Research Article
Preparation and Thermogravimetric and Antimicrobial Investigation of Cd (II) and Sn (II) Adducts of Mercaptopyridine, Amino Triazole Derivatives, and Mercaptothiazoline Organic Ligand Moieties

Ahmed Gaber,1 Walaa F. Alsanie,2 Robson F. de Farias,3 and Moamen S. Refat4

1Department of Biology, College of Science, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia
2Department of Clinical Laboratories Sciences, The Faculty of Applied Medical Sciences, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia
3Departamento de Química, Universidade Federal de Roraima, Cx. Postal 167, Boa Vista 69301-970, Roraima, Brazil
4Department of Chemistry, Faculty of Science, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia

Correspondence should be addressed to Moamen S. Refat; mrefat@yahoo.com

Received 9 December 2020; Revised 27 February 2021; Accepted 2 April 2021; Published 16 April 2021

Academic Editor: Guillermo Mendoza-Diaz

Copyright © 2021 Ahmed Gaber et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

The solid adducts of SnCl₂.(3amt).H₂O, SnCl₂.2(3amt).H₂O, CdCl₂.(3amt), CdCl₂.2(3amt), SnCl₂.(2mct).0.5H₂O, SnCl₂.2(2mct), CdCl₂.(2mct), CdCl₂.2(2mct).H₂O, SnCl₂.(4amt).1.5H₂O, CdCl₂.4(amt).H₂O, and CdCl₂.2(4amt) (where the 3amt, 4amt, 2mct, or 2mcp represent 3-amino-1,2,4-triazole, 4-amino-1,2,4-triazole, 2-mercaptothiazoline, and 2-mercaptopyridine simple organic chelates, respectively) were prepared using a solid-state route and investigated by CHN elemental analysis and infrared spectroscopy. Additionally, we investigated the thermogravimetric characterization and antimicrobial proprieties. It is verified that for 3amt and 4amt adducts, the coordination occurs through nitrogen atom. For 2mct compounds, the coordination occurs through nitrogen (Sn) or sulfur (Cd). For 2mcp adducts, both coordination sites nitrogen and sulfur are involved. By examination of TG curves, it is confirmed that for each hydrated compounds, the first mass loss step is linked with the release of water molecules followed by the release of ligand molecules and sublimation of the metal chloride. Furthermore, it is verified that, considering only the release of ligand molecules (3amp, 4amp, 2mct, or 2mcp) represent 3-amino-1,2,4-triazole, 4-amino-1,2,4-triazole, 2-mercaptothiazoline, and 2-mercaptopyridine simple organic chelates, respectively) were prepared using a solid-state route and investigated by CHN elemental analysis and infrared spectroscopy. Additionally, we investigated the thermogravimetric characterization and antimicrobial proprieties. It is verified that for 3amt and 4amt adducts, the coordination occurs through nitrogen atom. For 2mct compounds, the coordination occurs through nitrogen (Sn) or sulfur (Cd). For 2mcp adducts, both coordination sites nitrogen and sulfur are involved. By examination of TG curves, it is confirmed that for each hydrated compounds, the first mass loss step is linked with the release of water molecules followed by the release of ligand molecules and sublimation of the metal chloride. Furthermore, it is verified that, considering only the release of ligand molecules (3amp, 4amp, 2mct, or 2mcp), the cadmium adducts are always more stable than the correspondent tin adducts probably due to the formation of cross-linking bonds in these compounds. Finally, of these 16 adducts, 14 showed antimicrobial activities against different bacterial and fungal strains.

1. Introduction

In the past decades, the problem of multidrug-resistant microorganisms has reached an alarming level worldwide, and the synthesis of new antimicrobial compounds has become an urgent need to treat microbial infections. Organic compounds that include heterocyclic ring systems continue to attract significant interest due to their wide range of biological elements [1]. The nucleus 1,2,4-triazole is incorporated into a variety of important therapeutic agents, which mainly exhibit antimicrobial activities [1, 2]. Among the various five-membered heterocyclic systems, 1,2,4-triazoles and 1,3,4-thiadiazoles and their derivatives gain importance because they constitute the structural features of many bioactive compounds [2]. Triazole and thiadiazole rings are known to be included in the structure of various drugs [3, 4]. From these classes of heterocyclic compounds, the synthesis of novel derivatives of 1,2,4-triazole-3-thionate and 2-amino-1,3,4-thiadiazole has attracted great interest due to various biological properties such as antibacterial [5, 6], antifungal [7], antituberculosis [8, 9], interferon [10], antioxidant [11], antitumor [12], anti-inflammatory [13, 14], and anticonvulsant [15].

The thermogravimetry analysis technique is employed to identify the acceptability level regarding the coordination.
nature in between the central metal ions and different kinds of interesting biomolecule chelates, such as amino acids [16], caffeine molecule [17], or chemical materials that have a biological behavior as ethylene- and propylene-urea as well as ethylene-thiourea [18]. Moreover, the thermogravimetric information shows very close relationships with the caloriometric data [19] and the spectral data [20].

The main goal of this article is to investigate the synthesis, thermal analyses, and antimicrobial data of the sixteen solid adducts for the Cd (II) and Sn (II) metal ions coordinated with the 3amt, 4amt, 2mct, or 2mcp organic molecules. The molecular structural formulas of 3amt, 4amt, 2mct, and 2mcp are displayed in Figure 1. The sixteen solid adducts are SnCl2.(3amt).H2O, SnCl2.2(3amt).H2O, CdCl2. (3amt), CdCl2.2(3amt), SnCl2.(2mct).0.5H2O, SnCl2.2(2mc t), CdCl2.(2mct), CdCl2.2(2mct).H2O, SnCl2.(2mcp).1.5H2 O, SnCl2.2(2mcp).4H2O, CdCl2.(2mcp), CdCl2.2(2mcp), Sn Cl2.(4amt).4H2O, SnCl2.2(4amt).1.5H2O, CdCl2.(4amt). H2O, and CdCl2.2(4amt).

2. Materials and Methods

All used reagents were purchased from Sigma-Aldrich and were utilized with no additional purification.

All solid Cd(II) and Sn(II) adducts were prepared by the solid-state pathway by grinding stoichiometric amounts of metal halides and organic moieties (3amt, 4amt, 2mct, and 2mcp) in a mortar for 70 minutes at room temperature (27°C). The prepared solid adducts were dried under vacuum at room temperature for 24 h. This solid-state pathway was successfully used to enhance coordination reactions [21–24] as an alternative to conventional synthesis in solution. The synthesis is performed at room temperature, and where no solvent is used, any unwanted reaction to the metal cation is avoided. The infrared spectra result considering both free organic ligands and sixteen solid adducts proved that there are no free ligand particles after the grinding process.

C, H, and N elemental analysis were performed using a Perkin-Elmer 2400 analyzer. Infrared spectra of the solid adducts as a powder in situ KBr discs were scanned using a Gengis II FTIR apparatus within the 4000–400 cm−1 range, with a resolution of 4 cm−1. Thermogravimetric diagrams under N2 atmosphere were analyzed on the Shimadzu TG-50H apparatus with a heating rate of 15°C min−1.

Tin(II) and cadmium(II) contents were determined by gravimetry by the direct ignition of the adducts at 600°C for 3 h till constant weight. The residue was then weighted in the forms of SnO and CdO, respectively. The Mohr method uses chromate ions as an indicator in the titration of chloride ions with a silver nitrate standard solution. After all the chloride precipitated as white silver chloride, the first excess of titrant results in the formation of a silver chromate precipitate, which signals the end point.

Preparation of standard AgNO3 solution: 9.0 g of AgNO3 was weighed out, transferred to a 500 mL volumetric flask, and made up to volume with distilled water. The resulting solution was approximately 0.1 M. This solution was standardized against NaCl. Reagent-grade NaCl was dried overnight and cooled to room temperature. 0.25 g portions of NaCl were weighed into Erlenmeyer flasks and dissolved in about 100 mL of distilled water. In order to adjust the pH of the solutions, small quantities of NaHCO3 were added until effervescence ceased. About 2 mL of K2CrO4 was added, and the solution was titrated to the first permanent appearance of red Ag2CrO4.

The antimicrobial activity of all adducts were performed as previously explained in detail by Gaber et al. [21]. Escherichia coli and Pseudomonas aeruginosa were used as Gram-negative bacteria, whereas Bacillus subtilis and Staphylococcus aureus were used as Gram-positive bacteria. In addition, Aspergillus flavus and Candida albicans were used as fungal strains. Diameters of the inhibition zones around the hole were calculated [21].

3. Results and Discussion

Table 1 shows the data of the elemental analysis. These results are like the proposed formulas. Additionally, the main infrared bands are displayed in Figures 2–5. Before a discussion on the infrared data, it is important to note that considering (3amt and 4amt) and (2mct and 2mcp), organic molecules have a rich electron donor sites through the lone pair of electrons presented on the nitrogen and sulfur atoms, respectively. Moreover, for the four chelates, there is more than one potential coordination site, which makes them able, at the very first moment, to act as cross-links.

In case of 3amt adducts, the overall decrease observed for the symmetrical and asymmetrical N-H bands suggests that the NH2 group is engaged with the coordination. Furthermore, positive shifts observed for the δ1 bands reinforce this statement. It is worth noting that the observed shifts are more intense to Sn(II) adducts than to Cd(II), which is probably due to the higher acidity of Sn(II) (larger nuclear effective charge: 5.65 for Sn and 4.35 for Cd). It is verified that the symmetrical N-H bands are more sensitive to this acidity difference since a positive shift is observed for Cd(II) adducts, whereas a negative shift of this band is verified to Sn(II) adducts.

In case of 4amp approximation, the same general orientation is observed for asymmetric and symmetric N-H bands. This fact indicates that in this case, NH2 is involved to a slight degree in the metallic coordination. This hypothesis is reinforced by the fact that for 4amp, the ringed breathing
Table 1: Mass weight and carbon, hydrogen, and nitrogen elemental analysis data for the examined compounds.

| Adducts                                      | M.wt  | % C  | % H  | % N  |
|----------------------------------------------|-------|------|------|------|
| SnCl₂(3amt).H₂O                             | 291.68| 8.22 | 8.11 | 2.05 |
| SnCl₂(3amt).H₂O                             | 375.76| 12.77| 12.25| 2.66 |
| CdCl₂(3amt)                                 | 267.37| 8.97 | 8.89 | 1.49 |
| SnCl₂(2mct).0.5H₂O                          | 317.81| 11.33| 10.99| 1.89 |
| SnCl₂(2mct)                                 | 428.02| 16.82| 16.32| 2.33 |
| CdCl₂(2mct)                                 | 302.52| 11.90| 11.74| 1.65 |
| SnCl₂(2mcp).1.5H₂O                          | 327.77| 18.31| 18.19| 2.44 |
| SnCl₂(2mcp).4H₂O                            | 483.94| 24.80| 24.57| 3.72 |
| CdCl₂(2mcp)                                 | 345.68| 6.94 | 6.91 | 3.47 |
| SnCl₂(4amt).4H₂O                            | 384.76| 12.48| 12.34| 2.86 |
| SnCl₂(4amt).1.5H₂O                          | 285.37| 8.40 | 8.34 | 2.10 |

Table 2: Major infrared bands (cm⁻¹) for 3amt and its Cd(II) and Sn(II) adducts.

| 3amt       | CdCl₂-(3amt) | CdCl₂-2(3amt) | SnCl₂-(3amt) | SnCl₂-2(3amt) | Assignments  |
|------------|--------------|---------------|--------------|--------------|--------------|
| 3398 s     | 3419 ms      | 3440 ms       | 3312 w       | 3349 vw      | vₐₙ (N-H); NH₂ |
| 3326 mw    | 3356 w       | 3312 ms       | 3310 vm      | 3258 vw      |              |
| 3182 mw    | 3213 w       | 3212 ms       | 3155 mw      | 3155 ms      | vₑ (N-H); NH₂ |
| 1647 vs    | 1652 vs      | 1645 w, sh    | 1688 vs      | 1677 vs      | δₚ(NH₂)      |
| 1590 s     | 1573 w       | 1594 vs       | 1569 ms      | 1566 ms      | v(C-N)        |
| 1533 s     | 1537 s       | 1560 vs       | 1479 vs      |              | v(N-N)        |
| 1418 s     | 1429 ms      | 1374 vs       | 1405 mw      | 1415 w       | Ring breathing bands |
| 1389 ms    | 1373 w       | 1332 ms       | 1395 ms      | 1326 s       |              |
| 1275 vs    | 1283 s       | 1248 vs       | 1257 ms      | 1263 w       | ρₛ (NH₂)     |
| 1217 vs    | 1250 vw      | 1213 ms       | 1125 vw      | 1125 ms      | v(C-N)        |
| 1045 vs    | 1088 w       | 1083 vs       | 1048 s       | 1066 s       | ρₒ (NH₂)     |
| 945 vs     | 1057 s       | 1011 vs       | 951 vs       | 950 vs       |              |
| 873 vs     | 901 s        | 884 s         | 860 ms       | 867 s        | ρₜ (NH₂)     |
| 830 s      | 740 ms       | 747 sh        | 748 w, sh    | 773 ms       |              |
| 729 vs     | 693 vs       | 726 ms        | 694 vs       | 642 ms       |              |

Table 3: Major infrared bands (cm⁻¹) for 4amt and its Cd(II) and Sn(II) adducts.

| 4amt       | CdCl₂-(4amt) | CdCl₂-2(4amt) | SnCl₂-(4amt) | SnCl₂-2(4amt) | Assignments  |
|------------|--------------|---------------|--------------|--------------|--------------|
| 3312 w     | 3467 ms      | 3303 s        | 3417 w       | 3418 w, br   | vₐₙ (N-H); NH₂ |
| 3197 w     | 3199 s       | 3198 s        | 3127 ms      | 3129 w, br   | vₑ (N-H); NH₂ |
| 319 w      | 3136 s       | 3105 s        |              |              |              |
| 1647 vs    | 1618 vs      | 1618 vs       | 1623 vs      | 1685 w       | δₚ(NH₂)      |
Table 3: Continued.

| 4amt | CdCl₂-(4amt) | CdCl₂-2(4amt) | SnCl₂-(4amt) | SnCl₂-2(4amt) | Assignments |
|------|--------------|--------------|--------------|--------------|-------------|
| 1533 s | 1543 s | 1537 s | 1539 ms | 1631 vs | 1529 s |
| 1475 ms | 1474 mw | 1470 w | 1465 vw | 1465 vw | Ring breathing bands |
| 1404 s | 1398 s | 1394 w | 1402 vw | 1412 vw |
| 1188 s | 1209 s | 1209 ms | 1207 ms | 1205 s | \( \rho_t \) (NH₂) |
| 1074 s | 1145 vw | 1078 s | 1164 vw | 1075 s |
| 1016 vw | 1025 s | 1015 s | 1034 s | 1033 s | \( \rho_w \) (NH₂) |
| 959 ms | 980 vs | 984 ms | 934 ms | 935 ms |
| 873 s | 908 w | 894 ms | 871 ms | 875 ms | \( \rho_l \) (NH₂) |
| 672 w | 874 s | 845 vw | 690 w, sh | 661 ms |
| — | 689 ms | 686 ms | | |

\( s \) = strong; \( w \) = weak; \( m \) = medium; \( sh \) = shoulder; \( v \) = very; \( br \) = broad; \( \nu \) = stretching; \( \delta \) = bending.

Table 4: Major infrared bands (cm\(^{-1}\)) for 2mct and its Cd(II) and Sn(II) adducts.

| 2mct | CdCl₂-(2mct) | CdCl₂-2(2mct) | SnCl₂-(2mct) | SnCl₂-2(2mct) | Assignments |
|------|--------------|--------------|--------------|--------------|-------------|
| 2852 w | 3258 s | 3258 s | 3443 s, br | 3447 ms, br | \( \nu_s \) (C-H); –CH₂ |
| — | 2948 vw | 3136 ms | 3206 ms | 3207 w |
| — | 2998 w | 3144 w | 2997 w | 2997 w |
| — | 2947 w | 2929 w | 2845 w | |

\( s \) = strong; \( w \) = weak; \( m \) = medium; \( sh \) = shoulder; \( v \) = very; \( br \) = broad; \( \nu \) = stretching; \( \delta \) = bending.

Table 5: Major infrared bands (cm\(^{-1}\)) for 2mcp and its Cd(II) and Sn(II) adducts.

| 2mcp | CdCl₂-(2mcp) | CdCl₂-2(2mcp) | SnCl₂-(2mcp) | SnCl₂-2(2mcp) | Assignments |
|------|--------------|--------------|--------------|--------------|-------------|
| — | 3458 ms, br | 3448 ms, br | 3421 ms, br | 3423 ms, br | \( \nu_(O-H) \); H₂O |
| — | 3196 ms | 3172 ms | 3216 ms | 3073 vw |
| — | 3126 ms | 3087 s | 3135 w | |

\( s \) = strong; \( w \) = weak; \( m \) = medium; \( sh \) = shoulder; \( v \) = very; \( br \) = broad; \( \nu \) = stretching; \( \delta \) = bending.
bands exhibit a negative shift (compared to free chelates and synthesized solid adducts), whereas positive shifts are observed in 3amp. Therefore, for 4amp adducts, the two “isolated” nitrogen atoms are the main coordination sites.

Suggested coordination modes for 3amp and 4amp molecules are shown schematically in Figure 2.

As expilciatory examples, the infrared spectra of 3amp solid adducts are shown in Figure 3.

Figure 2: Schematic representation of the suggested coordinative characteristics for (a) 3amt and (b) 4amt adducts.

Figure 3: Infrared spectra for (a) CdCl₂-(3amt), (b) CdCl₂-2(3amt), (c) SnCl₂-(3amt).H₂O, and (d) SnCl₂-2(3amt).H₂O.
Figure 4: Thermogravimetric curves for the 16 solid adducts.

Table 6: TG data summary for Sn(II) and Cd(II) adducts with 3amt, 4amt, 2mcp, and 2mct.

| Adduct                  | Step | t_i (°C) | Degradation t_f (°C) | Process onset (°C) | Mass loss (%) |
|-------------------------|------|----------|----------------------|--------------------|---------------|
| SnCl_2.(3amt).H_2O      | 1    | 55       | 382                  | 238                | 26.7          |
|                         | 2    | 385      | 560                  | 473                | 35.5          |
|                         | 1    | 45       | 102                  | 76                 | 5.1           |
| SnCl_2.2(3amt).H_2O     | 2    | 160      | 445                  | 320                | 40.0          |
|                         | 3    | 447      | 606                  | 517                | 27.2          |
|                         | 1    | 280      | 331                  | 307                | 53.7          |
| CdCl_2.(3amt)           | 2    | 332      | 406                  | 348                | 12.8          |
|                         | 3    | 478      | 660                  | 569                | 29.3          |
|                         | 1    | 188      | 271                  | 234                | 24.1          |
| CdCl_2.(2mct)           | 2    | 348      | 448                  | 401                | 16.8          |
|                         | 3    | 449      | 681                  | 587                | 43.0          |
|                         | 1    | 31.4     | 83                   | 50                 | 3.3           |
| SnCl_2.(2mct).0.5H_2O   | 2    | 127      | 344                  | 237                | 43.2          |
|                         | 3    | 346      | 461                  | 404                | 16.8          |
|                         | 4    | 462      | 615                  | 513                | 6.2           |
|                         | 1    | 120      | 260                  | 204                | 44.0          |
|                         | 2    | 261      | 341                  | 288                | 10.2          |
| SnCl_2.(2mct)           | 3    | 342      | 430                  | 378                | 4.7           |
|                         | 4    | 431      | 595                  | 513                | 8.9           |
|                         | 5    | 596      | 656                  | 607                | 1.1           |
| CdCl_2.(2mct)           | 1    | 181      | 276                  | 223                | 30.2          |
|                         | 2    | 497      | 650                  | 583                | 41.3          |
|                         | 1    | 136      | 276                  | 207                | 43.2          |
| CdCl_2.(2mct).H_2O      | 2    | 277      | 485                  | 384                | 15.4          |
|                         | 3    | 486      | 628                  | 551                | 9.3           |
### Table 6: Continued.

| Adduct                  | Step | \( t_i \) (°C) | Degradation \( t_f \) (°C) | Process onset (°C) | Mass loss (%) |
|-------------------------|------|----------------|---------------------------|-------------------|--------------|
| \( \text{SnCl}_2.(2\text{mcp}).1.5\text{H}_2\text{O} \) | 1    | 110            | 199                       | 154               | 8.0          |
|                         | 2    | 200            | 355                       | 285               | 55.4         |
|                         | 3    | 421            | 654                       | 534               | 3.1          |
|                         | 1    | 103            | 221                       | 154               | 14.3         |
| \( \text{SnCl}_2.2(2\text{mcp}).4\text{H}_2\text{O} \) | 2    | 222            | 349                       | 302               | 82.1         |
|                         | 3    | 584            | 670                       | 585               | 1.8          |
|                         | 1    | 186            | 309                       | 244               | 23.3         |
| \( \text{CdCl}_2.(2\text{mcp}) \) | 2    | 310            | 459                       | 391               | 12.6         |
|                         | 3    | 460            | 647                       | 576               | 47.0         |
|                         | 4    | 648            | 751                       | 675               | 8.6          |
|                         | 1    | 139            | 274                       | 215               | 35.8         |
| \( \text{CdCl}_2.2(2\text{mcp}) \) | 2    | 276            | 349                       | 301               | 5.2          |
|                         | 3    | 351            | 482                       | 399               | 10.1         |
|                         | 4    | 483            | 649                       | 572               | 26.9         |
| \( \text{SnCl}_2.(4\text{amt}).4\text{H}_2\text{O} \) | 3    | 300            | 425                       | 355               | 37.0         |
|                         | 4    | 428            | 616                       | 515               | 10.2         |
|                         | 1    | 63             | 191                       | 133               | 6.8          |
|                         | 2    | 192            | 319                       | 257               | 39.2         |
| \( \text{SnCl}_2.2(4\text{amt}).1.5\text{H}_2\text{O} \) | 3    | 320            | 407                       | 358               | 18.2         |
|                         | 4    | 497            | 619                       | 545               | 14.2         |
|                         | 1    | 88             | 176                       | 123               | 6.7          |
|                         | 2    | 274            | 381                       | 328               | 21.1         |
| \( \text{CdCl}_2.(4\text{amt}).\text{H}_2\text{O} \) | 3    | 382            | 459                       | 407               | 7.9          |
|                         | 4    | 460            | 655                       | 583               | 51.3         |
|                         | 1    | 195            | 289                       | 256               | 17.9         |
|                         | 2    | 290            | 390                       | 324               | 23.7         |
| \( \text{CdCl}_2.2(4\text{amt}) \) | 3    | 392            | 456                       | 420               | 4.8          |
|                         | 4    | 457            | 679                       | 583               | 47.5         |

\( t_i \) and \( t_f \) are the initial and final temperatures of the thermal degradation process, respectively.

### Table 7: Antimicrobial activities (inhibition zone diameter, mm/µg sample) of papaverine and its metal complexes against Gram-positive bacteria, Gram-negative bacteria, and two types of fungi.

| The adducts                  | Gram-negative bacteria | Gram-positive bacteria | Fungi |
|------------------------------|------------------------|------------------------|-------|
|                              | \( \text{E. coli} \) | \( \text{P. aeruginosa} \) | \( \text{B. subtilis} \) | \( \text{S. aureus} \) | \( \text{A. flavus} \) | \( \text{C. albicans} \) |
| Control: DMSO                | 0.0                    | 0.0                    | 0.0   | 0.0   | 0.0   | 0.0   |
| Ampicillin (Antibacterial agent) | 22                    | 17                    | 0.0   | 20    | 0.0   | 0.0   |
| Amphotericin B (Antifungal agent) | 0.0                   | 0.0                   | 0.0   | 0.0   | 0.0   | 0.0   |
| \( \text{SnCl}_2.(3\text{amt}).\text{H}_2\text{O} \) | 10                  | 10                    | 12    | 11    | 0.0   | 23    |
| \( \text{SnCl}_2.2(3\text{amt}).\text{H}_2\text{O} \) | 15                  | 13                    | 13    | 15    | 0.0   | 14    |
| \( \text{CdCl}_2.(3\text{amt}) \) | 0.0                  | 0.0                   | 0.0   | 0.0   | 0.0   | 0.0   |
| \( \text{CdCl}_2.2(3\text{amt}) \) | 12                  | 12                    | 12    | 15    | 0.0   | 11    |
| \( \text{SnCl}_2.(2\text{mct}).0.5\text{H}_2\text{O} \) | 9                   | 0.0                   | 10    | 10    | 0.0   | 0.0   |
| \( \text{SnCl}_2.2(2\text{mct}) \) | 9                   | 9                     | 9     | 21    | 0.0   | 0.0   |
| \( \text{CdCl}_2.(2\text{mct}) \) | 16                  | 15                    | 14    | 20    | 0.0   | 14    |
| \( \text{CdCl}_2.2(2\text{mct}).\text{H}_2\text{O} \) | 26                 | 28                    | 26    | 32    | 0.0   | 35    |
| \( \text{SnCl}_2.(2\text{mcp}).1.5\text{H}_2\text{O} \) | 16                 | 15                    | 16    | 21    | 0.0   | 9     |
| \( \text{SnCl}_2.2(2\text{mcp}).4\text{H}_2\text{O} \) | 13                 | 14                    | 14    | 14    | 0.0   | 0.0   |
| \( \text{CdCl}_2.(2\text{mcp}) \) | 11                  | 10                    | 11    | 10    | 0.0   | 16    |
| \( \text{CdCl}_2.2(2\text{mcp}) \) | 14                 | 13                    | 12    | 16    | 0.0   | 13    |
| \( \text{SnCl}_2.(4\text{amt}).4\text{H}_2\text{O} \) | 9                  | 0.0                   | 0.0   | 0.0   | 0.0   | 0.0   |
| \( \text{SnCl}_2.2(4\text{amt}).1.5\text{H}_2\text{O} \) | 10                 | 9                     | 12    | 13    | 0.0   | 9     |
| \( \text{CdCl}_2.(4\text{amt}) \) | 0.0                 | 0.0                   | 0.0   | 0.0   | 0.0   | 0.0   |
| \( \text{CdCl}_2.2(4\text{amt}) \) | 9                  | 0.0                   | 0.0   | 9     | 0.0   | 0.0   |
In case of 2mct adducts, positive shifts of the νC=N band are observed for Sn(II) adducts, whereas negative shifts are verified to Cd(II) adducts. Such fact suggests a coordination through nitrogen to Sn(II) and a coordination through sulfur to Cd(II) in agreement with the fact that the nitrogen atom is a hard base and that Sn(II) is a harder acid than Cd(II).

For 2mcp adducts, the negative shifts observed for the ν(C-S); C-SH and ν(C=N) aromatic bands suggest that, in this case, both coordination sites N and S are involved in the coordination process for both cations considered.

The data of thermogravimetric curves for the 16 solid adducts are demonstrated in Figure 4. The main TG data are elucidated in Table 6. For each TG curve, the experimental mass losses (±5%) are similar to the proposed formulas. It is possible to verify that for all hydrated compounds, the first mass loss step is associated with the release of water molecules followed by the release of ligand molecules and sublimation of the metal chloride. Furthermore, it is verified that, considering only the release of ligand molecules (3amp, 4amp, 2mct, or 2mcp), the cadmium adducts are always more stable than the correspondent tin adducts. Since the infrared data suggest that the metal-to-ligand interaction is higher for tin adducts, this last result is an expected one, unless we take into account that the cadmium adducts generally polymerize [22–27] and so there is, probably for these compounds, the formation of cross-linking bonds, leading to more stable compounds, from a thermal point of view.

The antimicrobial effect of the adducts was measured against a variety of microorganisms including bacteria and fungus (Table 7 and Figure 5). The no-growth zones around the hole indicated the inhibiting activity of the adducts on the microbe. (These were calculated and compared with the ampicillin as an antibacterial agent or amphotericin B as an antifungal agent. The adduct CdCl₂.2(2mct).H₂O and CdCl₂.(2mct) showed the highest antimicrobial activities followed by SnCl₂.(2mct).1.5H₂O among all other adducts. On other hand, the CdCl₂.(3amt) and CdCl₂.(4amt).H₂O have no effect on any bacteria or fungal strains (Table 7). The antimicrobial activities of these adducts might be caused by a...
direct interaction of Cd (II) or Sn (II) ions with proteins, enzymes, nucleic acids, and membranes of microbe cells.

4. Conclusion

The adducts SnCl₂(3amt).H₂O, SnCl₂.2(3amt).H₂O, CdCl₂.3(amt), CdCl₂.2(3amt), SnCl₂.2(0.5H₂O), SnCl₂.2(2mct), CdCl₂.2(2mct), CdCl₂.2(2mct).H₂O, SnCl₂.2(2mcp).1.5H₂O, SnCl₂.2(2mcp).H₂O, CdCl₂.2(2mcp), CdCl₂.2(2mcp).SnCl₂.4(amt).H₂O, SnCl₂.2(2mct).SnCl₂.4(amt).1.5H₂O, CdCl₂.2(4amt).H₂O, and CdCl₂.2(4amt)—where 3amt = 3-amino-1,2,4-triazole; 4amt = 4-amino-1,2,4-triazole; 2mct = 2-mercaptothiazoline; and 2mcp = 2-mercaptopyridine—were synthesized by a solid-state route and characterized by CHN elemental analysis and infrared spectroscopy. A thermogravimetric study was also performed. It is verified that, for all compounds, the monoadducts are the most stable ones. Such fact agrees with a higher ionic and covalent character of the metal-ligand bond for such compounds. From the result, it can be concluded that 14 of the 16 compounds have a good biological activity against these microorganisms.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare no conflicts of interest.

Acknowledgments

The authors are grateful to Taif University for supplying essential facilities and acknowledge the support of Taif University Researchers Supporting Project (TURSP-2020/39), Taif University, Taif, Saudi Arabia.

References

[1] G. L. Almajan, S.-F. Barbuceanu, G. Bancescu, I. Saramet, and C. Draghici, "Synthesis and antimicrobial evaluation of some fused heterocyclic [1,2,4]triazolo[3,4-b][1,3,4]thiazidazole derivatives," European Journal of Medicinal Chemistry, vol. 45, no. 12, pp. 6139–6146, 2010.

[2] A. Demirbas, D. Sahin, N. Demirbas, and S. A. Karaoglu, "Synthesis of some new 1,3,4-thiadiazol-2-ylmethyl-1,2,4-triazole derivatives and investigation of their antimicrobial activities," European Journal of Medicinal Chemistry, vol. 44, no. 7, pp. 2896–2903, 2009.

[3] I. A. Imkan, S. Ullah, M. Imran et al., "Synthesis of bio-compatible triazole based non-ionic surfactant and its vesicular drug delivery investigation," Chemistry and Physics of Lipids, vol. 228, Article ID 104894, 2020.

[4] S. K. Yadav, R. K. Yadav, and U. Yadava, "Computational investigations and molecular dynamics simulations envisioned for potent antioxidant and anticancer drugs using indole-chalcone-triazole hybrids," DNA Repair, vol. 86, Article ID 102765, 2020.

[5] Z. Xu, "1,2,3-Triazole-containing hybrids with potential antibacterial activity against methicillin-resistant Staphylococcus aureus (MRSA)," European Journal of Medicinal Chemistry, vol. 206, Article ID 112686, 2020.

[6] F. Gao, T. Wang, J. Xiao, and G. Huang, "Antibacterial activity study of 1,2,4-triazole derivatives," European Journal of Medicinal Chemistry, vol. 175, pp. 274–281, 2019.

[7] G. Nuray and K. Ömer, "Synthesis and antimicrobial activity evaluation of new 1,2,4-triazoles and 1,3,4-thiadiazoles bearing imidazo[2,1-b]thiazole moiety," European Journal of Medicinal Chemistry, vol. 45, pp. 63–68, 2010.

[8] E. Ilkay, S. Günniz, S. R. Kückükgüzeld, and C. Erik De, "Synthesis of some novel thiourea derivatives obtained from 5-[(4-aminophenoxy)methyl]-4-alkyl-2,4-dihydro-3H-1,2,4-triazole-3-thiones and evaluation as antiviral/anti-HIV and anti-tuberculosis agents," European Journal of Medicinal Chemistry, vol. 43, pp. 381–392, 2008.

[9] S. Ilkay, S. Rollas, and K. Muammer, "Some 3-Thioxo/alkylthio-1,2,4-triazoles with a substituted thiourea moiety as possible antymycobacterials," Bioorganic & Medicinal Chemistry Letters, vol. 11, pp. 1703–1707, 2001.

[10] A. Charalabos, M. S. Ana Ciric, and M. Z. Panagiotis, "Sulfonamide-1,2,4-thiadiazole derivatives as antifungal and antibacterial agents: synthesis, biological evaluation, lipophilicity, and conformational studies," Chemical and Pharmaceutical Bulletin, vol. 58, pp. 160–167, 2010.

[11] S. Imtiaz, N. H. R. Shahid Hameed, A. Muhammad, Z. Reaz Uddin, and M. Ajmal Khan, "Synthesis, antioxidant activities and urease inhibition of some new 1,2,4-triazole and 1,3,4-thiadiazole derivatives," European Journal of Medicinal Chemistry, vol. 45, pp. 5200–5207, 2010.

[12] N. Rezki, M. A. Almehmadi, S. Ihmaid et al., "Novel scaffold hopping of potent benzothiazole and isatin analogues linked to 1,2,3-triazole fragment that mimic quinazoline epidermal growth factor receptor inhibitors: synthesis, antitumor and mechanistic analyses," Bioorganic Chemistry, vol. 103, Article ID 104133, 2020.

[13] S. Harish, A. Suroor, and M. A. Khan, "1,3,4-Oxadiazole/thiadiazole and 1,2,4-triazole derivatives of biphenyl-4-yloxy acetic acid: synthesis and preliminary evaluation of biological properties," European Journal of Medicinal Chemistry, vol. 43, pp. 2688–2698, 2008.

[14] F. Samir, H. M. Barseoum, and A. A. Marzouk, "Synthesis and anti-inflammatory activity of some pyrazole derivatives," Medicinal Chemistry Research, vol. 21, pp. 1722–1733, 2012.

[15] G. Rajesh and S. C. Jitendra Sainy, "Synthesis and biological evaluation of 2-amino-5-sulfanyl-1,3,4-thiadiazole derivatives as antidepressant, anxiolytics and anticonvulsant agents," Medicinal Chemistry Research, vol. 20, pp. 245–253, 2011.

[16] R. F. de Farias, L. M. Nunes, and C. Airoldi, "Thermodynamic study about histidine and cysteine-cadmium interactions," Journal of Thermal Analysis and Calorimetry, vol. 74, no. 3, pp. 923, 2003.

[17] R. F. de Farias, A. O. da Silva, and U. G. da Silva Jr., "Synthesis, characterization and TG-DSC study of cadmium halides adducts with caffeine," Thermochimica Acta, vol. 406, no. 1-2, pp. 245, 2003.

[18] R. F. de Farias, L. Martinez, and C. Airoldi, "Synthesis, characterization and a thermogravimetric study of copper, cobalt and tin mono- and bis-adducts with ethylenurea, ethylenethiourea and propyleneurea," Transition Metal Chemistry, vol. 27, p. 748, 2002.

[19] L. Martinez, R. F. de Farias, and C. Airoldi, "Thermochemical data on adducts of copper chloride with the amino acids lysine and glycine," Thermochimica Acta, vol. 395, p. 21, 2002.
[20] R. F. de Farias, C. Airoldi, and H. Scatena Jr., “Empirical correlations involving calorimetric, thermogravimetric and infrared data for zinc halides adducts,” Polyhedron, vol. 21, no. 17, p. 1677, 2002.

[21] A. Gaber, W. F. Alsanie, D. N. Kumar, M. S. Refat, and E. M. Saied, “Novel papaverine metal complexes with potential anticancer activities,” Molecules, vol. 25, no. 22, p. 5447, 2020.

[22] M. O. Machado, R. F. de Farias, and C. Airoldi, “Two different synthetic routes involving the reaction of dodecylamine or nicotinamide with crystalline lamellar vanadylphosphate,” Journal of Physics and Chemistry of Solids, vol. 65, no. 10, p. 1697, 2004.

[23] R. F. de Farias and A. O. da Silva, “Nickel(II) and cerium(IV) ethyleneurea, ethylenethiourea and propyleneurea adducts,” Transition Metal Chemistry, vol. 28, p. 21, 2003.

[24] R. F. de Farias and C. Airoldi, “The first VOPO4·2H2O intercalation compound synthesized through a solid-state reaction at room temperature,” Journal of Solid State Chemistry, vol. 166, no. 2, p. 277, 2002.

[25] R. F. de Farias and C. Airoldi, “Calorimetric study of the adducts CdBr2·nL (n = 1 and 2; L = ethyleneurea and propyleneurea),” Thermochimica Acta, vol. 390, no. 1-2, p. 213, 2002.

[26] R. F. Farias, G. C. Petrucelli, and C. Airoldi, “Thermochemical data on adducts of cyclic ureas and copper chloride: the resemblance to biological systems,” Thermochimica Acta, vol. 376, no. 1, p. 1, 2001.

[27] N. N. Greenwood and A. Earnshaw, Chemistry of the Elements, Butterworth-Heinemann, Cambridge, UK, 1984.