

## Synthesis and Anti-Bacterial Activity Profile of Cyclized Diazonium Compounds

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### Abstract

Synthesis of cyclised diazonium compounds and their *in vitro* activity against various microorganisms is described. Several primary aromatic amines were diazotised and the resulting diazotised compounds were coupled with active methylene compounds to give hydrazono derivatives. These were cyclized with hydrazine hydrate, phenyl hydrazine, urea and *o*-phenylene diamine to give pyrazolin-5-one, substituted pyrazolin-5-ones, pyrimidin-di-ones and benzodiazepinone derivatives, respectively. Some hydrazine derivatives were also produced by reduction of diazo compounds with Sn/HCl. The synthesized compounds were assessed for their antimicrobial profile against *Escherichia coli*, *Staphylococcus aureus*, *Bacillus cereus* and *Pseudomonas putida*. Chloramphenicol and tetracycline were used as standards for the comparison of activity. Some of the compounds were found to exhibit promising anti-bacterial activity.

**Keywords:** Diazotization, Pyrazolinone, Pyrimidinone, Benzodiazepine, Antimicrobial

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### INTRODUCTION

Heterocyclic compounds hold a special place among pharmaceutically important natural and synthetic materials. The remarkable ability of heterocyclic nuclei to serve both as biomimetics and active pharmacophores has largely contributed to their unique value as traditional key elements of numerous drugs.

Five or six membered ring compounds are ranked high among various classes of organic compounds in respect to the diverse biological activities. Pyrazolinone is a five membered lactam ring compound containing two nitrogens and ketone in the same molecule. Lactams are reported to have varying pharmacological activity. Some pyrazolinones are nonsteroidal anti-inflammatory agents used in the treatment of arthritis and other musculoskeletal and joint disorders. They also possess activities like antibacterial, antifungal, anti-inflammatory [1], antidiabetic, analgesic, antipyretic, antiviral and antineoplastic activity [2].

Pyrimidine ring structures also have received significant attention owing to their diverse range of biological properties. Pyrimidine nucleus is present in compounds used

clinically such as antibacterial agents, anticancer agent, antiviral agents, antifungal agents and antimalarial agents. Several important sulfonamide drugs are pyrimidine derivatives namely sulfadiazine, sulfamerazine and sulfadimidine. The nucleus also is an integral part of DNA and RNA, hence serves as an important part of nucleoside antibiotics, antibacterials and cardiovascular agents [3– 5]. The benzodiazepine nucleus is also a well-studied traditional pharmacophoric scaffold that has emerged as a core structural unit of various sedative-hypnotics, muscle relaxants, anxiolytics, antistaminic and anticonvulsant agents. Therefore diversely substituted benzodiazepine nuclei can serve as synthons for developing new drugs [6]. Keeping these facts in mind, we synthesized pyrazolin-5-one, substituted pyrazolin-5-ones, pyrimidin-di-ones and benzodiazepinone derivatives for probable antibacterial activity.

### EXPERIMENTAL

#### General:

Melting points were determined on Buchi-530 melting point apparatus and are reported uncorrected. IR spectra were recorded on a

Shimadzu-Prestige-21 FTIR and NMR on Bruker-400 MHz. All analytical samples were observed by thin layer chromatography, which was performed on EM Silica gel 60 F254 sheets (0.2 mm) using suitable solvent system. The spots were detected with a model UV lamp.

#### Diazotization of Primary Aromatic Amines

A mixture of aromatic amine (0.01 mole) in concentrated HCl (5 ml) was cooled to 0 - 5°C under ice. Cooled sodium nitrite solution (1.5 g in 10 mL of water) was added to it dropwise over 10 minutes. Addition of the solution was continued till the reaction mixture gives end point when tested with starch -iodide paper.

#### General Procedure for the Preparation of Hydrazone Derivatives (C)

The diazonium salt formed was then reacted with ethyl acetoacetate, which serves as a source of active methylene group (Figure 1). This proton of methylene group is very active and can replace anion from other compounds. Other compounds like ethyl malonate, ethyl acetone, ethyl cyanoacetate can also be used as a source of active methylene group.

#### PROCEDURE

To the diazotized compound, the cooled mixture of active methylene compound formed from ethyl acetoacetate (0.01 M) and sodium acetate (0.05 M) in ethanol (50 ml) was added drop-wise with stirring for about 15 minutes. The reaction mixture was left for 2 hours at room temperature. Recrystallization was done using suitable solvent [7].

#### Preparation of Pyrazoline-5-one Derivatives (D1-D8)

To compound 'C1 to C8' was added equimolar

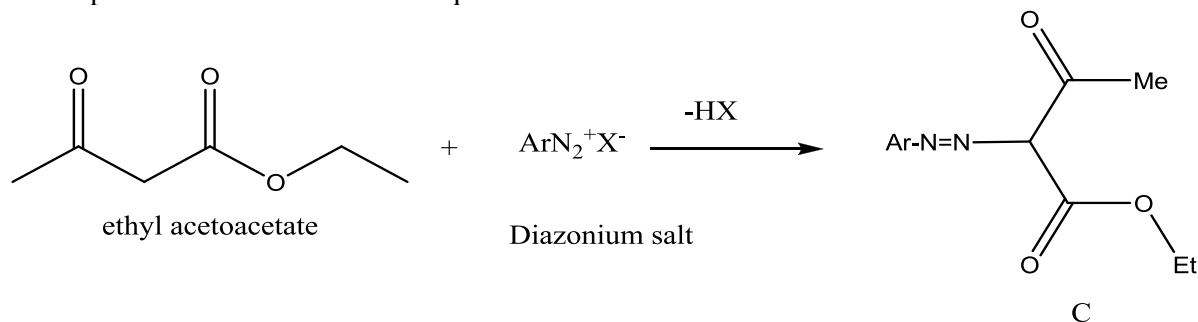


Fig. 1: Reaction of Diazonium Compounds with ethylacetoacetate.

Solution of hydrazine hydrate and 20 ml ethanol. The mixture was then refluxed for about 4 hours. The completion of reaction was monitored by TLC using suitable solvent system. The final product was recrystallised using ethanol [8, 10].

#### Preparation of 2-Phenyl Pyrazole-3-one Derivatives (E1-E8)

30mL of glacial acetic acid was added to 'C1 to C8' and stirred. To the resulting solution was added equimolar quantity of phenyl hydrazine and anhydrous sodium acetate. It was then refluxed for about 5 hours. The reaction mixture was poured in ice cool water and stored in refrigerator overnight. Filtered and recrystallized with suitable solvent [8, 10].

#### Preparation of Substituted 2, 4-Pyrimidine-dione Derivatives (F1-F8)

To compound 'C1 to C8' was added equimolar solution of urea and 40 ml ethanol. The mixture was then refluxed for about 3-4 hours. The completion of reaction was monitored by TLC using suitable solvent system. On completion of reaction, the mixture was cooled under ice and kept for about one hour. The final product was recrystallised using ethanol [9, 10].

#### Preparation of Substituted Benzodiazepine Derivatives (G1-G8)

To compounds 'C1 to C8' was added about 6 ml glacial acetic acid. Half molar of o-phenylene diamine was taken and dissolved in minimum quantity of glacial acetic acid. Both solutions were mixed and refluxed for about 6 hours, cooled and kept overnight. It was then filtered and recrystallized using acetic acid [11].

### Preparation of Hydrazine Derivatives (H1-H8)

To stannous chloride (2.1 gm), 2 ml HCl was added and cooled. This solution was slowly added to diazonium salt solution. It was kept for 2-3 hours, filtered and recrystallized.

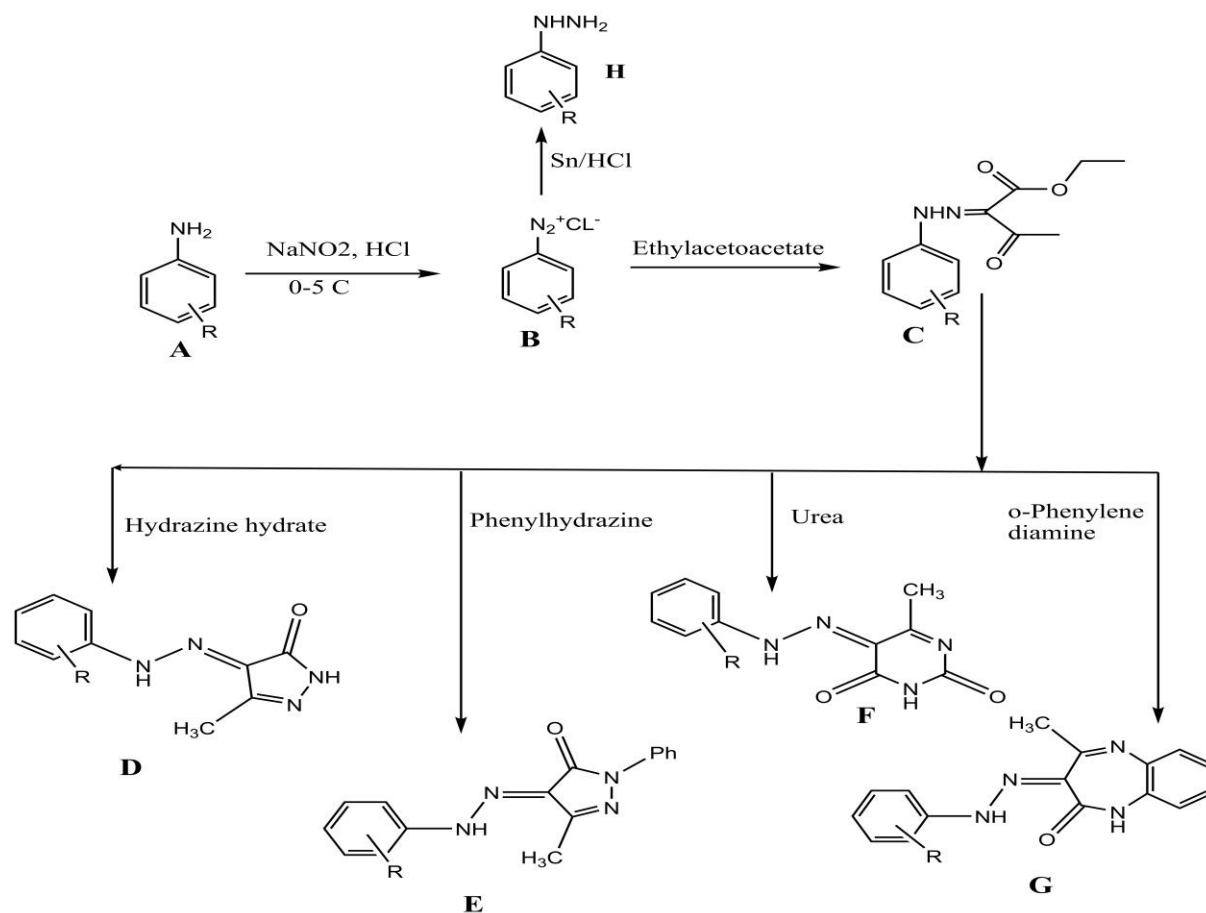
### Antimicrobial Screening

The synthesized compounds were screened for their antimicrobial activity against *Escherichia coli*, *Staphylococcus aureus*, *Bacillus cerius* and *Pseudomonas putida* by well plate method. The nutrient broth was prepared by dissolving 25 gm of Laurea Bertanni (LB) broth in 1000 ml of distilled water in a conical flask. 2% of agar powder was added to the

nutrient broth to prepare agar media. The solution was autoclaved at 121°C, 15psi for 15 minutes. The broth was then inoculated with culture as per USP guidelines and incubated for 15-18 hours at 37°C. 2% of agar powder was added to the nutrient broth to prepare agar media. Agar plates were prepared and wells were made in it for solvent, standard drug and for different concentrations (200 µg/ ml and 150 µg/ ml) of synthesized compounds. These were incubated at 37°C and zone of inhibition in cm was recorded after 12 hours. The experiments were performed in triplicate and average of the data is recorded in table 1 and synthesis of cyclized and reduced diazonium compounds is shown in Figure 2.

**Table 1:** Activity Profile of Synthesized Compounds.

Drug code	Zone of inhibition in cm			
	E.coli	B. cerius	S.aureus	P. putida
Standard drug	2.5	3.0	2.5	2.0
E5	0.4	2.1	1	Inactive
C5	1.1	Inactive	0.8	Inactive
G5	Inactive	1.9	Inactive	1.8
H2	3.0	2.8	1.9	2.2
D2	Inactive	Inactive	Inactive	Inactive
F2	Inactive	2.0	Inactive	1.7
C2	0.3	2.5	Inactive	Inactive
G2	2.1	Inactive	1.2	2.2
E2	Inactive	Inactive	Inactive	Inactive
C3	Inactive	1.2	1.9	Inactive
H3	2.5	0.6	Inactive	1.1
C1	Inactive	Inactive	Inactive	Inactive
D1	Inactive	1.5	Inactive	Inactive
F1	Inactive	2.5	Inactive	1.8
G1	Inactive	Inactive	Inactive	1.6
E1	0.2	1.9	0.8	-



**Fig. 2:** Reaction Scheme Showing Synthesis of Cyclized and Reduced Diazonium Compounds.

**Table 2:** Yield and Melting Points (in degrees celcius) of Synthesized Compounds.

S. No		1	2	3	4	5	6	7	8
	R	H	2-OCH3	3-Cl	4-Cl	4-CH3	3-CH3	4-OCH3	3-OCH3
<b>D</b>	%Y	95	96	64	60	15	5	47	5
	MP	180	215	200	180	187	-	178	-
<b>E</b>	%Y	91	97	17	5	80	43	25	95
	MP	120	132	170	5	148	152	121	153
<b>F</b>	%Y	92	97	26	67	72.5	5	60	5
	MP	142	101	160	162	77	-	65	-
<b>G</b>	%Y	90	15	62.5	22.3	57	70	5	82.7
	MP	127	123	-	110	98	85	-	-
<b>H</b>	%Y	75	92	95	51	72	94.8	90	92
	MP	120	160	180	-	120	120	178	151

## RESULTS AND DISCUSSION

All compounds synthesized were characterized using melting point, infrared, <sup>1</sup>H-NMR and mass spectroscopy. The yield and melting point of all the compounds is reported in table 2.

### **D1:5-Methyl-4-(phenyl-hydrazono)-2,4-dihydro-pyrazole-3-one:**

IR data: 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching) NMR: 0.9δ (CH<sub>3</sub>), 6.2-7.0δ<sub>5</sub> (Aromatic protons), 5.6 δ (N-H protons), m/z:201.31

### **D2:4-[(2-methoxy-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching), 1150 cm<sup>-1</sup> (C-O stretching), m/z:232.13 NMR: 0.9δ (CH<sub>3</sub>), 6.3-6.5 δ (Aromatic protons), 6 δ (N-H protons), 3.73δ (OCH<sub>3</sub>)

### **D3:4-[3-Chloro-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3380-3400 cm<sup>-1</sup> (N-H stretching), 2950-3120 cm<sup>-1</sup> (Aromatic C-H stretching), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching) NMR: 0.9 δ (CH<sub>3</sub>), 6.3-6.7 δ (aromatic protons), 6δ (N-H protons), m/z 236.08

### **D4:4-[4-Chloro-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3380-3400 cm<sup>-1</sup> (N-H stretching), 2950-3120 cm<sup>-1</sup> (Aromatic C-H stretching), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching) NMR: 0.9δ (CH<sub>3</sub>), 6.4-7.2 δ (aromatic protons), 6.2 δ (N-H protons), m/z 236.08

### **D5:4-[4-methyl-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3380-3400 cm<sup>-1</sup> (N-H stretching), 2950-3120 cm<sup>-1</sup> (Aromatic C-H stretching), 2750 cm<sup>-1</sup> (C-H stretching), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching)

NMR:0.9 δ (CH<sub>3</sub>), 2.34 δ (CH<sub>3</sub> of phenyl), 6.3-6.8δ (aromatic protons), 6δ (N-H protons), m/z 216.13

### **D6:4-[3-Chloro-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3380-3400 cm<sup>-1</sup> (N-H stretching), 2950-3120 cm<sup>-1</sup> (Aromatic C-H stretching), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching) NMR: 0.9δ (CH<sub>3</sub>), 2.34 δ (CH<sub>3</sub> of phenyl), 6.2-6.8δ (aromatic protons), 6 δ (N-H protons)

### **D7:4-[4-methoxy-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: : 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching), 1150 cm<sup>-1</sup> (C-O stretching) NMR: 0.9δ (CH<sub>3</sub>), 3.78 δ (OCH<sub>3</sub> of phenyl), 6.2-6.8δ (aromatic protons), 6δ (N-H protons), m/z:232.13

### **D8:4-[3-methoxy-phenyl)-hydrazono]5-methyl-2,4-dihydro-pyrazole-3-one:**

IR data: : 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching), 1150 cm<sup>-1</sup> (C-O stretching) NMR: 0.9 δ (CH<sub>3</sub>), 3.75δ (OCH<sub>3</sub> of phenyl), 6.2-6.8δ (aromatic protons), 5.6 δ (N-H protons), m/z:232.13

### **E1: 5-methyl-2-phenyl-4[phenyl-hydrazono]- 2,4-dihydro-pyrazole-3-one:**

IR data: 3458 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 1670-1690 cm<sup>-1</sup>(C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching). NMR: 0.9δ (CH<sub>3</sub> protons)6.2-6.8, 7.0-7.3 δ (Aromatic protons), 6.8δ (NH)

### **E2:4[(2-methoxy-phenyl)-hydrazono]- 5-methyl-2-phenyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3345 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 1670-1690 cm<sup>-1</sup>(C=O stretching), 1600-1700 cm<sup>-1</sup>



(C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), 1150  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9  $\delta$  (CH<sub>3</sub> protons), 3.73 $\delta$  (OCH<sub>3</sub>), 6.2-6.5, 7.0-7.6 $\delta$  (Aromatic protons), 6.9 $\delta$  (NH),  $m/z=308.13$

**E3:4-[3-chloro-phenyl]-hydrazono]-5-methyl-2-phenyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3455  $\text{cm}^{-1}$  (N-H stretching), 2930-3120  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), NMR: 0.9 $\delta$  (CH<sub>3</sub> protons), 6.2-6.5, 7.0-7.6 $\delta$  (Aromatic protons), 7.0 $\delta$  (NH),  $m/z=312.08$

**E4:4-[(4-chloro-phenyl)-hydrazono]-5-methyl-2-phenyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3455  $\text{cm}^{-1}$  (N-H stretching), 2930-3120  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching) NMR: 0.9  $\delta$  (CH<sub>3</sub> protons), 3.73 $\delta$  (OCH<sub>3</sub>), 6.2-6.5, 7.0-7.6  $\delta$  (Aromatic protons), 6.8 $\delta$  (NH),  $m/z=312.08$

**E5: 5-methyl-2-phenyl-4[p-tolyl]-hydrazono]-2, 4-dihydro-pyrazole-3-one:**

IR data: 3425  $\text{cm}^{-1}$  (N-H stretching), 2930-3320  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), 2850  $\text{cm}^{-1}$  (C-H stretching) NMR: 0.9, 2.35 $\delta$  (CH<sub>3</sub> protons), 6.3-6.81, 7.0-7.64 $\delta$  (Aromatic protons), 6.8 $\delta$  (NH),  $m/z=292.13$

**E6:5-methyl-2-phenyl-4[m-tolyl]-hydrazono]-2,4-dihydro-pyrazole-3-one:**

IR data: 3425  $\text{cm}^{-1}$  (N-H stretching), 2930-3320  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), 2850  $\text{cm}^{-1}$  (C-H stretching) NMR: 0.9, 2.35 $\delta$  (CH<sub>3</sub> protons), 6.2-6.8, 7.0-7.  $\delta$  (Aromatic protons), 6.8 $\delta$  (NH),  $m/z=292.13$

**E7:4[(4-methoxy-phenyl)-hydrazono]-5-methyl-2-phenyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3345  $\text{cm}^{-1}$  (N-H stretching), 2900-3100  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$

(C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), 1150  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub> protons), 3.73  $\delta$  (OCH<sub>3</sub>), 6.2-6.5, 7.0-7.6 $\delta$  (Aromatic protons), 6.8  $\delta$  (NH),  $m/z=308.13$

**E8:4[(3-methoxy-phenyl)-hydrazono]-5-methyl-2-phenyl-2,4-dihydro-pyrazole-3-one:**

IR data: 3345  $\text{cm}^{-1}$  (N-H stretching), 2900-3100  $\text{cm}^{-1}$  (Aromatic C-H stretching), 1670-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1565-1450  $\text{cm}^{-1}$  (C=C stretching), 1150  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub> protons), 3.73 $\delta$  (OCH<sub>3</sub>), 6.2-6.5, 7.0-7.6 $\delta$  (Aromatic protons), 6.84 $\delta$  (NH),  $m/z=308.13$

**F1:6-methyl-5(phenyl-hydrazono)-5-H-pyridine-2, 4-dione:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N=N=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 7, 10  $\delta$  (NH protons), 6.4-7.01 $\delta$  (Aromatic protons),  $m/z=230.22$

**F2:5[(2-methoxy-phenyl)-hydrazono]-6-methyl-5-H-pyridine-2, 4-dione:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N=N=C stretching), 1243  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9 (CH<sub>3</sub>), 7, 10 (NH protons), 6.3-6.51 (Aromatic protons), 3.5 (methoxy protons),  $m/z=260.09$

**F3:5[(3-chloro-phenyl)-hydrazono]-6-methyl-5-H-pyridine-2, 4-dione:**

IR data: 3345  $\text{cm}^{-1}$  (N-H stretching), 3153  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N=N=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 7.1, 10.2 $\delta$  (NH protons), 6.3-6.6  $\delta$  (Aromatic protons),  $m/z=264.04$

**F4:5[(4-chloro-phenyl)-hydrazono]-6-methyl-5-H-pyridine-2, 4-dione:**

IR data: 3345  $\text{cm}^{-1}$  (N-H stretching), 3153  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 7, 10  $\delta$  (NH protons), 6.3-6.6 $\delta$  (Aromatic protons), m/z=264.04

**F5:6-methyl-5-(p-tolyl-hydrazano)-5-H-pyridine-2, 4-dione:**

IR data: 3347  $\text{cm}^{-1}$  (N-H stretching), 3157  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching) NMR: 0.9, 2.3 $\delta$  (CH<sub>3</sub>), 7, 10  $\delta$  (NH protons), 6.3-6.6 $\delta$  (Aromatic protons), m/z=244.10

**F6:6-methyl-5-(m-tolyl-hydrazano)-5-H-pyridine-2, 4-dione:**

IR data: 3347  $\text{cm}^{-1}$  (N-H stretching), 3157  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching) NMR: 0.9, 2.3 $\delta$  (CH<sub>3</sub>), 7, 10  $\delta$  (NH protons), 6.3-6.6 $\delta$  (Aromatic protons), m/z=244.10

**F7:5[(4-methoxy-phenyl)-hydrazono]-6-methyl-5-H-pyridine-2, 4-dione:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching), 1243  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 7, 10  $\delta$  (NH protons), 6.3-6.51 $\delta$  (Aromatic protons), 3.5 $\delta$  (methoxy protons), m/z=260.09

**F8:5[(3-methoxy-phenyl)-hydrazono]-6-methyl-5-H-pyridine-2, 4-dione:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching), 1243  $\text{cm}^{-1}$  (C-O

stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 7, 10 $\delta$  (NH protons), 6.3-6.51 $\delta$  (Aromatic protons), 3.5 $\delta$  (methoxy protons), m/z=260.09

**G1:4-methyl-3(-phenyl-hydrazono)-1,3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3400-3600  $\text{cm}^{-1}$  (N-H stretching), 2900-3100  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1580-1450  $\text{cm}^{-1}$  (C=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 3.1 $\delta$  (CH<sub>2</sub>), 4, 7 $\delta$  (NH protons), 6.4-7.01 $\delta$  (Aromatic protons), m/z=264.14

**G2:3[(2-methoxy-phenyl)-hydrazono]-4-methyl-3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3400-3600  $\text{cm}^{-1}$  (N-H stretching), 2900-3100  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690  $\text{cm}^{-1}$  (C=O stretching), 1600-1700  $\text{cm}^{-1}$  (C=N stretching), 1580-1450  $\text{cm}^{-1}$  (C=C stretching), 1280  $\text{cm}^{-1}$  (C-O stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 3,2  $\delta$  (CH<sub>2</sub>), 3.9 $\delta$  (methoxy protons), 4, 7 $\delta$  (NH protons), 6.4-7.6 $\delta$  (Aromatic protons), m/z=308.13

**G3:3[(3-chloro-phenyl)-hydrazono]-4-methyl-3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 3,2  $\delta$  (CH<sub>2</sub>), 3.9  $\delta$  (methoxy protons), 4, 7  $\delta$  (NH protons), 6.4-8 $\delta$  (Aromatic protons), m/z=312.08

**G4:3[(4-chloro-phenyl)-hydrazono]-4-methyl-3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3424  $\text{cm}^{-1}$  (N-H stretching), 3052  $\text{cm}^{-1}$  (Aromatic C-H stretching), 2800- 2900  $\text{cm}^{-1}$  (aliphatic C-H stretching), 1688, 1676  $\text{cm}^{-1}$  (C=O stretching), 1592  $\text{cm}^{-1}$ , 1565  $\text{cm}^{-1}$ , 1481  $\text{cm}^{-1}$  (C=N, C=C stretching), 1284  $\text{cm}^{-1}$  (N-N=C stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 3,2 $\delta$  (CH<sub>2</sub>), 3.9 $\delta$  (methoxy protons), 4, 7  $\delta$  (NH

protons), 6.4-8 $\delta$  (Aromatic protons),  
m/z=312.08

**G5:4-methyl-3-(p-tolyl-hydrazono)-1,3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3345 cm<sup>-1</sup> (N-H stretching), 3153 cm<sup>-1</sup> (Aromatic C-H stretching), 2800- 2900 cm<sup>-1</sup> (aliphatic C-H stretching), 1688, 1676 cm<sup>-1</sup> (C=O stretching), 1592 cm<sup>-1</sup>, 1565 cm<sup>-1</sup>, 1481 cm<sup>-1</sup> (C=N, C=C stretching), 1284 cm<sup>-1</sup> (N-N=C stretching) NMR: 0.9  $\delta$  (CH<sub>3</sub>), 3.2 $\delta$  (CH<sub>2</sub>), 3.9  $\delta$  (methoxy protons), 4, 8 $\delta$  (NH protons), 6.4-7.6  $\delta$  (Aromatic protons), m/z=292.13

**G6:4-methyl-3-(m-tolyl-hydrazono)-1,3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3345 cm<sup>-1</sup> (N-H stretching), 3153 cm<sup>-1</sup> (Aromatic C-H stretching), 2800- 2900 cm<sup>-1</sup> (aliphatic C-H stretching), 1688, 1676 cm<sup>-1</sup> (C=O stretching), 1592 cm<sup>-1</sup>, 1565 cm<sup>-1</sup>, 1481 cm<sup>-1</sup> (C=N, C=C stretching), 1284 cm<sup>-1</sup> (N-N=C stretching) NMR: 0.9  $\delta$  (CH<sub>3</sub>), 3.2 $\delta$  (CH<sub>2</sub>), 3.9 $\delta$  (methoxy protons), 3.8, 7.5  $\delta$  (NH protons), 6.4-7.6 $\delta$  (Aromatic protons), m/z=292.13

**G7:3[(4-methoxy-phenyl)-hydrazono]-4-methyl-,3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching), 1280 cm<sup>-1</sup> (C-O stretching) NMR: 0.9 $\delta$  (CH<sub>3</sub>), 3.2 $\delta$  (CH<sub>2</sub>), 3.9 $\delta$  (methoxy protons), 4, 8 $\delta$  (NH protons), 6.4-7.6 $\delta$  (Aromatic protons), m/z=308.13

**G8:3[(3-methoxy-phenyl)-hydrazono]-4-methyl-,3-dihydro-benzo-1,4-diazepin-2-one:**

IR data: 3400-3600 cm<sup>-1</sup> (N-H stretching), 2900-3100 cm<sup>-1</sup> (Aromatic C-H stretching), 2818 (C-H stretching of CH<sub>3</sub>), 1650-1690 cm<sup>-1</sup> (C=O stretching), 1600-1700 cm<sup>-1</sup> (C=N stretching), 1580-1450 cm<sup>-1</sup> (C=C stretching), 1280 cm<sup>-1</sup> (C-O stretching) NMR :0.9 $\delta$  (CH<sub>3</sub>), 3.2 $\delta$  (CH<sub>2</sub>), 3.9 $\delta$  (methoxy protons), 4, 8  $\delta$  (NH protons), 6.4-7.5 $\delta$  (Aromatic protons), m/z=308.13

**H1:phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching), NMR: 2,4  $\delta$  (NH<sub>2</sub>, NH proton), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 108.07

**H2:2-methoxy phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching), 2856 cm<sup>-1</sup> (C-H stretching of methoxy), 1150 cm<sup>-1</sup> (C-O stretching) NMR: 2,4  $\delta$  (NH<sub>2</sub>, NH proton), 3.5  $\delta$  (methoxy protons), 6.8-7.38 $\delta$  (Aromatic protons), m/z : 138.08

**H3:3-chloro-phenyl hydrazine:**

IR data: 3000-3240 cm<sup>-1</sup> (aromatic C-H stretching), 3380-3500cm<sup>-1</sup>(N-H stretching) NMR: 2.3,4.1  $\delta$  (NH<sub>2</sub>, NH proton), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 142.03

**H4:4- chloro-phenyl hydrazine:**

IR data: 3000-3240 cm<sup>-1</sup> (aromatic C-H stretching), 3380-3500cm<sup>-1</sup>(N-H stretching) NMR: 2.3,4.1  $\delta$  (NH<sub>2</sub>, NH proton), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 142.03

**H5:4-methyl-phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching), 2856 cm<sup>-1</sup> (C-H stretching of methyl) NMR: 2.6,4  $\delta$  (NH<sub>2</sub>, NH proton), 2.35  $\delta$  (methyl protons), 6.7-7.58 $\delta$  (Aromatic protons), m/z : 122.17

**H6:3- methyl-phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching), 2856 cm<sup>-1</sup> (C-H stretching of methyl) NMR: 2,4  $\delta$  (NH<sub>2</sub>, NH proton), 2.35  $\delta$  (methyl protons), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 122.17

**H7:4-methoxy phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching), 2856 cm<sup>-1</sup> (C-H stretching of methoxy), 1150 cm<sup>-1</sup> (C-O stretching) NMR: 2,4  $\delta$  (NH<sub>2</sub>, NH proton), 3.5  $\delta$  (methoxy protons), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 138.08

**H8: 3-methoxy phenyl hydrazine:**

IR data: 3000-3100 cm<sup>-1</sup> (aromatic C-H stretching), 3400-3500cm<sup>-1</sup>(N-H stretching),



2856  $\text{cm}^{-1}$  (C-H stretching of methoxy), 1150  $\text{cm}^{-1}$  (C-O stretching)

NMR: 2,4  $\delta$  (NH<sub>2</sub>, NH proton), 3.5  $\delta$  (methoxy protons), 6.6-7.18 $\delta$  (Aromatic protons), m/z : 138.08

## CONCLUSION

Various cyclized diazonium compounds were synthesized possessing different heterocyclic ring systems in good yield. The well plate method for antibacterial activity showed significant reduction in bacterial growth in terms of zone of inhibition around the well. Chloramphenicol and tetracycline were used as standard drugs for the comparison. It was observed that phenyl group bearing no substituent, was inactive against E.coli and S. aureus. Benzodiazepine derivatives and reduced compounds displayed comparable activity to standard against all tested strains of micro-organisms.

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