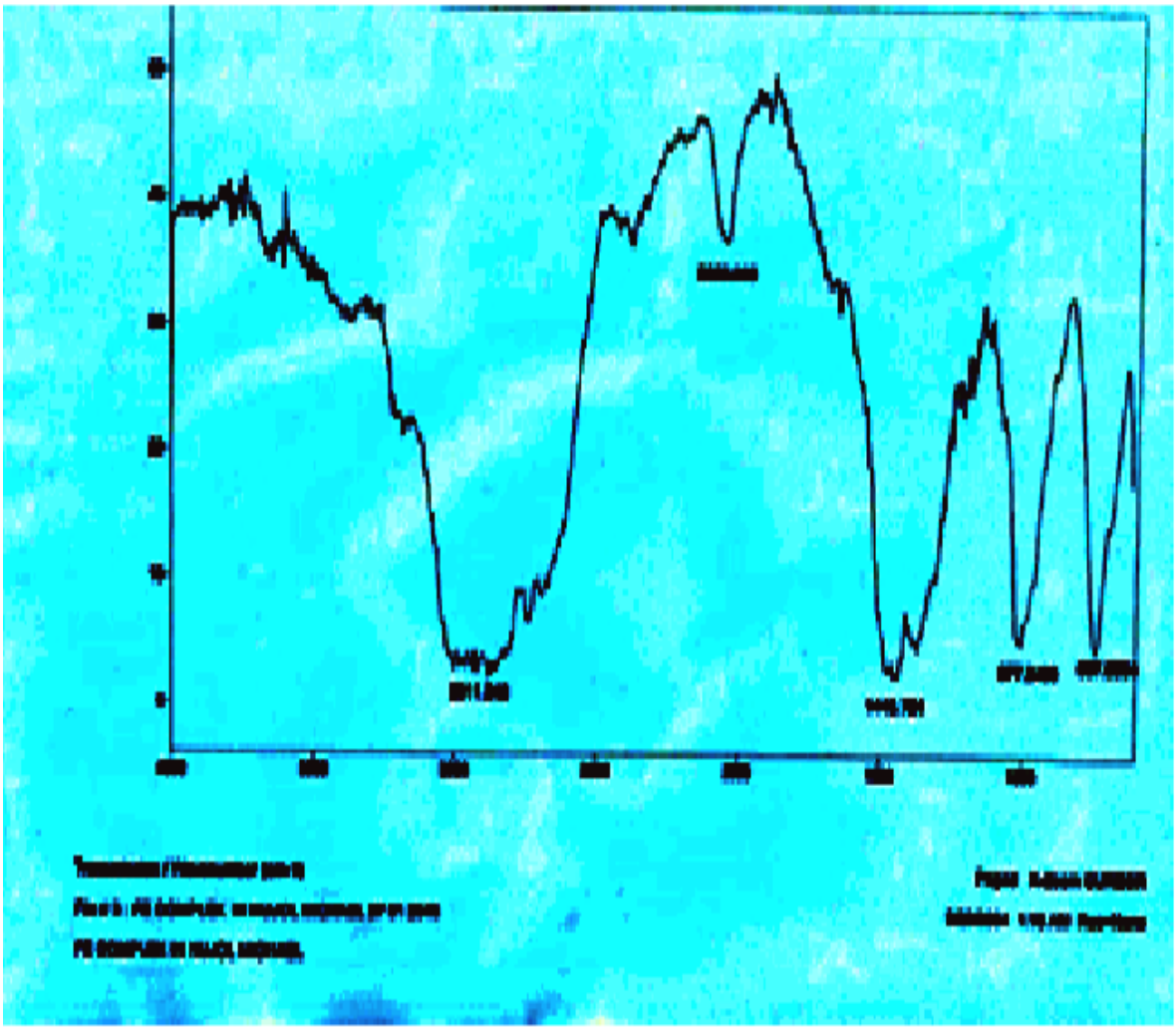

Figure 2: IR spectrum of Fe (II) complex

The electronic spectral data of the complex recorded in dimethylsulfoxide in the range 200–700 nm using a quartz cuvette of 1 cm path length presented in Table 3 confirms the formation of the iron(II) complex. The UV–vis spectral data values of the complex have shifts frequency when compared with the free ligands.

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**Table 3: Band positions (nm) and proposed transitions for Fe(II) complex**

| Complex | Band position | Assignment |
|---|---|---|
| Fe (II) complex | 203 | $^4T_{1g}(F)$ |
| | 253 | $^4T_{1g}(P)$ |
| | 269 | $^4T_{1g}(F)$ $^4A_{2g}$ |

From the antibacterial profile presented in Table 4; Fe (II) complex displays a significant inhibitory effect on the growth of tested bacteria species (Fig.3).


**Figure 3: Bacteria inhibition zone with Fe (II) complex**

**Table 4: Antibacterial activity of ligands, complex and ciprofloxacin-diameter of zone of inhibition (mm).**

| Tested compounds | *Staphylococcus aureus* (G+) | *Staphylococcus aubeus* (G+) | *Escherichia coli* (G-) |
|---|---|---|---|
| 1 | 0.0 | 0.0 | 0.0 |
| 2 | 0.0 | 0.0 | 3.6 |
| 3 | 48.1 | 48.6 | 20.2 |
| 4 | 23.7 | 32.2 | 14.5 |

Key: G+ = gram positive bacteria, G- = gram negative bacteria, IZ = inhibition zone, 1 = $(CH_2)_4N_4$, 2 = $C_4H_6O_4$, 3 = Fe(II) complex, 4 = Ciprofloxacin drug.

## DISCUSSION

The results obtained from solubility tests showed that the complex is soluble in coordinating solvents. The molar conductance measurement of complex was 2.75 $\Omega^{-1}\,cm^2\,mol^{-1}$ indicating that it possesses a non electrolytic nature. These observations are in agreement with octahedral Fe(II) complexes reported in the literature [10,11].

The experimentally found values for the elemental composition were very close with the calculated values [1]. These data are consistent with the general formulation as 1:1:1, mixed metal-ligands stoichiometry.

The electronic absorption spectrum of the iron (II) complex reveals three bands at 203, 253 and 269 $cm^{-1}$ assigned to $^4T_{1g}$ (F), $^4T_{1g}$(P) and $^4$T1g(F) $^4A_{2g}$ transitions which arise from an octahedral structure [10, 11].

The hexamethylenetetramine ligand (HMTA), was found to be active against only one of the 3 tested bacterial strains *(S. typhi)*; whereas the succinic acid has relatively low antibacterial activity [8].

The mixed ligands complex of Fe(II) with succinic acid and hexamethylenetetramine was synthesized and characterized by melting point determination, solubility, conductivity measurement, elemental analyses, UV-Vis and IR spectroscopy. From the molar conductance value, electronic and infrared spectral data octahedral geometry was proposed for this compound. The iron(II) complex was found to be a more potent antibacterial agent than the ligands and the ciprofloxacin drug. Based on these results, it could be proposed that the novel iron(II) complex can be better accommodated for pharmaceutical usage as a drug against bacteria infections. From the spectroscopic characterization, it is concluded that ligands act as a neutral bidentate through the HTMA nitrogen atom and carbonyl groups of the succinc acid.

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