

## RESEARCH ARTICLE

# Synthesis, Characterization and Bioactivity Study of Few Metal Complexes of Curcumin with 2-(1H-Benzimidazol-2-yl) Aniline

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Received: 09<sup>th</sup> June, 2022; Revised: 20<sup>th</sup> July, 2022; Accepted: 30<sup>th</sup> August, 2022; Available Online: 25<sup>th</sup> September, 2022

## ABSTRACT

We have synthesized many metal (II) complexes using curcumin L1 as the major ligand and 2-(1H-Benzimidazol-2-yl) aniline L2 as a supporting ligand. The complexes were characterized by spectroscopy methods such as; molar conductivity, elements microanalysis, Fourier-transform spectroscopy (FT-IR), UV-vis, and mass spectroscopy. Both curcumin ligands and L2 were found to be capable of binding to M(II) and metal ions via their two N atoms, according to the data. The formula for the complexes is the same.  $[M(L_1)(L_2)H_2OCl]$ , where M is Ni(II), Co(II), Cu(II), Cd(II), and Hg(II) (II). Octahedral complexes are proposed for the prepared compounds. The bio-actives suggested that the complexes are effective against bacteria and fungus on a mild to moderate level.

**Keywords:** Bacteria, Bidentate, Curcumin, Fungi, Supporting Ligand.

International Journal of Drug Delivery Technology (2022); DOI: 10.25258/ijddt.12.3.25

**How to cite this article:** Rashid, KW, Abdul Karem, LK, Rasheed, MK, Synthesis, Characterization and Bioactivity Study of Few Metal Complexes of Curcumin with 2-(1H-Benzimidazol-2-yl) Aniline. International Journal of Drug Delivery Technology. 2022;12(3):1076-1080.

**Source of support:** Nil.

**Conflict of interest:** None

## INTRODUCTION

Curcumin, (bis[4-hydroxy-3-methoxyphenyl]-1,6-heptadiene-3,5-dione), is an antioxidant found in turmeric, a yellow spice,<sup>1-3</sup> analgesic<sup>4,5</sup> as well as anticancer properties.<sup>5,6</sup> Certain research on curcumin have been conducted using the structure of ionic while keto-enol equilibrium present or when completely at the form of Keto with resulting attributes varying.<sup>7-9</sup> Curcumin's potent chelating capacity toward a wide variety of metal ions has been thoroughly studied; thus, curcumin may play a significant role treatments for mineral toxicity and overload chelation.<sup>10</sup> Curcuma is available in the keto-enol form. Curcuma was used for studying development of different complexes from metal.<sup>10-12</sup> Curcumin's stoichiometry with certain metal ions has also been observed.<sup>10, 12</sup> We synthesized, identified, and examined the bioactivity of curcumin complexes with As the five-coordinate.<sup>13</sup>

Metal, gold curcumin complexes with VO(II), Ga(III), and In(III) demonstrate therapeutic activity, Applications.<sup>14</sup> The complexes of curcumin with Eu, Ce, La, Y, Cr, and Pd demonstrated that curcumin coordinates with metal ions in the bidentate mode in the deprotonated form.<sup>15, 16</sup> Additionally, curcumin can form strong chelates with the transition metal ions Ni(II), Zn(II), Pd(II), Fe(III), Cr(III), and Mn(II).<sup>17-19</sup> Curcumin compounds also Zn(II) Cu(II), Mg(II), and Se(II) are synthesised mechanically avoiding using the typical

organic solvents so it's founds to the form of complexes with 1:1 ratio to Zn(II) Cu(II) Mg(II) complexes and 1:2 to Se(II) complexes.<sup>20</sup> Cobalt (III) complexes with curcumin and other ligands have been produced and described.<sup>21</sup> The preparation and identification of mixed ligand complexes from Schiff Base L<sub>3</sub> via the interaction of (istain with 2-aminopyridine), curcumin HL1, and azide ion N<sub>3</sub> with a variety of metal ions follows the general formula: Where  $[M L_1 L_3 N_3 H_2O]$ : M(II) = Co, Ni, Cu and  $[M L_1 L_3 N_3]$ , M(II) = (Mn, Zn, Cd, Hg). The proposed geometry for all complexes was octahedral.<sup>22</sup> This paper describer the synthesis and characterization of some transition metal ions mixed ligand complexes of curcuma L<sub>1</sub> and 2-(1H-Benzimidazol-2-yl) aniline L<sub>2</sub> and biological evaluation (*in-vitro* antibacterial activities) of the complexes were tested, in addition to the study of thermodynamic functions of Co complex. The behavior of two ligands toward M(II) ion was binary

## EXPERIMENTAL

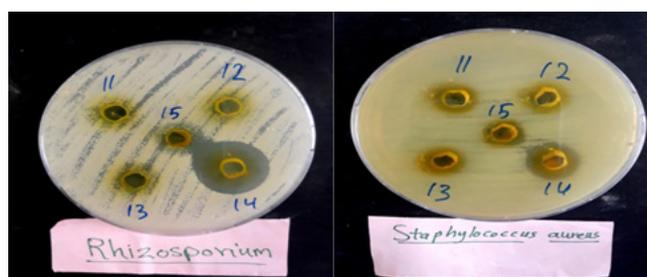
All chemicals were in its unprocessed form. The Stuart melting point instrument was used to record melting points. Atomic Absorption A was used to determine the metal content of the complexes. Japan A.A-67G Shimadzu is used in this procedure. The complexes' electrical conductivity was measured at the time pw 9527 Digital conductivity meter (Philips).

**Table 1:** The physical properties of the compounds

Compounds	M.Wt	$\Omega$	Theoretical % / practical %				
			C	H	N	M	Cl
L <sub>1</sub>	368	----	68.47 68.46	5.47 5.46	---	----	----
L <sub>2</sub>	209	----	74.62 74.61	5.30 5.29	20.08 20.07	---	----
[Co (L <sub>1</sub> )(L <sub>2</sub> )H <sub>2</sub> O C <sub>1</sub> ]	689.02	10	59.27 59.15	4.68 4.65	6.10 6.12	8.55 8.50	5.15 5.05
[Ni (L <sub>1</sub> )(L <sub>2</sub> )H <sub>2</sub> O C <sub>1</sub> ]	688.78	16	59.29 59.00	4.34 4.29	5.66 5.76	8.52 8.49	5.15 4.99
[Cu (L <sub>1</sub> )(L <sub>2</sub> )H <sub>2</sub> O C <sub>1</sub> ]	693.63	13	58.87 58.76	4.65 4.60	6.06 6.73	9.16 9.11	5.11 5.02
[Cd (L <sub>1</sub> )(L <sub>2</sub> ) H <sub>2</sub> O C <sub>1</sub> ]	742.50	8	55.00 54.91	4.34 4.22	5.66 5.82	15.14 15.01	4.77 4.68
[Hg (L <sub>1</sub> )(L <sub>2</sub> ) H <sub>2</sub> O C <sub>1</sub> ]	830.68	2	49.16 49.05	3.88 3.76	5.06 5.32	24.15 24.09	4.27 4.25

**Table 2:** The band of infrared for L1, L2 also their metal complexes.<sup>24</sup>

Compound	$\nu$ (O-H)	NH <sub>2</sub>	C-H arom. C-H alph.	C=O	(C=N)	C=C	( $\nu$ OH <sub>2</sub> ) <sub>aq.</sub> (M-N)	(M-O)
L <sub>1</sub>	----	3429 3346	3172 3045	---	1650	1612 1575	----	----
L <sub>2</sub>	3512-3417	---	3012	1627	---	1508	----	----
Co <sup>-1</sup>	3565-3481	3415 3307	3163, 3012 2933	1683	1623	1587	850 603	470
Ni <sup>-1</sup>	3581	3415 3234	3120 2933	1683	1639	1618 1510	844 596	489
Cu <sup>-1</sup>	3564	3413 3234	3120 2929	1683	1620	1565 1510	844 622	470
Cd <sup>-1</sup>	3558	3413 3234	3197 2925	1685	1620	1566 1508	621	476
Hg <sup>-1</sup>	3556	3413 3234	3196 2925	1676	1616	1564 1510	621	466



**Figure 1:** Diameter of inhibition of the second group of complexes of two types of *E. coli* and *Staphylococcus* and *Rhizosporium* fungi, where the numbers 11,12,13,14 and 15 means Cd, Ni, Co, Hg and Cu complexes

FT-IR Spectra were acquired using a Shimadzu<sup>24</sup> FT-IR 8400s as KBr discs. The electronic spectra of the synthesized compounds were determined in the range 1100-200 nm using a Shimadzu-UV- 160 Ultraviolet Visible-Spectrophotometer. Magnetic susceptibility was determined using the Faraday method at 298 K using a Bruker magnet BM6 device.

### SYNTHESIS OF THE TERNARY COMPLEXES OF CURCUMIN (L1) WITH 2-(1H-BENZIMIDAZOL-2-YL) ANILINE (L2).

The metal (II)– curcuma complex had synthesized under the mixture of equi -molar amounts of CoCl<sub>2</sub>.6H<sub>2</sub>O, NiCl<sub>2</sub>.6H<sub>2</sub>O, CuCl<sub>2</sub>.2H<sub>2</sub>O, CdCl<sub>2</sub>.2H<sub>2</sub>O, HgCl<sub>2</sub> 1.0 mmol and curcumin (0.160 g, 1.0 mmol) in ethanol and 0.209 g (1.0 mmole) 2-(1H-Benzimidazol-2-yl) aniline added and reaction continued for period of two hours of stirring, a precipitate was filtered to obtain the precipitate, washed with ethanol, and then dried.

### ANTIMICROBIAL RESEARCH

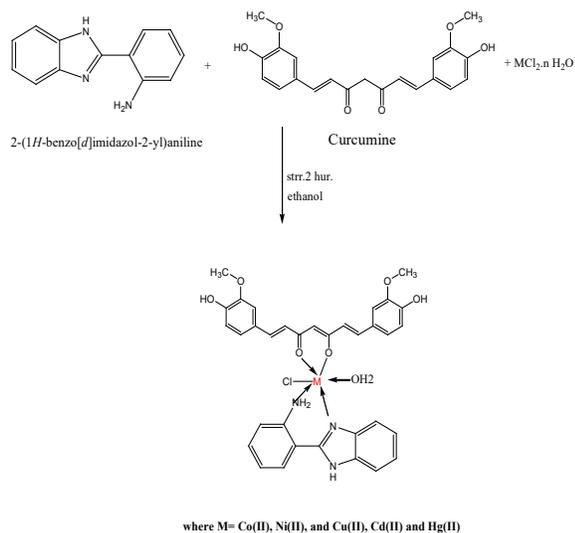
The antibacterial activity of the ligands and their complexes was determined using the agar diffusion technique<sup>24</sup> 100 mL of a 5 mg/mL solution in DMSO was evaluated. Gram-positive microorganisms such as *S. aureus* gram-negative bacteria such as *E. coli*, and fungus such as *Rhizosporium* were tested. On nutritional agar medium, bacteria and fungi were maintained. Different test microorganisms were added

**Table 3:** UV-visible spectrum of L1, L2 and their complexes

Compounds	$\lambda_{nm}$	$U\text{ cm}^{-1}$	$\epsilon_{max}$	Transition type
L <sub>1</sub>	246	40650	2211	$\pi \rightarrow \pi^*$
	341	29325	553	$\pi \rightarrow \pi^*$
	419	23866	35	$n \rightarrow \pi^*$
L <sub>2</sub>	273	36630	124	$\pi \rightarrow \pi^*$
	430	23255	2115	$n \rightarrow \pi^*$
Co	270	37037	139	L.F
	433	23094	1549	C.T
	551	19963	15	${}^4T_{1g(F)} \rightarrow {}^4T_{1g(P)}$
Ni	263	38022	2754	L.F
	313	31948	2626	L.F
	425	23529	153	C.T
	570	17543	18	${}^3A_{2g(F)} \rightarrow {}^3T_{1g(F)}$
Cu	270	37037	327	L.F
	434	23041	1476	C.T
	629	15898	11	${}^2E_g \rightarrow {}^2T_{2g}$
Cd	276	36231	20	L.F
	339	29498	23	L.F
	435	22988	1085	C.T
Hg	366	27322	50	L.F
	435	22988	2406	C.T

**Table 4:** Biological activity of L1, L2 and their complexes.<sup>36</sup>

NO	Compounds	<i>Escherichia coli</i>	<i>Staphylococcus aureus</i>	<i>Rhizosporium</i>
1	L <sub>2</sub>	9	13	---
2	L <sub>1</sub>	15	15	13
3	DMSO	---	--	--
4	Ampicillin	15	16	15
11	Cd	15	15	10
12	Ni	15	15	10
13	Co	15	15	13
14	Hg	18	19	25
15	Cu	14	13	13

**Scheme 1:** The synthesis route of mixed ligand

to the agar media. The diameter of the inhibitory zone (mm) was determined after 24 hours bacteria incubation at 30°C and fungal incubation at 28°C for 48 hours carefully weighed amount of ampicillin was dissolved in sterile distilled water and used like reference stander of Gram - positive & Gram-negative bacteria. The screening had done three times, and the mean standard deviation was calculated. Triple trials when the area of inhibition is bigger than 6 mm, a compound is deemed active.

## CONCLUSIONS AND DISCUSSIONS

Characterization of compounds containing several ligands curcumas and L<sub>2</sub> were reacted with metal(II) molar ratio 1:1,1 to generate the molecular formula  $[M(L_1)(L_2)H_2OCl]$  with M = Ni(II), Co(II), Cu(II), Cd(II), and Hg(II). At room temperature, all complexes were stable and soluble in DMSO. The development of complexes between L<sub>1</sub> and L<sub>2</sub> was confirmed by elemental analysis, FT-IR, and C.H.N. analysis, as well as UV-visible. The observed values of molar

conductance in DMSO 0.001M solution are within the (2-16).<sup>23</sup> Table 1 shows the physical properties of the complexes.

The FT-IR spectra of  $L_1$ , appears in  $1628\text{ cm}^{-1}$  stretching vibrations primarily ascribed to overlap stretched vibration of alkenes (C=C) carbonyls (C=O). Infrared spectroscopy the curcuma ligand reveals stretching vibrations at  $3200\text{--}3500\text{ cm}^{-1}$  because of O-H groups, C=C aromatic stretching vibrations  $1427\text{ cm}^{-1}$  and high intensity band  $1512\text{ cm}^{-1}$  due to mixing of vibrations comprising stretched carbonyl bond vibrations  $\nu(\text{C=O})$ .<sup>26</sup> The  $L_2$  spectra revealed an absorption band at  $3429$  and  $3346\text{ cm}^{-1}$  corresponds to asymmetric stretchy vibration of the elemental amine group (NH<sub>2</sub>-), whereas  $3045$ ,  $1650$ , and  $3172\text{ cm}^{-1}$  correspond to the stretch frequencies of the (CH) aromatic group, the C=N ring and the NH groups, respectively. The C=O band is visible at infrared spectra of complexes consider as indicated of the coordination with a metal ion.

The molecular weight of free curcuma varies between  $1627\text{ cm}^{-1}$  and  $1685\text{--}1576\text{ cm}^{-1}$  depends on metal employed.<sup>20,22</sup> The existence of a strong band at  $1277\text{ cm}^{-1}$ , ascribed for the phenolic group's (CO), confirmed the lack of -OH phenolic during the chelation process. Curcuma's two phenolic groups exhibited a broad band in the  $3417\text{--}3581\text{ cm}^{-1}$  range. The complexes' infrared spectra revealed additional bands at  $466\text{--}489$  and  $596\text{--}622\text{ cm}^{-1}$ , which correspond to the (M-O) and (M-N) stretching frequencies, respectively.<sup>27</sup> The band C=N caused by  $L_2$ 's ring vibrations shifting into  $1616\text{--}1639\text{ cm}^{-1}$  in the complexes, showing that  $L_2$  was involved at coordination. The factors analyzes also IR spectra indicates produced compounds contain the structure depicted in Scheme 1.

The magnetic moments are 2.35, 3.22, 1.72 to Co (II), Ni(II) and Cu(II) complexes respectively. The UV-visible spectrum of  $L_1$ ,  $L_2$  and their complexes shows in table 3. The spectrum of  $L_1$  and  $L_2$  appears peaks absorption in (246, 341) nm and 273 nm due to ( $\pi\rightarrow\pi^*$ ) transition respectively<sup>28</sup> while the peaks at the region 419 nm and 430 nm are because ( $n\rightarrow\pi^*$ ) respectively.<sup>29</sup> In the complexes appeared new absorption peaks in d-d transitions, the Co(II) complex show 551 nm is due to  ${}^4T_{1g(F)}\rightarrow{}^4T_{1g(P)}$  suggesting octahedral geometry of this complex<sup>30-33</sup>. Ni(II) complex show 570 nm attributed to  ${}^3A_{2g(F)}\rightarrow{}^3T_{1g(F)}$  This is consistent with the octahedral shape of this complexes.<sup>34</sup> Cu(II) complex show peak in 629 nm is attributed to  ${}^2E_g\rightarrow{}^2T_{2g}$  this is consistent with the octahedral shape of this complexes<sup>35</sup> while Cd and Hg complexes appears peak band in 435 nm is due to C.T transition.<sup>28,34</sup>

## BIOLOGICAL ACTIVITY

In this research, the efficacy of Ligands and its complexes prepared against two types of positive bacteria such as *Staphylococcus* and negative for the honor stain such as *E. coli* were studied in this study. The pathogenic bacteria are treated with this dye and when they respond to this dye and do not put it outside the cell wall, it is positive. As for the bacteria that do not respond to this stain, they are negative. This bacterium and this difference between the types of bacteria is due to

the character of its outer walls. One type of fungus, such as *Rhizosporium*, was also used. The 0.001 mol concentration of the prepared compounds was prepared in 10 mL of DMSO solvent and a sample of the solvent was conducted and the effect of bacterium growth studied under the same conditions to avoid control interference<sup>36-38</sup>. The solvent didn't show toxic effects of growth bacteria or fungi used. The plates were injected by making holes where the cork drill was added and 1-mL of solutions of these compounds was added in Each hole was incubated 24 hours in room temperature of  $37^\circ\text{C}$ , after which it was removed from the incubator and the damping diameter was then measured and we conclude the following;  $L_1$ , showed different efficacy against positive and negative bacteria and fungi while  $L_2$  no inhibition of fungi was shown, all complexes showed different efficacy against the two types of bacteria and fungi.<sup>39-41</sup>

## CONCLUSION

Mixing the Ni(II), Cu(II), Co(II), Hg(II), and Cd(II) a complexes from curcuma  $L_1$  and 2- (1H-Benzimidazol-2-yl ) aniline  $L_2$  was synthesized and characterized from the data of the analysis and spectroscopy of the compounds were shown the octahedral geometry for all complexes. The complexes show good bioactivity from the compounds against Bactria and fungi as shown in Figure 1.

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