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RESEARCH ARTICLE

SYNTHESIS AND CHARACTERIZATION OF NOVEL 3*H*-PHENOXAZIN-3-ONE, [1,4]BENZOXAZINO[2,3-*b*]PHENOXAZINE AND IMIDAZO[4,5-*b*]PHENOXAZIN-2(1*H*)-ONE DERIVATIVES

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Abstract

Novel heterocyclic derivatives of 3*H*-phenoxazin-3-one **6(a-d)**, [1,4]benzoxazino[2,3-*b*]phenoxazine **7(a-d)** and imidazo[4,5-*b*]phenoxazin-2(1*H*)-one **9(a-d)** are prepared by condensing 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4** with substituted *ortho*-aminophenols **5(a-d)** and urea **8** in ethanolic solution of fused sodium acetate. The structures of newly synthesized compounds are characterized by using elemental analysis, IR, ^{1}H NMR, ^{13}C NMR and mass spectral data.

Introduction:-

3*H*-Phenoxazin-3-ones and [1,4]benzoxazino[2,3-*b*]phenoxazines belong to the family of phenoxazines. 3*H*-Phenoxazin-3-ones are widely distributed in natural world being found in micro organisms and fungi[1-3] and exhibit antiviral[4], antibacterial[5], antimiocrobial[6,7], anticancer[8], antitumor[9], sun-induced antimelanogenesis[1] and phytotoxic[10] activities. [1,4]Benzoxazino[2,3-*b*]phenoxazine molecules and their derivatives are having good chromogenic properties. They form an important class of dyes of oxazine series[11-14] and are useful for colouring plastics, varnishes, lacquers, viscose, rubber and paper in different shades[15-17]. The sulfonated derivatives of this class have been widely used as direct cotton dye stuffs possessing high fastness against photochemical degradation[18]. These compounds exhibit variety of biological activities such as antifungal[19], antibacterial, muscle-relaxant and hypnotic agents[19,20]. Some recent applications regarding the preparation of semiconductors[21,22] and nonlinear optical wave guiding polymer films bearing a unique photovoltaic property[22-25] having enhanced their utility value.

A number of 3*H*-phenoxazin-3-one[26-28] and [1,4]benzoxazino[2,3-b]phenoxazine[29-31] derivatives have been reported for their wide range of applications and a typical reaction has been reported involving the condensation of urea with 1,4-benzoquinone[32]. Inview of this, the project is taken up and the present article deals with the synthesis and characterization of novel 3*H*-phenoxazin-3-one, [1,4]benzoxazino[2,3-*b*]phenoxazine and imidazo[4,5-*b*]phenoxazin-2(1*H*)-one heterocyclic derivatives.

Materials and Methods:-

All the reagents and solvents used are of laboratory grade. Melting points of all the compounds are determined in open capillary method and are uncorrected. All the new products are monitored by TLC using Merck brand silica gel-G plates for TLC. IR spectra are recorded in KBr pellets on Nexus 470 FTIR spectrometer. ^{1}H NMR spectra are recorded in $CDCl_3$ and DMSO-d_6 solvent on Varian Mercury 400 MHz spectrometer using TMS as internal

standard. ^{13}C NMR spectra are recorded on Varian Mercury 75 MHz spectrometer and ESI-Mass spectra are obtained on Shimadzu mass spectrometer.

Preparation of 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone (4): 2,5-Dihydroxy-1,4-benzoquinone **2** was prepared[33] from the oxidation of hydroquinone **1** by hydrogen peroxide in alkaline solution. The two hydroxyl groups were then protected by reaction with methanol under acidic conditions to give 2,5-dimethoxy-1,4-benzoquinone[34] **3**. Bromination of 2,5-dimethoxy-1,4-benzoquinone with N-bromosuccinimide in dimethyl formamide solvent was carried out at room temperature to obtain 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone[35] **4.** The structure of **4** was confirmed by its spectral analysis (Scheme 1).

General procedure for the synthesis of 3*H*-phenoxazin-3-one derivatives (6a-d): 6.135 mmol of substituted *ortho*-aminophenol **5(a-d)** was taken into 100 ml R.B flask and added 30 ml of ethanol followed by anhydrous sodium acetate (6.135 mmol). The mixture was stirred for 15 minutes at room temperature. To the mixture, 6.135 mmol of 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4** was added portion wise. Then the reaction mixture was heated at reflux temperature for 3-5 hours. The progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was cooled to room temperature, poured into water and extracted with EtOAc thrice (3x20 ml). The combined organic layers were washed with brine dried over anhydrous sodium sulfate and the organic layer was concentrated to obtain the crude product. The crude product was purified by silica gel column chromatography using variants of ethyl acetate petroleum ether mixture. The fractions containing the compound were subjected to distillation to remove solvent and to obtain the pure products **6(a-d)**. Formation of **6(a-d)** was confirmed by their spectral analysis (Scheme 1).

2-Bromo-1,4-dimethoxy-3*H*-phenoxazin-3-one (6a): Yield: 1.55 g, 75%, mp 199-201 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1651 (C=O), 1618 (C=N), 1259, 1030 (C-OCH$_3$), 1195 (C-O-C). ^{1}H NMR (400 MHz, CDCl$_3$) δ: 7.12-7.48 (4H, m, Ar-H), 3.88 (3H, s, OCH$_3$), 3.82 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 171.2, 153.9, 145.9, 138.7, 135.4, 134.5, 127.2, 126.6, 125.2, 119.6, 116.0, 115.5, 60.9, 60.3. ESI-MS, m/z 337 [M+H]$^+$. Anal. Calcd for $C_{14}H_{10}BrNO_4$; C, 49.98; H, 2.98; N, 4.15. Found C, 50.03; H, 3.02; N, 4.09.

2-Bromo-8-chloro-1,4-dimethoxy-3*H*-phenoxazin-3-one (6b): Yield 1.39 g, 61%, mp 221-223 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1653 (C=O), 1620 (C=N), 1261, 1038 (C-OCH$_3$), 1198 (C-O-C). ^{1}H NMR (400 MHz, CDCl$_3$) δ: 7.04-7.56 (3H, m, Ar-H); 3.88 (3H, s, OCH$_3$) 3.84 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 171.0, 153.0, 146.0, 139.3, 138.9, 135.3, 128.6, 127.0, 124.2, 118.1, 115.6, 115.4, 60.7, 60.2. ESI-MS, m/z 370 [M+H]$^+$. Anal. Calcd for $C_{14}H_9BrClNO_4$; C, 45.29; H, 2.49; N, 3.81. Found C, 45.35; H, 2.53; N, 3.88.

2-Bromo-1,4-dimethoxy-8-methyl-3*H*-phenoxazin-3-one (6c): Yield 1.58 g, 74%, mp 217-219 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1643 (C=O), 1608 (C=N), 1252, 1033 (C-OCH$_3$), 1192 (C-O-C). ^{1}H NMR (400 MHz, CDCl$_3$) δ: 6.96-7.49 (3H, m, Ar-H), 3.83 (3H, s, OCH$_3$); 3.78 (3H, s, OCH$_3$), 2.34 (3H, s, CH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 171.3, 152.5, 145.8, 138.6, 136.4, 135.6, 131.1, 127.5, 119.8, 118.1, 115.7, 115.2, 60.8, 60.1, 21.2. ESI MS, m/z 351 [M+H]$^+$. Anal. Calcd. for $C_{15}H_{22}BrNO_4$; C, 51.39; H, 3.51; N, 3.99. Found C, 51.43; H, 3.46; N, 4.02.

2-Bromo-1,4,8-trimethoxy-3*H*-phenoxazin-3-one (6d): Yield 1.62 g, 72%, mp 226-228 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1640 (C=O), 1605 (C=N), 1249, 1028 (C-OCH$_3$), 1188 (C-O-C). ^{1}H NMR (400 MHz, CDCl$_3$) δ: 6.92-7.05 (3H, m, Ar-H), 3.83 (3H, s, OCH$_3$), 3.82 (3H, s, OCH$_3$), 3.80 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 171.1, 152.0, 150.1, 145.5, 138.5, 136.7, 135.5, 127.4, 123.8, 116.5, 115.1, 107.8, 60.6, 60.1, 55.9. ESI-MS, m/z 366 [M+H]$^+$. Anal. Calcd for $C_{15}H_{12}BrNO_5$; C, 49.28; H, 3.26; N, 3.88. Found C, 49.15; H, 3.32; N, 3.84.

General procedure for the synthesis of [1,4]benzoxazino[2,3-b]phenoxazine derivatives (7a-d): 1.5 mmol of substituted *ortho*-aminophenol **5(a-d)** was taken into 50 ml R.B flask and added 15 ml of ethanol followed by anhydrous sodium acetate (1.5 mmol). The mixture was vigorously stirred for 15 minutes at room temperature. Later 1.5 mmol of substituted 2-bromo-1,4-dimethoxy-3*H*-phenoxazin-3-one **6(a-d)** was added portion wise. Then the mixture was heated at reflux temperature for 3-5 hours. The progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was cooled to room temperature, poured into water and extracted with EtOAc thrice (3x20 ml). The combined organic layers were washed with brine and dried over anhydrous sodium sulfate. On concentrating combined organic layers, afforded the crude product. The crude product was purified by silica gel column chromatography using variants of ethyl acetate-petroleum ether mixture. The fractions

containing the compound were concentrated by distilling out the solvent to obtain the pure products **7(a-d)**. Formation of **7(a-d)** and their structures were confirmed by their spectral analysis (Scheme 2).

6,13-Dimethoxy[1,4]benzoxazino[2,3-*b*]phenoxazine (7a): Yield 0.28 g, 54%, mp>300 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1619 (C=N), 1229, 1027 (C-OCH$_3$), 1162 (C-O-C). ^{1}H NMR (400 MH$_Z$, CDCl$_3$) δ: 7.02-7.48 (8H, m, Ar-H), 4.02 (6H, s, OCH$_3$).^{13}C NMR (75 MHz, CDCl$_3$) δ: 146.2, 136.1, 135.4, 132.4, 127.3, 127.1, 125.5, 122.6, 115.4, 60.3. ESI-MS, m/z 347 [M+H]$^+$. Anal. Calcd. For $C_{20}H_{14}N_2O_4$; C, 69.4; H, 4.04; N, 8.12. Found C, 69.34; H, 4.09; N, 8.07.

2,9-Dichloro-6,13-dimethoxy[1,4]benzoxazino[2,3-*b*]phenoxazine (7b): Yield 0.42 g, 68%, mp 298-300 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1620 (C=N), 1232, 1029 (C-OCH$_3$), 1163 (C-O-C) 1090. ^{1}H NMR (400 MHz, CDCl$_3$) δ: 7.08-7.42 (6H, s, Ar-H), 4.04 (6H, s, OCH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 145.6, 136.3, 132.8, 132.5, 132.1, 129.0, 127.1, 122.0, 116.9, 60.1. ESI-MS, m/z 415 [M+H]$^+$. Anal. Calcd. for $C_{20}H_{12}Cl_2N_2O_4$; C, 57.79; H, 2.88; N, 6.80. Found C, 57.86; H, 2.94; N, 6.72.

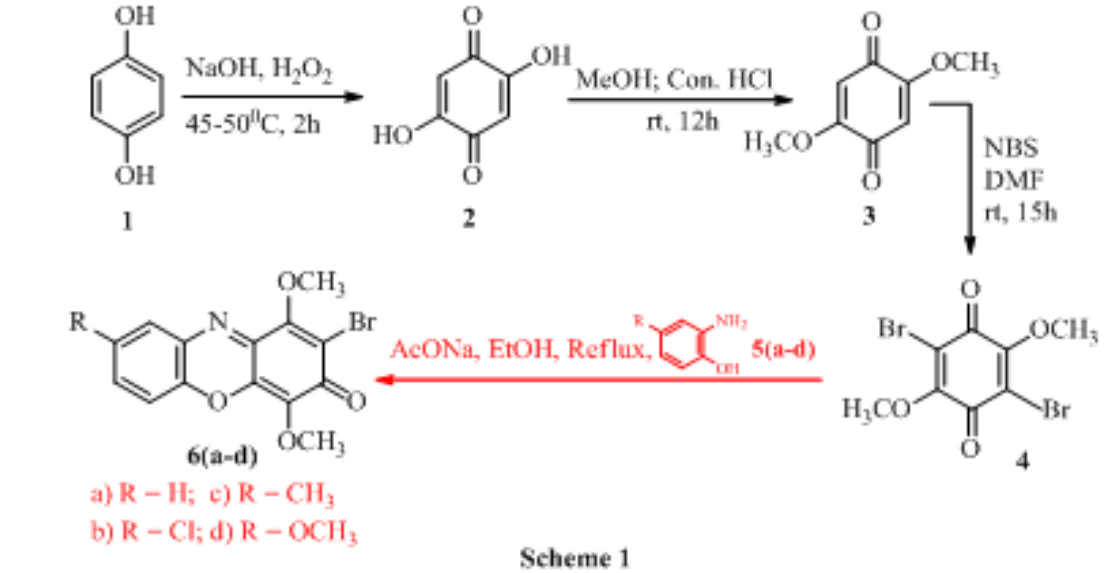
6,13-Dimethoxy-2,9-dimethyl[1,4]benzoxazino[2,3-*b*]phenoxazine (7c): Yield 0.35 g, 63%, mp>300 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1616 (C=N), 1226, 1024 (C-OCH$_3$), 1154 (C-O-C). ^{1}H NMR (400 MHz, CDCl$_3$) δ: 7.05-7.44 (6H, m, Ar-H), 3.98 (6H, s, OCH$_3$), 2.24 (6H, s, CH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 143.9, 136.5, 136.4, 132.3, 128.1, 127.3, 127.2, 122.71, 115.0, 60.2, 21.8. ESI-MS, m/z 375 [M+H]$^+$. Anal. Calcd. for $C_{22}H_{18}N_2O_4$; C, 70.63; H, 4.88; N, 7.51. Found C, 70.57; H, 4.84; N, 7.58.

2,6,9,13-Tetramethoxy[1,4]benzoxazino[2,3-*b*]phenoxazine (7d): Yield 0.39 g, 65%, mp>300 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 1609 (C=N), 1223, 1024 (C-OCH$_3$), 1151 (C-O-C), ^{1}H NMR (400 MHz, CDCl$_3$) δ: 6.69-7.08 (6H, m, Ar-H), 4.00 (6H, s, OCH$_3$), 3.82 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, CDCl$_3$) δ: 155.0, 142.7, 136.7, 136.0, 132.1, 127.4, 116.2, 113.5, 104.1, 60.6, 60.2. ESI-MS, m/z 407 [M+H]$^+$, Anal. Calcd. for $C_{22}H_{18}N_2O_6$; C, 57.79; H, 2,88; N, 6.80. Found C, 57.86; H, 2.94; N, 6.72.

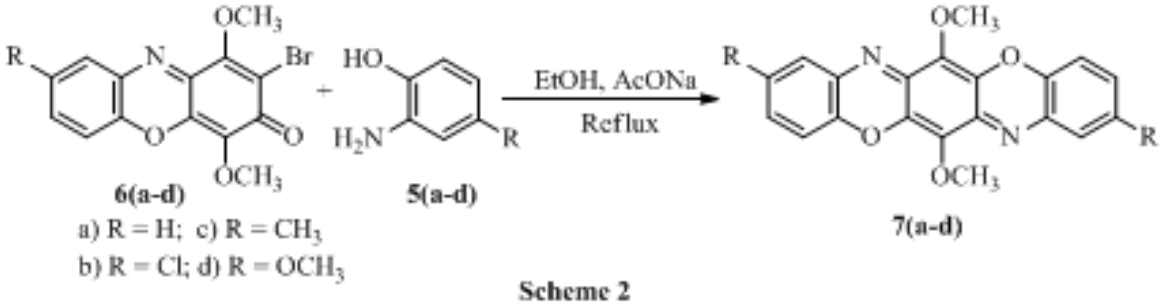
General procedure for the synthesis of 4,11-dimethoxyimidazo[4,5-*b*]phenoxazin-2(1*H*)-one derivatives (9a-d): To the stirred suspension of substituted 2-bromo-1,4-dimethoxy-3*H*-phenoxazin-3-one **6(a-d)** (1.5 mmol) and anhydrous sodium acetate (1.5 mmol) in ethanol 10 ml was added 1.5 mmol of urea **8** at room temperature and stirred for 5 minutes. Then the reaction mixture was refluxed for 3-6 hours. The progress of the reaction was monitored by TLC. After completion of the reaction, the reaction mixture was cooled to room temperature, poured into water and extracted with ethyl acetate thrice (3x20 ml). The combined organic layers were washed with brine and dried over anhydrous sodium sulfate. The organic layers were concentrated to obtain the crude product. The crude product was purified by silica gel column chromatography using variants of ethyl acetate-petroleum ether mixture. Combined fractions containing the compound were freed from the solvent to obtain the pure products **9(a-d)**. Structures of **9(a-d)** were confirmed by their spectral analysis (Scheme 3).

4,11-Dimethoxyimidazo[4,5-*b*]phenoxazin-2(1*H*)-one (9a): Yield 0.3 g, 68%, mp 244-246 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 3221 (NH), 1676 (C=O), 1618 (C=N), 1597 (NH, bend) 1221, 1032 (C-OCH$_3$), 1158 (C-O-C). ^{1}H NMR (400 MHz, DMSO-d$_6$) δ: 7.70 (1H, br.s, NH), 7.06-7.36 (4H, m, Ar-H), 4.08 (3H, s, OCH$_3$), 3.96 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, DMSO-d$_6$) δ: 163.4, 145.1, 144.0 136.7, 135.4, 134.1, 133.3, 131.2, 127.1, 126.5, 125.6, 116.4, 115.4, 60.9, 60.3. ESI-MS, m/z 298 [M+H]$^+$. Anal. Calcd for $C_{15}H_{11}N_3O_4$; C, 60.56; H, 3.71; N, 14.18. Found C, 60.63; H, 3.69; N, 14.15.

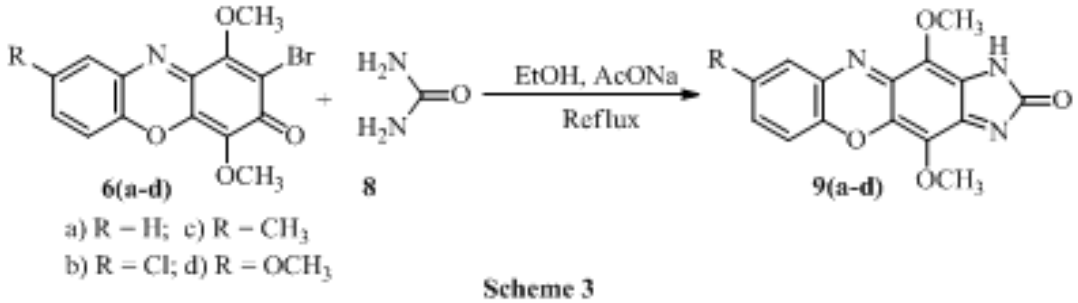
8-Chloro-4,11-dimethoxyimidazo[4,5-*b*]phenoxazin-2(1*H*)-one(9b): Yield 0.3 g, 61%, mp 251-253 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 3236 (NH), 1678 (C=O), 1620 (C=N), 1602 (NH, bend), 1228, 1036 (C-OCH$_3$), 1162 (C-O-C). ^{1}H NMR (400 MHz, DMSO-d$_6$) δ: 7.72 (1H, br.s, NH), 7.14-7.42 (3H, m, Ar-H), 4.08 (3H, s, OCH$_3$), 4.02 (3H, s, OCH$_3$). ^{13}C NMR (75 MHz, DMSO-d$_6$) δ: 163.2, 144.6, 138.3, 136.4, 135.2, 133.1, 131.6, 131.2, 128.6, 125.5, 121.1, 117.6, 115.6, 60.7, 60.2. ESI-MS, m/z 332 [M+H]$^+$. Anal.Calcd for $C_{15}H_{10}ClN_3O_4$; C, 54.26; H, 2.98; N, 12.78. Found C, 54.33; H, 3.01; N, 12.65.



Scheme 1



Scheme 2



Scheme 3

4,11-Dimethoxy-8-methylimidazo[4,5-*b*]phenoxazin-2(1*H*)-one (9c): Yield 0.29 g, 63%, mp 242-244 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 3229 (NH), 1674 (C=O), 1615 (C=N), 1595 (NH, bend), 1220, 1033 (C-OCH_3), 1155 (C-O-C). ^{1}H NMR (400 MHz, DMSO-d_6) δ: 7.66 (1H, s, CH_3), 7.10-7.44 (3H, m, Ar-H), 4.06 (3H, s, OCH_3), 4.01 (3H, s, OCH_3), 2.28 (3H, s, CH_3). ^{13}C NMR (75 MHz, DMSO-d^6) δ: 163.5, 142.2, 138.1, 136.6, 135.3, 135.1, 133.2, 133.0, 131.1, 125.4, 123.6, 116.5, 114.6, 60.8, 60.1, 21.2. ESI-MS, m/z 312 $[M+H]^+$. Anal. Calcd for $C_{16}H_{13}N_3O_4$; C, 61.69; H, 4.24; N, 13.48. Found C, 61.72; H, 4.20; N, 13.52.

4,8,11-Trimethoxyimidazo[4,5-*b*]phenoxazin-2(1*H*)-one (9d): Yield 0.35 g, 72%, mp 239-241 ^{0}C. IR (KBr) ν_{max} cm^{-1}: 3225 (NH), 1671 (C=O), 1612 (C=N), 1593 (NH, bend), 1220, 1030 (C-OCH_3), 1151 (C-O-C). ^{1}H NMR (400MHz, DMSO-d_6) δ: 7.64 (1H, br.s, NH), 6.88-7.17 (3H, m, Ar-H), 4.04 (3H, s, OCH_3), 4.00 (3H, s, OCH_3), 3.86 (3H, s, CH_3). ^{13}C NMR (75 MHz, DMSO-d_6) δ: 163.1, 156.5, 140.1, 138.2, 136.5, 135.5, 135.1, 133.2, 131.3, 125.4, 116.0, 109.0, 99.2, 60.6, 60.1, 55.9. ESI-MS, m/z 328 $[M+H]^+$. Anal. Calcd for $C_{16}H_{13}N_3O_5$; C, 58.81; H, 4.01; N, 12.86. Found C, 58.70; H, 4.01; N, 12.83.

Results and Discussion:-

In this article, we have synthesized twelve new heterocyclic compounds **6(a-d)**, **7(a-d)**, **9(a-d)**. All reactions were carried out in ethanol in the presence of fused sodium acetate at reflux temperatures. The synthesis of 3*H*-phenoxazin-3-one derivatives **6(a-d)** have been achieved by condensation of 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4** with substituted *ortho*-aminophenols **5(a-d)**. The synthesis of [1,4]benzoxazino[2,3-b]phenoxazine derivatives **7(a-d)** have been achieved by condensation of **6(a-d)** with another mole of **5(a-d)** and imidazo[4,5-*b*]phenoxazin-2(1*H*)-one derivatives **9(a-d)** are synthesized by condensation of 3*H*-phenoxazin-3-one derivatives **6(a-d)** with urea **8**. The structures of all the newly synthesized compounds have been supported by elemental analysis, IR, ^{1}H NMR, ^{13}C NMR and mass spectral studies.

The infrared spectrum of the compound **6a** shows C=O stretching frequency at 1651 cm^{-1}, C-OCH_3 stretching frequencies at 1259 $cm^{-1}$and 1030 cm^{-1}. These functional groups are also present in the parent compound 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4**. The C=N and C-O-C stretching frequencies are observed at 1618 cm^{-1} and 1195 cm^{-1} along with aromatic C-C and C-H absorption bands respectively. They are absent in the IR spectrum of 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4** and confirm the 2-bromo-1,4-dimethoxy-3*H*-phenoxazin-3-one **6a** structure. The ^{1}H NMR spectrum of the compound **6a** shows multiplet in between δ 7.12-7.48 for the four aromatic protons corresponding to phenoxazin part. Two singlets appearing at δ 3.82 and δ 3.88 belong to six protons of two methoxy groups and confirm the structure of 2-bromo-1,4-dimethoxy-3*H*-phenoxazin-3-one **6a** for the product. The ^{13}C NMR spectrum of the compound **6a** showed the presence of carbonyl (C=O), C-OCH_3, imine (-N=C<), aromatic and methoxy carbons. The peak appeared at δ 171.2 belong to carbonyl (C=O) carbon, the peaks at δ 153.9 and δ 145.9 are for C-OCH_3 (C_1 & C_4) carbons and the peaks which appeared at δ 138.7, 135.4 belong to C-O-C (4a) and imine (C-10a) carbons respectively. The aromatic carbon peaks appeared between δ 115.5 and 134.5 and the peaks appeared at δ 60.9, 60.3 belong to two methoxy carbons. The ESI mass spectrum of the compound **6a** shows a peak at m/z 337 $[M+H]^+$, indicating molecular weight of the new derivative. The IR, ^{1}H NMR, ^{13}C NMR and Mass spectral data fit the structure proposed indicating the formation of the compound **6a** in the reaction as expected.

The infrared spectrum of the compound **7a** shows absorption bands at 1619 cm^{-1} (C=N), 1229 cm^{-1}, 1027 cm^{-1} (C-OCH_3) and 1162 cm^{-1} (C-O-C). The main difference of this spectrum from that of its parent compound is the conspicuous loss of oxazin-3-one carbonyl (C=O) absorption band. The ^{1}H NMR spectrum of the compound **7a** has a multiplet at δ 7.02-7.48 in the aromatic region corresponding to eight protons. A singlet appeared at δ 4.02 integrating for six protons belongs to two methoxy groups. The ^{13}C NMR spectrum of the compound **7a** showed the peaks corresponding to C-OCH_3, C-O-C, imine, aromatic and methoxy carbons. The peaks showed up at δ 146.2, 136.1, 135.4 and 60.3 belong to C-OCH_3(C-6,13), C-O-C (C-5a,12a), imine (C-6a,13a) and methoxy carbons respectively. Rest of the peaks appeared between δ 115.4 and 132.4 belong to aromatic carbons. The absorptions confirm the formation of 6,13-dimethoxy[1,4]benzoxazino[2,3-b]phenoxazine **7a**. The ESI mass spectrum of the compound **7a** shows a peak at m/z 347 corresponding $[M+H]^+$ indicating the molecular weight. The IR, ^{1}H NMR, ^{13}C NMR and Mass spectral data accrued confirm the formation of the compound **7a**.

The absorption in the infrared spectrum of the compound **9a** at 1618 cm^{-1} (C=N), 1597 cm^{-1} (N-H), 1221 cm^{-1}, 1032 cm^{-1} (C-OCH_3), 1158 cm^{-1} (C-O-C) are in accordance with the structure. The characteristic bands of the spectrum include 1676 cm^{-1} (C=O, amide) 3221 cm^{-1} (NH). The main difference of this spectrum from that of its parent compound is the loss of oxazin-3-one carbonyl (C=O) and the appearance of amide carbonyl (C=O) and NH frequency bands and they confirm the compound **9a** structure. A multiplet in the ^{1}H NMR spectrum of the compound **9a** at δ 7.06-7.36 in the aromatic region corresponds to four protons of benzoxazin part. Two singlets at δ 3.96 and δ 4.08 accounting for three protons each indicate the presence of six protons of two methoxy groups. The NH proton of imidazolone appears as a broad singlet at 7.70. This confirms the structure proposed for the compound **9a**. The ^{13}C NMR spectrum of the compound **9a** represent amide (-NH-C=O), C-OCH_3, imine (N=C<), aromatic and methoxy carbons. The peak showed at δ 163.4 belongs to amide carbon. The peaks appearing at δ 145.1 and δ 144.0 belong to C-OCH_3 (C-4,11) carbons and the peaks appeared at δ 136.7, 135.4, and 134.1 belong to (C-4a), imine carbons C-3a and C-10a respectively, while the peaks appeared in the range δ 115.4 and 133.3 correspond to aromatic carbons. The peaks appeared at δ 60.9 and δ 60.3 belong to two methoxy carbons. The peak for $[M+H]^+$ appearing at m/z 298 in the ESI mass spectrum of the compound **9a** confirm the molecular weight. The spectral data accrued from the IR, ^{1}H NMR, ^{13}C NMR and Mass spectral data indicates the formation of the compound **9a** in the reaction.

Conclusion:-

All the reactions were carried out in ethanol in the presence of fused sodium acetate at reflux temperature. Different 3*H*-phenoxazin-3-one **6(a-d)** derivatives are successfully prepared by the condensation of 2,5-dibromo-3,6-dimethoxy-1,4-benzoquinone **4** with substituted *ortho*-aminophenols **5(a-d)**, symmetric [1,4]benzoxazino[2,3-*b*]phenoxazine **7(a-d)** derivatives are successfully obtained by the condensation of 3*H*-phenoxazin-3-ones **6(a-d)** with substituted *ortho*-aminophenols **5(a-d)** and imidazo[4,5-*b*]phenoxazin-2(1*H*)-one derivatives **(9a)** are synthesized by condensation of **6(a-d)** with urea **8**. All the products were obtained in good yields. Structures of new compounds are evidenced by analytical and spectral data.

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