Egyptian Journal of Chemistry

http://ejchem.journals.ekb.eg/

Characterization and biological effectiveness of synthesized complexes of Palladium (II) from imine compounds

Shaimaa Hammad Hafidh,[a,*] Rasim Farraj Muslim[b,*] and Mohammed Ahmed Awad[a]

[a] *Department of chemistry, college of education for pure sciences, University Of Anbar, Anbar, Iraq*

[b] *Department of environmental, college of applied sciences-Hit, University Of Anbar, Anbar, Iraq**

Abstract

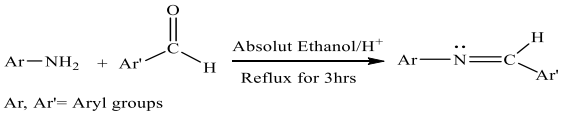
This article involved synthesize a novel Palladium (II) complexes as follows: The imine compounds were prepared by reacting of aldehydes with nitrogen compounds: 4-methylbenzene-1,2-diamine, naphthalene-1,8-diamine, and 4-chloro-5-methyl benzene -1,2-di amine and sublimated into ethanol. The reaction was continued by the thin layer chromatography (TLC), where the imine compounds were determined by spectrophotometry of FT-IR UV-Visible, ^{1}H-NMR. The palladium complexes were synthesized by the reaction of the prepared imine compounds (after they were dissolved) using absolute ethanol and palladium salt ($PdCl_2$), which was dissolved by using absolute ethanol with 4 drops of 11.6 N of HCl acid, then the mixture was raised for 3 hours. When the amine compounds reacted with the Palladium ion (II), this interaction leads to formation of palladium complexes, the reaction was continued by the thin layer chromatography (TLC), the complexes were spectrophotometrically characterized by measurement the ultraviolet-visible, infrared rays, mass spectrometry and molar conductivity. The geometric shape of the complexes had been proven, which the palladium complexes have a square planer shape. The biological activity of some synthesized complexes was determined using two different types of bacteria (Gram-Positive and Gram-Negative), namely ***Staphylococcus Aureus*** and ***Escherichia coli***. The results show that some concentrations have an intense inhibitory effect on the target bacteria.

Keywords: Imine compounds, complex of Palladium (II);

1. Introduction

The organometallic compounds are important synthesized compounds in organic chemistry [1-3] .The nitrogen atom of ligands (**Imine compounds**) have the ability to form complex [4, 5]. The Condensation reaction is considered one of the most important and common methods of preparing amine compounds, which it occurs by the direct condensation between primary aromatic amines and aldehydes or ketones, the reaction is done by using glacial acetic acid as a catalyst, leading to displacement of the water molecule to produce imine compound [6-10] below in scheme 1:

The Imine compounds are organic compounds that contain the active group (-HC=N-), Imine produced by the interactions between ketones or aldehydes with primary amines, which considered important organic compounds in the synthesis process of cyclic compounds and organometallic compounds, Imine compounds are effective materials in the medical field [11]. For the first time, imine compounds were prepared by Hugo Schiff [12]. Imine compounds are used as ligand compounds, where it's more soluble with metallic salts [13]. Imine compounds are one of the compounds that can be used to make a Palladium (II) complex [14].



Scheme 1. General reaction of synthesized imine compounds

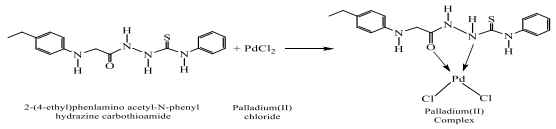
One of the important palladium complexes was prepared from the reaction of 2-(4-ethyl) phenyl amino acetyl-N-phenyl hydrazine carbothioamide

*Corresponding author e-mail: shy19u4009@uoanbar.edu.iq, dr.rasim92hmts@uoanbar.edu.iq

Receive Date: 04 June 2021, Revise Date: 25 June 2021, Accept Date: 05 July 2021

DOI: 10.21608/EJCHEM.2021.79085.3876

compounds with the palladium (II) salt, the diagnosis was made by spectroscopic methods to suggest the geometry of the prepared complex, where the shape was a square planer with dsp^2 hybridization, there were effects of the synthesized complex on the growth process of ***Bacillus Escherichia*** (Gram-Positive) and ***Escherichia coli*** (Gram-negative), this complex was shown to have anti-activity for these types of bacteria, the following figure shows the formula for the synthesized complex (palladium (II) complex) [15]:



Scheme 2. Synthesis of palladium (II) complex

2. Experimental

2.1. Synthesis of imine compounds

0.004 mol (0.5gm) of 3,4-diaminotoluene was dissolved in (15 mL) of EtOH and mixed with 0.008mol (1.15gm) of para-chloro benzaldehyde, which was also dissolved in (12 mL) of EtOH. The solution was cooled after the refluxing process (3 hours) and filtered. The precipitant was recrystallized from EtOH to obtain the imine (S_2), at the same process the other imine compounds were synthesized [16].

2.2. Synthesis the complexes of palladium (II)

0.001mol (0.4gm) from S_2 was dissolved in (20 mL) EtOH and mixed with 0.0005mol (0.09gm) of $PdCl_2$ which was also dissolved in (15 mL) of EtOH by assisting few drops of HCl (11.6 N). The solution was cooled after the refluxing process (3 hours) and filtered. The precipitant was recrystallized from EtOH to obtain the (P_2) complex, at the same process, all the palladium (II) complexes were synthesized [4].

2.3. Biological Evaluation

Antibacterial activity of the palladium complexes was done by using Mueller Hinton Agar against ***Staphylococcuus Aureus*** and ***Escherichia coli***. The holes have a diameter of 6 mm. The biological activity was dependent on calculated of the inhibition zone.

3. DISCUSSION THE RESULTS

3.1. Imine compounds

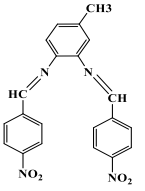
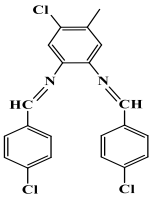
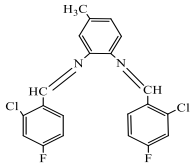
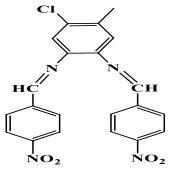
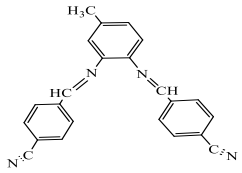
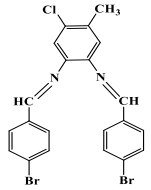
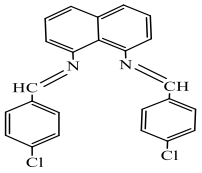
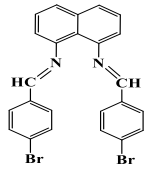
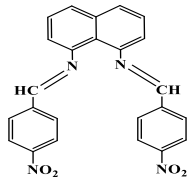
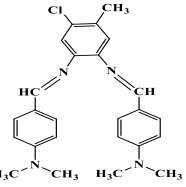
Imines were synthesized from the reaction between aldehyde compounds and diamine compounds, the reaction showed in the following equations (scheme 3):

The mechanism of imine compound synthesis includes the addition of the proton from glacial acid to the carbonyl group (C=O) of aromatic aldehyde, which leads to the formation of the intermediate compound (carbonium ion). The second step includes the attack of the nitrogen atom of the amino group (-NH_2) in the aromatic amines as a nucleophilic attack on the carbon of the carbonium ion to form the intermediate compound (N-Subtituted Hemiaminal). In the third step, the proton is withdrawn from the nitrogen atom by the water molecule to form a carbinolamine. The fourth step involves adding a proton from the acid to the hydroxyl group. The fifth step involves the displacement of a water molecule from the compound. The final step is the displacement of a proton of nitrogen to yield the stable imine compound, as shown in Scheme 4:

Table 1 . Properties of synthesized imine compounds

Comp. Code	Molecular Formula	m.p. ^oC	Yield %	Colour	M. wt	Time of Reaction	R_f
S_2	$C_{21}H_{16}N_4O_4$	64-66	64	Light yellow	122.17	2 hrs.	0.8
S_3	$C_{21}H_{14}Cl_2F_2N_2$	100-102	52	Light nutty	122.17	2 hrs.	0.4
S_4	$C_{23}H_{16}N_4$	98-100	71	Yellow	122.17	1 hr.	0.8
S_5	$C_{24}H_{16}Cl_2N_2$	130-132	50	Light nutty	158.2	3 hr.	0.5
S_6	$C_{24}H_{16}N_4O_4$	240-242	55	Orange	158.2	3 hrs.	0.7
S_9	$C_{21}H_{15}Cl_3N_2$	118-120	65	Light nutty	154.6	1 hrs.	0.8
S_{10}	$C_{19}H_{13}ClN_4O_4$	235-238	75	Light nutty	154.6	30 min	0.6
S_{11}	$C_{21}H_{15}Br_2ClN_2$	178-180	50	Light nutty	154.06	3 hrs.	0.8
S_{12}	$C_{24}H_{16}Br_2N_2$	118-120	82	Green yellow	158.2	3 hrs.	0.7
S_{13}	$C_{25}H_{27}ClN_4$	194-196	51	Dark nutty	154.06	2 hrs.	0.7

Table 2 . Structure and nomenclature of synthesized imine compounds

Code	Structure nomenclature	Code	Structure nomenclature
S_2	 **(1*E*,1'*E*)-*N*,*N*'-(4-methyl-1,2-phenylene)bis(1-(4-nitrophenyl)methanimine)**	**S_9**	 **(1*E*,1'*E*)-*N*,*N*'-(4-chloro-5-methyl-1,2-phenylene)bis(1-(4-chlorophenyl)methanimine)**
S_3	 *N*,*N*'-(4-methyl-1,2-phenylene)bis(1-(2-chloro-4-fluorophenyl)methanimine)	**S_{10}**	 **(1*E*,1'*E*)-*N*,*N*'-(4-chloro-5-methyl-1,2-phenylene)bis(1-(4-nitrophenyl)methanimine)**
S_4	 4,4'-((1*E*,1'*E*)-((4-methyl-1,2-phenylene) bis(azaneylylidene)) bis(methaneylylidene))dibenzonitrile	**S_{11}**	 **(1*E*,1'*E*)-*N*,*N*'-(4-chloro-5-methyl-1,2-phenylene)bis(1-(4-bromophenyl)methanimine)**
S_5	 **(1*E*,1'*E*)-*N*,*N*'-(naphthalene-1,8-diyl)bis(1-(4-chlorophenyl)methanimine)**	**S_{12}**	 **(1*E*,1'*E*)-*N*,*N*'-(naphthalene-1,8-diyl)bis(1-(4-bromophenyl)methanimine)**
S_6	 **(1*E*,1'*E*)-*N*,*N*'-(naphthalene-1,8-diyl)bis(1-(4-nitrophenyl)methanimine)**	**S_{13}**	 **4,4'-((1*E*,1'*E*)-((4-chloro-5-methyl-1,2-phenylene)bis(azaneylylidene))bis(methaneylylidene))bis (*N*,*N*-dimethylaniline)**

3.1.1. Ultra violet - Visible of imines

The compounds were dissolved in DMSO and characterized by Uv-Visible spectra, the transitions of S_2 showed the transition at (238 nm and 296nm) of the type $\pi \rightarrow \pi^*$ caused by C=C bonds of aromatic structure, transition at (480 nm) of the type $n \rightarrow \pi^*$ for C=N [17]. Table 3 showed all the types of transitions and wavelengths for all synthesized imines. See figures 1-4 for some imines compounds.

3.1.2. FT-IR of imine compounds

The FT-IR spectrum of the compound S_2 showed the absorption absorption band at (1600cm^{-1}) for C=N imine, absorption band at (1440cm^{-1}) for C=C aromatic, stretch absorption band at (3070 cm^{-1}) refers to aromatic C-H, absorption band (3109cm^{-1}) refers to C-H of imine, absorption band at (2856 and 2922cm^{-1}) refers to symmetric and asymmetric respectively of aliphatic C-H [18, 19]. Table 4 showed the other stretch absorption bands of all synthesized imine compounds. See the figures 5-8 of some imines compounds.

Table 3. The Ultra violet - Visible of synthesized imines

Comp.	Max/nmλ	Transition
S_2	238	π-π* of Aromatic C=C
	296	π-π*
	480	n-π*of C=N
S_3	238	π-π* of Aromatic C=C
	296	π-π*
	438	n-π*of C=N
S_4	238	π-π* of Aromatic C=C
	295	π-π*
	475	n-π*of C=N
S_5	237	π-π* of Aromatic C=C
	295	π-π*
	460	n-π*of C=N
S_6	238	π-π* of Aromatic C=C
	295	π-π*
	435	n-π*of C=N
S_9	235	π-π* of Aromatic C=C
	297	π-π*
	480	n-π*of C=N
S_{10}	240	π-π* of Aromatic C=C
	295	π-π*
	560	n-π*of C=N
S_{11}	235	π-π* of Aromatic C=C
	295	π-π*
	435	n-π*of C=N
S_{12}	235	π-π* of Aromatic C=C
	295	π-π*
	380	n-π*of C=N
S_{13}	235	π-π* of Aromatic C=C
	295	π-π*
	465	n-π*of C=N

3.1.3. ^{1}H-NMR spectra of imine compounds

The ^{1}H - NMR of imine (S_2) showed the chemical shift (δ ppm): singlet signal at (δ= 2.45) refers to the methyl group (-CH_3), singlet at (δ= 10.14) refers to the proton of imine group and a multiplet at (δ=7.96-8.42) refers to the protons of aromatic system. Table 5 shows the ^{1}H-NMR of the synthesized imines [20, 21]. See the figures 9-12 of some imines compounds

3.2. Complexes of Palladium (II)

The structure, formula, and other properties of synthesized Palladium recorded in the table (6), Palladium complexes were characterized with molar conductivity, atomic absorption and (Uv-Visible, FT-IR, Lc-mass) spectroscopies, The good yield of the synthesized Palladium complexes was for P_9 90%, the highest melting points of synthesized Palladium complexes were for compounds P_{11} and P_{13} (>300 ^{o}C). Table 7 showed the time of reaction and Rf (the ratio between the distance of the complexes and the mixtures of solvents in the thin layer chromatography) of all synthesized Palladium (II) complexes.

3.2.1. Ultra violet - Visible of complexes of the Palladium (II)

Palladium complexes were dissolved in solvent (DMSO) and showed the transitions: P_2 showed the transition at (230 nm and 295nm) of the type π→ π* caused by C=C bonds of aromatic structure, transition at (438nm) of the type n→ π* for C=N, transition at (880 nm) of the $^1A_1g \rightarrow {}^1A_2g$ transition, all transition proved the shape of P_2 was Square planer [17]. Table 8 showed all the transitions for all synthesized Palladium complexes. See the figures 13-16 of some Palladium (II) complexes

3.2.2. FT - IR spectra complexes of Palladium (II)

The FT-IR of the synthesized complexes of Palladium (II) appeared the absorption: Complex (P_2) showed the band at (1606 cm^{-1}) for C=N imine, band at (1460cm^{-1}) for C=C aromatic, band at (3070 cm^{-1}) refers to aromatic C-H, band at (3120cm^{-1}) refers to C-H of imine, band at (2800 and 2924 cm^{-1}) refers to symmetric and an asymmetric respectively of aliphatic C-H, the band at (599 cm^{-1}) refers to presence of Palladium – Nitrogen band (M-N) [18]. Table 9 showed the other absorption bands of all synthesized Palladium (II) complexes. See the figures 17-20 of some Palladium (II) complexes.

Table 4. The FT-IR of synthesized imine compounds

Comp.	υC-N	υC=C Ar	υC=N	υ=C-H Aliphatic		υ=C-H Ar	υ=C-H Imine	Other Groups
				Symmetric	Asymmetric			
S_2	1109	1440	1600	2856	2922	3070	3109	NO_2: 1344, 1517
S_3	1118	1456	1602	2862	2922	3072	3111	Cl: 1041, F: 1259
S_4	1118	1440	1610	2922	2974	3072	3090	CN: 2227
S_5	1124	1419	1598	-----	-----	3034	3068	Cl : 1087
S_6	1165	1512	1600	-----	-----	3068	3078	NO_2: 1348,1512
S_9	1093	1446	1600	2972	2924	3010	3120	Cl : 1008
S_{10}	1105	1595	1610	2922	2080	3109	3109	NO_2: (1344, 1514)/Cl:999
S_{11}	1109	1446	1627	2922	2890	3070	3140	Br: 686, Cl: 1006
S_{12}	1165	1421	1597	-----	-----	3041	3070	Br: 644
S_{13}	1195	1442	1612	2889	2804	3030	3075	Cl: 999

Table 5. The ^{1}H-NMR spectra of some synthesized imine compounds

Comp. No.	Group	No. of proton	Chemical shift (ppm)δ	Type of Single
S_2	-C**H**$_3$	3	2.45	singlet
	Aromatic protons	11	7.96-8.42	multiplet
	2(-C**H**=N-)	2	10.14	singlet
S_3	-C**H**$_3$	3	2.53	singlet
	Aromatic protons	9	6.73-7.97	multiplet
	2(-C**H**=N-)	2	10.20	singlet
S_4	-C**H**$_3$	3	2.53	singlet
	Aromatic protons	11	7.07-8.62	multiplet
	2(-C**H**=N-)	2	10.10	singlet
S_5	Aromatic protons	14	6.48-8.06	multiplet
	2(-C**H**=N-)	2	10.85	singlet
S_6	Aromatic protons	14	6.50-8.29	multiplet
	2(-C**H**=N-)	2	10.20	singlet
S_9	-C**H**$_3$	3	1.08	singlet
	Aromatic protons	11	6.96-8.18	multiplet
	2(-C**H**=N-)	2	10.00	singlet
S_{10}	-C**H**$_3$	3	2.46	singlet
	Aromatic protons	11	6.70-8.87	multiplet
	2(-C**H**=N-)	2	10.20	singlet
S_{11}	-C**H**$_3$	3	2.61	singlet
	Aromatic protons	11	6.90-8.11	multiplet
	2(-C**H**=N-)	2	10.00	singlet
S_{12}	Aromatic protons	14	6.48-7.88	multiplet
	2(-C**H**=N-)	2	10.00	singlet
S_{13}	-C**H**$_3$	3	2.60	singlet
	4 (–NC**H**$_3$)	12	3.07	singlet
	Aromatic protons	10	6.63-7.98	multiplet
	2(-C**H**=N-)	2	9.67	singlet

3.2.3. Lc - mass of the complexes of Palladium (II) [4]

The Lc - mass spectra showed the fragments of the complexes and proved the synthesizing process and the square planer shape, the tables 10-19 showed the fragments of the complexes. See the figures 21-24 of some Palladium (II) complexes and scheme 5-14.

3.2.4. Molar conductance measurements of the complexes of Palladium (II)

The measured of molar conductivity of the complexes uses to know the ionic formulas of complexes, the complexes concentration are 1×10^{-3} molarity, the measured completed by dissolving the samples in DMF at the room temperature. If the value of conductance is more than 70 ($Ohm^{-1}. cm^{-1}. mol^{-1}$ x 10^{-6}), this proves that the negative charge of chloride is outside of the coordination [22]. See table 20.

3.2.5. Atomic absorption of the Palladium Complexes [23]

Standard solutions of metal chlorides were used for titration. A specific weight of the solid compounds was digested with a mixture (5 mL) of concentrated nitric acid and perchloric acid, this issue was repeated several times until completely dissolve of all organic matter. The reaction was evaporated to drying. After cooling, the remaining salt was dissolved in deionized water where the proportion of the presence of palladium in the synthesized complexes was measured and compared with the theoretically calculated ratio, as shown in the following table 21.

3.2.6. Biological activity of some Palladium complexes

Antibacterial activity of Palladium complexes was done by using Mueller Hinton Agar against ***Staphylococcuus Aureus*** and ***Escherichia Coli***. The Palladium complexes dissolved in DMSO Solvent, the holes diameter was 6 mm, the concentrations of Palladium complexes are 50% and 100% [24], the inhibition zone recorded in table 22. See pictures 1 and 2.

Table 6. Properties of synthesized complexes of Palladium (II)

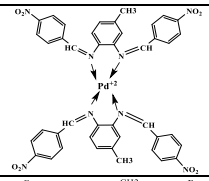
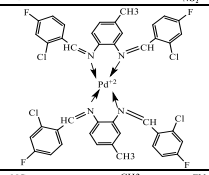
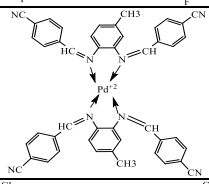
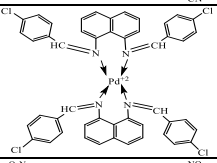
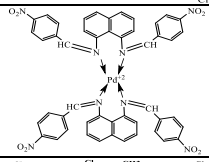
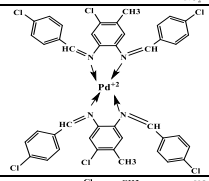
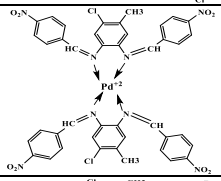
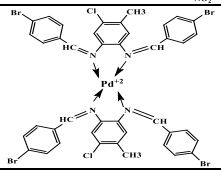
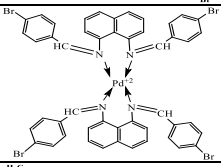
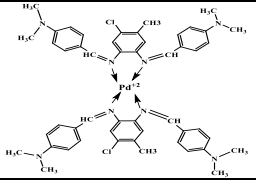
Comp. code	Structure	Formula	m.p. °C	Yield%	Color
P_2		**$C_{42}H_{32}N_8O_8Pd$**	**280-284**	**58**	**Gray**
P_3		**$C_{42}H_{28}Cl_4F_4N_4Pd$**	**196-200**	**52**	**Gray**
P_4		**$C_{46}H_{32}N_8Pd$**	**>300**	**60**	**Gray**
P_5		**$C_{48}H_{32}Cl_4N_4Pd$**	**230-234**	**55**	**Brawn**
P_6		**$C_{48}H_{32}Cl_2N_8O_8Pd$**	**260-263**	**30**	**Gray**
P_9		**$C_{42}H_{30}Cl_6N_4Pd$**	**230-234**	**90**	**Gray**
P_{10}		**$C_{42}H_{30}Cl_2N_8O_8Pd$**	**220-223**	**68**	**Green yellow**
P_{11}		**$C_{42}H_{30}Br_4Cl_2N_4Pd$**	**>300**	**40**	**Light nutty**
P_{12}		**$C_{42}H_{32}Br_4N_4Pd$**	**206-209**	**97**	**Brown**
P_{13}		**$C_{50}H_{54}Cl_2N_8Pd$**	**300>**	**31**	**Gray**

Table 7. Rf of synthesized Palladium complexes

Comp. Code	Wt. of ligand(g)	Wt. of Salt(g)	Wt. of Complex(g)	M.wt of complex (g/mol)	Time of Reaction	R_f
P_2	0.4	0.09	0.26	883	2 hrs.	0.7
P_3	0.25	0.06	0.16	912	2 hrs.	0.7
P_4	0.35	0.04	0.26	803	3 hrs.	0.7
P_5	0.1	0.02	0.17	913	3 hrs.	0.8
P_6	0.25	0.05	0.09	1026	3 hrs.	0.5
P_9	0.25	0.05	0.18	909	3 hrs.	0.8
P_{10}	0.4	0.09	0.34	952	2 hrs.	0.8
P_{11}	0.1	0.02	0.04	1087	3 hrs.	0.4
P_{12}	0.25	0.05	0.3	1090	2 hrs.	0.8
P_{13}	0.25	0.06	0.1	944	2 hrs.	0.5

Table 8. Transitions and Wavelengths and the suggested Structure of synthesized Palladium (II) complexes

Comp. No	Max/nmλ nm	Transition	Suggested Structure
P_2	230 and 290	π-π^* of Aromatic C=C	Square planer
	435	n→π* of C=N - C.T	
	880	$^1A_1g \rightarrow {}^1A_2g$	
P_3	232 and 295	π-π^* of Aromatic C=C	Square planer
	420	n→π* of C=N - C.T	
	883	$^1A_1g \rightarrow {}^1A_2g$	
P_5	230 and 296	π-π^* of Aromatic C=C	Square planer
	470	n→π* of C=N - C.T	
	886	$^1A_1g \rightarrow {}^1A_2g$	
P_6	230 and 295	π-π^* of Aromatic C=C	Square planer
	435	n→π* of C=N - C.T	
	887	$^1A_1g \rightarrow {}^1A_2g$	
P_9	235 and 295	π-π^* of Aromatic C=C	Square planer
	465	n→π* of C=N - C.T	
	886	$^1A_1g \rightarrow {}^1A_2g$	
P_{10}	235 and 295	π-π^* of Aromatic C=C	Square planer
	440	n→π* of C=N - C.T	
	880	$^1A_1g \rightarrow {}^1A_2g$	
P_{11}	234 and 296	π-π^* of Aromatic C=C	Square planer
	420	n→π* of C=N - C.T	
	886	$^1A_1g \rightarrow {}^1A_2g$	
P_{12}	232 and 294	π-π^* of Aromatic C=C	Square planer
	480	n→π* of C=N - C.T	
	890	$^1A_1g \rightarrow {}^1A_2g$	
P_{13}	234 and 294	π-π^* of Aromatic C=C	Square planer
	480	n→π* of C=N - C.T	
	886	$^1A_1g \rightarrow {}^1A_2g$	

C.T=Charge transfer

Table 9. FT - IR spectra of the synthesized complexes of Palladium (II)

Comp.	υC-N	υC=C Ar	υC=N	υ=C-H Aliphatic Symmetric	υ=C-H Aliphatic Asymmetric	υ=C-H Ar	υ=C-H Imine	υM-N	Other Groups
P_2	1112	1460	1606	2800	2924	3070	3120	599	NO_2: 1346, 1525
P_3	1132	1454	1602	2892	2926	3078	3221	597	F:1215, Cl: 900
P_4	1130	1523	1633	2856	2924	3060	3217	480	CN: 2231
P_5		1595	1637	-----	-----	3051	3180	586	Cl:964
P_6	1107	1417	1589	2856	2924	3050	3080	505	NO_2: 1519,1344
P_9	1093	1456	1616	-----	-----	3064	3200	503	Cl: 1012
P_{10}	1112	1446	1608	2860	2922	3075	3095	592	NO_2:(1523,1442)/ Cl:1016
P_{11}	1070	1454	1633	2890	2926	3060	3072	590	Br: 719, Cl: 1010
P_{12}	1078	1587	1633	-----	-----	3040	3065	493	Br: 823
P_{13}	1122	1454	1612	2858	2920	3015	3040	476	Cl:941

Table 10. Lc -Mass fragments of P_2

Fragments	m / z
$M^+ = C_{42}H_{32}N_8O_8Pd^{2+}$	883
$C_{15}H_{12}N_2Pd^{2.2+}$	327
$C_{11}H_{10}N_4Pd^{6.2+}$	306
$C_2H_2N_2Pd^{4\bullet 2+}$	158
$C_8H_7N_2^{3\bullet}$	131

Table 11. Lc -Mass fragments of P_3

Fragments	m / z
$M^+ = C_{42}H_{28}Cl_4N_4Pd^{2+}$	913
$C_{15}H11ClFN_2Pd^{\cdot 2+}$	376
$C_{14}H_{10}ClFN^{\cdot}$	245
$C_2H_2N_2^{4.2+}$	158
$C_8H_7N_2^{3\cdot}$	131
$C_7H_6N^{3\cdot}$	105

Table 12. Lc -Mass fragments of P_4

Fragments	m / z
$M^+ = C_{46}H_{32}N_8Pd^{+2}$	803
$C_{35}H_{24}N_5Pd^{5.\ 2+}$	628
$C_9H_8N_4Pd^{8.\ 2+}$	277
$C_{14}H_{11}N^{2\cdot}$	193
$C_2H_2N_2Pd^{4.\ +2}$	163

Table 13. Lc -Mass fragments of P_5

Fragments	m / z
$M^+ = C_{48}H_{32}Cl_4N_4Pd^{2+}$	913
$C_{18}H_{12}ClN_2Pd^{\cdot 2+}$	393
$C_{10}H_6N_2Pd^{4\cdot 2+}$	259
$C_{10}H_6N_2^{4\cdot}$	150
$C_7H_5ClN^{\cdot}$	136
$C_6H_4Cl^{\cdot}$	110

Table 14. LC-Mass fragments of P_6

Fragments	m / z
$M^+ = C_{48}H_{32}N_8O_8Pd^{2+}$	955
$C_{30}H_{20}N_4Pd^{4\cdot 2+}$	550
$C_{10}H_8N_5O_2Pd^{7\cdot 2+}$	333
$C_{11}H_7N_2Pd^{3\cdot 2+}$	275
$C_8H_6N_3O_2^{3\cdot}$	173
$C_7H_5N^{2\cdot}$	103

Table 15. Lc -Mass fragments of P_9

Fragments	m / z
$M^+ = C_{42}H_{30}Cl_6N_4Pd^{2+}$	910
$C_{23}H_{15}Cl_3N_4Pd^{4\cdot 2+}$	553
$C_{14}H_{10}ClNPd^{2\cdot 2+}$	335
$C_8H_6ClN_2Pd^{3\cdot 2+}$	273
$C_9H_7ClN_2^{2\cdot}$	173
$C_7H_5ClN_2^{4\cdot}$	153

Table 16. Lc -Mass fragments of P_{10}

Fragments	m / z
$M^+ = C_{42}H_{30}Cl_2N_8O_8Pd^{2+}$	952
$C_{21}H_{15}N_5O_4Pd^{4\cdot 2+}$	502
$C_{18}H_{14}Cl_2N_4Pd^{4\cdot 2+}$	467
$C_{14}H_{10}N_3O_2^{3\cdot}$	250
$C_{14}H_{11}N^{2\cdot}$	191
$C_8H_6N_3^{3\cdot}$	173
$C_7H_5N^{2\cdot}$	100

Table 17. Lc -mass fragments of P_{11}

Fragments	m / z
$M^+ = C_{42}H_{30}Br_4Cl_2N_4Pd^{2+}$	1088
$C_{21}H_{13}BrClN_3Pd^{4\cdot 2+}$	532
$C_{15}H_{11}N^{3\cdot}$	221
$C_8H_6BrN_2^{3\cdot}$	207
$C_7H_5^{3\cdot}$	93

Table 18. Lc -mass fragments of P_{12}

Fragments	m / z
$M^+ = C_{48}H_{32}Br_4N_4Pd^{2+}$	1091
$C_{25}H_{17}Br_2N_3Pd^{2\cdot 2+}$	622
$C_{20}H_{14}BrN_4Pd^{5\cdot 2+}$	498
$C_{10}H_6N_2Pd^{4\cdot 2+}$	262
$C_{11}H_7N^{2\cdot}$	158
$C_7H_5N^{2\cdot}$	102

Table 19. Lc -mass fragments of P_{13}

Fragments	m / z
$M^+ = C_{50}H_{54}Cl_2N_8Pd^{2+}$	944
$C_{25}H_{27}ClN_4Pd^{\cdot 2+}$	519
$C_{21}H_{16}N_3^{3\cdot}$	310
$C_{14}H_{10}N^{3\cdot}$	194
$C_7H_5ClN_2^{4\bullet}$	157
$C_7H_5N^{4\cdot}$	105

Table 20. Molar conductance measurements of the complexes of Palladium (II)

No.	Complexes	Concentration Molarity	Temperature °C	Molar Condactivity Λm (Ohm^{-1}.Cm^2.mol^{-1})*10^{-6}
P_2	$[Pd(S_2)_2]^{+2}$	$1x10^{-3}$	28	77
P_3	$[Pd(S_3)_2]^{+2}$	$1x10^{-3}$	28	77
P_4	$[Pd(S_4)_2]^{+2}$	$1x10^{-3}$	28	71
P_5	$[Pd(S_5)_2]^{+2}$	$1x10^{-3}$	28	75
P_6	$S_6)_2]$ $[Pd(^{+2}$	$1x10^{-3}$	28	80
P_9	$[Pd(S_9)_2]^{+2}$	$1x10^{-3}$	28	77
P_{10}	$[Pd(S_{10})]^{+2}$	$1x10^{-3}$	28	80
P_{11}	$[Pd(S_{11})]^{+2}$	$1x10^{-3}$	28	84
P_{12}	$[Pd(S_{12})]^{+2}$	$1x10^{-3}$	28	75
P_{13}	$[Pd(S_{13})_2]^{+2}$	$1x10^{-3}$	28	79

Table 21. Ratio of Theoretical and Practically of Palladium (II)

No.	Practically	Theoretical
P_2	10.17	11.15
P_3	11.24	10.82
P_4	10.69	12.17
P_5	12.05	11.66
P_6	12.02	11.14
P_9	11.98	11.70
P_{10}	11.78	11.18
P_{11}	8.17	9.19
P_{12}	8.13	9.16
P_{13}	11.28	11.27

Table 22. Inhibition diameter (mm) of some Palladium complexes

No.	*Staphylococcuus Aureus*		*Escherichia Coli*	
	Concentration			
	50%	100%	50%	100%
P_2	10mm	10mm	18mm	12mm
P_3	11mm	15mm	14mm	10mm
P_4	16mm	15mm	16mm	17mm
P_9	12mm	12mm	16mm	0
P_{10}	12mm	12mm	15mm	9mm

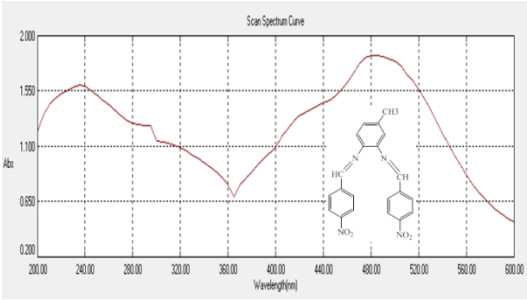


Figure 1. Uv-Visible spectra of S_2

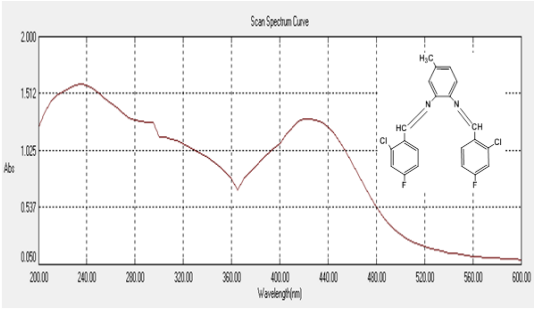


Figure 2. Uv-Visible spectra S_3

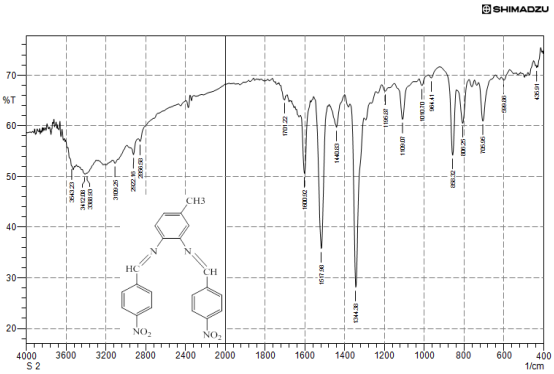


Figure 11. FT-IR spectra of S_2

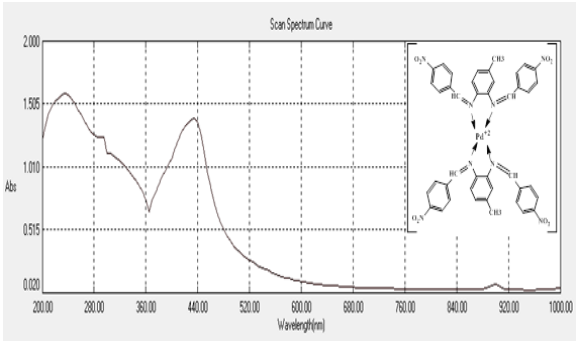


Figure 31. Uv-Visible spectra of P_2

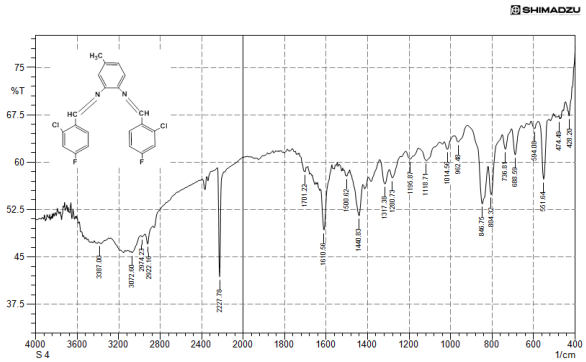


Figure 12. FT-IR spectra of S_3

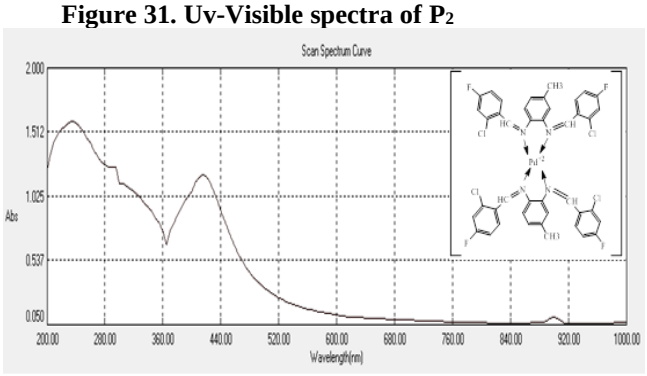


Figure 32. Uv-Visible spectra of P_3

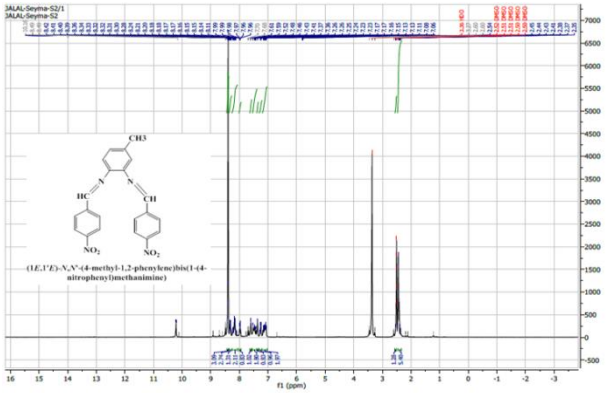


Figure 21. ^{1}H-NMR spectra of S_2

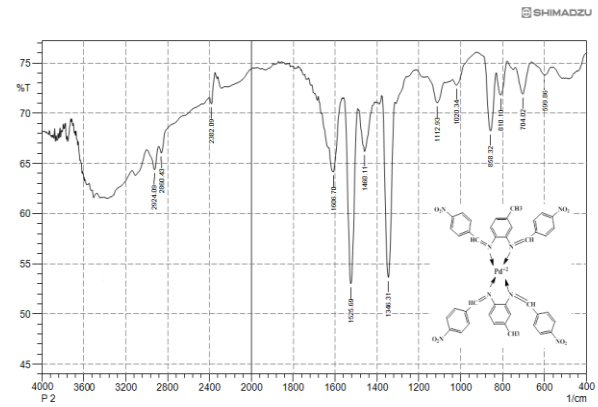


Figure 41. FT-IR spectra of P_2

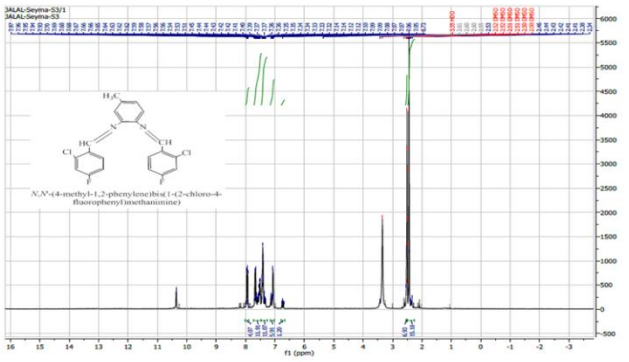


Figure 42. FT-IR spectra of P_3

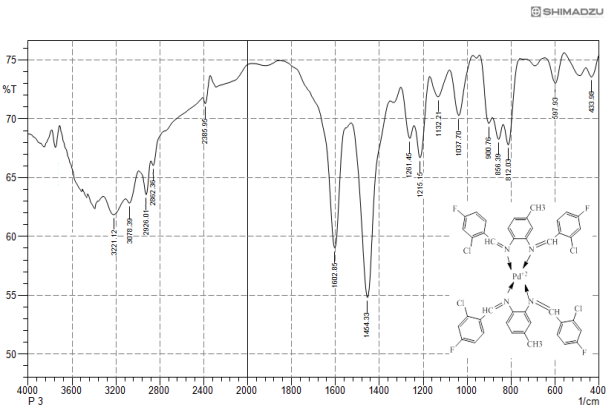


Figure 22. ^{1}H-NMR spectra of S_3

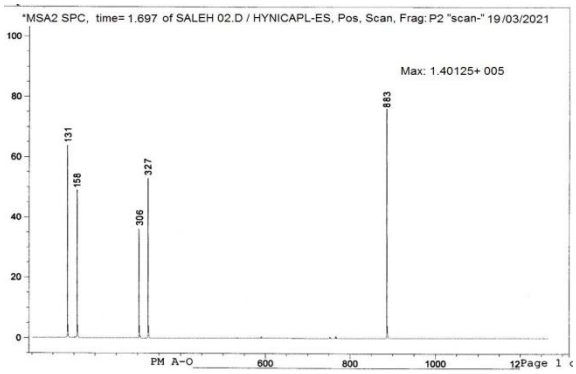


Figure 51. LC-Mass spectra of P_2

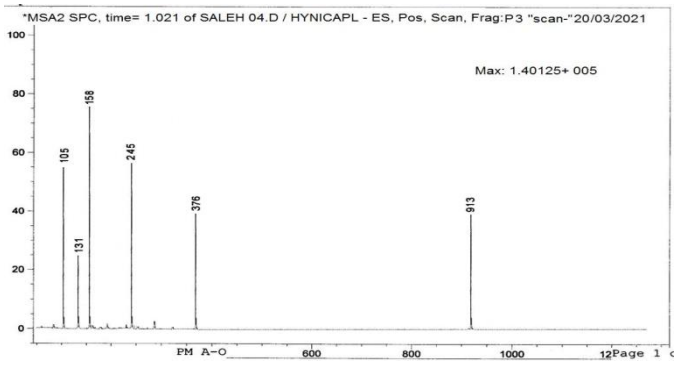
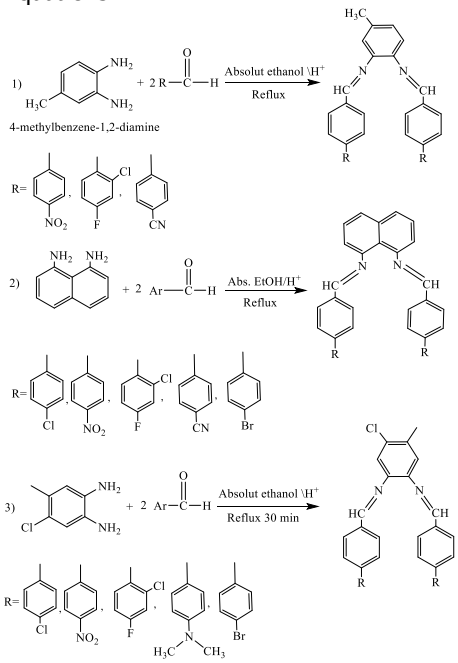
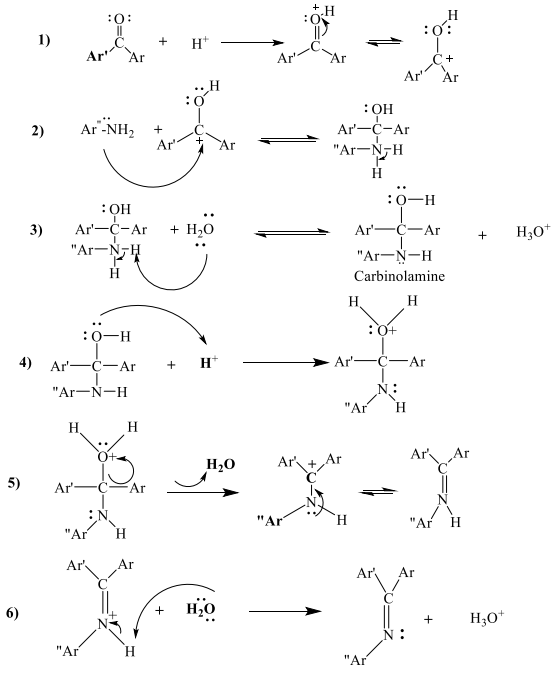


Figure 52. LC-Mass spectra of P_3

Equations



Scheme 3. The general equation for imine compounds synthesis



Scheme 4. Mechanism of imine compounds synthesizing

4. Conclusions

In this study, the intermediate compounds (imine compounds) in synthesizing the palladium complexes was confirmed by the disappearance of the infrared bands and the proton spectrum signals of the amine group (-NH2) in the primary aromatic amines, the disappearance of the carbonyl (C=O) bands in the aromatic aldehydes of the primary materials and the appears of new bands and signals of the imine group (-C=N-). The result of the thin layer chromatography (TLC) and the spectrophotometrically characterized by measuring ultraviolet-visible and infrared rays, mass spectrometry and molar conductivity measurement proved the geometric shape of palladium (II) complexes exhibits a square planer. The antibacterial activity of Palladium complexes was done against ***Staphylococcuus Aureus*** and ***Escherichia Coli***, where our obtained results proved that the best activity of Palladium (II) complexes was for P_4 (16 mm, 50%) against ***Staphylococcuus Aureus*** and was for P_2 (18 mm, 50%) against ***Escherichia Coli***.

Acknowledgments

We wish to extend our grateful to everyone who provided us with a helping hand in the measurements, and those who provided laboratories to complete our practical part. Special thanks to the President of Anbar University and the Dean of the College of Applied Sciences-Hit for their continuous support to the researchers.

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