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Synthesis and Antimicrobial studies of few New Substituted 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-ones

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Abstract

Synthesis of substituted 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-b]quinazolin-9(3H)ones [4a-4t] is carried out by coupling of diazonium salts of aromatic amines [3a-3d] with 2-methylpyrazolo[5,1-b]quinazolin-9(4H)-ones [2a-2e]. The structures of compounds [4a-4t] are confirmed by IR and ^{1}HNMR, ^{13}CNMR and mass spectral studies. Further, they were screened in vitro for antibacterial activity against Escherichia coli and Salmonella typhi. Antifungal activity is evaluated against Aspergillus niger and Penicillium chrysogenum using Paper disc diffusion method. Few compounds have shown potential antibacterial and antifungal activities.

Keywords: Substituted aryldiazenyl-pyrazol-quinazolinones, 3-methyl-1H-pyrazol-5(4H)-one, aryl diazonium salt, antibacterial and antifungal activity.

Introduction

The literature study reveals that the nitrogen containing heterocyclic moiety associated with various aromatic compounds posses vast stream pharmacological applications as well as they have alarming important in dyeing industries due to their coloring properties.

The 2-methylpyrazolo[5,1-*b*]quinazolin-9(4*H*)-ones are widely used in dyes and pigments as an intermediates. Various types of dyes and pigments are currently used in different industries[1]. Dyes are the versatile chemicals which are used by number of chemical industries like textile, printing, paper, food and cosmetics industries[2]. Most important characteristics of reactive azo dyes is their ability to form covalent bond with fabric on which they are applied[3].

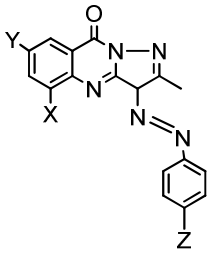
The general structure of azo dyes includes –N=N- substituted by variety of groups on aromatic ring making it a complex structure. Among various dyes, azo dyes find the largest commercial applications[4]. The class azo dyes covers 65-75% of dye stuff available in world. According to statically data survey one million tones of azo dyes are producing annually in the industrial sectors based on chemical compositions[5-6]. There are various classes of azo dyes and reactive dyes are most important in them. The 80-85% of reactive dye have azo groups as chromegen[7-8]. Now a day's production rate as well as usage ratio of dyes has increased dramatically according to consumer's demand. Therefore, the derivatives of pyrazolo [5,1-*b*] quinazolines are widely used in dyes and pigments industries. These azo compounds also exhibit multiple pharmaceutical and biological applications and are known for their, antimicrobial[9-10], antifungal properties[11], potential anti metabolite agents[12-14], potent amino acid antagonist[15]. These compounds exhibit anti diabetic, antiseptic, anticancer, anti asthmatic, anti allergic and immuno suppressant activities[16-20].

The aim of present research focused on synthesis 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-ones. Further, they were screened in vitro for antimicrobial properties.

Material and Method

All the reagents were of analytical reagent grade and were used without further purification. All the products were synthesized and characterized by their spectral analysis. All chemical and solvents were purchased from S.D. Fine chemicals (India). Melting points were taken in open capillary tube. IR spectra (KBr,ν, cm^{-1}) were recorded on Perkin-Elmer Spectrophotometer and ^{1}H NMR 400 MHz ($CDCl_3$) and chemical shifts are given in δ (ppm), ^{13}C NMR 7 MHz ($CDCl_3$). The mass spectra were performed using VG 2AB-3F spectrometer (70 ev), (M^{+1}). All reactions were followed by TLC (Silica gel, aluminum sheets 60 F_{254}, Merck).

Experimental: All the chemical and solvents used were of A.R. grade from E-Merck and S.D. fine Ltd. Melting points were determined in an open capillary tube and are uncorrected. The purity of the compound has been checked by TLC. IR spectra were recorded in $CHCl_3$ on a Shimadzu FTIR-8300 spectrophotometer. The ^{1}H NMR (300 MHz) and ^{13}C NMR (70 MHz) were run on a Bruker Avance DPX-250 spectrometer in $CDCl_3$ using tetramethylsilane as an internal standard. Chemical shift values are given in δ scale. Mass spectra were recorded on VG 2AB-3F spectrometer (70 ev), (M^{+1}). The in vitro biological screenings of the investigated compounds were tested against the bacterial species by agar cup method and fungal species by the poison plate method.



X/Y= H/H, Cl/H, H/Cl, H/CH_3, H/NO_2 , Z= H, Cl, CH_3, OCH_3

2-methyl-3-(aryldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-ones [4a -4t]

Scheme-1

Synthesis of substituted 2-methylpyrazolo[5,1-*b*]quinazolin-9(4*H*)ones [2a-2e]: 2-methylpyrazolo[5,1-*b*]quinazolin-9(4*H*)ones[2a-2e] are prepared by earlier known method.[21] 3-methyl-1*H*-pyrazol-5(4*H*)-one (0.05 mole) is introduced in 50 ml of xylene and heated to 120-130^{0}c. 1*H*-benzo[*d*][1,3]oxazine-2,4-diones [1a-1e] (0.05 mole) were slowly added to the mixture with continues stirring. After the evolution of carbon dioxide is stoped, the temperature is raised to 140-150^{0}c with simultaneous azeotropic removal of water. The heating is continued till no further water is formed. The reacting mixture is cooled, filtered, washed with methanol and then with warm water to obtain [2a-2e]

Diazotisation of amines: A solution of aromatic amines (10mmol) and 8ml of 3M HCl was heated gently, and then water (10ml) was added in order to dissolve the solid. The mixture was cooled to 0^{0}C in an ice bath with constant stirring. Freshly prepared solution of 1M sodium nitrate (10ml) was added drop wise. Precaution is taken to maintain the temperature below 5^{0}C. The solution was kept in ice bath and used immediately for the next step [3a-3d].

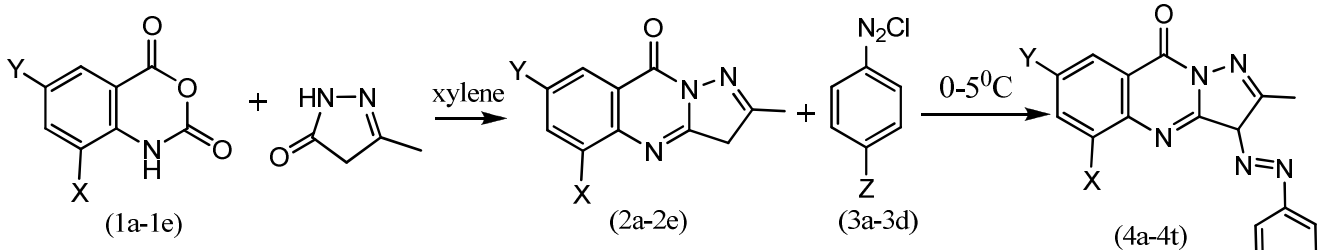
Synthesis of substituted 2-methyl-3-(aryldiazenyl) pyrazolo [5,1-b]quinazolin-9(3H)-ones [4a-4t]: A mixture of 2-methylpyrazolo[5,1-b]quinazolin-9(4H)-ones [2a-2e] (0.01 mole) is dissolved in alcohol and kept it for 0-5^{0}C and then add it drop wise to ice cooled solution of diazonium salts of aromatic amines [2a-2d] with constant stirring. After half an hour the coloured precipitate of 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-ones [4a-4t] is formed. It is washed with cold water, dried and crystallized with absolute alcohol.

Characterization of Compounds (4a-4t): 2-methyl-3-(phenyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4a): Colour: Orange, Yield = 81%, M.P.: 230^{o}C, IR (KBr, cm^{-1}): 3030 (C=C-H), 1683 (C=O), 1640 and 1622 (C=N), 1610 and 1514 (Aromatic C=C), 1430 (N=N). H^{1}NMR: 1.43 (s,1H), 1.90 (s,3H), 7.05-7.40 (m,5H), 7.60- 8.05 (m,4H). C^{13} NMR: 14.3, 66.1, 122.3, 122.5, 122.5, 125.3, 126.3, 126.5, 127.5, 129.5, 129.5, 133.7, 147.1, 151.2, 155.3, 160.5, 166.2. Mass Spectra [M$^+$]: 304.

5-chloro-2-methyl-3-(phenyldiazenyl)pyrazolo[5,1-*b*] quinazolin -9(3*H*)-one (4b): Colour: Orange Yield = 73%, M.P.: 241^{o}C, IR (KBr, cm^{-1}): 3032 (C=C-H), 1685 (C=O), 1642 and 1624 (C=N), 1612 and 1516 (Aromatic C=C), 1456 (N=N). H^{1}NMR: 1.46 (s,1H), 1.91 (s,3H), 7.05-7.40 (m,5H), 7.55- 7.95 (m,3H). C^{13} NMR: 14.4, 66.2, 122.4, 122.7, 122.7, 124.9, 125.5, 128.4, 129.5, 129.5, 132.7, 133.7, 151.3, 155.4, 160.6, 161.6, 166.3. Mass Spectra [M$^+$]: 338.

7-chloro-2-methyl-3-(phenyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4c): Colour: Orange, Yield = 77%, M.P.: 248^{o}C, IR (KBr, cm^{-1}): 3034 (C=C-H), 1686 (C=O), 1644 and 1622 (C=N), 1614 and 1515(Aromatic C=C), 1456 (N=N). H^{1}NMR: 1.53 (s,1H), 1.93 (s,3H), 7.07-7.47(m,5H), 7.49 (d,1H), 7.77 (d,1H), 7.97 (s,1H). C^{13} NMR: 14.5, 66.3, 122.6, 122.9, 122.9, 125.5, 127.9, 127.9, 129.5, 129.5, 133.2, 133.7, 145.3, 151.5, 155.7, 160.5, 166.6. Mass Spectra [M$^+$]:338.

2,7-dimethyl-3-(phenyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4d): Colour: Dark brown, Yield = 84%, M.P.: 253^{o}C, IR (KBr, cm^{-1}): 3036 (C=C-H), 1688 (C=O), 1649 and 1626(C=N), 1618 and 1513 (Aromatic C=C), 1444 (N=N). H^{1}NMR: 1.57 (s,1H), 1.93 (s,3H), 2.39 (s,3H), 7.07-7.47(m,5H), 7.40(d,1H), 7.51 (d,1H), 7.79 (s,1H). C^{13} NMR: 14.8, 21.7, 66.5, 121.1, 122.9, 122.9, 125.6, 125.6, 125.5, 129.7, 129.7, 133.5, 137.6, 144.3, 151.3, 155.3, 160.6, 166.7. Mass Spectra [M$^+$]:318.



(1a-1e) (2a-2e) (3a-3d) (4a-4t)

X / Y = H / H, Cl / H, Cl /H, Me / H, and -NO_2 / H

Z= H, Cl, Me, OMe

Synthesis of substituted 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-b]quinazolin-9(3H)-ones[4a-4t]

Scheme-2

2-methyl-7-nitro -3 - (phenyldiazenyl)pyrazolo [5,1-*b*] quinazolin -9(3*H*)-one (4e): Colour: Reddish brown, Yield= 88% M.P.: 267°C, IR (KBr, cm^{-1}):3040 (C=C-H), 1690 (C=O), 1650 and 1629 (C=N), 1620 and 1518 (Aromatic C=C), 1460 (N=N). H^1NMR : 1.60 (s,1H), 1.98 (s,3H), 7.08-7.49 (m,5H), 7.81(d,1H), 8.40 (d,1H), 8.61 (s,1H). C^{13} NMR: 15.3, 66.5, 118.4, 121.6, 122.9, 122.9, 123.5, 125.9, 128.9, 129.7, 129.7, 143.6, 151.5, 153.7, 155.9, 160.7, 166.9.Mass Spectra [M^+]:349.

3-((4-chlorophenyl)diazenyl)-2-methylpyrazolo[5,1-b[5,1-*b*]quinazolin-9(3*H*)-one (4f): Colour: Orange, Yield = 75% M.P.: 232°C, IR (KBr, cm^{-1}): 3036 (C=C-H), 1682 (C=O), 1649 and 1626 (C=N), 1622 and 1512 (Aromatic C=C),1438 (N=N). H^1NMR: 1.41 (s,1H), 1.92 (s,3H), 7.31 and 7.51(dd,4H), 7.60-8.05 (m,4H). C^{13} NMR: 15.1, 66.3, 122.2, 124.3, 124.3, 126.9, 126.9, 127.5, 129.5, 129.6, 131.5, 133.3, 149.5, 146.7, 155.5, 160.6, 166.3.

Mass Spectra [M^+]:337: **5-chloro-3-((4-chlorophenyl) diazenyl)-2-methylpyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4g):** Colour: Orange, Yield = 82%, M.P.: 238°C, IR (KBr, cm^{-1}): 3040 (C=C-H), 1684 (C=O), 1645 and 1620 (C=N), 1624 and 1516 (Aromatic C=C), 1458 (N=N). H^1NMR: 1.44 (s,1H), 1.95 (s,3H), 7.25 and 7.53 (dd,4H), 7.59-7.93 (m,3H). C^{13} NMR: 15.3, 66.6, 122.5, 124.5, 124.5, 124.3, 128.9, 129.6, 129.6, 131.7, 132.8, 133.8, 149.5, 155.8, 160.5, 161.3, 166.5. Mass Spectra [M^+]:372**.**

7-chloro-3-((4-chlorophenyl)diazenyl)-2-methylpyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4h): Colour: Orange, Yield = 4.5 gm, M.P.: 243°C, IR (KBr, cm^{-1}): 3045 (C=C-H), (Aromatic C-H) 1684 (C=O), 1648 and 1624 (C=N), 1614 and 1516 (Aromatic C=C), 1458 (N=N). H^1NMR: 1.49 (s,1H), 1.95 (s,3H), 7.25 and 7.53 (dd,4H), 7.51(d,1H), 7.79 (d,1H), 7.98 (s,1H). C^{13} NMR: 15.4, 66.7, 122.5, 124.5, 124.3 127.9, 127.9, 129.1, 129.1, 131.5, 132.6, 133.8, 145.5, 149.5, 155.8, 160.5, 166.5. Mass Spectra [M^+]:372.

3-((4-chlorophenyl)diazenyl)-2,7-dimethylpyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4i): Colour: Reddish brown, Yield = 76%, M.P.: 249°C, IR (KBr, cm^{-1}): 3038 (C=C-H), 1682 (C=O), 1643 and 1626 (C=N), 1626 and 1518 (Aromatic C=C), 1446 (N=N). H^1NMR: 1.55 (s,1H), 1.97 (s,3H), 2.39 (s,3H) 7.29 and 7.55 (dd,4H), 7.41(d,1H), 7.54 (d,1H), 7.79 (s,1H). C^{13} NMR: 15.6, 21.7, 65.5, 120.3, 124.4, 124.4, 125.5, 127.5, 129.7, 129.7, 131.6, 133.3, 137.5, 143.7, 149.7, 155.9, 160.8, 166.7. Mass Spectra [M^+]:352.

3-((4-chlorophenyl)diazenyl)-2-methyl-7-nitropyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4j): Colour: Dark brwon, Yield = 83%, M.P.: 251°C, IR (KBr, cm^{-1}): 3048 (C=C-H), 1688 (C=O), 1649 and 1629 (C=N), 1610 and 1518 (Aromatic C=C) 1460 (N=N). H^1NMR: 1.58 (s,1H), 1.99 (s,3H), 7.31 and 7.57 (dd,4H), 7.81(d,1H), 8.42 (d,1H), 8.60 (s,1H). C^{13} NMR: 15.5, 66.5, 119.3, 121.8, 123.5, 124.7, 124.7, 128.8, 129.5, 129.5, 131.7, 143.3, 149.8, 153.7, 155.5, 160.5, 166.9. Mass Spectra [M^+]:383.

2-methyl-3-(p-tolyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9 (3*H*)-one (4k): Colour: Orange, Yield =84%, M.P.: 235°C, IR (KBr, cm^{-1}): 3035 (C=C-H), 1683 (C=O), 1641 and 1627 (C=N), 1622 and 1518 (Aromatic C=C), 1444 (N=N). H^1NMR: 1.46 (s,1H), 1.95 (s,3H), 2.43 (s,3H),7.33 and 7.83 (dd,4H), 7.69-8.10 (m,4H). C^{13} NMR: 15.3, 21.6, 66.2, 122.3, 122.7, 122.7, 126.3, 126.4, 127.1, 129.2, 129.2, 133.1, 135.1, 146.4, 147.5, 155.3, 160.5, 166.2.Mass Spectra [M^+]:318.

5-chloro-2-methyl-3-(p-tolyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4l): Colour: Orange, Yield =78%, M.P.: 244°C, IR (KBr, cm^{-1}): 3038 (C=C-H), 1688 (C=O), 1643 and 1626 (C=N), 1614 and 1511 (Aromatic C=C), 1458 (N=N). H^1NMR: 1.49 (s,1H), 1.91 (s,3H), 7.29 and 7.84 (dd,4H), 7.55- 7.95 (m,3H). C^{13} NMR: 15.3, 21.5, 66.3, 122.0, 122.8, 122.8, 124.5, 128.5, 129.8, 129.8, 132.2, 133.2, 135.7, 147.6, 155.3, 160.7, 161.6, 166.3. Mass Spectra [M^+]:352.

7-chloro-2-methyl-3-(p-tolyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4m): Colour: Orange, Yield = 84%, M.P.: 250°C, IR (KBr, cm^{-1}): 3040(C=C-H), 1688(C=O), 1646 and 1624 (C=N), 1616 and 1518 (Aromatic C=C), 1458 (N=N). H^1NMR: 1.52 (s,1H), 1.94 (s,3H), 2.44 (s,3H), 7.28 and 7.83 (dd,4H), 7.49 (d,1H), 7.77 (d,1H), 7.99 (s,1H). C^{13} NMR: 15.1, 21.6, 66.6, 122.0, 122.8, 122.8, 127.9, 127.9, 129.8, 129.5, 132.3, 133.7, 135.5, 145.3, 147.6, 155.9, 160.5, 166.6.Mass Spectra [M^+]:352.

2, 7-dimethyl-3-(p-tolyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4n): Colour: Dark brown, Yield = 75%, M.P.:257°C, IR (KBr, cm^{-1}): 3044 (C=C-H), 1690 (C=O), 1650 and 1624(C=N), 1615 and 1515 (Aromatic C=C), 1446 (N=N).H^1NMR: 1.56 (s, 1H), 1.96(s, 3H), 2.41(s, 3H), 7.28 and 7.83 (dd, 4H), 7.40 (d, 1H), 7.52(d, 1H), 7.77 (s, 1H). C^{13} NMR: 15.5, 21.6, 21.6, 65.7, 120.3, 122.8, 122.8, 125.5, 127.6, 129.8,129.8, 133.5, 135.6, 137.3, 143.7, 147.7, 155.9, 160.5, 166.3. Mass Spectra [M^+]:332**.**

2-methyl-7-nitro-3-(p-tolyldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4o): Colour: Reddish brown, Yield = 80%, M.P.: 262°C, IR (KBr, cm^{-1}): 3049 (C=C-H), 1686 (C=O), 1647 and 1630 (C=N), 1613 and 1520(Aromatic C=C), 1458 (N=N). H^1NMR: 1.59(s,1H), 1.99 (s,3H), 2.63 (s,3H), 7.29 and 7.84 (dd,4H), 7.81 (d,1H), 8.45 (d,1H), 8.63 (s,1H). C^{13} NMR: 15.4, 21.3 65.6, 119.3, 121.5, 122.4, 122.4, 123.2, 128.9, 129.9, 129.6 135.9 143.7, 147.6, 153.36, 155.58, 160.9, 166.22. Mass Spectra [M^+]:363.

3-((4-methoxyphenyl)diazenyl)-2-methylpyrazolo[5,1-*b*]quinazolin-9(3*H*)-one (4P): Colour: Orange, Yield = 84%, M.P.: 238°C, IR (KBr, cm^{-1}): 3040 (C=C-H), 1680 (C=O), 1641 and 1627 (C=N), 1612 and 1511 (Aromatic C=C), 1446 (N=N). H^1NMR: 1.40 (s,1H), 1.93 (s,3H), 3.89 (s,3H),7.03 and 7.33 (dd,4H), 7.69-8.10 (m,4H). C^{13} NMR: 15.3, 55.5, 65.3, 114.4, 114.4, 121.7, 123.9, 123.9, 126.8, 126.5, 127.7, 133.8, 143.6, 146.6, 155.4, 157.2, 160.6, 166.5. Mass Spectra [M^+]:334.

5-chloro-3-((4-methoxyphenyl)diazenyl)-2-methylpyrazolo [5,1-*b*]quinazolin-9(3*H*)-one (4q): Colour: Orange, Yield = 85%, M.P.: 249^{o}C, IR (KBr, cm^{-1}): 3045 (C=C-H), 1683 (C=O), 1645 and 1628 (C=N), 1617 and 1515 (Aromatic C=C), 1458 (N=N). H^{1}NMR: 1.47 (s,1H), 1.96 (s,3H), 3.89 (s,3H),7.04 and 7.26 (dd,4H), 7.55- 7.95 (m,3H). C^{13} NMR: 15.3, 55.5, 65.3, 114.4, 114.4, 122.2, 123.9, 123.9, 124.4, 128.3, 132.1, 133.7, 143.5, 155.9, 157.3, 160.5, 161.6, 166.6. Mass Spectra [M^{+}]:368.

7-chloro-3-((4-methoxyphenyl)diazenyl)-2-methylpyrazolo [5,1-*b*]quinazolin-9(3*H*)-one (4r): Colour: Orange, Yield = 88%, M.P.: 255^{o}C, IR (KBr, cm^{-1}): 3056 (C=C-H), 1687 (C=O), 1649 and 1620 (C=N), 1610 and 1520 (Aromatic C=C), 1456 (N=N). H^{1}NMR: 1.54 (s,1H), 1.97 (s,3H), 3.89 (s,3H), 7.03 and 7.31 (dd,4H), 7.49 (d,1H), 7.77 (d,1H), 7.99 (s,1H). C^{13} NMR: 15.1, 55.3, 65.6, 114.4, 114.4, 122.5, 123.9, 123.9, 127.5, 127.5, 132.4, 133.8, 143.6, 145.5, 155.3, 157.3, 160.5, 166.5. Mass Spectra [M^{+}]:368.

3-((4-methoxyphenyl)diazenyl)-2,7-dimethylpyrazolo[5,1-*b*] quinazolin-9(3*H*)-one (4s): Colour: Dark brown, Yield = 86%, M.P.: 264^{o}C, IR (KBr, cm^{-1}): 3050 (C=C-H), 1685 (C=O), 1650 and 1624 (C=N), 1616 and 1515 (aromatic C=C), 1460 (N=N). H^{1}NMR: 1.56 (s,1H), 1.99 (s,3H), 2.41(s,3H), 3.89 (s,3H), 7.05 and 7.33 (dd,4H), 7.40 (d,1H), 7.52 (d,1H), 7.77 (s,1H). C^{13} NMR: 15.5, 21.6, 55.5, 65.6, 114.4, 114.4, 120.3, 123.9, 123.9, 125.6, 127.6, 137.3, 133.4, 143.5, 143.6, 155.3, 157.3, 160.5, 166.6. Mass Spectra [M^{+}]:348.

3-((4-methoxyphenyl)diazenyl)-2-methyl-7-nitropyrazolo [5,1-*b*] quinazolin-9(3*H*)-one (4t): Colour: Reddish brown, Yield = 80%, M.P.: 269^{o}C, IR (KBr, cm^{-1}): 3047(C=C-H), 1690 (C=O), 1648 and 1624 (C=N), 1607 and 1518 (aromatic C=C) 1460 (N=N). H^{1}NMR: 1.59 (s,1H), 1.99 (s,3H), 3.89 (s,3H), 7.05 and 7.29 (dd,4H), 7.81 (d,1H), 8.45(d,1H), 8.63 (s,1H). C^{13} NMR: 15.3 , 55.5, 65.6, 114.4, 114.4, 118.6, 121.5, 123.3, 123.3, 123.5, 128.3, 143.5, 143.5, 153.3, 155.3, 157.5, 160.5, 166.6. Mass Spectra [M^{+}]:379.

Biological Activity: Antibacterial Activity: The antibacterial activity was measured by agar cup method using Nutrient agar (Himedia) as medium[22]. The bacterial cultures selected were, cultures viz. *Escherichia coli, Salmonella typhi.* A plate with ethanol was prepared as blank (negative control) Solution of penicillin in ethanol (0.1%) was also placed on the seeded nutrient agar surface as standard reference antibiotic (positive control). Incubation plates were observed for the zone of inhibition of bacterial growth around the agar cup. Results were recorded by measuring the zone of inhibition in millimeter (mm) using zone reader (table-1).

Antifungal Activity: Antifungal activity was performed by Poison plate method. The medium used was Potato Dextrose Agar (Himedia)[22]. A plate with ethanol was prepared as blank (negative control) similarly a plate with 1% Gresiofulvin was prepared as standard reference plate (positive control).

Aspergillus niger and *Penicillium chrysogenum* were selected as test fungal cultures. After incubation plates were observed for the growth of inoculated fungi. Results were recorded (table-1) as moderate growth of fungi (**++**), reduced growth of fungi (**+**) and no growth of inoculated fungi (-) antifungal activity.

Results and Discussion

All the reactions were performed by conventional methods. The intermediates diazonium salts solutions [2a-2d] was added drop wise to the ice cooled solution of 2-methylpyrazolo[5,1-*b*]quinazolin-9(4*H*)-ones[3a-3e] dissolved in alcohol. Addition is done with constant stirring and maintaining the temperature below 5^{0}C. The colored precipitate of 2-methyl-3-(aryldiazenyl)pyrazolo[5,1-*b*]quinazolin-9(3*H*)-ones, [4a-4t] were washed with cooled water, dried, crystallized with absolute alcohol.

Assignment of significant peaks observed in IR, ^{1}HNMR, ^{13}CNMR spectra of the compounds [4a-4t] is clarified in analytical data. The IR spectra of compound [4a-4t] show high intensity bands at 1650-1640 and 1630-1622 cm^{-1} is assigned to ν (C=N) vibrations. The band around 1610 and 1510 cm^{-1} is assigned to combination of ν (C=C) of the aromatic ring. A high intensity band in the region 1690-1680 cm^{-1} is assigned for carbonyl group. The band around 1460-1430 cm-1 indicates the presence of azo linkage for ν (N=N).

Each one of the ^{1}H NMR spectra of [4a-4t] revealed singlet for 3H between 2.39-2.51 ppm and for 1H between 1.60-1.40 ppm are assigned to C^{2}-metyl group and C^{3} hydrogen respectively. Peaks round 8.61-7.05 ppm are assigned to aromatic protons. ^{1}H NMR spectra of [4f-4t] showed double doublet confirming para substitution at C^{3}-aryldiazenyl moiety. Compounds [4a-4e] lack this double doublet peak. Compound [4p-4t] revealed peak at 3.97-3.94 ppm assigned to methyl proton of –OCH$_{3}$. ^{13}CNMR showed peaks around 163 ppm indicates carbonyl carbon. Assignment given to peaks observed in IR, ^{1}HNMR, ^{13}CNMR spectra and also molecular ion peaks in mass spectra justifies the structure of compounds [4a-4t].

The synthesized compounds were evaluated for anti-bacterial and anti-fungal activity with different strains of bacteria and fungi. Results are shown in table-1. All have shown lesser activity against *E. coli and B. typhi* compared with penicillin taken as standard. The activity of few compounds was satisfactory and has also shown activity against *S. typhi* and fungi. Antifungal activity observed against *Aspergillus niger* was satisfatory than *Penicillium chrysogenum.*

Table-1
Anti Microbial activity

Compound	Zone of Inhibition (diameter in mm)		Growth of Fungi	
	E. coli	***S. typhi***	***A. niger***	***P. chrysogenum***
Penicillin	24	18	-	-
(3a)	13	-	+	++
(3b)	20	10	-	-
(3c)	20	11	+	++
(3d)	12	-	+	++
(3e)	21	10	-	+
(3f)	14	-	+	++
(3g)	19	12	-	-
(3h)	20	9	+	++
(3i)	13	-	+	++
(3j)	20	12	-	+
(3k)	13	-	+	++
(3l)	19	7	-	-
(3m)	20	8	+	++
(3n)	14	-	+	++
(3o)	21	9	-	+
(3p)	15	-	+	++
(3q)	20	8	-	-
(3r)	21	9	+	++
(3s)	14	-	+	++
(3t)	21	13	-	+

Moderate growth (**++**), Reduced growth (**+**) and No growth (-) of fungi

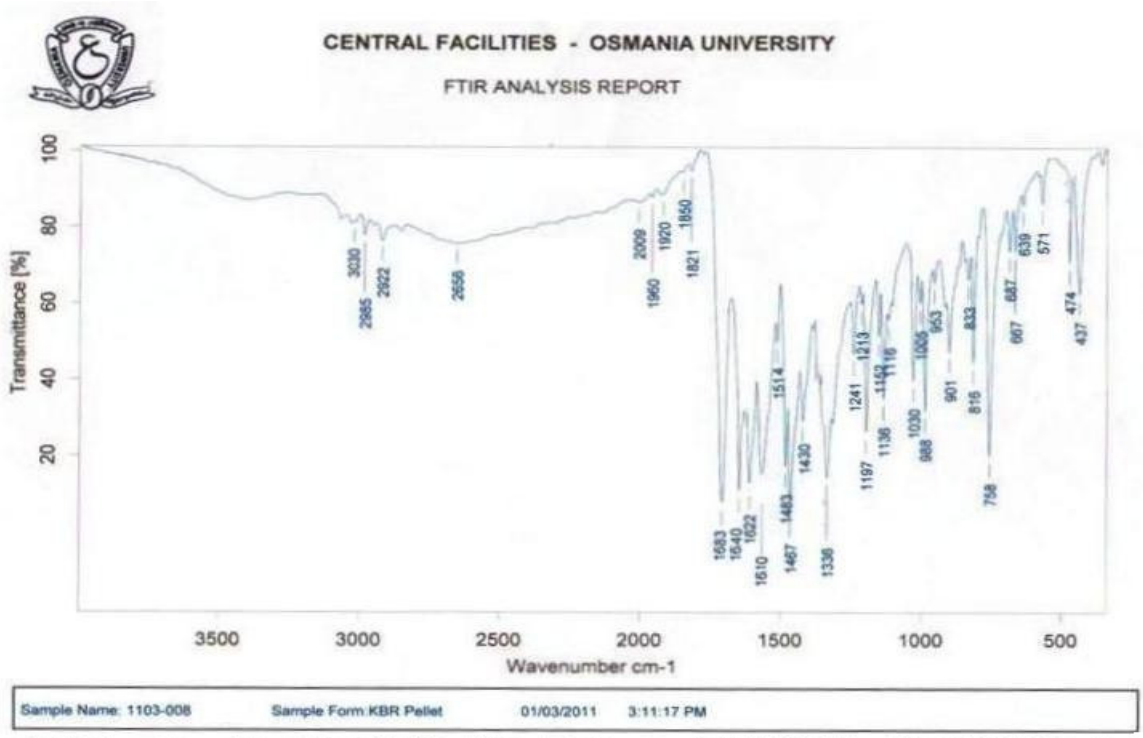


Fig. 2.1: IR spectra of 2-methyl-3-(phenyldiazenyl)pyrazolo[5,1-b]quinazolin-9(3H)-one (4a)

Figure-1
IR spectra of 2-methyl-3-(phenyldiazenyl) pyrazolo [5,1-b]quinazolin-9(3H)-one (4a)

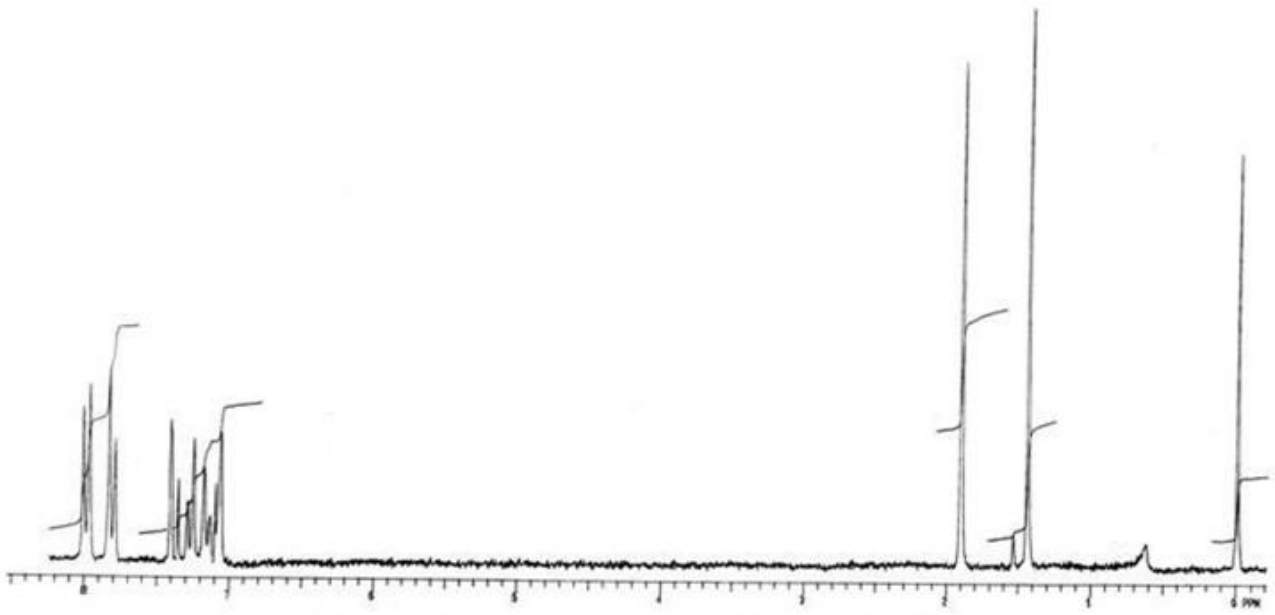


Figure-2
HNMR spectrum of 2-methyl-3-(phenyldiazenyl) pyrazolo [5,1-b]quinazolin-9(3H)-one (4P)

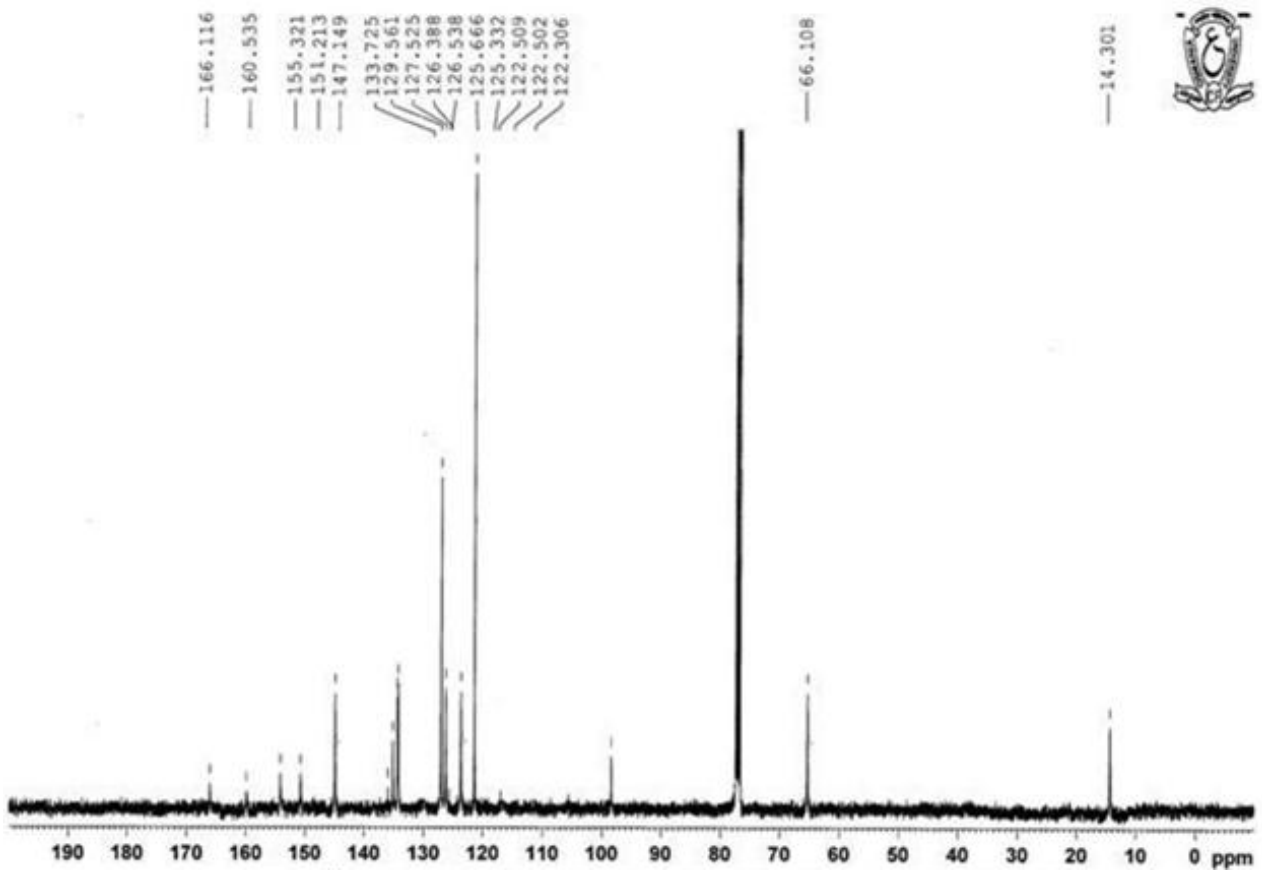


Figure-3
CNMR of 2-methyl-3-(phenyldiazenyl) pyrazolo [5,1-b]quinazolin-9(3H)-one (4P)

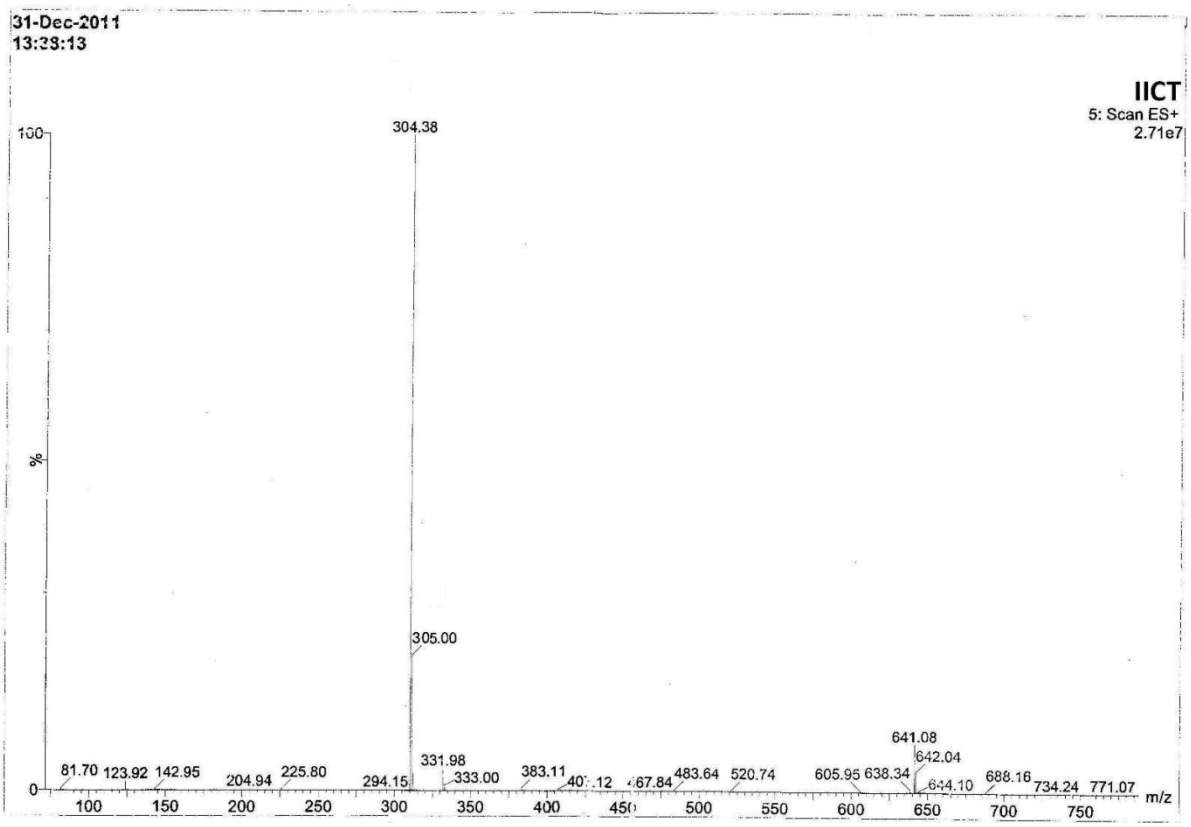


Figure-4
Mass spectrum of 2-methyl-3-(phenyldiazenyl) pyrazolo [5,1-b]quinazolin-9(3H)-one (4P)

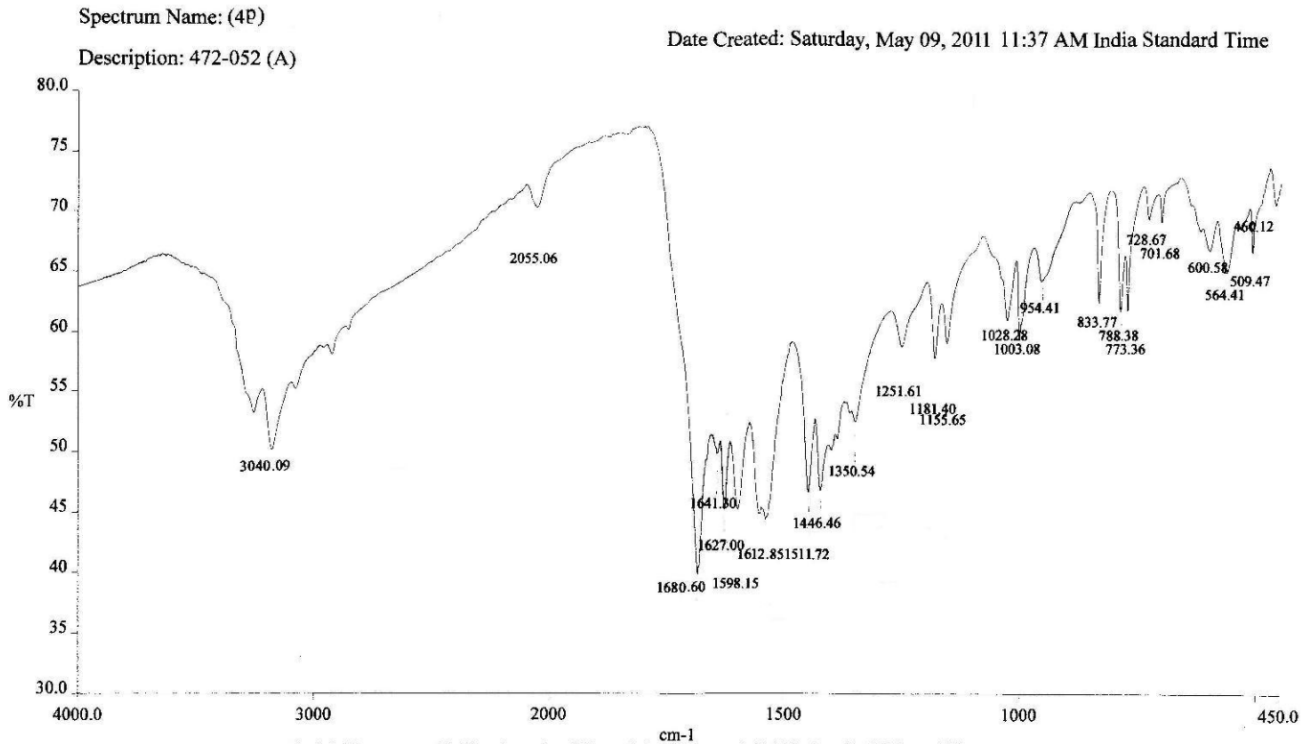


Figure-5
IR spectrum of 3-((4-methoxyphenyl) diazenyl)-2-methylpyrazolo[5,1-b]quinazolin-9(3H)-one (4P)

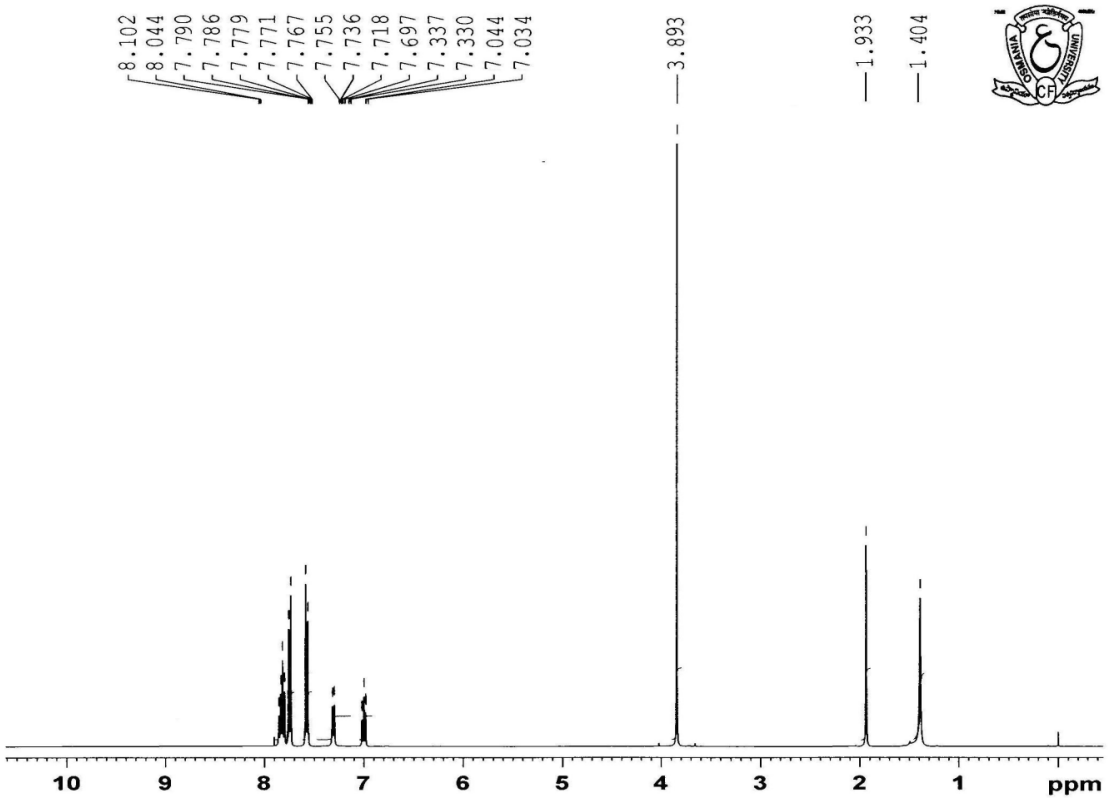


Figure-6
HNMR spectrum of 3-((4-methoxyphenyl) diazenyl)-2-methylpyrazolo[5,1-b]quinazolin-9(3H)-one (4P)

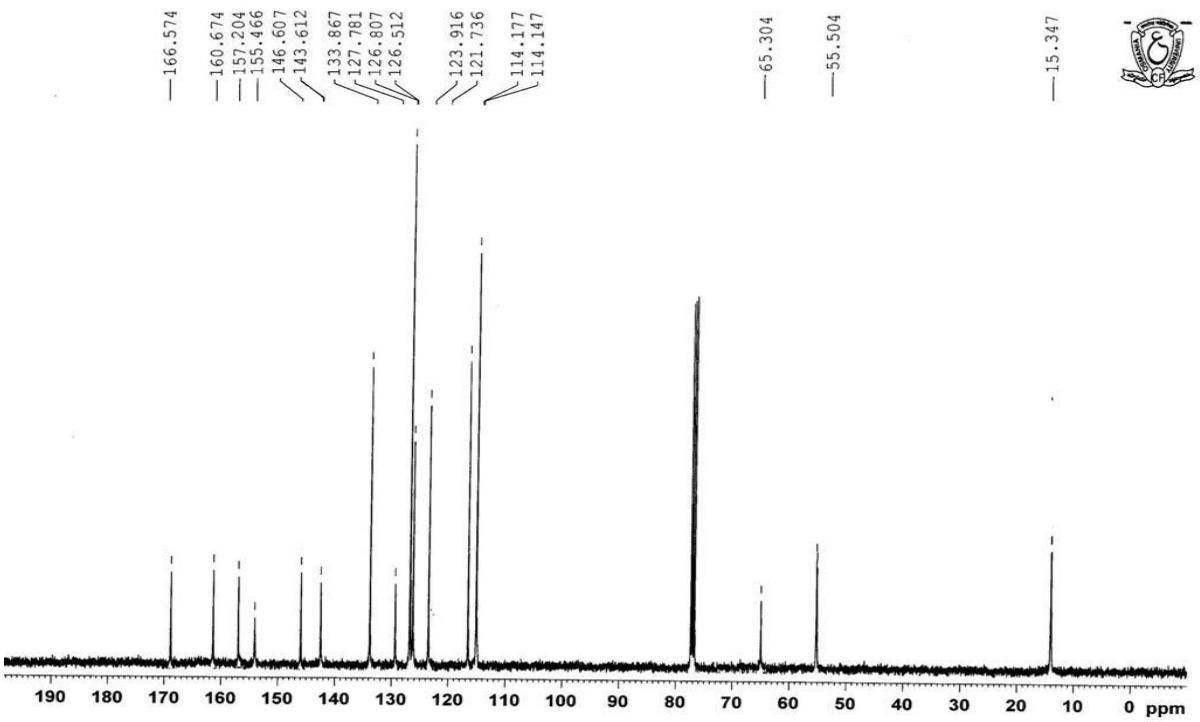


Figure-7
CNMR spectrum of 3-((4-methoxyphenyl) diazenyl)-2-methylpyrazolo[5,1-b]quinazolin-9(3H)-one (4P)

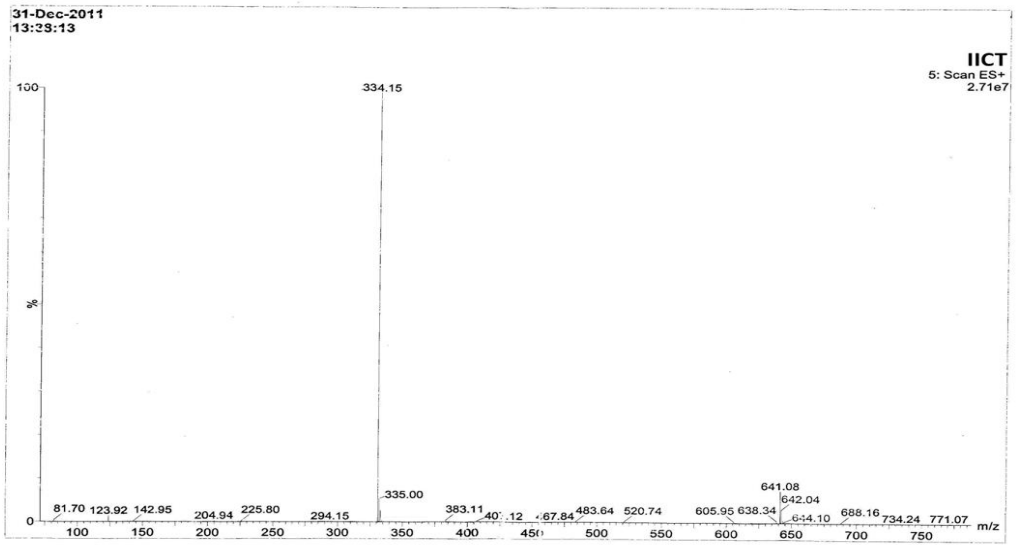


Figure-8

Mass spectrum of 3-((4-methoxyphenyl) diazenyl)-2-methylpyrazolo[5,1-b]quinazolin-9(3H)-one (4P)

Conclusion

It may be concluded from results that better antimicrobial activity is due to the presence of halogen and methoxy group as substituent.

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