Mechanochemical Synthesis, Characterization, And Antimicrobial Studies of Metal Complexes of Transition Metals with Benzimidazole

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Abstract- Mechanochemical syntheses of Benzimidazole with transition metals; have been carried out by using direct mixing and grinding methods. The solid synthesized complexes were characterized based on their physical properties such as melting point or decomposition temperature, color, thin layer chromatography, spectroscopic studies like infra-red spectroscopy and UV-Visible spectroscopy; metal complexes for antimicrobial activities screened against some selected pathogens. The bacteria include Xanthomonas axonopodis, Streptococcus feacalis, Pseudomonas aeruginosa, Chromobacterium violaceum, and Erwinia carotovora. Fungi pathogens were: Collectotrichum factum, Phytophthora palmivora, Ceratocysti sparadora, and Helminthosporium toxins. The results obtained have presumed that metal complexes were pure, single component, non – polar in nature, and coordination of metals to the ligand is through the nitrogen atoms of the amide group in the ligand while, the results of antimicrobial activities have also shown that metal complexes were more bioactive than the parent ligand.

Indexed Terms- Mechanochemical, Benzimidazole, Metal complexes, Anti-microbial activities

I. INTRODUCTION

Mechanochemical synthesis is the form of molecular nanotechnology used for preserving the originality of the components that are useful for applications in the fields of medical and technological sciences for saving lives due to its simplicity, environmentally friendly, and low-cost technology effect (Gajovic *et al.*, 2009 and Ni *et al.*, 2009). Mechanochemical synthesis is becoming more intensely studied because it can support reactions between solids quickly and quantitatively, with either no added solvent or only nominal amounts. It has been a sideline approach to chemical synthesis and solution-based methods.

This study shows that mechanochemical is not only a general way to make co-crystals but also that it could give products not accessible by the solution-based method as the solution-based method suffers some set back such as long reaction time, an excess of organic solvent, lower products yield, and harsh refluxing conditions. According to Sara *et al.* (2009), the mechanochemical method is a solvent-free reaction that leads to a new environmental concern that saves resources and energy.

Benzimidazole is a heterocyclic aromatic organic compound with a privileged structure that possesses many pharmacological properties in several fields such as analgesic, anti-inflammatory, anti-bacterial, antifungal, anti-viral, anti-helminthic, anti-convulsion, anti-cancer, and anti-hypertensive agents in medicinal chemistry (Barbital *et al.*, 2012). Benzimidazoles are often bioactive where their fungicides are commercialized. They act by binding to the fungal microtubules and stopping hyphal growth; they also bind to the spindle microtubules and block nuclear division (Mavrova *et al.*, 2006).

Recent research on the usefulness of mechanochemical synthesis was reported according to Kemp *et al. (*2009) using Schiff base derived from (benzaldehyde and sulphonamide) with some metal salts such as Cu(II), Ni(II), and Co(II) to give neutral complexes of configuration $M(L)(H_2O)_2Cl_2$. The mechanochemical for the synthesis and characterization of 2, 4- dinitrophenyl hydrazine metal complexes have been established accordingly (Rotimi *et al.,* 2017). Solid-state synthesis of 1-Ethnoxycarbonyl (-4-substituted semi carbides) was satisfied under free-solvent conditions; the products through using TLC (Thin Layer Chromatography); and reactions were complete within a short period (Mallakpour *et al*., 2003).

Microorganisms are found almost everywhere on earth, especially in (liquid water, moisture, hot spring, ocean, inside rocks, human skin, cow's stomach, and sponges) used for washing dishes (Madigan and Matinka, 2006). There is a need to develop a more efficient, reliable, and versatile method for the preparation and synthesis of metal complexes of Benzimidazole.

To this end, we report the mechanochemical synthesis, characterization, and antimicrobial activity studies of transition metal complexes with Benzimidazole.

II. EXPERIMENTAL

- Materials and Instrumentation

All reagents and chemicals were of analytical grade and used as obtained from Aldrich. The ligand used is benzimidazole. The metal salts used include cobalt (II) hexahydrate ($CoCl_2.6H_2O$), copper pentahydrate ($CuSO_4.H_2O$), Nickel (II) heptahydrate, ($NiSO_4.7H_2O$), and zinc (II) heptahydrate ($ZnSO_4.7H_2O$). IR spectra of the samples in KBr pellets were obtained in this range (4000 – 400 cm^{-1}) using an FTIR spectrometer. The melting points of the metal complexes were determined using a Gallenkamp melting points apparatus. Metal analyses were determined using complexometric titration; UV-Visible spectra were determined using an Aquamate v4.60 spectrophotometer. Antimicrobial activities were cultured at the Pest Control Laboratory Section, Department of Microbiology, Federal University of Technology, Akure. Nigeria.

- Experimental Procedures

Syntheses of the complexes:
The methods described by Pichon and James were modified and employed using the mechanochemical method to synthesize metal-ligand complexes.

- Synthesis of [Co (C6H7N2)2Cl2. 6H2O] complex

Equimolar concentrations of the ligand and metal were weighed carefully into a mortar containing 2mmol (0236g) benzimidazole and 2mmol (0.467g) cobalt chloride hexahydrate, pounded, and grounded thoroughly for twenty (20) minutes to obtain a signal blue powder. Then, the powder was removed from the mortar and stored in a desiccator for three (3) days, and the amount weighed was taken and calculated in percentage; also, for the synthesis of the following metal complexes [$CuC_6H_7N_2)_2SO_4.5H_2O$] (0.50g); [Ni ($C_6H_7N_2)_2SO_4.7H_2O$] (0.578g); [Zn ($C_6H_7N_2)SO_4.7H_2O$] (0.574g) viz-a-viz, which was later stored or kept in a desiccator for three days and amount weighed, and calculate the percentage yield in gram.

- Antimicrobial test

The ligands and the metal complexes for antimicrobial activities were analyzed using 0.5g\100ml at 1000ppm concentration in-vitro, and their antifungal activity against four fungi viz: *Fusariumoxysporium, Collectotrichum phomoiides, Helminthosporium toxins, and Percularia oryzae and bacteria including Klebsiella pneumonia, Cadinadaalbicans, Salmonella typhi, Pseudomonas aeruginosa, Clostridium sporogenous, and Bacillus subtilis;* in-vitro antibacterial and antifungal activities, using the disc diffusion method. Meanwhile, the diameter zone of inhibition in the ligand and metal complexes were cultured for their antibacterial and antifungal activities according to Bukhari *et al. (2005);* Ramon *et al.* (2003) for streptomycin sulfate and mancozeb.

The antifungal activity of the complexes was cultured by the plate method for five days and was suspended in potato dextrose agar (PDA) medium and autoclaved at 1200ºC 15 minutes. The percentage inhibition of antifungal activities was calculated using the formula given below.

$$\text{PERCENTAGE OF INHIBITION} = \frac{X-Y}{X} * 100$$

$$Where\ X = area\ of\ colony\ in\ control\ plate\ (without\ sample)$$

$$Y = area\ of\ colony\ in\ test\ plate(\ Vashi\ et\ al, 2012)$$

III. RESULTS

The physical properties of metal complexes and ligand were shown in Tables 1. Table 2 shows the ratio of different solvent used for the solubility tests of the metal complexes. Tables 3 redeemed the prominent regions of FTIR spectra and UV-Visible of mixed ligand complexes while the evaluation of antifungal activities of the metal complexes are shown in Tables 4. And finally, evaluation of antibacterial screening of the metal complexes are shown in Table 5.

Table 1: Physical Properties of the Metal Complexes and Ligand

Complexes	Colour	% yield	Melting Point (ºC)	TLC	% Metal
$[CoLCl.H_2O]\ H_2O$	Blue	98.2	202 - 204	0.9	11.36
$[CuLSO_4.H_2O]H_2O$	Light blue	98.1	248 - 250	0.05	11.81
$[NiLSO_4H_2O]H_2O$	Green	97.6	250 - 252	0.6	10.56
$[ZnLSO_4.H_2O]$	White	98.7	245 - 247	0.07	12.9
LIGAND	White		178		

LIGAND - $C_7H_6N_2$

Table 2: Solubility Tests of the Metal Complexes and Solvent

Solvent	$[CoLCl.H_2O]H_2O$	$[CuLSO_4.H_2O]H_2O$	$[NiLSO_4.H_2O]H_2O$	$ZnLSO_4.H_2O$
Ethylacetate	Soluble	Insoluble	Insoluble	Insoluble
Chloroform	Insoluble	Insoluble	Insoluble	Insoluble
Toluene	Insoluble	Insoluble	Insoluble	Insoluble
n- hexane	Insoluble	Insoluble	Insoluble	Insoluble
DMSO	Soluble	Soluble	Soluble	Soluble
Distilled water	Insoluble	Soluble	Insoluble	Soluble

DMSO -Dimethylsulphoxide.

Table 3: FTIR and Uv- visible prominent regions of the ligand and metal complexes

Complexes	R –OH	NH	M- L	H_2O	SO_4^{2-}	UV- Visible
Ligand	------	3056		738	-	253
$[CoLCl.H_2O]H_2O$	3416	--	623	754	-	462,433,305
$[CuLSO4.H_2O]H_2O$	33,553,450	-	613	773	1112	457,254
$[NiLSO4.H_2O]H_2O$	3494	3206	619	767	1117	457,438,359
$[ZnLSO4.H_2O]$	--	3188	617	777	1106	336,390

Table 4: Anti-fungal Activities of the ligand, standard (control) and metal complexes in percentage

Complexes	C.F	P .P	C. P	H.T	P. O
$[CoLCl.H_2O]H_2O$	23.33	21	23	25	36.6
$[CuLSO4.H_2O]H_2O$	83.33	89	90	91	83.33
$[NiLSO4.H_2O]H_2O$	56.57	56	63	30	41.66
$[ZnLSO4.H_2O]$	56.67	55	73	50	35
Macrozeb (S.T)	60.64	62.34	66.66	68	70
Ligand	20	18.5	20	21.25	23

C.F- *Collectotrichum factum;* P.P- *Collectotrichum acutatum;* C.P- *Collectotrichum pacutatum,* H.T- *Helminthosporium toxins;* P.O- *Percularia oryzae*

Table 5: Anti-bacterial Activities of the ligand, standard (control) and metal complexes after 24 hours of incubation at 37.^{O}C in mm

Complexes	A	B	C	D	E
[CoLCl.H_2O]H_2O	36	30	27	25	23
[CuLSO4.H_2O]H_2O	31	25	27	39	46
[NiLSO4.H_2O]H_2O	22	19	26	17.5	22
[ZnLSO4.H_2O]	16	13	16	20	12
S. T	60	67	65	58	65
LIGAND	15	10	12	10	10.5

A- *Xanthomonas axonopodis; B- Streptococccus feacalis;* C- *Pseudomonas aeruginosa;*
D- *Chromobacterium violaceum;* E- *Erwinia carotovora; S.T.- Streptomycin sulphate* (Control)

IV. DISCUSSION

The physical properties of benzimidazole with metal complexes were reported and recorded in Table 1; from this result, the metal complexes demonstrate different colors on complexation from the ligand. In addition, the results also showed that all the metal complexes were dominant with the ligand, except for zinc which is white due to its complete electronic configuration of 3d10; the relatively sharp resolution in the melting points have also demonstrated that metal complexes are pure and it is a confirmation of metal complex formation. The levels of percentage yields for metal complexes at different levels of melting point by mechanochemical synthesis proved to be very high but are environmentally friendly as no pollution was recorded (Mallokpour *et al.*, 2003). While the solubility results showed that metal complexes are soluble in DMSO, showing that all complexes are non-polar compounds. The results of Thin Layer Chromatographic studies for the metal complexes confirmed the formation of only one complex or compound as only a spot formed on the chromatograms for each of the metal complexes (Table 1). All the above properties showed that the metal complexes are pure (Rotimi *et al.*, 2017). The percentage yield results obtained by mass as determined by complexometric titrations for metals also confirm the presence and coordination of metals to the ligand mechanochemically.

However, all-metal complexes synthesized are soluble in dimethylsulphoxide (DMSO), while others are insoluble in the available organic transparent solvents such as distilled water, as shown in Table 2.

The results of UV-Visible spectroscopic studies of the ligand and metal complexes in methanol were in the range of 200nm-800nm (Table 3); from this study, the electronic spectra of benzimidazole showed bands at 253nm due to low energy π - π^* transition of the aromatic system within organic ligand (Ismail *et al.*, 2015*).* The presence of metal complexes coordination in the benzimidazole complexes such as Co (II), Ni (II), Cu (II), and Zn (II) had also shifted and shown bands (λmax) around 423, 457, 457, 336 nm respectively (Rotimi *et al.*, 2017 and Olagboye *et al., 2015*).

The spectra of transition metal complexes depend on the transition of unpaired electrons from the ground state to the excited state, while most of the transition metal complexes are colored, and the color observed is due to d-d electronic transition in the visible regions between 400-800 nm. The atomic overlap in metal-ligand bond (ML) allows d-electrons to penetrate the central atom to the ligand (Saha *et al.*, 2002); overlap shifts in transition then affect the ligands on the energies of the d-orbital of the metal ions. In addition to this, transition metal complexes of benzimidazole are colorful, and they can absorb strongly within 400-800nm visible regions due to the presence of d-d absorption band, indicating the involvement of the chromophoric group in the ligand except for colorless Zinc metal and absorbed below 400nm due to complete filled d-orbital (Ajibade and Kolawole, 2007 and Chandraleka and Chandramohan, 2014).

The examination results of FTIR spectroscopy studies on both the ligand and metal complexes were listed and recorded in Table 3. From the analysis of the FTIR, it shows that the absorptions take place between 4000-350 cm^{-1} due to vibration energy (William *et al*., 1993). All the metal complexes have characteristic bands between 3494-374cm^{-1}; the absorption band is around 410 cm^{-1} in the ligand and with a shift to 419 cm^{-1} in the cobalt complex, but not found in the other metal complexes, which indicate the presence of chloride ions in the coordination sphere of [CoLClH$_2$O]. On the other hand, the prominent bands around 1112, 1117, and 1109 cm^{-1} for the other metal complexes may be the presence of sulfate ions SO$_4^{2-}$ in the inner coordination sphere of [CuLSO$_4$H$_2$O], [NiLSO$_4$H$_2$O], and [ZnLSO$_4$H$_2$O], as also confirmed by gravimetric analyses (William and Kemp, 1993 and Silverstein *et al.*, 1991). The absorption band at 738 cm^{-1} in the spectra with the commonest shift to higher bands such as 754, 773, 767, and 777 cm^{-1}. It shows the presence of inner coordinated water or bonded water. Other bands are seen at the following sets of vibration energy levels 623, 613, 619, and 616 cm^{-1} in the spectral of metal complexes but not found in the spectra of the ligand. These bands show the coordination of metals to the ligand (ML).

However, the strong band at 1770 cm^{-1} in the ligand were assigned to NH-stretching with hypochromic shifts to 1772 and 1778 cm^{-1} in Ni (II) and Zn (II) complexes, but with a bathochromic to 1637 and 1624 cm^{-1} in Co (II) and Cu (II) complexes respectively. Similarly, the characteristic band at 3056 cm^{-1} in the ligand ascribed or assigned to N-H of the amide group experienced a shift to higher frequencies to 3206 and 3188 cm^{-1} in Ni (II) and Zn (II) complexes but with the bands' disappearance or absence in the Co (II) and Cu (II) complexes which are due to N-H stretching and bending vibrations (Salim *et al.*, 2002 and Sani, 2011). It confirms the coordination of metal(s) to the adjacent nitrogen atom of the ligand (Barros – Garcia *et al.*, 2005). Also, another conspicuous band's absorptions at 3494, 3455, and 3355 cm^{-1} by Co (II), Cu (II), and Ni (II) complexes whereas, they are not found in both the ligand and Zn (II) complex. It can be attributed to the presence of water of crystallization molecules in the outer coordination sphere of metal complexes [CoLClH$_2$O]H$_2$O, [CuLSO$_4$H$_2$O]H$_2$O, and [NiLSO$_4$H$_2$O]H$_2$O except for Zn (II) [ZnLSO$_4$H$_2$O] (Rotimi *et al.*, 2017). Table 4 reports the antifungal activities of the ligand and its metal complexes alongside the control (Mazroceb) screened against five different microorganisms. Fungi pathogens are: *Collectotrichum factotum, Phytophthora palmivora, Ceratocystis paradora, and Helminthosporium toxins* and bacteria used are *Xanthomonas axonopodis, Streptococcus feacalis, Pseudomonas aeruginosa, Chromobacterium violaceum, and Erwinia carotovora.* The antifungal data revealed that the metal complexes performed moderately on the tested organisms with excellent inhibitive properties among the sulfur-containing complexes (Iqbal, 2009). The increase in antifungal activities of metal ligands complexes can be related to the effect of metal ions on the cell process. The toxicity of metal chelate increases as the metal ions concentration increases (Ismail *et al.*, 2015). The study of antibacterial activities on metal complexes have shown that metal complexes are far more bioactive than the ligand while standard *Streptomycin* appeared to be better than the complexes; based on bacterial data, it is evident that the processes of chelation dominantly affect the biological behavior of the complexes that are potent against some selected bacteria stains (Choudhary *et al.*, 2011; Shaker *et al.*, 2010 and Srivastava, 1997). The levels of microbial activities in the metal complexes were in these order: Cu > Ni > Zn > Co. Muchmore, the metal complexes for the production of garden fungicides in agriculture (Vashi *et al.*, 2012); the complexation of the metal (II) ions with the ligand could also be responsible for the mutation as a result of the fungi cell wall adaptation to the chemical effects of the complexes that are more prominent with the Cu (II) complexes (Osunlaja *et al.*, 2011).

CONCLUSION

Mechanochemical synthesis of transition metal complexes with benzimidazole has been done using solvent-free methods and then characterized by the following parameters: melting points, TLC, Uv – visible, and FTIR spectroscopy. The results of the spectroscopic studies of metal complexes indicated that the coordination of metals to the ligand was through the adjacent nitrogen atom of benzimidazole; also, confirmed the d – d electronic transitions accepted in the Zinc (II) complex due to complete filled d – orbital electronic configuration. The antimicrobial activities of the ligand and metal complexes show evidence that the metal complexes are more potent than the parent ligand that can be used in agriculture not only that mechanochemical synthesis is environmentally safe, low cost, and not time-consuming, unlike the conventional solvent a refluxing method that is prone to environmental pollution.

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