

## 二核双席夫碱铜(II)配合物的合成、晶体结构及抗菌活性研究

仇晓阳

(商丘师范学院功能材料制备工程研究中心, 商丘 476000)

**摘要:** 合成了一对结构类似的双核铜配合物,  $[\text{CuL}^1]_2$  (**1**)和 $[\text{CuL}^2]_2$  (**2**), 其中  $\text{L}^1$  是双席夫碱配体  $N,N'$ -二(5-氟水杨基)-1,3-丙二胺( $\text{H}_2\text{L}^1$ )的二价阴离子,  $\text{L}^2$  是  $N,N'$ -二(5-氟水杨基)-1,2-乙二胺( $\text{H}_2\text{L}^2$ )的二价阴离子, 通过元素分析、红外光谱以及单晶 X-射线衍射表征了它们的结构。配合物 **1** 以单斜晶系  $P2_1/c$  空间群结晶, 其晶体学参数  $a=1.348\ 8(1)\ \text{nm}$ ,  $b=0.681\ 4(1)\ \text{nm}$ ,  $c=1.708\ 7(1)\ \text{nm}$ ,  $\beta=108.903(3)^\circ$ ,  $V=1.485\ 7(2)\ \text{nm}^3$ ,  $Z=4$ ,  $R_1=0.048\ 0$ ,  $wR_2=0.114\ 1$ ,  $\text{Goof}=1.115$ 。配合物 **2** 以单斜晶系  $C2/c$  空间群结晶, 其晶体学参数  $a=2.756\ 8(2)\ \text{nm}$ ,  $b=0.703\ 6(1)\ \text{nm}$ ,  $c=1.454\ 7(1)\ \text{nm}$ ,  $\beta=94.758(2)^\circ$ ,  $V=2.811\ 8(5)\ \text{nm}^3$ ,  $Z=4$ ,  $R_1=0.047\ 2$ ,  $wR_2=0.113\ 9$ ,  $\text{Goof}=1.094$ 。X-射线分析表明 2 个化合物都是中心对称的双核配合物, 其中 Cu 原子都是四方锥配位构型。通过 MTT 法研究了这 2 个配合物的抗菌(枯草芽孢杆菌, 金黄色葡萄球菌, 大肠杆菌和荧光假单胞菌)和抗真菌(白假丝酵母菌和黑曲霉菌)活性。

**关键词:** 席夫碱; 铜配合物; 双核配合物; 晶体结构; 抗菌活性

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## Synthesis, Crystal Structures and Antimicrobial Activity of Dinuclear Copper(II) Complexes with bis-Schiff Bases

QIU Xiao-Yang

(Engineering Research Center of Functional Material Preparation, Shangqiu Normal University, Shangqiu, Henan 476000, China)

**Abstract:** A pair of structurally similar dinuclear copper(II) complexes,  $[\text{CuL}^1]_2$  (**1**) and  $[\text{CuL}^2]_2$  (**2**), where  $\text{L}^1$  is the dianionic form of the bis-Schiff base  $N,N'$ -bis(5-fluorosalicylidene)-1,3-propanediamine ( $\text{H}_2\text{L}^1$ ), and  $\text{L}^2$  is the dianionic form of  $N,N'$ -bis(5-fluorosalicylidene)-1,2-ethanediamine ( $\text{H}_2\text{L}^2$ ), have been synthesized and characterized by elemental analysis, infrared spectra and single-crystal X-ray diffraction. Complex **1** crystallizes as the monoclinic space group  $P2_1/c$ , with unit cell dimensions  $a=1.348\ 8(1)\ \text{nm}$ ,  $b=0.681\ 4(1)\ \text{nm}$ ,  $c=1.708\ 7(1)\ \text{nm}$ ,  $\beta=108.903(3)^\circ$ ,  $V=1.485\ 7(2)\ \text{nm}^3$ ,  $Z=4$ ,  $R_1=0.048\ 0$ ,  $wR_2=0.114\ 1$ ,  $\text{Goof}=1.115$ . Complex **2** crystallizes as the monoclinic space group  $C2/c$ , with unit cell dimensions  $a=2.756\ 8(2)\ \text{nm}$ ,  $b=0.703\ 6(1)\ \text{nm}$ ,  $c=1.454\ 7(1)\ \text{nm}$ ,  $\beta=94.758(2)^\circ$ ,  $V=2.811\ 8(5)\ \text{nm}^3$ ,  $Z=4$ ,  $R_1=0.047\ 2$ ,  $wR_2=0.113\ 9$ ,  $\text{Goof}=1.094$ . X-ray analysis indicates that the complexes are centrosymmetric dinuclear species, with the Cu atoms in square pyramidal coordination. The complexes were evaluated for their antibacterial (*Bacillus subtilis*, *Staphylococcus aureus*, *Escherichia coli*, and *Pseudomonas fluorescens*) and antifungal (*Candida albicans* and *Aspergillus niger*) activities by MTT (3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl tetrazolium bromide) method. CCDC: 940737, **1**; and 972404, **2**.

**Key words:** Schiff base; copper complex; dinuclear complex; crystal structure; antimicrobial activity

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E-mail: xiaoyang\_qiu@126.com

Schiff bases are a kind of biological active compounds bearing the  $-N=CH-$  functional groups, which can be prepared by the condensation reactions of carbonyl-containing compounds with primary amines. The compounds have been attracted considerable attention for their wide range of biological activities, such as antibacterial<sup>[1-2]</sup>, antifungal<sup>[3]</sup>, antitumor<sup>[4-5]</sup>, anti-inflammatory<sup>[6-7]</sup>, and cytotoxic<sup>[8-9]</sup>. It was reported that Schiff bases bearing electron-withdrawing groups can improve their antimicrobial activities<sup>[10-11]</sup>. Rai and co-workers reported a series of fluoro, chloro, bromo, and iodo-substituted compounds, and found that they have significant antimicrobial activities<sup>[12-13]</sup>. Compared to mono-Schiff bases, the number of bis-Schiff bases and their complexes is much less. As a continuation of work on the exploration of novel antimicrobial agents and preparation of new bis-Schiff base complexes, in this paper, two new dinuclear copper(II) complexes,  $[CuL^1]_2$  (**1**) and  $[CuL^2]_2$  (**2**), where  $L^1$  is the dianionic form of the bis-Schiff base  $N,N'$ -bis(5-fluorosalicylidene)-1,3-propanediamine ( $H_2L^1$ ), and  $L^2$  is the dianionic form of  $N,N'$ -bis(5-fluorosalicylidene)-1,2-ethanediamine ( $H_2L^2$ ), Scheme 1, were prepared and investigated their antibacterial activities.

## 1 Experimental

### 1.1 General methods and materials

Copper nitrate, 5-fluorosalicylaldehyde, pentane-1,3-diamine and ethane-1,2-diamine were purchased Sigma-Aldrich and used as received. All other reagents were of analytical reagent grade. Elemental analyses of C, H and N were carried out in a Perkin-Elmer automated model 2400 Series II CHNS/O analyzer. FT-IR spectra were obtained on a Perkin-Elmer 377 FT-IR spectrometer with samples prepared as KBr pellets. Molar conductance was measured with a Shanghai DDS-11A conductometer. X-ray diffraction was carried out on a Bruker APEX II CCD

diffractometer.

### 1.2 Synthesis of the Schiff bases

The Schiff bases were synthesized as follows. To the methanolic solution (30 mL) of 5-fluorosalicylaldehyde (0.02 mol, 2.96 g) was added a methanolic solution (20 mL) of pentane-1,3-diamine (0.01 mol, 0.074 g) or ethane-1,2-diamine (0.02 mol, 0.060 g) with stirring. The mixtures were stirred for 30 min at room temperature to give yellow precipitation. The solvent was evaporated to give yellow crystalline product, which was recrystallized from methanol and dried in vacuum containing anhydrous  $CaCl_2$ .

For  $H_2L^1$ : Yield 93%. Characteristic IR data ( $cm^{-1}$ ): 3 317 (m), 1 641 (s). Anal. Calcd. for  $C_{17}H_{16}F_2N_2O_2$ (%): C, 64.1; H, 5.1; N, 8.8. Found(%): C, 63.9; H, 5.0; N, 8.9. For  $H_2L^2$ : Yield 91%. Characteristic IR data ( $cm^{-1}$ ): 3 332 (m), 1 639 (s). Anal. Calcd. for  $C_{16}H_{14}F_2N_2O_2$ (%): C, 63.2; H, 4.6; N, 9.2. Found(%): C, 63.1; H, 4.7; N, 9.1.

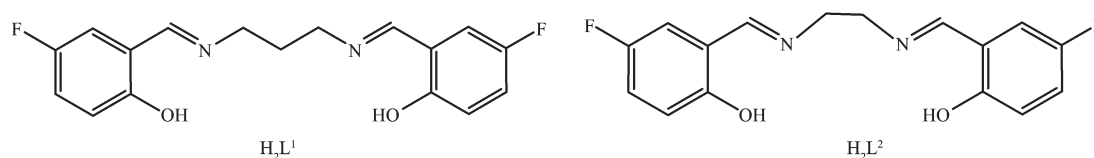
### 1.3 Synthesis of the complexes

The Schiff base ligands (0.1 mmol each) and copper nitrate trihydrate (24.2 mg, 0.1 mmol) were mixed in methanol (10 mL). The mixture was refluxed for 1 h and then cooled to room temperature. Single crystals of the complexes, suitable for X-ray diffraction, were grown from the solution upon slow evaporation within a few days. The crystals were isolated by filtration, washed with methanol and dried in vacuum containing anhydrous  $CaCl_2$ .

For **1**: Yield 53%. Characteristic IR data ( $cm^{-1}$ ): 1 605 (s). Anal. Calcd. for  $C_{34}H_{28}Cu_2F_4N_4O_4$  (%): C, 53.8; H, 3.7; N, 7.4. Found(%): C, 53.6; H, 3.8; N, 7.4. For **2**: Yield 62%. Characteristic IR data ( $cm^{-1}$ ): 1 607 (s). Anal. Calcd. for  $C_{32}H_{24}Cu_2F_4N_4O_4$  (%): C, 52.5; H, 3.3; N, 7.7. Found(%): C, 52.6; H, 3.3; N, 7.5.

### 1.4 X-ray crystallography

X-ray diffraction was carried out at a Bruker APEX II CCD area diffractometer equipped with



Scheme 1 Schiff bases

Mo  $K\alpha$  radiation ( $\lambda=0.071\ 073\ \text{nm}$ ). The collected data were reduced with SAINT<sup>[14]</sup>, and multi-scan absorption correction was performed using SADABS<sup>[15]</sup>. The structures of the complexes were solved by direct method, and refined against  $F^2$  by full-matrix least-squares method using SHELXTL<sup>[16]</sup>. All of the non-hydrogen atoms were refined anisotropically. All hydrogen atoms were placed in calculated positions and constrained to ride on their parent atoms. The crystallographic data and refinement parameters for the compounds are listed in Table 1. Selected bond lengths and angles are listed in Table 2.

CCDC: 940737, **1**; and 972404, **2**.

### 1.5 Antimicrobial assay

The antibacterial activities of the compounds were tested against *B. subtilis*, *S. aureus*, *E. coli*, and *P. fluorescence* using MH (Mueller-Hinton) medium.

The antifungal activities of the compounds were tested against *C. albicans* and *A. niger* using RPMI-1640 medium. The MIC values of the tested compounds were determined by a colorimetric method using the dye MTT<sup>[17]</sup>. A stock solution of the compound ( $150\ \mu\text{mol}\cdot\text{L}^{-1}$ ) in DMSO was prepared and graded quantities ( $75\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $37.5\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $18.8\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $9.4\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $4.7\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $2.3\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $1.2\ \mu\text{mol}\cdot\text{L}^{-1}$ ,  $0.59\ \mu\text{mol}\cdot\text{L}^{-1}$ ) were incorporated in specified quantity of the corresponding sterilized liquid medium. A specified quantity of the medium containing the compound was poured into micro-titration plates. Suspension of the microorganism was prepared to contain approximately  $1.0\times 10^5\ \text{cfu}\cdot\text{mL}^{-1}$  and applied to microtitration plates with serially diluted compounds in DMSO to be tested and incubated at  $37\ ^\circ\text{C}$  for 24 h and 48 h for bacterial and fungi, respectively. Then the MIC values were

Table 1 Crystallographic information for the complexes

| Complex   | <b>1</b>  | <b>2</b>  |
|---|---|---|
| Formula   | $\text{C}_{34}\text{H}_{28}\text{Cu}_2\text{F}_4\text{N}_4\text{O}_4$ | $\text{C}_{32}\text{H}_{24}\text{Cu}_2\text{F}_4\text{N}_4\text{O}_4$ |
| Formula weight                                  | 759.7   | 731.6   |
| Crystal shape / color                           | Block / blue  | Block / blue  |
| $T / \text{K}$                                  | 298(2)  | 298(2)  |
| Crystal size / mm                               | $0.27\times 0.23\times 0.23$  | $0.20\times 0.17\times 0.17$  |
| Crystal system                                  | Monoclinic  | Monoclinic  |
| Space group                                     | $P2_1/c$  | $C2/c$  |
| $a / \text{nm}$                                 | 1.348 8(1)  | 2.756 8(2)  |
| $b / \text{nm}$                                 | 0.681 4(1)  | 0.703 6(1)  |
| $c / \text{nm}$                                 | 1.708 7(1)  | 1.454 7(1)  |
| $\beta / (^\circ)$                              | 108.903(3)  | 94.758(2)   |
| $V / \text{nm}^3$                               | 1.485 7(2)  | 2.811 8(5)  |
| $Z$   | 4   | 4   |
| $D_c / (\text{g}\cdot\text{cm}^{-3})$           | 1.698   | 1.728   |
| $\mu (\text{Mo } K\alpha) / \text{mm}^{-1}$     | 1.505   | 1.587   |
| $F(000)$  | 772   | 1 480   |
| Measured reflections                            | 13 766  | 12 503  |
| Unique reflections                              | 2 765   | 2 906   |
| Observed reflections ( $I\geq 2\sigma(I)$ )     | 2 120   | 2 321   |
| Min. and max. transmission                      | 0.686 7 and 0.723 4   | 0.741 9 and 0.774 1   |
| Parameters                                      | 217   | 208   |
| Goodness of fit on $F^2$                        | 1.115   | 1.094   |
| $R_1, wR_2$ ( $I\geq 2\sigma(I)$ ) <sup>a</sup> | 0.048 0, 0.114 1  | 0.047 2, 0.113 9  |
| $R_1, wR_2$ (all data) <sup>a</sup>             | 0.069 8, 0.122 9  | 0.063 7, 0.127 4  |

<sup>a</sup>  $R_1 = \sum \|F_o\| - |F_c| / \sum \|F_o\|$ ,  $wR_2 = [\sum w(F_o^2 - F_c^2) / \sum w(F_o^2)]^{1/2}$

**Table 2** Selected bond lengths (nm) and angles (°) for the complexes with estimated standard deviations (e.s.d.s) in parentheses

| <b>1</b>   |            |            |            |            |            |
|------------|------------|------------|------------|------------|------------|
| Cu1-O1     | 0.190 8(3) | Cu1-O2     | 0.193 1(3) | Cu1-N1     | 0.198 3(4) |
| Cu1-N2     | 0.199 9(4) | Cu1-O2A    | 0.271 9(5) |            |            |
| O1-Cu1-O2  | 82.13(13)  | O1-Cu1-N1  | 91.20(14)  | O2-Cu1-N1  | 172.64(14) |
| O1-Cu1-N2  | 168.23(14) | O2-Cu1-N2  | 90.73(14)  | N1-Cu1-N2  | 96.36(15)  |
| O1-Cu1-O2A | 94.55(15)  | O2-Cu1-O2A | 92.42(15)  | N1-Cu1-O2A | 84.98(15)  |
| N2-Cu1-O2A | 95.08(15)  |            |            |            |            |
| <b>2</b>   |            |            |            |            |            |
| Cu1-O1     | 0.190 7(3) | Cu1-O2     | 0.194 3(2) | Cu1-N1     | 0.195 7(3) |
| Cu1-N2     | 0.195 2(3) | Cu1-O2A    | 0.241 7(3) |            |            |
| O1-Cu1-O2  | 91.03(11)  | O1-Cu1-N2  | 170.18(12) | O2-Cu1-N2  | 91.38(12)  |
| O1-Cu1-N1  | 92.56(12)  | O2-Cu1-N1  | 171.83(12) | N2-Cu1-N1  | 83.88(13)  |
| O1-Cu1-O2A | 96.21(11)  | O2-Cu1-O2A | 87.33(10)  | N2-Cu1-O2A | 93.41(11)  |
| N1-Cu1-O2A | 99.56(11)  |            |            |            |            |

Symmetry code: **1**: A: 2-x, 1-y, 2-z; **2**: A: 1/2-x, 1/2-y, -z.

visually determined on each of the microtitration plates, 50  $\mu\text{L}$  of PBS (phosphate buffered saline 0.01  $\text{mol}\cdot\text{L}^{-1}$ , pH=7.4) containing 2 mg of  $\text{MTT}\cdot\text{mL}^{-1}$  was added to each well. Incubation was continued at room temperature for 4~5 h. The content of each well was removed, and 100  $\mu\text{L}$  of isopropanol containing 5% 1  $\text{mol}\cdot\text{L}^{-1}$  HCl was added to extract the dye. After 12 h of incubation at room temperature, the optical density was measured with a microplate reader at 550 nm.

## 2 Results and discussion

### 2.1 Synthesis and characterization

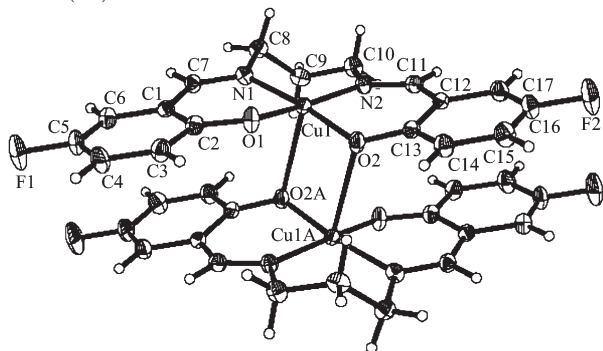
The Schiff bases were readily prepared by the condensation reaction of 2:1 molar ratio of 5-fluorosalicylaldehyde with propane-1,3-diamine and ethane-1,2-diamine, respectively, in methanol. Complexes **1** and **2** were prepared by the reaction of the Schiff base ligands with copper nitrate in methanol, followed by recrystallization. Elemental analyses of the complexes are in accordance with the molecular structures proposed by the X-ray analysis. FT-IR spectra of both complexes are of similar type. The complexes show typical C=N absorptions at 1 605  $\text{cm}^{-1}$  for **1** and 1 607  $\text{cm}^{-1}$  for **2**. Both complexes are stable in air at room temperature. The molar conductivity of the complexes

measured in absolute methanol at concentration of 1  $\text{mmol}\cdot\text{L}^{-1}$  are 15.0  $\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$  for **1** and 17.5  $\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$  for **2**, indicating the non-electrolytic nature of the complexes in solution<sup>[18]</sup>.

### 2.2 Structure description of the complexes

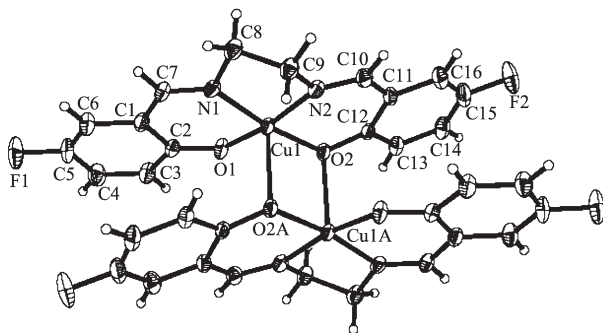
The molecular structures of complexes **1** and **2** are shown in Fig.1 and 2, respectively. Both complexes are centrosymmetric dinuclear copper(II) species, with the inversion center located at the midpoint of the two Cu atoms. The Cu...Cu distances are 0.326 7(2) nm in **1** and 0.317 1(2) nm in **2**. Each Cu atom of the complexes is coordinated in a square pyramidal geometry, with two phenolate oxygen and two imino nitrogen of the Schiff base ligand defining the basal plane, and with one phenolate oxygen of the other symmetry related Schiff base ligand occupying the apical position. The Cu atoms in **1** and **2** deviate from the corresponding least-squares planes defined by the four basal donor atoms by 0.009 9(3) nm and 0.013 8(3) nm, respectively. The distortion of the square pyramidal coordination can be observed from the bond distances and bond angles. The *cis* bond angles in the basal planes are from 82.13(13)° to 96.36(15)° in **1**, and from 83.88(13)° to 92.56(12)° in **2**, and the *trans* bond angles are 168.23(14)° and 172.64(14)° in **1**,

and  $170.18(12)^\circ$  and  $171.83(12)^\circ$  in **2**. The angles among the apical and basal donor atoms are in the range  $84.98(15)^\circ \sim 95.08(15)^\circ$  in **1** and  $87.33(10)^\circ \sim 99.56(11)^\circ$  in **2**. The Cu-O and Cu-N bonds in the basal



Thermal ellipsoids are drawn at the 30% probability level;  
Symmetry operation to generate related atoms:  $2-x, 1-y, 2-z$

Fig.1 A perspective view of complex **1** with the atom labeling scheme



Thermal ellipsoids are drawn at the 30% probability level;  
Symmetry operation to generate related atoms:  $1/2-x, 1/2-y, -z$

Fig.2 A perspective view of complex **2** with the atom labeling scheme

planes of both complexes are comparable to each other, and also similar to those observed in Schiff base copper(II) complexes<sup>[19-20]</sup>. However, the apical bonds in both complexes are much longer than the basal ones, indicating they are loosely coordinate to the Cu atoms. The two benzene rings of the Schiff base ligands form dihedral angles of  $5.0(5)^\circ$  for **1** and  $17.8(6)^\circ$  for **2**.

### 2.3 Antimicrobial activity

The copper complexes and the free Schiff bases were screened for antibacterial activity against two Gram (+) bacterial strains (*Bacillus subtilis* and *Staphylococcus aureus*) and two Gram (-) bacterial strains (*Escherichia coli* and *Pseudomonas fluorescence*) by MTT method. The MIC (minimum inhibitory concentration) values of the compounds against four bacteria are listed in Table 3. Penicillin G was used as the standard drug. The Schiff bases showed strong activity against the bacteria *B. subtilis* and *S. aureus*, medium activity against *E. coli*, and no activity against *P. fluorescence*. The copper complexes, in general, have stronger activities against *B. subtilis*, *S. aureus* and *P. fluorescence*, but weaker activity against *E. coli* when compared with the free Schiff bases. Except for *P. fluorescence*, there is no obvious difference between the two complexes against the bacteria. It is interesting that the complexes showed most effective activity against all the bacteria, which is more effective than Penicillin G.

Table 3 MIC values ( $\mu\text{mol} \cdot \text{L}^{-1}$ ) of the tested materials

| Tested material               | <i>B. subtilis</i> | <i>S. aureus</i> | <i>E. coli</i> | <i>P. fluorescence</i> | <i>C. albicans</i> | <i>A. niger</i> |
|-------------------------------|--------------------|------------------|----------------|------------------------|--------------------|-----------------|
| H <sub>2</sub> L <sup>1</sup> | 4.7                | 4.7              | 37.5           | >150                   | >150               | >150            |
| H <sub>2</sub> L <sup>2</sup> | 4.7                | 4.7              | 18.8           | >150                   | >150               | >150            |
| <b>1</b>                      | 1.2                | 2.3              | 75             | 37.5                   | >150               | >150            |
| <b>2</b>                      | 1.2                | 2.3              | 75             | 75                     | >150               | >150            |
| Penicillin G                  | 2.3                | 4.7              | >150           | >150                   | >150               | >150            |
| Ketoconazole                  | >150               | >150             | >150           | >150                   | 4.7                | 18.8            |

The antifungal activities of the complexes and the Schiff bases were also evaluated against two fungal strains (*Candida albicans* and *Aspergillus niger*) by MTT method. Ketoconazole was used as a reference material. However, both the copper complexes and the Schiff bases have no obvious activity against

the fungal strains.

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