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A Comparative Study of some New Heterocyclic Compounds Containing the 1,2,4-Triazole Moiety

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ABSTRACT

The present study reports the synthesis of a range of compounds including oxazepines (5a-e), benzoxazepines (6a-e), imidazolidines (7a-e) and (8a-e), and tetrazoles (9a-e) containing 1,2, -4-triazole. These compounds were synthesized from benzilic acid hydrazide (1) using both traditional and green approaches, with the latter utilizing ultrasound technology to increase the yield and speed up reaction times. Thus, the bezilic acid hydrazide was first converted to potassium2-(2-hydroxy-2,2-diphenylacetyl) hydrazine-1–carbodithioate) (2) followed by treatment with hydrazine hydrate to produce (4-amino -5- mercapto- 4H-1,2,4-triazole-3-yl) diphenyl methanol (3). Compound (3) was then reacted with various substituted benzaldehydes to produce Schiff base compounds (4a-e). The cyclization of compound (4a-e) with many reagents provides the product with oxazepines، benzoxazepines, imidazolidineand tetrazoles compounds. The results proved that the use of ultrasound technology proved to be a more efficient method compared to traditional approaches, as it resulted in shorter reaction times and high-quality products. The purity of all synthesized compounds was determined using thin-layer chromatography (TLC), and the structures were confirmed through physical and spectral analysis including FT-IR and ^{1}H NMR.

Keywords: Hydrazones, 1,2,4-triazole, Potassium salt, Oxazepine, Imidazolidine, Oxadiazole**.** Tetrazole.

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INTRODUCTION

1,2,4-triazole and its derivatives have considerable biological activity as antibacterial (Gao *et al*., 2019) antifungal (Ezabadi *et al*., 2008) and antitumor (El-Sherief *et al.*, 2018). Also, for the treatment of pain and inflammatory diseases (Tariq *et al*., 2018). A large number of 1,2,4- triazoles have been used in therapeutically interesting drugs, including antivirals (Mohamed *et al*., 2021), analgesics (Almasirad *et al*., 2014) and some triazoles can be used in the other tissue culture system for different plants (Kawada *et al.*, 2019). Derivatives of imidazole-4-one are frequently employed as precursors in the production of organic substances, such as medicines and photographic chemicals (Bayoumi, 2012; El-Hady and Abubshait, 2017) and their antibacterial activity (Nath *et al.*, 2020). Also, the imidazole derivatives have different pharmacological activities in addition to their agricultural use as an insecticide and herbicide (Luczynski and Kudelko, 2022). Numerous experts from all over the world have worked to create 1,3-oxazepine derivatives and report on their synthesis and biological activity in the scientific literature (Hariprasad and Srinivasa, 2015; Muslim, 2019). It has been reported that tetrazoles They possess antiviral, analgesic, anti-inflammatory, and anticancer activities (Alagarsamy *et al*., 2018; Hasan, 2019). It has been reported to use some Schiff bases that carry triazole rings with biological activities (Dawood *et al*., 2020; Taha, 2017).

EXPERIMENTAL

All components and chemical reagents from the Aldrich, BDH, and Fluka firms were used without purification. The Stuard-SMP30 was used to determine each melting point and is incorrect. Recordings of FTIR spectra were made using the FTIR. Spectrophotometer, equipped with a 600 brot detector. Tech engineering management. The 1H-NMR spectra were acquired on a JEOLEEA 400 MHZFT-NMR instrument by employing TMS as the instrument's internal standard and utilizing DMSO-d6 as the solvent. TLC assay presents all reactions and purity of compounds. The esterification of benzilic acid prepared the methyl benzilate in the presence of sulfuric acid. When treated with hydrazine hydrate, gave benzilic acid hydrazide according to the reported method (AL-Obaidy *et al*., 2020). Thus, in amino acids the carboxyl function can be esterified with an excess of methanol under acidic conditions.

Synthesis of Potassium 2-(2-Hydroxy-2,2-Diphenylacetyl) Hydrazine-1-Carbodithioate (2) (Wang *et al*., 2010).

To a cold stirred solution benzilic acid hydrazide (1) (0.01mols) in absolute ethanol (30mL) containing potassium hydroxide (0.8g, 0.015 mols), carbon disulfide (0.7g ,0.015 mols) was added. The reaction mixture was stirred at room temperature for nine hours, cooled, the resulting yellow precipitate was then filtered, washed with dry ether, and recrystallized from ethanol to yield a dark yellow product with a melting point of 198 to 200°C.

Synthesis of (4-Amino-5-Mercapto-4H-1,2,4-Triazol-3-yl) Diphenylmethanol (3) (Mohammed *et al*., 2021).

A suspension of salt (2) (0.1mol) in (20 mL) absolute ethanol (0.2 mol) of hydrazine hydrate and 5 mL of This mixture was refluxed for 4 hours, during which time the colour changed to light green. The reaction mixture was then cooled and poured into distilled water (50 mL), the solution was acidified with 10% hydrochloric acid. The resulting solid was filtered, washed with cold water, and recrystallized from methanol to give a pale-yellow compound (3). Yield 55%, m.p. (190-192ºC).

Synthesis Of (Z)-(4-(Benzylideneamino)-5-Mercapto-4H-1,2,4-Triazol-3-yl) Diphenylmethanol (4a- e):

Conventional Method (A) (Mohammed *et al*., 2021)

A mixture of substituted benzaldehyde (0.01mol), (0.01mol) of 4-amino-5-mercapto-4H-1,2,4-triazolyl) diphenyl methanol of (3) in an absolute ethanol (25 mL) and glacial acetic acid (0.2 mL) were added. The mixture was heated under reflux conditions for (0.5- 4 hrs.). The resulting material was filtered, cooled, dried, and recrystallized from a suitable solvent to give the resulting hydrazones (4a-e). The physical properties are listed in (Table 1).

Method B (Ultrasound Technique)

A mixture solution of (0.01 mol) 4-amino-5-mercapto-4H-1,2,4-triazolyl) diphenyl methanol of (3), aromatic aldehyde (0.001 mol), and zirconyl chloride hexahydrate in ethanol (15 mL) The solution was subjected to sonication using Unisonic PTY.LTD type FXp12. Then raise the temperature to 20-35o C, after sonication for about (20min.), and Monitored by TLC. The reaction mixture was filtered, and recrystallization from a suitable solvent yield's pure products (4a-e). Physical data are listed in (Table 1).

Table 1: Physical Properties for Compounds (4a-e).

Comp. No.	R	M.p.(^{o}C)	Yield% Method		Colour	Time of Reaction hrs. (Method A)	Crystallization solvent
			A	B			
4a	4- NO_2	246-248	75	92	Yellow	0.5	Ethanol
4b	4- Cl	278-280	75	94	Brown	1	aq. Ethanol
4c	4-Br	216-218	74	94	Dark yellow	4	Acetone
4d	2-NH_2,	233-235	68	90	Pale yellow	4	Methanol
4e	4-OCH_3	246-247	67	89	Yellow	2.5	Methanol

Preparation of Oxazepines Derivatives (5a-e) and(6a-e) (garcía *et al*., 2019), Synthesis of Substituted 1,2,5-Oxadiazine Compounds (6a-e) (7a-d) Conventional Method (a)

A mixture of (0.01mol) hydrazones of (4a-e) and the (0.01mol) appropriate acid anhydride, succinic anhydride or phthalic anhydride, in (20 ml) THF was refluxed for about (14hrs.), cooled, the solid precipitate formed was filtered, dried, and then recrystallized from benzene and ethanol (2:1) to give compounds (5a-e) and (6a-e), respectively. The physical properties of these prepared compounds are presented in (Tables 2 and 3), respectively.

(Ultrasound Technique) Method (B)

A mixture of (0.001mol) compounds (4a-e), phthalic anhydride or succinic anhydride and (0.1gm) of zirconyl chloride hexahydrate were irradiated by an ultrasonic generator in a water bath at (40-50 °C) for 25min. (Monitored by TLC). On completion of this reaction, the formed solid was filtered off, washed with ethanol, dried, and recrystallized from a suitable solvent to give pure compounds (5a–e) and (6a-e). The Physical data are recorded in (Tables 2 and 3), respectively.

Table 2: The Physical data for compounds (5a-e)

Comp No.	R	MP (C)	Yield% Method A	Yield% Method B	Colour	Crystallization solvent
5a	4-NO_2	208-209	80	87	Yellow	Ethanol
5b	4-Cl	200-201	75	91	Brown	Methanol
5c	4-Br	190-192	68	88	Yellow	aq. Ethanol
5d	2-NH_2	188-190	78	92	Yellow	Ethanol
5e	4-OCH_3	194-195	72	86	Pale Yellow	Chloroform

Table 3: The Physical data for compounds (6a-e)

Comp No.	R	MP (C)	Yield% Method A	Yield% Method B	Colour	Crystallization solvent
6a	4-NO_2	218-220	85	93	Pale Yellow	aq. Ethanol
6b	4-Cl	250-251	70	89	Brown	Acetone
6c	4-Br	190-192	65	86	Pale Yellow	Ethanol
6d	2-NH_2	188-190	70	90	Yellow	Methanol
6e	4-OCH_3	194-195	65	85	White-Yellowish	Ethanol

Preparation of Imidazolidine Derivatives (7a-e) and (8a-e) (Botros *et al*., 2013).

Conventional Method (A)

A mixture of (0.01mol) hydrazones (4a-e) and (0.01mol) of appropriate ester of α-amino acid, (glycine or alanine) in (10 mL) THF, was refluxed for about (12hrs.). Pour the mixture into ice water. The solid product formed is filtered, dried and recrystallized from ethanol and THF (1:3) to give the titled tetrazoles derivatives. (7a-d) and (8a-e), respectively, the physical data are shown in (Tables 4 and 5).

(Ultrasound Technique) Method (B)

A mixture of (0.01mol) compounds (4a-e), (0.01mol) of glycine or alanine and (0.1gm) of zirconyl chloride hexahydrate. The final mixture was ultrasonically irradiated at 25 °C for about 30 min. After the reaction is completed, the mixture is filtered. The remainder was washed with 95% ethanol. The physical data are shown in (Tables 4 and 5), respectively.

Table 4: The Physical data for compounds (7a-e)

Comp No.	R	MP (C)	Yield% Method A	Yield% Method B	Colour
7a	4-NO2	172-173	65	91	Dark yellow
7b	4-Cl	229-231	70	88	Dark brown
7c	4-Br	163-165	65	87	Pale Yellow
7d	2-NH_2	212-214	70	89	Yellow
7e	4-OCH3	203-205	65	87	White

Table 5: The Physical data for compounds (8a-e)

Comp No.	R	MP (C)	Yield % Method		Colour
			A	B	
8a	4-NO2	180-182	60	85	Brown
8b	4-Cl	199-200	55	84	Brown
8c	4-Br	166-168	66	82	Yellowish White
8d	2-NH_2	191-192	54	78	Yellow
8e	4-OCH3	205-207	50	75	Yellow

Preparation of tetrazole derivatives (9a-e).

Conventional Method (A) (Wittenberger, 1994).

Sodium azide (0.02mol) was added to a stirring solution of hydrazones(4a-e) (0.01mol) in dry tetrahydrofuran (15 mL). The mixture was refluxed approximately (3 h) with stirring and cooled at room temperature. The precipitate was filtered, washed with water. It is recrystallized from petroleum ether to give compounds (9a-e). The physical data are shown in (Table 6).

Method B (Ultrasound Technique)

A mixture of (0.01mol) compounds (4a-e), (0.01mol) sodium azide and (0.1gm) of zirconyl chloride hexahydrate. The reaction mixture was irradiated with ultrasound at 25ºC for about 40 mi. The reaction was monitored by TLC technology. After the reaction was completed, the precipitate was filtered, washed with ethanol and it is recrystallized from petroleum ether to give tetrazoles (9a-e). The physical data is shown in (Table 6).

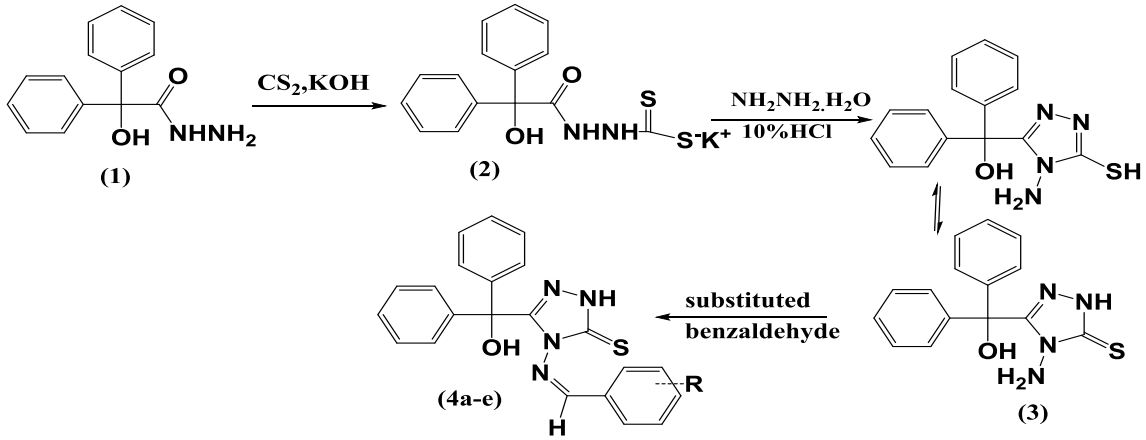
Table 6: Physical data for compounds (9a-e)

Comp No.	R	MP (C)	Yield % Method		Colour
			A	B	
9a	4-NO_2	199-200	60	79	White
9b	4-Cl	220-221	55	74	Brown
9c	4-Br	190-191	54	81	Yellow
9d	2-NH_2	175-176	50	82	Yellow
9e	4-OCH_3	210-211	48	73	Yellow

RESULTS AND DISCUSSION

Continuation of our efforts to study and develop Schiff bases and as part of this ongoing work, we have recently synthesized several new hydrazones. New compounds containing 1,2,4 –triazole (4a-e) were obtained by condensation reaction of 4-amino-5-mercapto-4H-1,2,4-triazole-3-yl) diphenylmethanol (3) with some substituted benzaldehyde as shown in Scheme 1, obtained hydrazone compounds (4a-e) are solids at room temperature and their spectral data (FT-IR,1H-NMR) in addition to physical properties and monitor reactions were checked by thin-layer chromatography. The FT-I.R. spectrum of the triazole (3) showed broad bands of (O.H.) group at (3385cm^{-1}), two bands (NH_2) group at (3258 cm^{-1}) and a band of (C=N) group; at (1651cm^{-1}) and another band at (1078 cm^{-1}) due to C=S groups. This assignment was further supported by 1H-NMR spectrum data which showed multiple bands at δ(6.69-6.96ppm) for (10H) aromatic protons, a singlet band at (5.36 ppm) for (O.H.) protons and broadband at (4.65 ppm) for two (NH2) protons. The structures of the target hydrazones (4a-e) were elucidated using (FT-IR and 1H-NMR). The FT-IR spectra for compounds (4a-e) showed the following stretching bands; (1652-1671cm^{-1}) due to the (C=N) bond for heterocyclic ring, (1598-1632cm^{-1}) due to (C=N) bond for

hydrazones, (1065-1071 cm $^{-1}$) for C=S) group, (3198-3302cm^{-1}) for the (NH) bond (3342-3390cm $^{-1}$) due to (NH) group. The 1H-NMR spectra for compounds (4a,4b,4d) in (DMSO-d6) representation for these hydrazones (4a-e) showed the peaks for (CH=N, NH, and aromatic protons) and other groups in these compounds is in full agreement with the proposed structures 21 which confirmed in (Table 7).

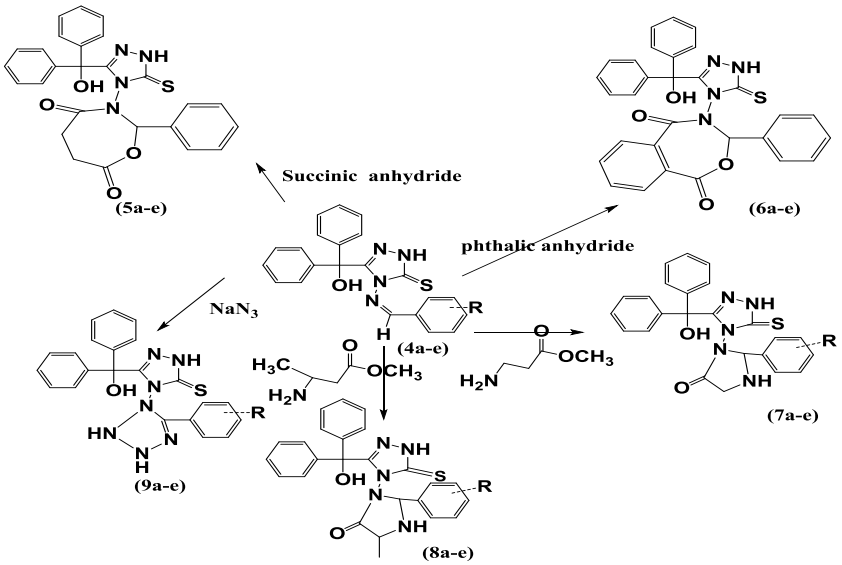


Scheme 1: Synthesis of Schiff bases compounds (4a-e)

Table 7: The spectral data for compounds (4a-e)

Comp. No.	R	FT.IR ν(cm^{-1})					^{1}H -NMR δ(ppm) DMSO-$_6$
	4- NO_2	C=N (cyclic)	C=N (hydrazone)	C=S	NH	OH	
4a	4- Cl	1655	1632	1071	3198	3371	5.6(s,1H, OH),7.55(s,1H, NH),6.14(s1H, CH=N),6.64-7.53(m,14H, ArH),
4b	4-Br	1663	1605	1067	3221	3342	5.59(s,1H, OH),7.71(s,1H, NH),6.11(s1H, CH=N),6.59-7.64(m,14H, ArH),
4c	2-NH_2,	1652	1598	1075	3271	3388	5.6 2(s,1H, OH),7.48(s,1H, NH),6.08(s1H, CH=N),6.57-7.47 (m,14H, ArH),
4d	4- OCH_3	1661	1610	1067	3264	3390	5,37(s,2H, NH_2),5.6(s,1H, OH),7.56(s,1H, NH),6.05(s1H, CH=N),6.76-7.6m,14H, ArH),),
4e	4- NO_2	1671	1618	1069	3302	3365	3.793(s,3H, OCH_3),5.6O (s,1H, OH),7.80(s,1H, NH),6.06(s1H, CH=N),6.76-7.55(m,14H, ArH),),

Compounds (5a-e) and (6a-e) were prepared by reacting Schiff base compounds (4a-e) with succinic anhydride and phthalic anhydride, respectively, which are shown in Scheme 2. These products were characterized by physical and spectral data using. FT-IR and ^{1}H-NMR, thus the IR spectra for compound (5a-e) and (6a-e) showed the following frequencies arrange (1648-1671) and (1651-1674) cm^{-1} due to (C=O lactam) group and at (1728-1746) and (1732-1752) cm^{-1} for (C=O lactone) group. While ^{1}H-NMR (DMSO-d_6) spectral data of compounds (5a-e) and (6a-e) are summarized in (Tables 8 and 9), respectively.



Scheme 2: Synthesis of titled compounds

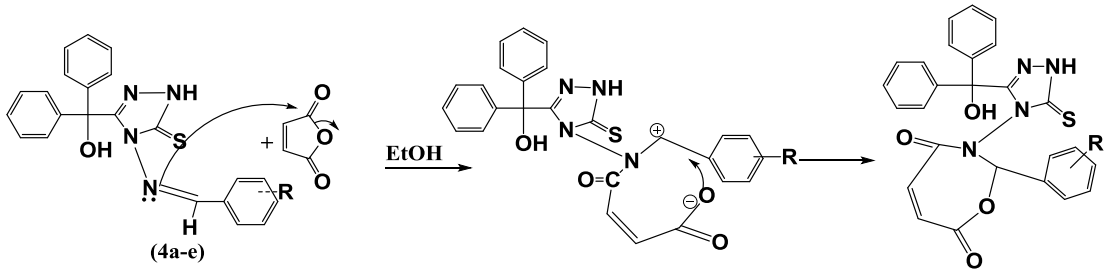
Table 8: The spectral data for compounds (5a-e)

Comp. No.	FT.IR ν(cm^{-1})				^{1}H -NMR δ(ppm) DMSO-$_6$
	C=O (lactone)	C=O (lactam)	C=S	CH Ar.	
5a	1732	1665	1084	3054	5.68(s,1H,CH=OCO),5.85(s,1H,OH),6.12(s,H,CH=CON),6.63(bs,1H,CH-substituted aromatic ring),7.53(s,1H,NH),7.55-8.06.(s,13H,ArH).
5b	1735	1665	1074	3062	5.34(s,1H,CH=OCO),5.52(s,1H,OH),5.98(s,H,CH=CON),6.63(bs,1H,CH-substituted aromatic ring),,7.48(s,H,NH),7.62-8.18.(s,13H,ArH)
5c	1728	1671	1069	3066	5.32(s,1H,CH=OCO),5.56(s,1H,OH),5.99(s,H,CH=CON),6.62(bs,1H,CH-substituted aromatic ring),,7.49(s,H,NH),7.64-8.16.(s,13H,ArH)
5d	1746	1654	1062	3088	5.45(s,2H,NH_2),5.53(s,1H,CH=OCO),6.03(s,1H,OH),6.08(s,H,CH=CO-N),7.43(s,H,NH),6.93-7.42.(s,13H,ArH)
5e	1737	1648	1075	3075	3.82(s,3H,OCH_3),5.61(s,1H,CH=OC0),5.92(s,1H,OH),6.05(s,H,CH=CON),7.42(s,H,NH),7.16-7.12.(s,13H,ArH)

Table 9: The spectral data for compounds (6a-e)

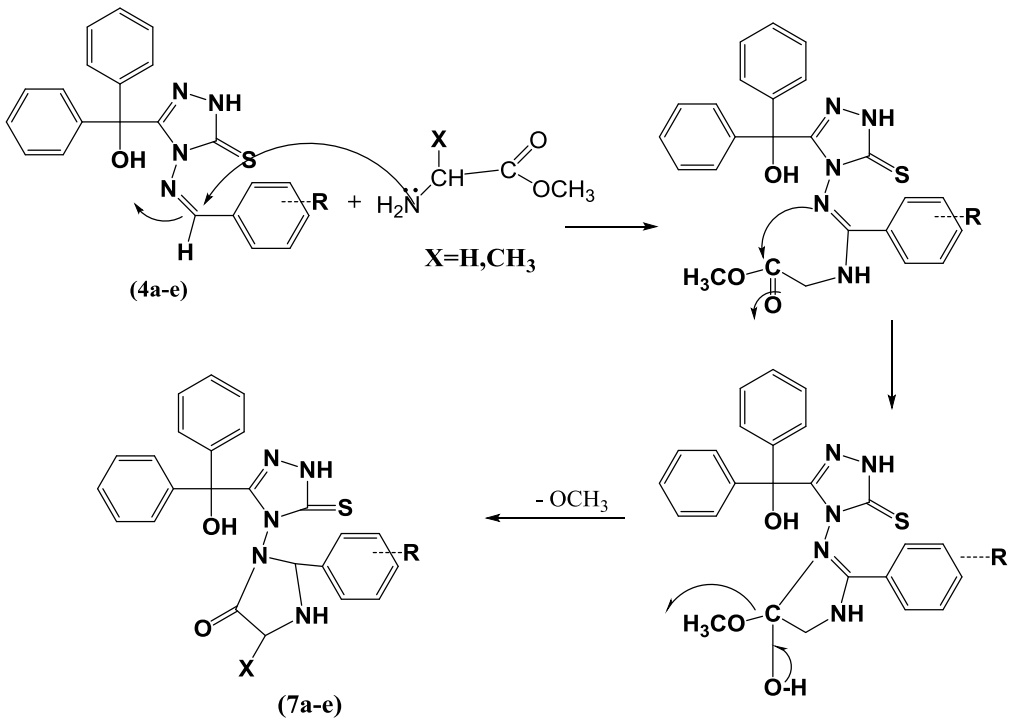
Comp. No.	FT.IR ν(cm^{-1})				^{1}H –NMR δ(ppm) DMSO-$_6$
	C=O (lactone)	C=O (lactam)	C=S	CH Ar.	
6a	1741	1672	1096	3061	5.38(s,1H,CH=OCO),5.62(s,1H,OH),6.91(s,H,CH=CON),6.51(bs,1H,CH-substituted aromatic ring),7.32(s,1H,NH),7.36-8.06.(s,13H,ArH).
6b	1733	1658	1082	3075	5.28(s,1H,CH=OCO),5.44(s,1H,OH),5.74(s,H,CH=CON),6.07(bs,1H,CH-substituted aromatic ring),,7.28(s,H,NH),7.42-7.98.(s,13H,ArH)
6c	1732	1674	1078	3082	5.32(s,1H,CH=OCO),5.43(s,1H,OH),5.71(s,H,CH=CON),6.08(bs,1H,CH-substituted aromatic ring),7.30(s,H,NH),7.39-7.95.(s,13H,ArH)
6d	1752	1668	1067	3081	5.45(s,2H,NH_2),5.53(s,1H,CH=OCO),6.03(s,1H,OH),6.08(s,H,CH=CON),7.43(s,H,NH),6.93-7.42.(s,13H,ArH)
6e	1732	1651	1084	3082	3.74(s,3H,OCH_3),5.43(s,1H,CH=OC0),5.85(s,1H,OH),6.01(s,H,CH=CON),7.39s,H,NH),7.42-8.11(s,13H,ArH)

The mechanism for obtaining derivatives of 1,3-oxazepine-4,7-dione that include a cycloaddition is achieved through ring formation that results from adding π electrons of π bonds and forming new bonds (Allamy and Mejbel, 2022).



Scheme 3: Mechanism of formation of 1,3-oxazepine-4,7-dione

Imidazoline compounds (7a-e) and (8a-e) which prepared through the reaction of ester of amino acids (glycine and alanine) with substituted hydrazones (4a-e). The reaction mechanism includes a nucleophilic attack of the amine group in the amino acids on the carbon atom, of the azomethine hydrazone group, following intra cyclization with the withdrawal of a water molecule shown below (Mehedi and Tepe, 2020).



Scheme 4: Mechanism of formation of imidazoline compounds

The structure of compounds (7a-f) and (8a-e) were confirmed by FT-IR and 1HNMR data. FT-IR spectra showed the stretching vibration bands in the region, (1684-1710) and (1659-1714) cm^{-1} due to (C=O imidazole) group, and (3174-3269) and (3211, 3281) cm^{-1} respectively. While1 H-NMR (DMSO-d6) spectra for compounds (7a,7b, 7c and 7e) showed the peaks for (CH_2- imidazole rings) at the range (3.41-371) ppm and at the range (7.28-7.79 ppm) representation for (N.H.) group of imidazole ring, while the (N.H.) of triazole ring appeared at the range (6,98-7,12) ppm. In addition to the other peaks representing the aromatic region, which are shown in the (Table 10).

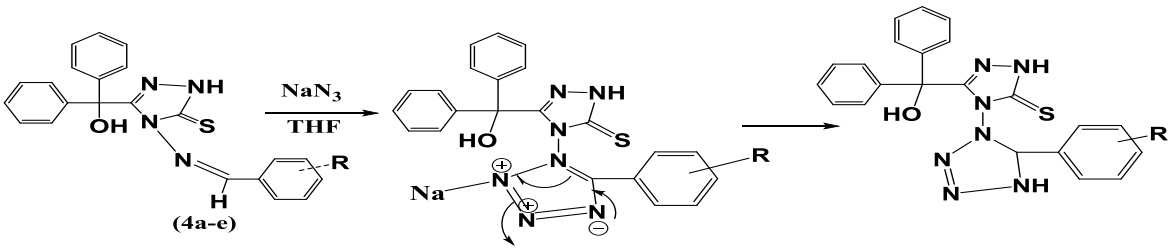
Table 10: The spectral data for compounds (7a-e)

Comp. No.	FT.IR ν(cm^{-1})					^{1}H - NMR δ(ppm) DMSO-$_6$
	C=O (imidazole)	C-N	C=S	CH Ar.	NH	
7a	1698	1243	1065	3175	317.l4	3.28(s,2H, CH_2imidazole),5.95 (s,1H, OH), 6.12 (s,1H, CH-Ar), 6.43(s,1H, NH traizole ring), 7.26 (s,1H, NH imidazole ring),7.44-8.08 (m,10H, ArH),
7b	1701	1154	1077	3073	3241	3.36 (s, 2H, CH_2imidazole),5.76 (s,1H, OH),6.05 (s,1H, CH-Ar), 6.45(s,1H, NH traizole ring), 7.37(s,1H, NH imidazole ring), 7.46-8.13 (m,10H, ArH).
7c	1710	1166	1085	3098	3197	3.39(s,2H, CH_2imidazole),5.78(s,1H, OH),6.02(s,1H, CH-Ar), 6.47(s,1H, NH traizole ring),7.38 (s,1H, NH imidazole ring),7.43-8.12 (m,10H, ArH).
7d	1684	1188	1101	3071	3174	3.41 (s,2H, CH_2, imidazole), 4.82 (d,2H, NH_2 imidazole), 5.8. (s,1H, OH), 6.15(s,1H, CH-Ar), 7.18 (s,1H, NH traizole ring), 7.28 (s,1H, NH imidazole ring), 7.43-8.12 (m,10H, ArH),
7e	1707	1213	1069	3088	3267	3.19 (s,2H, CH_2imidazole), 3.61 (s, 3H, OCH_3), 5.94 (s,1H, OH), 6.02 (s,1H, CH-Ar), 6.98(s,1H, NH traizole ring), 7.27 (s,1H, NH imidazole ring), 7.31-7.97 (m,10H, ArH),

Table 11: The spectral data for compounds (8a-e)

Comp., No.	FT-IR ν(cm^{-1})					^{1}H -NMR δ(ppm) DMSO-$_6$
	C= O (imidazole)	C-N	C=S	CH Ar.	NH	
8a	1703	1231	1084	3072	3233	1.78(s,3H, CH_3),3.59(s,1H, CH-imidazole), 5.85(s,1H, OH),5.53(s,1H, NH traizole ring), 6.05 (s,1H, CH-Ar),6.49 (s,1H, NH imidazole ring), 6.77-7.53 (m,10H, ArH).
8b	1711	1122	1083	3064	3229	1.88(s,3H, CH_3),3.64(s,1H, CH imidazole), 5.76 (s,1H,OH),5.85(s,1H, NH traizole ring), 6.15 (s,1H,CH-Ar), 6.44 (s,1H,NH imidazole ring), 6.84-7.09 (m,10H, ArH).
8c	1697	1158	1077	3108	3211	1.79(s,3H, CH_3), 3.61(s,1H, CH imidazole), 5.79(s,1H, OH),5.83(s,1H, NH traizole ring),6.17(s,1H, CH-Ar),6.41 (s,1H, NH imidazole ring), 6.82-7.11(m,10H, ArH).
8d	1659	1217	1124	3075	3246	1.69(s,3H, CH_3),3.38(s,2H, CH_2imidazole),4.82(d,2H, NH_2 imidazole),5.68. (s,1H, OH),5.97(s,1H, CH-Ar),7.18(s,1H, NH traizole ring),7.28(s,1H, NH imidazole ring),7.35-7.89 (m,10H, ArH).
8e	1714	1251	1082	3091	3281	1.85(s,3H, CH_3),3.59(s,1H, CHimidazole), 3.83(s,3H, OCH_3), 5.85(s,1H, OH),5.53(s,1H, NH traizole ring) 6.12 (s, 1H, CH-Ar),6.49 (s,1H, NH imidazole ring), 6.83-7.36 (m,10H, ArH).

Tetrazole compounds were prepared by reacting one of the hydrazone derivatives (4a-e) with sodium azide in the presence of tetrahydrofuran as a solvent. The expected reaction mechanism can be illustrated as shown in the scheme 5 (Sribalan *et al*., 2018).



Scheme 5: Mechanism of formation tetrazole compounds (9a-e)

The FT-IR. Spectra of compounds (9a-e) exhibited characteristic absorption bands at (1058-1105) cm^{-1} due to the (-N-N=N- tetrazole) ring, (1169-1211) cm^{-1} for the (C-N) group and other bands. They represent and refer to the other aggregates in these compounds, as shown in (Table 12).

Table 12: The spectral data for compounds (9a-e)

Comp. No.	FT.IR ν(cm^{-1})					^{1}H -NMR δ(ppm) DMSO-$_6$
	-N- N= N	C-N	C=S	CH Ar.	NH	
9a	1058	1166	1078	3098	3245 3344	5.04(s,1H, CH-Ar-),5.32(s,1H, OH),6.53(s,1H, NH tetrazole),7.74(s,1H, NH traizole),6,55-7.68(m,14H, ArH).
9b	1061	1174	1077	3085	3198 3289	5.16(s,1H, CH-Ar-), 5.38(s,1H, OH), 5.97(s,1H, NH tetrazole),7.86(s,1H, NH traizole), 6,55-7.81 (m,14H, ArH).
9c	1105	1208	1052	3076	3269 3412	5.19 (s,1H, CH-Ar-), 5.38(s,1H, OH), 6.03(s,1H, NH tetrazole), 8.12(s,1H, NHtraizole), 6,38-7.83 (m,14H, ArH).
9d	1076	1211	1094	3088	3258 3390	4.63 (d, 2H, NH_2), 5.21 (s,1H, CH-Ar-), 41(s,1H, OH), 6.60 (s,1H, NH tetrazole), 7.64 (s,1H, NHtraizole), 6,61-7.78 (m,14H, ArH).
9e	1067	1192	1065	3081	3264 3421	3.79(s,3H, OCH_3), 5.11(s,1H, CH-Ar-), 5.29 (s,1H, OH), 6.42 (s,1H, NH tetrazole),7.71(s,1H, NHtraizole), 6,49-7.69(m,14H, ArH).

CONCLUSIONS

To conclude, our study successfully synthesized a new series of hydrazones (4a-e) and characterized them using FT-IR and 1H-NMR. Further cyclization of these hydrazones using various anhydrides, amino acids, and sodium azide resulted in the synthesis of oxazines, imidazolines, and tetrazole compounds, respectively. Notably, our use of benzilic acid hydrazide and traditional methods, combined with ultrasound technology, resulted in significantly reduced reaction times and high reaction yields, all without the use of solvents. These findings hold promising implications for the development of new, efficient synthetic strategies for the preparation of diverse and valuable compounds.

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دراسة مقارنة لتحضير بعض المركبات الحلقية الجديدة المحتوية على 4,2 ، 1-تريازول

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الملخص

تم تحضير سلسلة من من مركبات الاوكسازيبين (oxazepines 5a-e) و مركبات البنزوكسازيبين (a-e6) و مركبات الايميدازولين (a-e7 ومركبات التترازول) (9a-e8) ,(a-e)، والتي تحتوي على 4,2,1- ترايازول باستخدام المادة الاولية هيدرازيد حامض البنزيليك (1) باستخدام الطريقة التقليدية وطرق الكيمياء الخضراء التي تمثلها الموجات فوق الصوتية لزيادة نواتج التفاعل وتقليل وقت التفاعل، تم تحويل هيدرازيد الحامض اولا إلى ملح البوتاسيوم الخاص به (م -2 (2-هيدروكسي-2،2-ثنائي فينيل أسيتيل) هيدرازين-1-كاربوديثيوات) (2) والذي تم تحويله إلى (4-أمينو- 5- مركابتو- 1-H،2،4-تريازول-3-يل) ثنائي فينيل ميثانول (3) بتفاعله مع الهدرازين المائي. تمت معالجة مركب 4،2،1-تريازول (3) باستخدام معوضات البنزالديهايد يعطي ناتج يمثل قواعد شيف (a-e4). أثبتت النتائج أن استخدام تقنيات الموجات فوق الصوتية أفضل بكثير من الطرق التقليدية، حيث أن أوقات رد الفعل منخفضة ومنتج ممتاز. تم فحص نقاوة جميع المركبات، وتم مراقبة التفاعلات بواسطة كروماتوجرافيا الطبقة الرقيقة (TLC)، وتم تشخيص المركبات المحضرة حديثًا في هذه الدراسة عن طريق التحليل الفيزيائي والطيفي المتضمن الاشعة تحت الحمراء (FT-IR)، الرنين المغناطيسي النووي (H NMR1) لبعض هذه المركبات.

الكلمات الدالة: هيدرزون،4،2،1-ترايازول، ملح البوتاسيوم، اوكسازيبين، ايميدازولين، تترازول.