DocID: https://connectjournals.com/03896.2020.20.4719

Biochem. Cell. Arch. Vol. 20, No. 2, pp. 4719-4726, 2020 | www.connectjournals.com/bca | *ISSN 0972-5075*

SYNTHESIS AND CHARACTERIZATION OF TWO MOIETIES OF-1,3-OXAZEPINE-1,5-DIONE COMPOUNDS VIA 4,4'-DIAMINODIPHENYLMETHANE IMINES AS PRECURSORS

Maha Saleh Mahdi and Abdullah Hussein Kshash*

Department of Chemistry, Education College for Pure Science, University of Anbar, Ramadi, Iraq.
*e-mail: drabdullah@uoanbar.edu.iq

(Received 11 May 2020, Revised 30 July 2020, Accepted 9 August 2020)

ABSTRACT : New derivatives of 1,3-oxazepine(1b–7b)(1c–7c) were synthesized by the reaction of Schiff bases (1a–7a) with Succinicand Maleic anhydrideusing dry benzene as a solvent. Schiff bases (1a–7a)were prepared by condensation of 4,4'-Diaminodiphenylmethanewith methoxy and halobenzaldehydeusing absolute ethanol as a solvent in the presence of glacial acetic acid as a catalyst. All synthesized compounds were monitored by TLC and characterized using spectroscopy techniques such as (FT-IR), (^{1}H-NMR) and (^{13}C-NMR), they revealed that all synthesized compounds were conformity of the proposed structures.

Key words : Schiff bases, oxazepine, succinic anhydride, maleic anhydride.

INTRODUCTION

Schiff bases are organic compoundsidentified by the general formula (RR'C=NR'') where R,R' and R'' are alkyl, aryl, cycloalkyl or heterocyclic groups (Brodowska and £odyga-chruscinska, 2014), which have been preparedby the German scientist Hugo Schiff (1864) by condensation reaction of amines and aldehydes or ketones, they were known as Imine and Azomethines compounds (Abirami and Nadaraj, 2014). Schiff bases considered asintermediates compounds in organic synthesis, especially of heterocyclic compounds (Williamson *et al*, 2014; Kapadiya *et al*, 2014; Al-Sultani, 2016; Rasheed *et al*, 2015; Alexander *et al*, 2016), organometallic complexes (Al-Shemary *et al*, 2016) and their applications in analytical chemistry (Cimerman *et al*, 2000), corrosion inhibitors (Mohammed, 2011), plant growth regulators (Ye *et al*, 2007). Most imine and their organometallic complexes exhibit significant biological activities (Arulmurugan and Kavitha, 2010) and pharmacological applications such as analgesic, anti-inflammatory, anthelmintic, anticancer, antimicrobial, antioxidant and antidepressant activities (Hussain *et al*, 2016). Oxazepine compounds are the class of seven-membered heterocyclic organic compounds that contain oxygen, nitrogen, and carbonyl group in the heptane ring (Kareem and Ghanim, 2015; Muslim *et al*, 2018). 1,3-oxazepines were prepared byreaction of imine and carboxylic anhydrides by polar mechanism (Muslim *et al*, 2018). Oxazepines have a wide spectrum of biological activities and pharmacological applications such as anti-convulsant (Suha *et al*, 2015), anti-tumor and colorectal adenocarcinoma (Sunil *et al*, 2014), anti-bacterial (Ahmed *et al*, 2015), anti-oxidant and anti-inflammatory (Alexander *et al*, 2016), besides their uses as corrosion inhibitors (Hamak and Eissa, 2013) and liquid crystal components (Mohammad *et al*, 2016).

EXPERIMENTAL SECTION

2.1 Materials

4,4'-Diaminodiphenylmethane, 2-Methoxy Benzaldehyde, 3- MethoxyBenzaldehyde, 4- Methoxy Benzaldehyde, 3-Chloro Benzaldehyde, 4-Chloro Benzaldehyde, 3-Bromo Benzaldehyde, 4-Bromo Benzaldehyde, Succinicanhydride, and Maleicanhydride were supplied from Sigma-Aldrich, all solvents were supplied from Scharlau and Romal. Melting points were recorded on Electro-Thermal Melting Point Apparatus. FT-IR spectra were recorded on the Bruker-Tensor 27 spectrophotometer in the range of 4000-600 cm^{-1}. ^{1}H-NMR, ^{13}C-NMR spectra were recorded on Bruker-500 MHz spectrometer using DMSO-d_6 as a solvent and TMS as internal standard.

2.2 Synthesis of Schiff Bases1a–7a

A mixture of 4,4'-Diaminodiphenylmethane(10mmol) and substituted Benzaldehyde (20mmol) dissolved in absolute ethanol (50 mL) with 5 drops of glacial acetic

acid as a catalyst, they were placed in the round-bottom flask (100mL) equipped with a condenser and stirring magnetic bar, the mixture refluxed for 3hr then left to cool down to room temperature, the obtainedprecipitate was filtered and recrystallized twice from absolute ethanol (Kshash and Mokhlef, 2017).

2.3 Characterization of Schiff Bases 1a–7a

[1a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(2-methoxyphenyl)methanimine) : Brown gummy, (74% yield), m.p. gummy. IR (ν cm^{-1}):3073 cm^{-1} (C–H aromatic), 3025 cm^{-1} (C–H alkene), 2960-2837 cm^{-1} (C–H aliphatic), 1618 cm^{-1} (C=N), 1593-1485 cm^{-1} (C=C aromatic).^{1}H-NMR(500 MHz, DMSO-d_6): 8.86 (s, C<u>H</u>=N),8.05-7.01 (m, $H_{aromatic}$), 3.93(s, –C<u>H</u>$_2$–), 3.84 (s, –OC<u>H</u>$_3$).

[2a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene)) bis(1-(3-methoxyphenyl)methanimine) : White solid, (72% yield), m.p. 80-82 °C. IR (ν cm^{-1}): 3069 cm^{-1} (C–H aromatic), 3011cm^{-1} (C–H alkene), 2965-2837 cm^{-1} (C–H aliphatic), 1626 cm^{-1} (C=N), 1582-1486 cm^{-1} (C=C aromatic).^{1}H-NMR(500 MHz, DMSO-d_6): 8.58(s, C<u>H</u>=N),7.49-7.09(m, $H_{aromatic}$), 3.99(s, –C<u>H</u>$_2$–), 3.34 (s, –OC<u>H</u>$_3$).

[3a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(4-methoxyphenyl)methanimine) : White solid, (76% yield), m.p. 163-165 °C. IR (ν cm^{-1}): 3076 cm^{-1} (C–H aromatic), 3015 cm^{-1} (C–H alkene), 2965-2838 cm^{-1} (C–H aliphatic), 1620 cm^{-1} (C=N), 1597-1507 cm^{-1} (C=C aromatic).^{1}H-NMR(500 MHz, DMSO-d_6): 8.51 (s, C<u>H</u>=N),7.87-7.05 (m, $H_{aromatic}$), 3.96 (s, –C<u>H</u>$_2$–), 3.32 (s, –OC<u>H</u>$_3$).

[4a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(2-chlorophenyl)methanimine) : White solid, (84% yield), m.p. 121-123 °C. IR (ν cm^{-1}): 3063cm^{-1} (C–H aromatic), 3020 cm^{-1} (C–H alkene), 2989-2910 cm^{-1} (C–H aliphatic), 1616 cm^{-1} (C=N), 1588-1501cm^{-1} (C=C aromatic).^{1}H-NMR(500 MHz, DMSO-d_6): 8.84 (s, C<u>H</u>=N),8.15-7.22 (m, $H_{aromatic}$), 3.99 (s, –C<u>H</u>$_2$–).

[5a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(3-chlorophenyl)methanimine) : Yellow solid, (87% yield), m.p. 130-132 °C. IR (ν cm^{-1}): 3063 cm^{-1} (C–H aromatic), 3025 cm^{-1} (C–H alkene), 2979-2920 cm^{-1} (C–H aliphatic), 1618 cm^{-1} (C=N), 1597-1498 cm^{-1} (C=C aromatic).^{1}H- 500 MHz, DMSO-d_6): 8.62 (s, C<u>H</u>=N),7.95-7.23 (m, $H_{aromatic}$), 3.99 (s, –C<u>H</u>$_2$–).

[6a] (1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(2-bromophenyl) methanimine) : White solid, (80% yield), m.p. 106-108 °C. IR (ν cm^{-1}): 3052 cm^{-1} (C–H aromatic), 3022 cm^{-1} (C–H alkene), 2946-2923 cm^{-1} (C–H aliphatic), 1615 cm^{-1} (C=N), 1588-1500 cm^{-1} (C=C aromatic).^{1}H-NMR(500 MHz, DMSO-d_6): 8.77 (s, CH=N),8.13-7.22 (m, $H_{aromatic}$), 4.00 (s, –CH_2–).

[7a](1*E*,1'*E*)-*N*,*N*'-(methylenebis(4,1-phenylene))bis(1-(3-bromophenyl)methanimine) : Yellow solid, (78% yield), m.p. 126-128°C. IR (ν cm^{-1}): 3053 cm^{-1} (C–H aromatic), 3016 cm^{-1} (C–H alkene), 2942-2919 cm^{-1} (C–H aliphatic), 1625 cm^{-1} (C=N), 1596-1499 cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 8.61(s, C<u>H</u>=N), 8.10-7.21 (m, $H_{aromatic}$), 3.99 (s, –C<u>H</u>$_2$–).

2.4 Synthesis of 1,3-Oxazepines-4,7-dione derivatives

A mixture of Succinicanhydride (20mmol) and Schiff Bases(10mmol) in dry benzene (50 mL) was placed in the round-bottom flask (100mL) equipped with a condenser and stirring magnetic bar, the reaction mixture refluxed for 5hr then stirred overnight at room temperature, the obtained precipitate was filtered, dried and recrystallized twice from chloroform (Kshash, 2020).

2.5 Characterization of 1,3- Oxazepines-4,7-dione derivatives (1b–7b)

[1b] 3,3'-(methylenebis(4,1-phenylene))bis(2-(2-methoxyphenyl)-1,3-oxazepane-4,7-dione) : Yellow solid, (71% yield), m.p. 206-208°C. IR (ν cm^{-1}): 3062cm^{-1} (C–H aromatic), 2999-2843cm^{-1} (C–H aliphatic), 1694 cm^{-1} (C=O lactone), 1655 cm^{-1} (C=O lactam), 1596-1525cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.90 (s, O–C<u>H</u>–N),8.01-7.03(m, $H_{aromatic}$),3.87 (s, –C<u>H</u>$_2$–), 3.81 (s, –OC<u>H</u>$_3$), 2.54-2.51(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.85(<u>C</u>O–O), 169.88(<u>C</u>O–N),159.22-119.13 ($C_{aromatic}$), 119.07 (O–<u>C</u>H–N),55.78 (–O<u>C</u>H_3), 39.96 (–<u>C</u>H_2–), 30.01-28.86(<u>C</u>H_2–<u>C</u>H_2)**.**

[2b]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-methoxyphenyl)-1,3-oxazepane-4,7-dione) : White solid, (75% yield), m.p. 222-224 °C. IR (ν cm^{-1}): 3081cm^{-1} (C–H aromatic), 2989-2835cm^{-1} (C–H aliphatic), 1693 cm^{-1} (C=O lactone), 1661 cm^{-1} (C=O lactam), 1596-1525cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.87 (s, O–C<u>H</u>–N),7.51-7.06(m, $H_{aromatic}$), 3.89 (s, –C<u>H</u>$_2$–), 3.80 (s, –OC<u>H</u>$_3$), 2.53-2.50(m, C<u>H</u>$_2$–C<u>H</u>$_2$)**,** ^{13}C-NMR (500 MHz, DMSO-d_6): 173.84 (<u>C</u>O–O),169.87 (<u>C</u>O–N),154.74-121.12 ($C_{aromatic}$), 119.06 (O–<u>C</u>H–N),55.24 (–O<u>C</u>H_3), 39.95 (–<u>C</u>H_2–), 30.00-28.85(<u>C</u>H_2–<u>C</u>H_2).

[3b]3,3'-(methylenebis(4,1-phenylene))bis(2-(4-methoxyphenyl)-1,3-oxazepane-4,7-dione) : Yellow solid, (70% yield), m.p. 207-209 °C. IR (ν cm^{-1}): 3076cm^{-}

[1] (C–H aromatic), 2999-2840cm^{-1} (C–H aliphatic), 1692 cm^{-1} (C=O lactone), 1660 cm^{-1} (C=O lactam), 1596-1524cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.88 (s, O–C<u>H</u>–N),7.87-7.04(m, $H_{aromatic}$), 3.88 (s, –C<u>H</u>$_2$–), 3.80 (s, –OC<u>H</u>$_3$), 2.53-2.50(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.84 (<u>C</u>O–O),169.86 (<u>C</u>O–N),159.25-119.12 ($C_{aromatic}$), 119.05 (O–<u>C</u>H–N),55.40 (–O<u>C</u>H$_3$),39.95(–<u>C</u>H$_2$–), 30.99-28.84(<u>C</u>H$_2$–<u>C</u>H$_2$).

[4b]3,3'-(methylenebis(4,1-phenylene))bis(2-(2-chlorophenyl)-1,3-oxazepane-4,7-dione) : White solid, (70% yield), m.p. 182-184 °C. IR (ν cm^{-1}): 3064cm^{-1} (C–H aromatic), 2987-2865cm^{-1} (C–H aliphatic), 1694 cm^{-1} (C=O lactone), 1657 cm^{-1} (C=O lactam), 1595-1525cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.85 (s, O–C<u>H</u>–N),8.14-7.08(m, $H_{aromatic}$), 3.99 (s, –C<u>H</u>$_2$–),2.54-2.49(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.86 (<u>C</u>O–O),169.89 (<u>C</u>O–N),155.80-121.30 ($C_{aromatic}$), 119.08 (O–<u>C</u>H–N),39.85 (–<u>C</u>H$_2$–), 30.01-28.86(<u>C</u>H$_2$–<u>C</u>H$_2$).

[5b]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-chlorophenyl)-1,3-oxazepane-4,7-dione) : White solid, (76% yield), m.p. 184-186 °C. IR (ν cm^{-1}): 3064cm^{-1} (C–H aromatic), 2990-2884cm^{-1} (C–H aliphatic), 1693 cm^{-1} (C=O lactone), 1656 cm^{-1} (C=O lactam), 1595-1525cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.85 (s, O–C<u>H</u>–N),7.57-7.08(m, $H_{aromatic}$), 3.99 (s, –C<u>H</u>$_2$–),2.52-2.44(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.86 (<u>C</u>O–O),169.89 (<u>C</u>O–N),158.70-121.32 ($C_{aromatic}$), 119.07 (O–<u>C</u>H–N),39.85 (–<u>C</u>H$_2$–), 30.01-28.86(<u>C</u>H$_2$–<u>C</u>H$_2$)**.**

[6b]3,3'-(methylenebis(4,1-phenylene))bis(2-(2-bromophenyl)-1,3-oxazepane-4,7-dione) : White solid, (63% yield), m.p. 230-232 °C. IR (ν cm^{-1}): 3078cm^{-1} (C–H aromatic), 2990-2920cm^{-1} (C–H aliphatic), 1693 cm^{-1} (C=O lactone), 1653 cm^{-1} (C=O lactam), 1595-1528cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.87 (s, O–C<u>H</u>–N),8.13-7.09(m, $H_{aromatic}$),3.90 (s, –C<u>H</u>$_2$–),2.53-2.50(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.83 (<u>C</u>O–O),169.86 (<u>C</u>O–N),158.05-119.12 ($C_{aromatic}$), 119.05 (O–<u>C</u>H–N),39.85(–<u>C</u>H$_2$–), 30.99-28.84(<u>C</u>H$_2$–<u>C</u>H$_2$).

[7b]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-bromophenyl)-1,3-oxazepane-4,7-dione) : White solid, (67% yield), m.p. 190-192 °C. IR (ν cm^{-1}): 3065cm^{-1} (C–H aromatic), 2997-2908cm^{-1} (C–H aliphatic), 1693 cm^{-1} (C=O lactone), 1655 cm^{-1} (C=O lactam), 1596-1524cm^{-1} (C=C aromatic). ^{1}H-NMR(500 MHz, DMSO-d_6): 9.87 (s, O–C<u>H</u>–N),8.09-7.09(m, $H_{aromatic}$), 3.90(s, –C<u>H</u>$_2$–), 2.53-2.50(m, C<u>H</u>$_2$–C<u>H</u>$_2$), ^{13}C-NMR(500 MHz, DMSO-d_6): 173.82 (<u>C</u>O–O),169.85 (<u>C</u>O–N),158.53-119.12 ($C_{aromatic}$), 119.04 (O–<u>C</u>H–N),39.85 (–<u>C</u>H$_2$–), 30.98-28.83(<u>C</u>H$_2$–<u>C</u>H$_2$).

2.6 Characterization of 1,3- Oxazepines-4,7-dione derivatives (1c-7c).

[1c]3,3'-(methylenebis(4,1-phenylene))bis(2-(2-methoxyphenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (69% yield), m.p. 204-206 °C. IR (ν cm^{-1}): 3082cm^{-1} (C–H aromatic), 3034 cm^{-1} (C–H alkene), 2948-2840cm^{-1} (C–H aliphatic), 1699 cm^{-1} (C=O lactone), 1632 cm^{-1} (C=O lactam), 1575-1505cm^{-1} (C=C aromatic). ^{1}H-NMRspectrum (500 MHz, DMSO-d_6): 10.38 (s, O–C<u>H</u>–N), 7.54-7.14(m, $H_{aromatic}$), 6.47-6.28(m, C<u>H</u>=C<u>H</u>), 3.91 (s, –C<u>H</u>$_2$–), 3.86 (s, –OC<u>H</u>$_3$), ^{13}C-NMR(500 MHz, DMSO-d_6): 166.79 (<u>C</u>O–O),163.08 (<u>C</u>O–N),156.93-122.44 ($C_{aromatic}$), 136.46 and 124.14 (<u>C</u>H=<u>C</u>H), 119.74 (O–<u>C</u>H–N),55.78 (–O<u>C</u>H$_3$), 40.01 (–<u>C</u>H$_2$–).

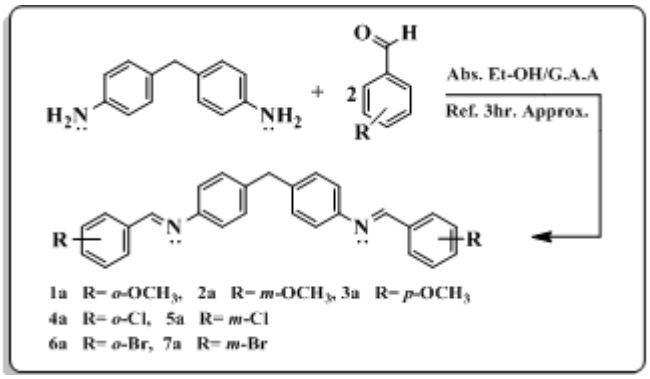
[2c]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-methoxyphenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (78% yield), m.p. 174-176 °C. IR (ν cm^{-1}): 3080cm^{-1} (C–H aromatic), 3022 cm^{-1} (C–H alkene), 2988-2836cm^{-1} (C–H aliphatic), 1699 cm^{-1} (C=O lactone), 1627 cm^{-1} (C=O lactam), 1578-1505cm^{-1} (C=C aromatic). ^{1}H-NMRspectrum (500 MHz, DMSO-d_6): 10.37(s, O–C<u>H</u>–N), 7.56-7.08(m, $H_{aromatic}$), 6.47-6.28(m, C<u>H</u>=C<u>H</u>), 3.92 (s, –C<u>H</u>$_2$–), 3.83 (s, –OC<u>H</u>$_3$), ^{13}C-NMR(500 MHz, DMSO-d_6): 166.77 (<u>C</u>O–O),163.06 (<u>C</u>O–N),159.95-119.76 ($C_{aromatic}$), 136.45and 121.12 (<u>C</u>H=<u>C</u>H), 119.72 (O–<u>C</u>H–N),55.40 (–O<u>C</u>H$_3$), 41.51(–<u>C</u>H$_2$–).

[3c]3,3'-(methylenebis(4,1-phenylene))bis(2-(4-methoxyphenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (70% yield), m.p. 172-174 °C. IR (ν cm^{-1}): 3071cm^{-1} (C–H aromatic), 3041 cm^{-1} (C–H alkene), 2954-2834cm^{-1} (C–H aliphatic), 1698 cm^{-1} (C=O lactone), 1628 cm^{-1} (C=O lactam), 1573-1506cm^{-1} (C=C aromatic). ^{1}H-NMRspectrum (500 MHz, DMSO-d_6): 10.39 (s, O–C<u>H</u>–N), 7.90-7.02(m, $H_{aromatic}$), 6.48-6.29(m, C<u>H</u>=C<u>H</u>), 3.91 (s, –C<u>H</u>$_2$–), 3.82 (s, –OC<u>H</u>$_3$), ^{13}C-NMR(500 MHz, DMSO-d_6): 166.78 (<u>C</u>O–O),163.08 (<u>C</u>O–N),161.82-119.77 ($C_{aromatic}$), 136.47 and 121.02 (<u>C</u>H=<u>C</u>H), 119.73 (O–<u>C</u>H–N),55.39 (–O<u>C</u>H$_3$), 41.52 (–<u>C</u>H$_2$–).

[4c]3,3'-(methylenebis(4,1-phenylene))bis(2-(2-chlorophenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (72% yield), m.p. 190-192 °C. IR (ν cm^{-1}): 3090cm^{-1} (C–H aromatic), 3032 cm^{-1} (C–H alkene), 2979-2880cm^{-1} (C–H aliphatic), 1699 cm^{-1} (C=O lactone), 1631cm^{-1} (C=O lactam), 1507-1443cm^{-1} (C=C aromatic).

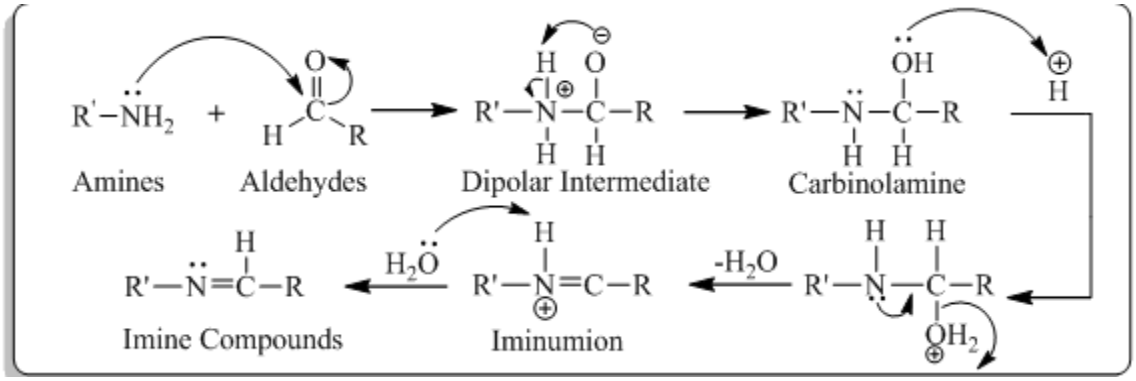
^{1}H-NMR spectrum (500 MHz, DMSO-d_6): 10.39(s, O–C<u>H</u>–N), 8.15-7.16(m, $H_{aromatic}$), 6.48-6.29(m, C<u>H</u>=C<u>H</u>), 3.93 (s, –C<u>H</u>$_2$–),^{13}C-NMR(500 MHz, DMSO-d_6): 166.79 (<u>C</u>O–O),163.12 (<u>C</u>O–N),155.70-119.80 ($C_{aromatic}$), 136.46 and 121.23 (<u>C</u>H=<u>C</u>H), 119.76 (O–<u>C</u>H–N), 40.08 (–<u>C</u>H$_2$–).

[5c]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-chlorophenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (54% yield), m.p. 194-196 °C. IR (ν cm^{-1}): 3070cm^{-1} (C–H aromatic), 3040 cm^{-1} (C–H alkene), 2983-2875cm^{-1} (C–H aliphatic), 1722 cm^{-1} (C=O lactone), 1626 cm^{-1} (C=O lactam), 1567-1500cm^{-1} (C=C aromatic). ^{1}H-NMR spectrum (500 MHz, DMSO-d_6): 10.39 (s, O–C<u>H</u>–N), 7.95-7.12(m, $H_{aromatic}$), 6.48-6.29(m, C<u>H</u>=C<u>H</u>), 3.93 (s, –C<u>H</u>$_2$–),^{13}C-NMR(500 MHz, DMSO-d_6): 166.77 (<u>C</u>O–O),163.08 (<u>C</u>O–N),158.63-119.77 ($C_{aromatic}$), 136.51 and 121.25 (<u>C</u>H=<u>C</u>H), 119.73 (O–<u>C</u>H–N),40.06 (–<u>C</u>H$_2$–).

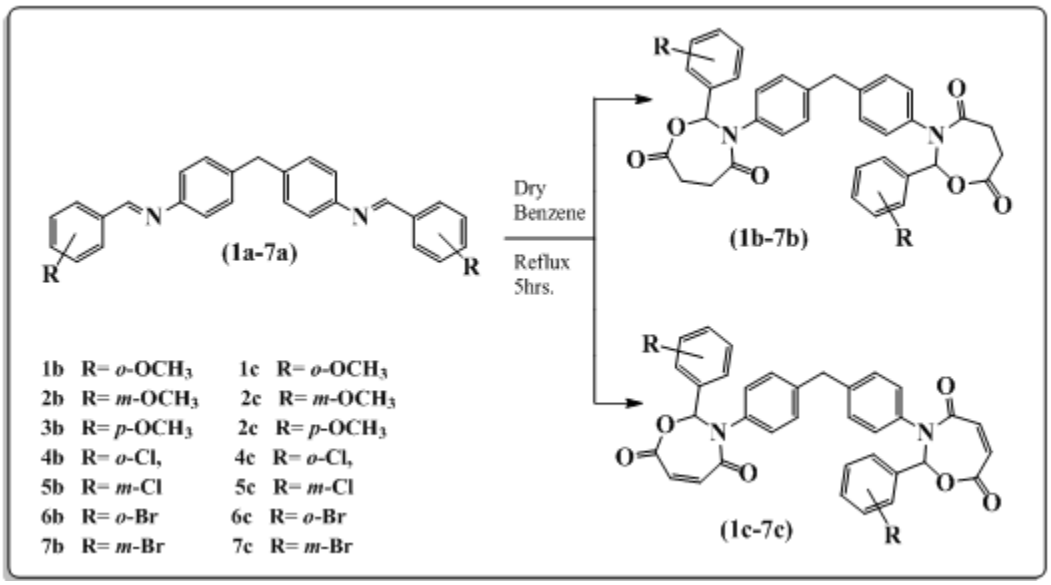


Scheme 1 : Synthetic route for synthesized Schiff Bases (1a-7a).

[6c]3,3'-(methylenebis(4,1-phenylene))bis(2-(2-bromophenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (62% yield), m.p. 191-193 °C. IR (ν cm^{-1}): 3071cm^{-1} (C–H aromatic), 3041 cm^{-1} (C–H alkene), 2983-2862cm^{-1} (C–H aliphatic), 1722 cm^{-1} (C=O lactone), 1628 cm^{-1} (C=O lactam), 1574-1501cm^{-1} (C=C aromatic). ^{1}H-NMR spectrum (500 MHz, DMSO-d_6): 10.39 (s, O–C<u>H</u>–N), 7.59-7.12(m, $H_{aromatic}$), 6.48-6.27(m, C<u>H</u>=C<u>H</u>), 3.93 (s, –C<u>H</u>$_2$–),^{13}C-NMR(500 MHz, DMSO-d_6): 166.81 (<u>C</u>O–O), 163.12 (<u>C</u>O–N),150.51-119.78 ($C_{aromatic}$), 136.48 and 121.22 (<u>C</u>H=<u>C</u>H), 119.19 (O–<u>C</u>H–N),10.02 (–<u>C</u>H$_2$–).



Scheme 2 : Proposed mechanism for the formation of azomethine compounds.



Scheme 3 : Synthetic route for synthesized Oxazepines compounds.

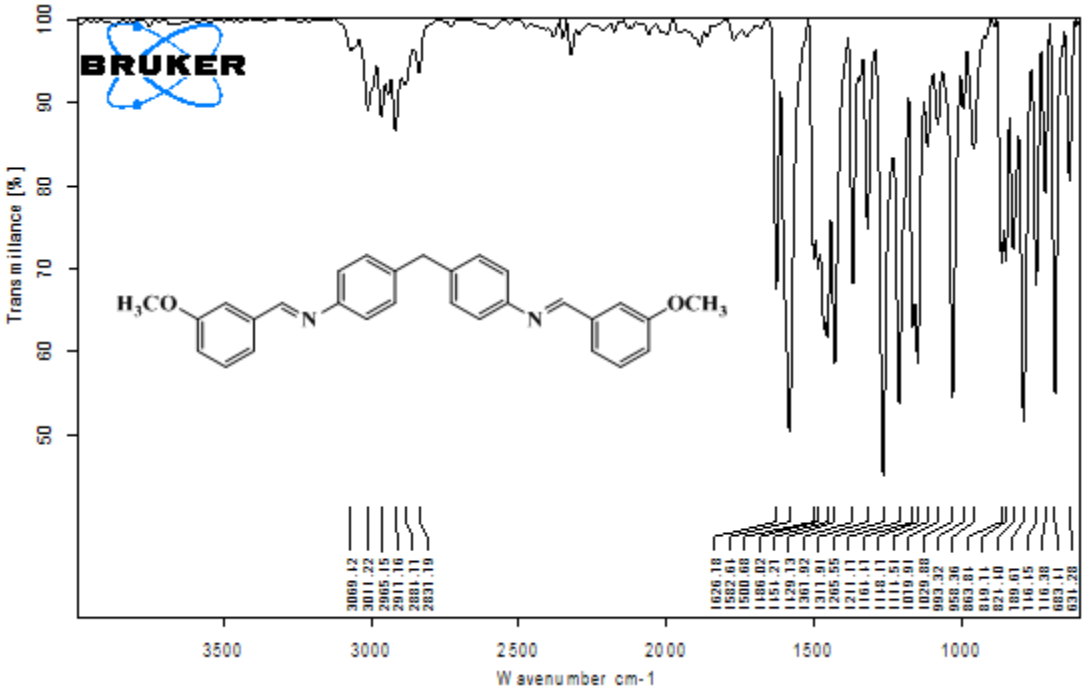


Fig. 1 : FT-IR spectra for (2a) compound as illustration.

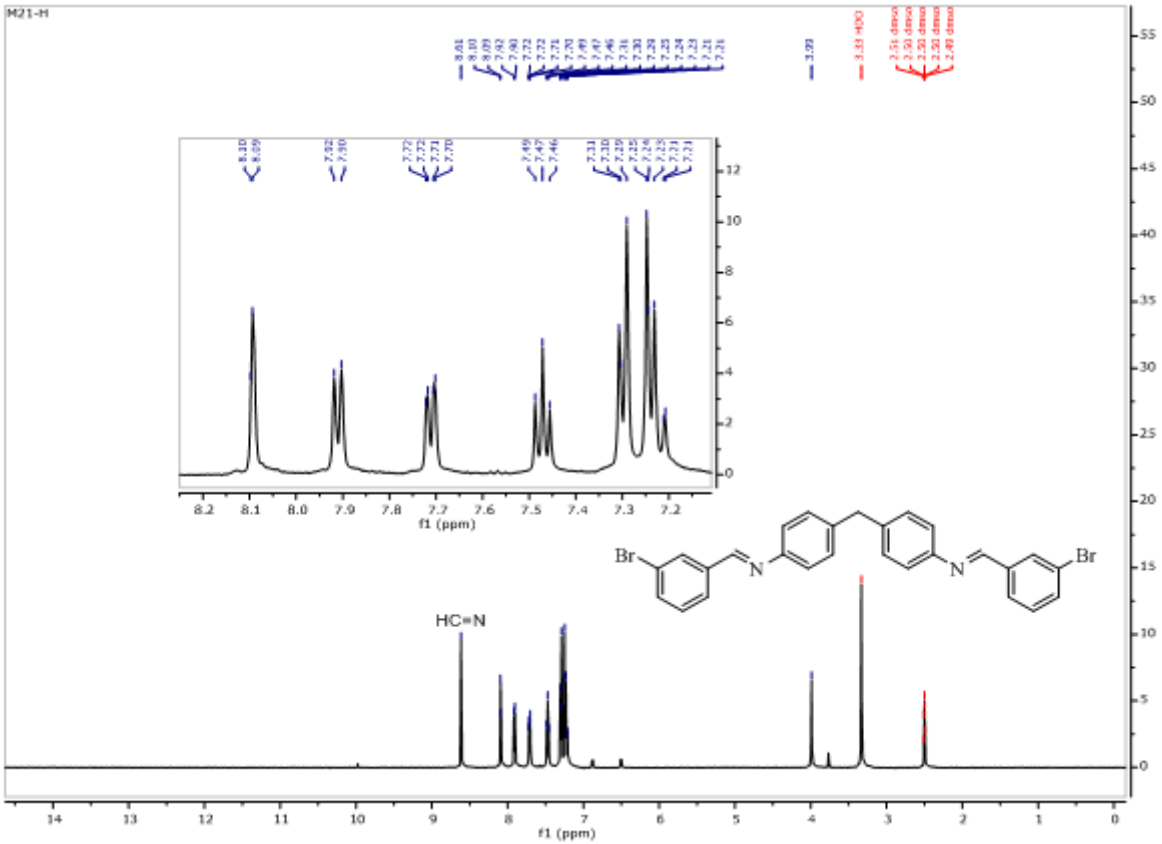


Fig. 2 : ^{1}H-NMR spectra for Compound (7a).

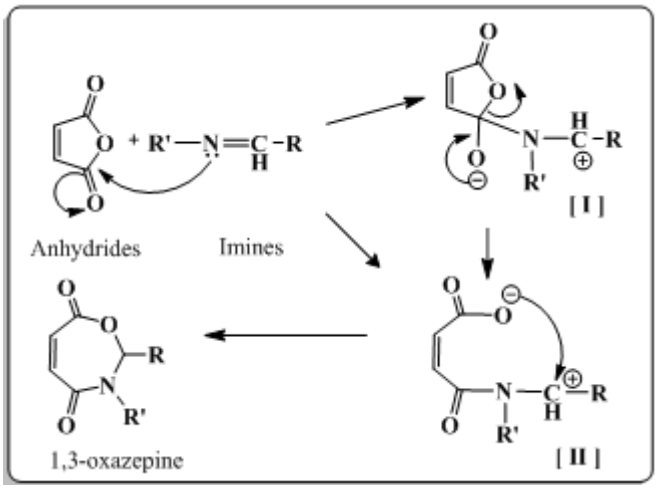
[7c]3,3'-(methylenebis(4,1-phenylene))bis(2-(3-bromophenyl)-2,3-dihydro-1,3-oxazepine-4,7-dione) : Yellow solid, (60% yield), m.p. 192-194°C. IR (ν cm^{-1}): 3069cm^{-1} (C–H aromatic), 3038 cm^{-1} (C–H alkene), 2983-2880cm^{-1} (C–H aliphatic), 1721cm^{-1} (C=O lactone), 1625 cm^{-1} (C=O lactam), 1545-1499cm^{-1} (C=C aromatic). ^{1}H-NMRspectrum (500 MHz, DMSO-d_6): 10.39 (s, O–C<u>H</u>–N), 8.09-7.12(m, $H_{aromatic}$), 6.48-6.29(m, C<u>H</u>=C<u>H</u>), 3.92 (s, –C<u>H</u>$_2$–),^{13}C-NMR(500 MHz, DMSO-d_6): 166.78 (<u>C</u>O–O),163.09 (<u>C</u>O–N),158.53-

119.78 ($C_{aromatic}$), 136.51 and 121.26 (CH=CH), 119.74 (O–CH–N),40.02 (–CH_2–).

RESULTS AND DISCUSSION

Schiff Bases (1a-7a) compounds were prepared by the condensation reaction of 4,4'-Diaminodiphenylmethane with methoxy and halo benzaldehyde using absolute ethanol as a solvent and glacial acetic acid as a catalyst (Scheme 1).

The formation mechanism of Schiff Bases explained by literature and suggested to be occured via nucleophilic addition of the amino group to the carbonyl group to forms a hemiaminal, followed by dehydration to produce the Schiff Bases (Muzammil *et al*, 2015) as shown in Scheme 2.



Scheme 4 : The mechanism formation of 1,3-oxazepine derivatives.

The FT-IR spectra for prepared Schiff bases showed that the disappearance of the stretching frequency absorption bands of (-NH_2) group at (3530-3225cm^{-1}) and (C=O) group at (1720-1700cm^{-1}) in amines and aldehydes respectively and the appearance of the aromatic (C-H) at (3076-3052 cm^{-1}), (=C-H) at (3025-3011 cm^{-1}), aliphatic (C-H) at (2989-2837 cm^{-1}), imine groups (C=N) at (1626-1615 cm^{-1}) and (C=C) of the aromatic ring at (1597-1485 cm^{-1}) (Silverstein *et al*, 2005).

The ^{1}H-NMR spectra of the prepared Schiff Bases showed signals of the following groups at δ ppm range: (=C-H) at range (2H, s, 8.86-8.51), aromatic protons at range (16H, m, 8.15-7.01), -CH_2- at range (2H, s, 4.00-3.93), Ar-OCH_3- at range (3H, s, 3.84-3.32) (Timothy, 2009).

1,3-oxazepine derivatives have been synthesized by reaction of Schiff Bases with Succinic and Maleic anhydride as shown in Scheme 3.

The reaction is take place by nucleophilic attack of nitrogen atomfor azomethine group on the electrophilic carbonyl group of the cyclic anhydride to give a dipolar intermediate [I], which give intermediate [II], which undergoes internal cyclization to give the target molecule (Scheme 4) (Ahmed *et al*, 2015).

FT-IR spectra for 1,3-Oxazepine derivatives showed that the disappearance of stretching absorption bands of (-C=N) group at (1626-1615 cm^{-1}) and that of (C=O) group at (1825-1780cm^{-1}) for Schiff Bases and carboxylic anhydrides respectively and appearance of the stretching absorption bands of aromatic (C-H) at (3044-3041 cm^{-}

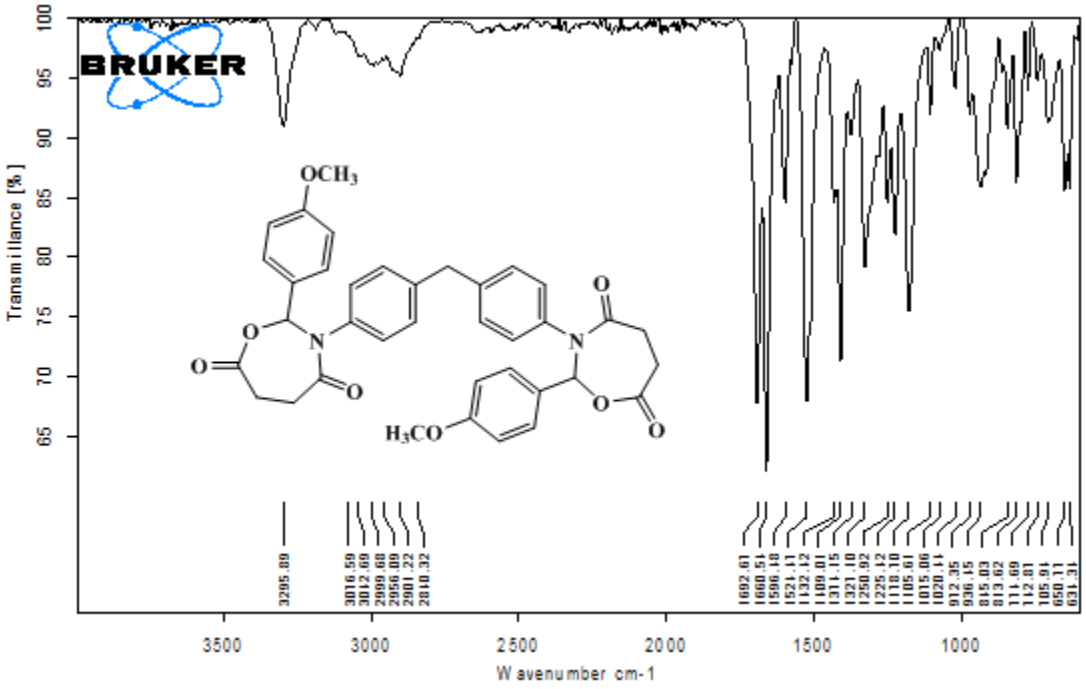


Fig. 3 : FT-IR spectra for (3b) compound as illustration.

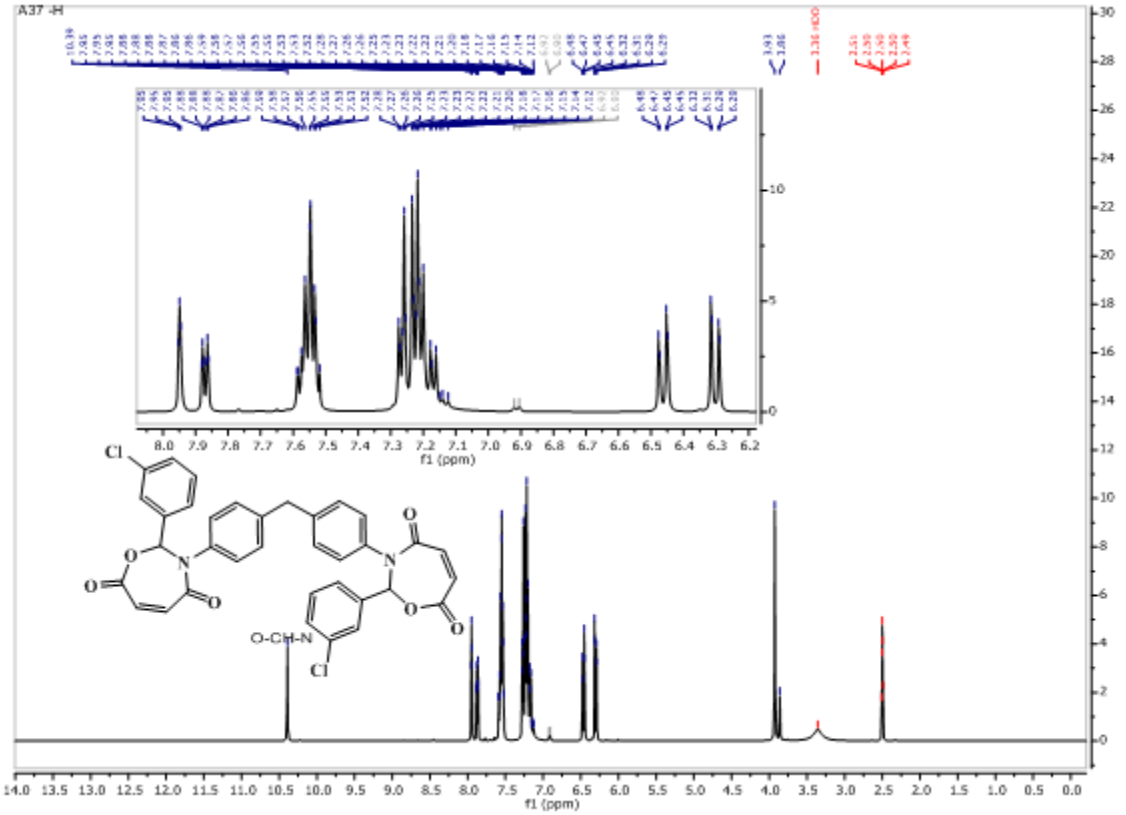


Fig. 4 : ^{1}H-NMR spectra for Compound (5c).

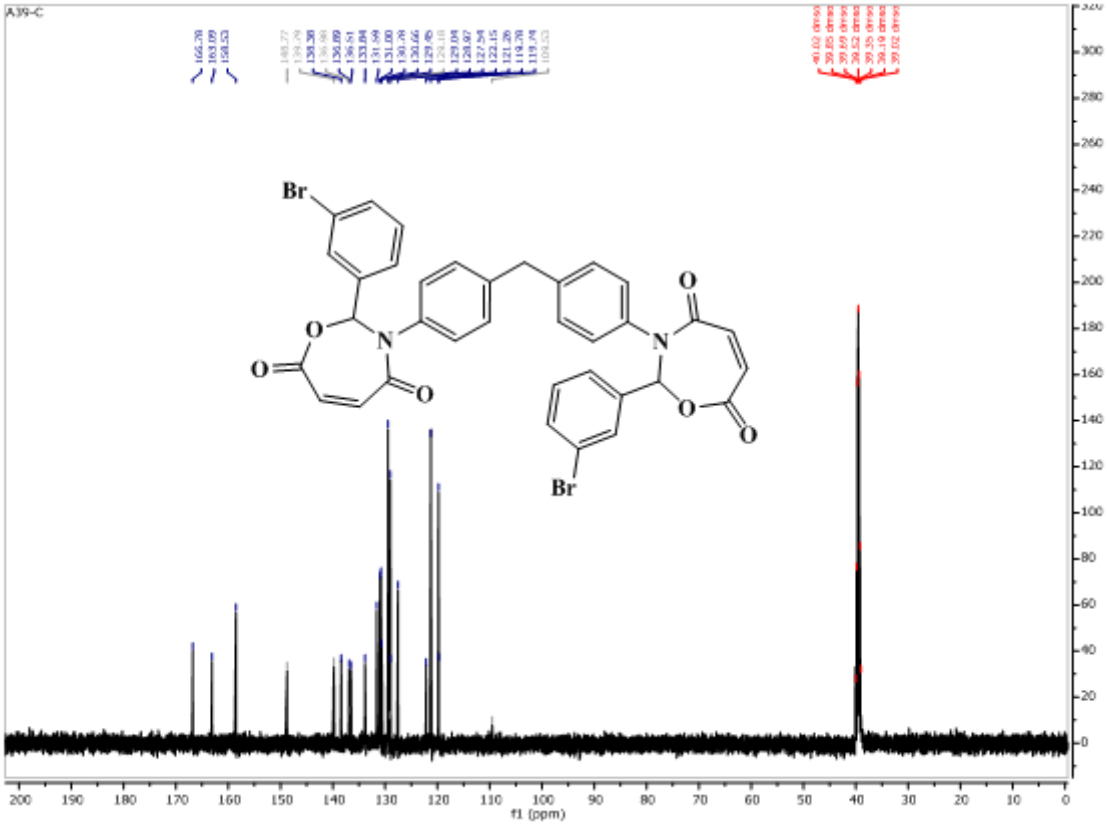


Fig. 5 : ^{13}C-NMR spectra for Compound (7c).

[1]), (=C-H) at (3041-3022 cm^{-1}) aliphatic, (C-H) at (2999-2834 cm^{-1}), lactone (C=O) at (1722-1629 cm^{-1}), lactam (C=O) at (1661-1625 cm^{-1}), aromatic ring (C=C) at (1596-1443 cm^{-1}) (Silverstein *et al*, 2005). Figure (3) FT-IR spectra for (3b) compound as illustration.

^{1}H-NMR spectra of the prepared 1,3-oxazepine derivatives showed signals of the following groups at δ ppm range: O-CH-N at range (2H,s, 10.39-9.85), aromatic protons at range (16H, m,8.15-7.02),CH=CH at range (4H, d,6.27-6.48),-CH_2- at range (1H, s, 3.99-3.87), Ar-OCH_3 at range (6H, s,3.86-3.80), CH_2-CH_2 at range (2H, s,2.54-2.44) (Timothy, 2009). Figure (4)^{1} H-NMR spectra for(5c)compound as illustration.

^{13}C-NMR spectra of the prepared 1,3-oxazepine derivatives showed signals of the following groups at δ ppm range: O-(CO) at range (2C, 173.86-166.77), N-(CO) at range (2C, 169.89-163.06), aromatic carbons at range (24C, 161.82-119.12),CH=CH at range (4C, 136.51-136.45 and 124.14-121.02), O-CH-N at range (2C, 119.76-119.04), -CH_2- at range (2C, 41.52-39.85),Ar-OCH_3 at range (2C, 55.78-55.24), CH_2-CH_2 at range (8H, 31.01-28.80) (BalciM, Basic 1H- And 13C-NMR Spectroscopy, 2004). Figure (5)^{13}C-NMR spectra for(7c)compound as illustration.

CONCLUSION

Two moieties of 1,3-oxazepines were successfully synthesized in the same molecule via reactions of Schiff bases as a precursor with Succinic and Maleic anhydride, all reactions have been monitored by TLC and synthesized compounds identified byFT-IR, ^{1}H.NMR and ^{13}C.NMR spectral.

REFERENCES

Abirami M and Nadaraj V (2014) Synthesis of Schiff Base under Solvent-free Condition: As a Green Approach. *IJ Chem. Tech. Res.* **6**(4), 2534-2538.

Ahmed A, Mahdi S, Hussein A, Hamed A and Yousif E (2015) Antibacterial Study of Some Oxazepine Derivatives. *J. Al-Nahrain Univ.* **18**(4), 22-26.

Alexander A K, Joseph L and George M (2016) Synthesis of Novel 5, 6-Benz 1, 3 -Oxazepine 4, 7-Dione Derivatives and Screeninc For AntibacterialL, Antixidant and Anti-inflammatory Activities. *Europ. J. Pharmaceut. Med. Res.* **3**(7), 330-336.

Al-Shemary R K, Al-Khazraji A M A and Niseaf A N (2016) Preparation, spectroscopic study of Schiff base ligand complexes with some metal ions and Evaluation of antibacterial activity. *The Pharma Innovation J.* **5**(1), 81-86.

Al-Sultani K T A (2016) Synthesis, Identification and Evaluation The Biological Activity For Some New Heterocyclic Compounds Derived From Schiff Bases. *IOSR-JAC* **9**(5), 01-11.

Arulmurugan S, Kavitha H P and Venkatraman B R (2010) Biological Activities of Schiff Base and its Complexes: A Review. *RJC* **3**(3), 385-410.

Brodowska K and £odyga-chruscinska E (2014) Schiff bases-interesting range of applications in various fields of science. *CHEMK* **68**(2), 129-134.

Cimerman Z, Miljani S and Gali N (2000) Schiff Bases Derived from Aminopyridines as Spectrofluorimetric Analytical Reagents. *CCACAA* **73**(1), 81-95.

Hamak K F and Eissa H H (2013) Synthesis, Characterization, Biological Evaluation and Anti Corrosion Activity of Some Heterocyclic Compounds Oxazepine Derivatives from Schiff Bases. *Organic Chem. Curr. Res.* **2**(3), 1-7.

Hussain Z, Fadhil Z, Adil H, Khalaf M, Abdullah B and Yousif E (2016) Schiff's Bases Containing Sulphamethoxazole Nucleus. *RJPBCS* **7**(3), 1500-1510.

Kapadiya K M, Namera D H, Kavadia K M and Khunt R C (2014) Synthesis of benzthiazole derivatives grouping with substituted azetidinone ring and its functional behavior. *ILCPA* **30**, 223-232.

Kareem A F and Ghanim H T (2015) Synthesis and Identification some of 1,3-Oxazepine Derivatives. *JAPBR* **5**(1), 45-56.

Kshash A H and Mokhlef M G (2017) Synthesis, Characterization and DFT Study of 4,42 -Oxydianiline Imines as Precursors of Tetrahalo-1,3-oxazepine-1,5-dione, Indones. *J. Chem.* **17**(2), 330-335.

Kshash A H (2020) Synthesis and characterization of tetrachloro-1,3-oxazepine derivatives and evaluation of their biological activities. *Acta Chimica Slovenica* **67** (1), 113–118.

Mohammed M Q (2011) Synthesis and characterization of new Schiff bases and evaluation as Corrosion inhibitors. *J. Basrah Researches* **37**(4A), 116-130.

Muslim R F, Tawfeeq H M, Owaid M N and Abid O H (2018) Synthesis, characterization and evaluation of antifungal activity of seven-membered heterocycles. *Acta Pharmaceutica Sciencia* **56** (2), 39–57.

Mohammad A T, Yeap G Y and Osman H (2016) Smectogenic and nematogenic liquid crystals of a new series of heterocyclic derivatives bearing an ester terminal chain: Synthesis, characterization, and theoretical study. *Mole. Crystals and Liquid Crystals* **630**, 44-57.

Muzammil K, Trivedi P and Khetani D (2015) Synthesis and Characterization of Schiff base m-nitro aniline and their complexes. *Research J. Chem. Sci*. **5**(5), 52-55.

Silverstein R M, Webster F X and Kiemle D J (2005) *Spectrometric Identification of Organic compounds*. 7th Ed, John- Wiley & Sons, INC., pp. 72-126, (2005).

Suha M, Nerkar A G, Chikhale H U and Sawant D S (2015) *In Silico* Screening, Synthesis and Pharmacological Screening of Quinazolinones Containing Oxazepinone Ring as NMDA Receptor Antagonists for Anticonvulsant Activity. *J. Young Pharmacists* **7**(1), 21-27.

Sunil D, Ranjitha C, Rama M and Pai K S R (2014) Oxazepine Derivative as an Antitumor Agent and Snail1 Inhibitor against Human Colorectal Adenocarcinoma. *Int. J. Innovative Res. in Science Engineering and Tech.* **3**(8), 15357-15363.

Timothy D W Claridge (2009) High-Resolution NMR Techniques In Organic Chemistry, 2th Ed , Elsevier.

Williamson K S, Michaelis D J and Yoon T P (2014) Advances in the Chemistry of Oxaziridines. *Chem. Rev.* **114**, 8016-8036.

X, Chen Z, Zhang A and Zhang L (2007) Synthesis and Biological Evaluation of Some Novel Schiff's Bases from 1,2,4-Triazole. *Molecules* **12**, 1202-1209.

Ye Rasheed M K, AL-Hiti W F and Rabei S M (2015) Synthesis and Characterization of Some 1,3-Oxazine -6-One , 1,3-Oxazine -6,6-Dione and N-Bromo Amines Derivatives. *J. Applicable Chem.* **4**(6), 1725-1731.