Egyptian Journal of Chemistry
http://ejchem.journals.ekb.eg/

New Derivatives From 4-Amino Anti-Pyrine And Vanillin, Synthesis Characterization And Antibacterial Activity

Hawraa M Alabidi[a], Ali M Farhan[b], Hutham Mahmood Yousif Al-labban[c]* and Ahmed Abduljabbar Jaloob Aljanaby[d]
[a,c]*Department of Chemistry, Faculty of Science, University of Kufa, Iraq*
[b]*Ministry of Education, Al-Najaf, Iraq*
[d]*Department of Biology, Faculty of Science, University of Kufa, Iraq*

Abstract

Schiff base, Azo-Schiff base, nickel (II), and copper (II) complexes have been prepared successfully from 4-amino antipyrine and vanillin and diagnosed by UV-visible, FT-IR, ^{1}H-NMR, and ^{13}C-NMR. The derivative compound Azo-Schiff with a 200 mg/ml concentration has good antibacterial activity with a diameter of inhibition zone 16.11 ± 0.1035 mm and 13.21 ± 0.4044 against *Staphylococcus aureus* (*S. aureus*) and *Pseudomonas aeruginosa* (*P. aeruginosa*), respectively. Therefore, this compound may be considered a raw material for manufacturing treatments against these bacterial infections.

Keywords: 4-Amino antipyrine, Azo schiff base, Complexes, Biological activity, S. *aureus*, *P. aeruginosa*.

1. Introduction

Heterocycles are a hugely important class of molecules, accounting for more than half of all known organic compounds [1]. Heterocycles nitrogen-containing are important structural units for drug development [2]. Among the heterocyclic various structures, pyrazoles are considered to be very essential bioactive systems that are widely available in biological active groups of compounds [3]. Antipyrine is the first pyrazolone-derived compound that was first synthesized by Knorr [4, 5]. Because of its promising applications, the chemistry of antipyrine molecules has received a great deal of attention in medical and synthetic chemistry, as well as in materials science [6], so it is used as antipyretic, analgesic [7] non-steroidal anti-inflammatory drug [8]. 4-Amino-1,5-dimethyl-2-phenylpyrazole-3-one known as 4-amino antipyrine is an antipyrine derivative that has revealed a broad variety of biological activities, e.g. antimicrobial activity, painkiller, antiviral activity, as a result of the present free amino group which is used to prepare azo methane compound via condensation with carbonyl compounds (aldehyde or ketone) moreover it is treated with metals to synthesized metal complexes [9, 10, 11] . In recent years that it is also showed that certain drugs displayed increased activity when presented as metal complexes rather than organic compounds [12, 13]. This study aims to synthesize a new Azo-Schiff derived from 4-Amino antipyrine, coordinate with metals, and evaluation of antibacterial activity against *S. aureus* and *P. aeruginosa*.

2. Methods

2.1 Apparatuses

Shimadzu UV-1650 UV-Vis Spectrophotometer, Japan and IR spectra performed electronic spectra on Shimadzu FTIR8400 by KBr disk in the region (400-4000) cm-^{1}, ^{1}H-NMR, and ^{13}C-NMR of ligand recorded by using Burker Ultra Sheild (100MHz) and 720(WTW) measured molar conductivity.

*Corresponding author e-mail: huthamm.sadiq@uokufa.edu.iq.; (Hutham Mahmood Yousif Al-labban).
Receive Date: 24 April 2022, Revise Date: 05 May 2022, Accept Date: 07 May 2022.
DOI: 10.21608/EJCHEM.2022.135727.5977

2.2 Chemical synthesis

2.2.1 Synthesis of Schiff base derivative of 4-Amino antipyrine ((*E*)-4-((4-hydroxy-3-methoxybenzylidene) amino)-1,5-dimethyl-2-phenyl-1,2-dihydro-*3H*-pyrazol-3-one) [14].

A solution of vanillin (4-Hydroxy -3-methoxy benzaldehyde) (160 mg, 0.73 mmol) and glacial acetic acid (1 ml) in EtOH (20 ml) was added to the solution of 4-Amino antipyrine (110 mg 0.73 mmol) in (15 ml) EtOH the resulting mixture under reflux was heated for 5 h. After cooling, by filtration, the solid product was recrystallized from ethanol after collected to give the pale yellow desired imine derivative. FT-IR (KBr) (cm^{-1}): 3417 (OH) , 3105 (C-H ,arom) , 2993 and 2833 (C-H aliph) , 1666 (C=O , amide) , 1625 (C=N, arom) , 1581(C=C, arom) . ^{1}H-NMR (400 MHz, DMSO-d6) , 9.62 (s ,br, 1H , OH-arom) , 9.48 (s,1H, HC=N) , 7.52-7.57 (tr ,1H , H4-arom B) , 7.41-7.44 (d ,2H , H2,6-arom B) , 7.38 (d ,2H , H3,5-arom B), 7.22 (s ,1H , H2-arom A) , 7.19 (d ,1H , H6-arom A) , 6.86-6.89 (d ,1H , H5-arom A) , 3.86 (s,*3H*, OMe) , 2.69 (s,*3H*, N-Me) , 2.03 (s,*3H*, MeC=C) . ^{13}C-NMR (100 MHz, DMSO-d6) 160.4(C=O, Amide) , 155.5 (C=N) , 152.2 (C4-arom A) , 149.7 (C3-arom A), 148.4 (C1-arom B), 135.2 (C3-Pyr) , 129.6 (C1-arom A), 127.1(C3,5-arom B), 124.7(C2,6-arom B), 122.6 (C6-arom A) , 117.5 (C4-arom B), 115.9 (C2-arom A) , 110.0 (C5-arom A + C4-Pye) , 56.0 (OMe), 36.1 (Me2-Per) , 10.6 (Me3-Per).

2.2.2 Synthesis of Azo-Schiff derivative (4-(((E)-3-((E)-(1,5-dimethyl-3-oxo-2-phenyl-2,3-dihydro-1H-pyrazol-4-yl)diazenyl)-4-hydroxy-5-methoxybenzylidene)amino)-1,5-dimethyl-2-phenyl-1,2-dihydro-*3H*-pyrazol-3-one) [15].

The azo compound was prepared by dissolving the 4-Amino antipyrine (105 mg, 0.74 mmol) in a mixture of 2 ml HCl Conc. and 15 ml absolute EtOH and stirred for (20) min in an ice bath then an ice-cold solution of $NaNO_2$ (2 mg, 0.74 m moles) 10 ml was added dropwise on a period of (30) min, the resulting solution becomes a brown, then add dropwise to another ice-cooled solution of Schiff base derivative (1) (250 mg, 0.74 mmol) in 15 ml of alkaline EtOH, with continuous stirring at(0-5) ºC, later left overnight. the mixture was treated with dilute HCl and ammonia solution to obtain a neutral solution. The solid product was filtered, washed with cold distilled water, and dried. 180-182 FT-IR (KBr) (cm^{-1}): 3417 (OH) , 3064 (C-H ,arom) , 2931 and 2837 (C-H aliph) , 1647 (C=O , amide) , 1635 (C=N, arom) , 1589(C=C, arom), 1421(N=N) . . ^{1}H-NMR (400 MHz, DMSO-d6) , 14.28 (s ,br, 1H , OH-arom) , 9.77 (s,1H, HC=N) , 8.35 (s ,1H , H6-arom A) , 8.17-8.20 (tr ,1H , H4-arom C) , 8.04-8.10 (tr ,1H , H4-arom B) , 7.78-7.83 (d ,2H , H2,6-arom C) , 7.62-7.68 (d ,2H , H2,6-arom B) , 7.39 (s ,1H , H2-arom A) , 7.38 , 6.82-6.85 (d ,2H , H3,5-arom C) , 6.66-6.69 (d ,2H , H3,5-arom B) , 3.42 (s,*3H*, OMe) , 2.66 (s,*3H*, N-Me) , 2.09 (s,*3H*, Me-C=C) ^{13}C-NMR (100 MHz, DMSO-d6) 158.4(C=O, Amide) , 156.9 (C=N) , 153.5 (C5-arom A) , 152.2 (C4-arom A) , 149.7 (C3-arom A), 148.4 (C1-arom C) , 146.5 (C1-arom B), 138.7 ($C4^{--}$ -Pye) , 136.4 (C3-Pyr) , 132.2 (C6-arom A) , 131.6 ($C3^{--}$ -Pye) , 129.1 (C1-arom A) , 128.5 (C3,5-arom C), 127.3(C3,5-arom B) , 126.7 (C2,6-arom B), 125.8 (C2,6-arom C) , 123.9 (C4-arom C) , 122.8 (C4-arom B) , 118.4 (C4-Pye) , 114.8 (C2-arom A), 55.6 (OMe), 20.8 ($Me2^{--}$ -Per) , 20.6 (Me2-Per) , 17.2 ($Me3^{--}$ -Per) , 16.4 (Me3-Per).

2.2.3 Synthesis of Cu (II) and Ni (II) complexes [16]

The ion complexes were obtained by adding equimolar amounts of Cu (II) and Ni (II) salts (1 mmole) in 10 ml of ethyl alcohol to an ethanolic solution of the (2 mmole) azo-schiff derivative and refluxing for 2 h. The product solid complexes were filtered off, washed and recrystallized from ethanol, and dried in air. Cu Complex : FT-IR (KBr) (cm^{-1}): 3400 (OH) , 2924 and 2850 (C-H aliph) , 1635 (C=O , amide) , 1674 (C=N, arom) , 1591(C=C, arom) , 1433 (N=N) , 536 (Cu-N) , 474 (Cu-O). Ni Complex : FT-IR (KBr) (cm^{-1}): 3400 (OH) , 2927 and 2850 (C-H aliph) , 1639 (C=O , amide) , 1660 (C=N, arom) , 1589 (C=C, arom) , 1427 (N=N) , 532 (Ni-N) , 476 (Ni-O).

2.2.4 Characterization of ligand and its complexes

The complexes were insoluble in water but soluble in DMF, DMSO and EtOH solvents. The ligand was Dark brown crystals. Table 1 showed some physical properties of the prepared ligand and its complexes.

Table 1. Physical properties of the Azo-Schiff derivative

Compound	Formula	Color	Yield (%)	M.P (°C)
Schiff base derivative	$C_{19}H_{19}N_3O_3$	pale yellow	82 %	141-143
Azo-Schiff derivative (L)	$C_{30}H_{29}N_7O_4$	Deep brown	74 %	180-182
$[CuL_2Cl_2]$	$[Cu(C_{30}H_{28}N_7O_4)_2Cl_2]$	Deep orange	70 %	126-128
$[NiL_2Cl_2]$	$[Ni(C_{30}H_{28}N_7O_4)_2Cl_2]$	Pale red	75 %	106-107

M.P: melting point

2.3 Anti-bacterial activity

Three derivative compounds (Cu, Ni complexes, and Azo-Schiff) have been used as antibacterial compounds in antibacterial activity tests according to the agar well diffusion method [17, 18]. Two concentrations: 100 and 200 (mg/ml) were made from each derivative compounds [19]. Two aerobic pathogenic bacteria were selected: *S.aureus* and *P.aeruginosa* were provided kindly from the department of microbiology faculty of science university of Kufa. Four wells were made in Muller-Hinton agar surface by crock poorer 5mm swabbed with two pathogenic bacteria according to 0.5 MacFarland turbidity. Thirty microliters of each dilution were transferred to each well, left at 20°C for two hours, and incubated at 37°C for 24h. Three replicates were done for each test. The inhibition zone around each well was measured in millimeters [20].

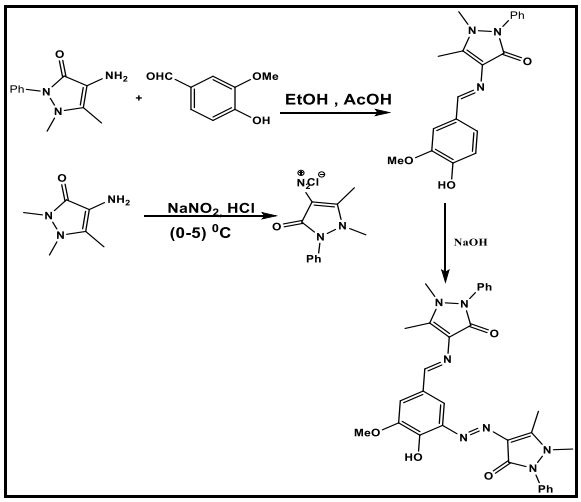
2.4 Statistical analysis

Graph Pad prism V.10 windows software has been used in statistically analysis. Mean± standers error (SE) have been used to compare between diameters of inhibitions zones [21, 22].

3. Results and discussion

3.1 Chemistry

4-AAP was key intermediate for prepared azo methane compound after treated it with 4-Hydroxy-3-methoxy benzaldehyde (Vanillin) in acidic medium, then the azo compound synthesized after coupled with diazonium salt that prepared from another 4-AAP molecule [23, 24]. The synthesized derivative of the azo-Schiff compound was outline in scheme 1.



Scheme 1. Preparation steps of the azo-Schiff compound

The structures of new compounds were identified by spectral methods (FT-IR, ^{1}H-NMR, ^{13}C-NMR), The ^{1}H-NMR spectrum of 1 showed two signals at lower field 9.62 and 9.48 ppm resonated to protons of hydroxyl and azo methane groups respectively, while the protons of aromatic rings appeared as singlet, doublet and triplet signals at range 7.57-6.86 ppm, whereas the spectrum of azo compound 2 showed proton of the hydroxyl group at 14.28 ppm due to intramolecular hydrogen-bonded while the imine proton showed singlet at 9.77 ppm, furthermore the aromatic protons displayed at 8.35-6.66 ppm the more lower field than Schiff base spectrum this due to the effect of substituent groups but the protons of methoxy and methylamine appear approximately at the same chemical shift of spectrum 1 (Fig.1).

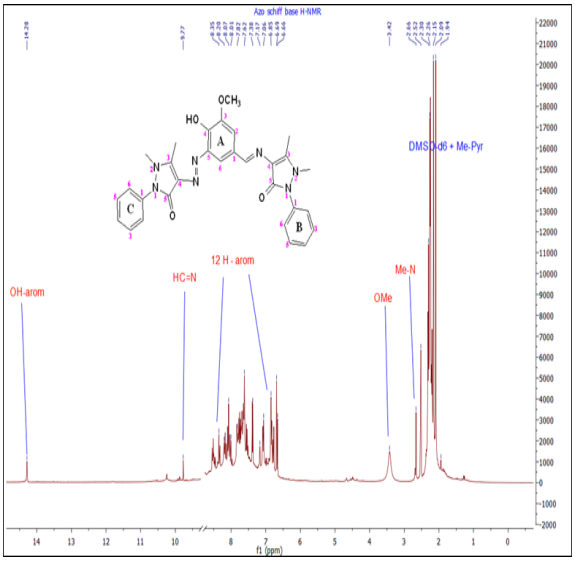


Fig.1. ^{1}H-NMR spectrum of Azo-Schiff compound

The ^{13}C-NMR spectrum showed a signal at 160.4 and 158.4 ppm assigned for carbonyl lactam of derivative 1,2 respectively, while the resonances of (C=N) group of compounds 1,2 appeared at 155.5 and 156.9 ppm respectively, moreover, compounds 1,2 showed signals at region 110-152.2 and 114.9-153.5 ppm respectively, which were assigned to the carbon atoms of aromatic ring and antipyrine, the other signals of derivatives 1,2 showed approximately the same chemical shift as shown in Fig.2.

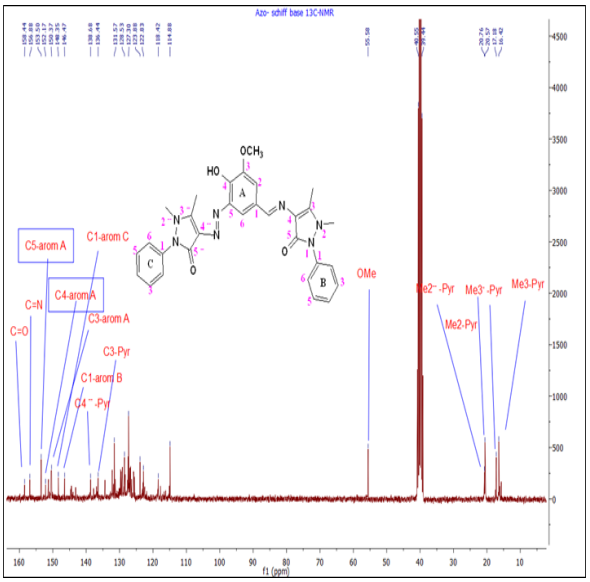


Fig.2. ^{13}C-NMR spectrum of Azo-Schiff compound

3.2 UV-visible spectrum

UV-Vis Spectra of alcohol solution for Azo-Schiff derivative showed tow bands at (234,340) nm attributed to (π –π*) transitions of the aromatic rings, Intra charge transfer, and also other band in (480) nm resonated to (n-π*) transitions (Fig.3). All of these bands appear blue shift in the complexes spectra as a results of coordination [25, 26]. The values are explained in the Fig. 4, 5 and Table 2.

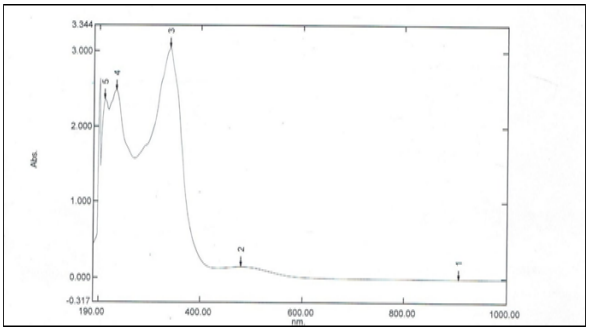


Fig. 3. UV-visible spectrum of Azo-Schiff compound

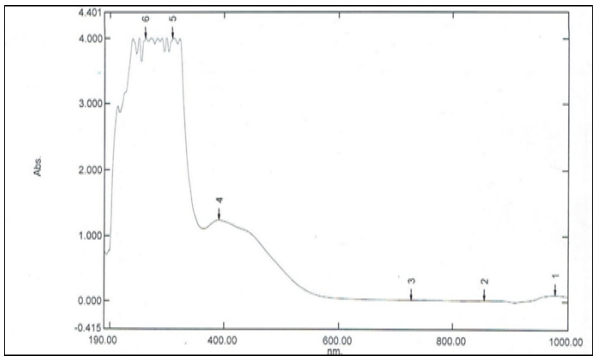


Fig.4. UV-visible spectrum of Cu-Complex

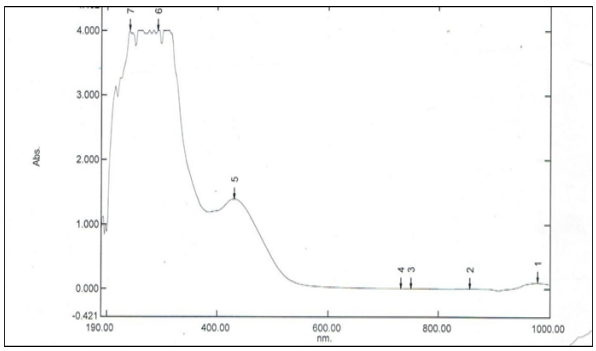


Fig.5. UV-visible spectrum of Ni-Complex

Table 2. Electronic spectra and molar conductance of free ligand

Compound	λ nm	Assignment	Molar Conductivity **(S.mol^{-1}.cm^{2})**	Geometry
Azo-Schiff derivative (L)	480 340 234	n→ π* π→π*	–	–
[CuL2]	390 310 262	Ligand field Charge transfer π→π*	20.4	Octahedral
[NiL2]	430 292 242	Ligand field Charge transfer π→π*	19.2	Octahedral

3.3 Molar Conductivity

Conductivity measurements of the complexes in solvent (DMSO) as shown in Table 2, the values indicate that the complexes are non- ionic character, where the values suggest that no anions present outside the coordination spheres [27].

Suggested chemical structure of the complexes:
According to spectrophotometric results and molar conductivity suggested that the complexes have octahedral geometry as Fig.6.

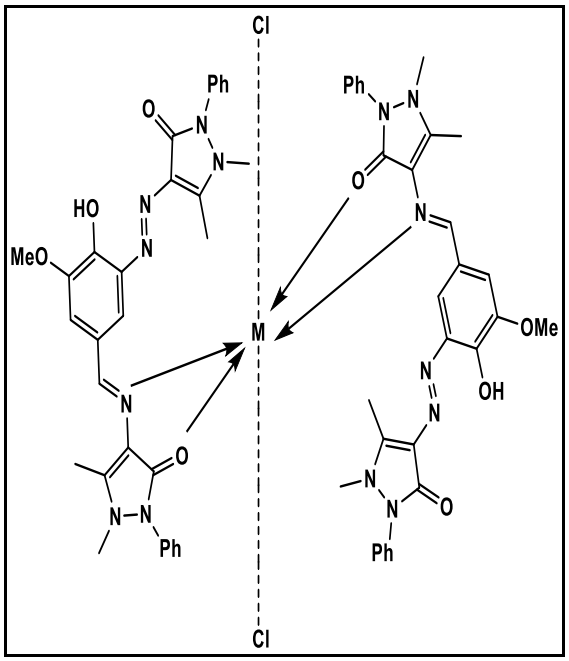


Fig.6. Chemical structure of complexes

3.4 Biological activity

The results proved (Table 3) that the derivative compound Azo-Schiff with a 200 mg/ml concentration has good antibacterial activity with a diameter of inhibition zone of 16.11 ± 0.1035 mm and 13.21 ± 0.4044 against S. *aureus* and *P. aeruginosa*, respectively (Fig. 7). The results of the current study are in agreement with many previous studies that concluded azo compounds have an excellent inhibitory effect against pathogenic bacteria and fungi [28, 29, 30].

Fig.7. Antibacterial activity of three derivative compounds against *S. aureus* on to Muller-Hinton agar surface after incubated at 37 °C for 24h. 1: the Ni complex, 2: the Azo-Schiff compound, and 3: the Cu complex.

Table 3. Antibacterial activity of three derivative compounds against pathogenic bacteria

Derivative compounds	Aerobic pathogenic bacteria			
	S. aureus (Gram-positive)		*P. aeruginosa* (Gram-negative)	
	Con. mg/ml	M±SE(mm) R=4	Con. mg/ml	M±SE(mm) R=4
Cu complex	100	8.5500 ± 0.25736	100	8.3487 ± 0.27837
	200	9.3433 ± 0.12441	200	8.7000 ± 0.27025
Ni complex	100	9.4833 ± 0.20185	100	9.3833 ± 0.22981
	200	9.7200 ± 0.24987	200	9.6333 ± 0.23709
Azo-Schiff	100	11.377 ± 0.18114	100	10.693 ± 0.10349
	200	16.11 ± 0.1035	200	13.21 ± 0.4044
AMC	30 µg	16.13 ± 0.2152	AMC	12.92 ± 0.3312

Con.: Concentration, M±SE: Mean± standard error, R: Numbers of replicates, AMC: Amoxiclav(standard)

4. Conclusions:

Schiff base, Azo-Schiff base, nickel (II), and copper (II) complexes have been prepared successfully from 4-amino antipyrine and vanillin and diagnosed by UV-visible, FT-IR, ^{1}H-NMR, and ^{13}C-NMR. The Azo-Schiff base compound has an excellent inhibitory effect against S. *aureus* and *P. aeruginosa*. Therefore, these compounds may be considered a raw material for manufacturing treatments against these bacterial infections.

Declaration of competing interest

There is no conflict of interest in this study.

Funding and assistance

An independent study, this one did not receive any financial assistance**.**

References

[1] Eftekhari-Sis, B., Zirak, M. and Akbari, A. 2013. Arylglyoxals in synthesis of heterocyclic compounds. *Chemical reviews*, *113*(5), 2958-3043 (2013).

[2] Fadda, A.A., Rabie, R., Bondock, S. and Etman, H.A. Derivational, Structural, and Biological Studies of Some New Pyrazolyl, Isoxazolyl, Pyrimidinyl, Pyridazinyl, and Pyridopyridazinyl from 4-Substituted Antipyrine. *Journal of Heterocyclic Chemistry*, *54*(2), 1304-1310 (2017)

[3] Maroyi, A. Review of ethnomedicinal, phytochemical and pharmacological properties of Lannea schweinfurthii (Engl.) Engl. Molecules, 24(4), 732 (2019)

[4] Elattar, K.M. and Fadda, A.A. 2016. Chemistry of antipyrine. *Synthetic Communications*, *46*(19), 1567-1594 (2016)

[5] Bayrak, H., Cebeci, Y.U. and Karaoğlu, Ş.A. Synthesis of Novel Antipyrine Derivatives Possessing Remarkable Antimicrobial Activities. ChemistrySelect, 4(44), 12906-12908 (2019)

[6] Rammohan, A., Reddy, G.M., Krinochkin, A.P., Kopchuk, D.S., Savchuk, M.I., Shtaitz, Y.K., Zyryanov, G.V., Rusinov, V.L. and Chupakhin, O.N. A facile synthesis of triazine integrated antipyrine derivatives through ecofriendly approach. Synthetic Communications, 51(2), 256-262 (2021)

[7] Ei Ashry, E.S.H., Awad, L.F., Ibrahim, E.I. and Bdeewy, O.K. Synthesis of antipyrine derivatives derived from dimedone. Chinese Journal of Chemistry, 25(4), 570-573 (2007)

[8] Premnath, D., Enoch, I.V., Selvakumar, P.M., Indiraleka, M. and Vennila, J.J. Design, synthesis, spectral analysis, in vitro anticancer evaluation and molecular docking studies of some fluorescent 4-amino-2, 3-dimethyl-1-phenyl-3-pyrazolin-5-one, ampyrone derivatives. Interdisciplinary Sciences: Computational Life Sciences, 9(1), 130-139 (2017)

[9] Rosu, T., Negoiu, M., Pasculescu, S., Pahontu, E., Poirier, D. and Gulea, A. Metal-based biologically active agents: Synthesis, characterization, antibacterial and antileukemia activity evaluation of Cu (II), V (IV) and Ni (II) complexes with antipyrine-derived compounds. European Journal of Medicinal Chemistry, 45(2), 774-781 (2010)

[10] Adam, R.W., Al-Labban, H.M.Y., Aljanaby, A., A., J., and Abbas, N.,A. Synthesis, Characterization and Antibacterial Activity of Some Novel 1, 2, 3-Triazol-Chalcone Derivatives from N-Acetyl-5H-Dibenzo [b, f] Azepine-5-Carboxamide. Nano Biomed. Eng, 11(2), 99-110 (2019)

[11] Al-labban, H., M., Y., and Aljanaby, A.A.J. An overview of some heterocyclic organic compounds; synthesis, characterization, thermal properties and antibacterial activity. International Journal of Pharmaceutical Research, Supplementary Issue 1, 2394-2399 (2020)

[12] Kant, R. and Maji, S. Recent advances in the synthesis of piperazine based ligands and metal complexes and their applications. Dalton Transactions, 50(3), 785-800 (2021)

[13] Khurana, P., Pulicharla, R. and Brar, S.K. Antibiotic-metal complexes in wastewaters: fate and treatment trajectory. Environment International, 157, p.106863 (2021)

[14] Abdul-Rida, N.A., Farhan, A.M., Al-Masoudi, N.A., Saeed, B.A., Miller, D. and Lin, M.,F. A novel pregnene analogs: synthesis, cytotoxicity on prostate cancer of PC-3 and LNCPa-AI cells and in silico molecular docking study. Molecular diversity, 25(2), 661-671 (2021)

[15] Özkınalı, S., Gür, M., Şener, N., Alkın, S. and Çavuş, M.,S. Synthesis of new azo schiff bases of pyrazole derivatives and their spectroscopic and theoretical investigations. Journal of Molecular Structure, 1174, 74-83 (2018)

[16] Anitha, C., Sumathi, S., Tharmaraj, P. and Sheela, C.D. Synthesis, characterization, and biological activity of some transition metal complexes derived from novel hydrazone azo schiff base ligand. International Journal of Inorganic Chemistry, 2011(2012)

[17] Aljanaby, A.A.J., 2018. Antibacterial activity of an aqueous extracts of Alkanna tinctoria roots against drug resistant aerobic pathogenic bacteria isolated from patients with burns infections. Russian Open Medical Journal, 7(1) (2018)

[18] Majeed, H.T. and Aljanaby, A.A.J., 2019. Antibiotic susceptibility patterns and prevalence of some extended spectrum beta-lactamases genes in gram-negative bacteria isolated from patients infected with urinary tract infections in Al-Najaf City, Iraq. Avicenna journal of medical biotechnology, 11(2), 192 (2019)

[19] Aljanaby, A.A.J. and Aljanaby, I.A.J. Profile of antimicrobial resistance of aerobic pathogenic bacteria isolated from different clinical infections in Al-Kufa central hospital-Iraq during period from 2015 to 2017. Research Journal of Pharmacy and Technology, 10(10), 3264-3270 (2017)

[20] Yuan, G., Guan, Y., Yi, H., Lai, S., Sun, Y. and Cao, S. Antibacterial activity and mechanism of plant flavonoids to gram-positive bacteria predicted from their lipophilicities. Scientific reports, 11(1), 1-15 (2021)

[21] Alhasnawi, H.M.R.J. and Aljanaby, A.A.J. Evaluation of Galectin-3 and CD19 in *Helicobacter pylori* patients infected with stomach cancer. Gene Reports, 26.101520 (2022)

[22] Aljanaby, A.A.J., Al-Faham, Q.M.H., Aljanaby, I.A.J. and Hasan, T.H., 2022. Immunological role of cluster of differentiation 56 and cluster of differentiation 19 in patients infected with mycobacterium tuberculosis in Iraq. Gene Reports, 101514 (2022)

[23] Joseph, J., Nagashri, K. and Rani, G.A.B. Synthesis, characterization and antimicrobial activities of copper complexes derived from 4-aminoantipyrine derivatives. Journal of Saudi Chemical Society, 17(3), 285-294 (2013)

[24] Erturk, A.G. Synthesis, structural identifications of bioactive two novel Schiff bases. Journal of Molecular Structure, 1202, 127299 (2020)

[25] Özdemir, Ö. Synthesis of new luminescent bis-azo-linkage Schiff bases containing amino-phenol and its derivative. Part I: Studying of their tautomeric, acidochromic, thermochromic, ionochromic, and photolüminesence properties. Journal of Photochemistry and Photobiology A: Chemistry, 380, 111868 (2019)

[26] Pervaiz, M., Sadiq, S., Sadiq, A., Younas, U., Ashraf, A., Saeed, Z., Zuber, M. and Adnan, A. Azo-Schiff base derivatives of transition metal complexes as antimicrobial agents. Coordination Chemistry Reviews, 447, 214128 (2021)

[27] Wong, C.L., Soriano, A.N. and Li, M.H. Diffusion coefficients and molar conductivities in aqueous solutions of 1-ethyl-3-methylimidazolium-based ionic liquids. Fluid Phase Equilibria, 271(1-2), 43-52 (2008)

[28] Al-Hamdani, A.A.S., Balkhi, A.M., Falah, A. and Shaker, S.A. Synthesis and investigation of thermal properties of vanadyl complexes with azo-containing Schiff-base dyes. Journal of Saudi Chemical Society, 20(5), 487-501 (2016)

[29] AbouEl-Enein, S.A., Emam, S.M., Wagdy, R.M. and Abouzayed, F.I. Spectral and thermal investigation of novel biologically active (N-(1, 5-dimethyl-3-oxo-2-phenyl-2, 3-dihydro-1H-pyrazol-4-yl)-2-(1, 5-dimethyl-3-oxo-2-phenyl-2, 3-dihydro-1H-pyrazol-4-yl-amino)-2-oxo-cetimidic acid) ligand and its metal complexes. Journal of Molecular Structure, 1215, 128230 (2020)

[30] Kargar, H., Aghaei-Meybodi, F., Behjatmanesh-Ardakani, R., Elahifard, M.R., Torabi, V., Fallah-Mehrjardi, M., Tahir, M.N., Ashfaq, M. and Munawar, K.S. Synthesis, crystal structure, theoretical calculation, spectroscopic and antibacterial activity studies of copper (II) complexes bearing bidentate Schiff base ligands derived from 4-aminoantipyrine: influence of substitutions on antibacterial activity. Journal of Molecular Structure, 1230, 129908 (2021)