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Synthesis and Characterization of Some 1,3-Oxazine -6-One , 1,3-Oxazine -6,6-Dione and N-Bromo Amines Derivatives

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ABSTRACT

This study includes synthesis and characterization of new derivatives of 1,3-Oxazine-6-one, 1,3-Oxazine-6,6-dione and N-Bromo amines, via Schiff's bases reactions through one step process in inert solvents. Some different Schiff bases [A3,A4, A7, A10] synthesized from reaction of different amines with aldehydes such as (Salicylaldehyde, Glutaraldehyde) in absolute ethanol or methanol under reflux. Heterocyclic rings of the 1,3-oxazine-6-one derivatives prepared by the reaction of 3-chloropropanoic acid with Schiff's bases[A3,A4, A7, A10] in dry benzene. The 1,3-oxazine-6,6-dione derivatives prepared by the reaction of two moles of 3-Chloropropanoic Acid with Schiff bases [A3,A4, A7, A10] by using dry benzene as solvent, Synthesis of some N-Bromo amine derivatives by the reaction of Schiff's bases with 2,4,4,6-TBCD (2,4,4,6-tetrabromocyclohexa-2,5-dienone) in dry benzene, The prepared compounds were characterized by melting point , FT-IR , UV-Vis and ^{1}H- NMR spectra.

Keywords: Schiff bases, 1,3-Oxazine -6-one, 1,3-Oxazine -6,6-dione N-bromo amines derivatives.

INTRODUCTION

Schiff's Bases act important intermediate compounds in the preparation of some of the biological activity Compounds such as (β-Lactams) and Heterocyclic Compounds [1-4] as well as pharmaceutical materials, anti-bacterial [5,6], anticancer [7-10] and some of which are effective against cardiovascular cramps and others have effective anti-TB. [11]. Oxazines are six heterocyclic compounds contain nitrogen and oxygen atoms in the same ring, and use in organic synthesis as starting material. Oxazines have biological activity [12] and broad industrial applications such as pigments and pharmaceutical industry, antimicrobial, antiphlogistic [13], antineoplastic [14], anti TB. [15], antibacterial [16], antifungal [17], antimalarial [18], enzyme inhibitors, analgesic and antidepressant [19– 1]. N-bromo compounds have bromine atom attached to nitrogen and have much applications as antibacterial, antifungal and anti HIV [22-27].

MATERIALS AND METHODS

Melting points were recorded with (Stuart) 30 Melting point Apparatus and were uncorrected, UV-Visible spectra were recorded with Schimadsu (UV-1800) spectrophotometer Infrared spectra were recorded as KBr pellets on a Thermo-Fisher spectrometer. ^{1}H-NMR spectra were recorded on Bruker-500 MHz Spectrometer using DMSO $-d_6$ as a solvent and TMS (Tetramethylsilane $(CH_3)_4Si$) as internal standard.

Synthesis of Salicylaldehydyde Schiff bases [1-6]: A Solution of (0.01 mol) of (Ethylenediamine**,** P-phenylenediamine) in (40 mL) absolute ethanol was added to (0.02mol) salicylaldehyde in (20 mL) absolute ethanol then the mixture was refluxed for 2h, the mixture was cooled to room temperature, filtered , dried and recrystallized from absolute ethanol [28], physical properties are given in table 1.

Synthesis of glutaraldehyde Schiff base[1-6]: Schiff base were synthesized by the condensation of (0.01 mol) of glutaraldehyde and (0.02 mol) of (2-Aminopyridine, 2-aminobenzoic acid) (1:2 mol ratio), in 100 mL of absolute ethanol, the mixture was refluxed for 2 h. Yellow solid precipitate of Schiff base obtained was filtered, washed with distilled water dried, recrystallized and finally preserved in a desiccators[28].

Synthesis of Heterocyclic Compounds

Synthesis of 1,3-Oxazine -6-one Derivatives (B3, B4, B7, B10): A mixture (0.01 mol) of Schiff bases (A3, A4, A7, A10) with (0.01 mol, 1.085 g) of (3-Chloropropanoic acid) in (20 mL) dry benzene and two drops of (Ammonia), the mixture was refluxed for 6h,the solvent was evaporated then the formed precipitate was recrystallized from absolute ethanol physical properties are given in table3 [29].

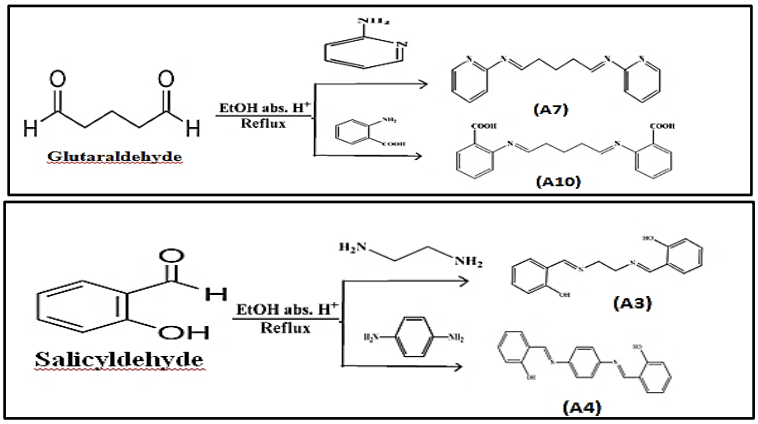
Synthesis of 1,3-Oxazine -6,6-dione Derivatives (C3,C4, C7, C10): A mixture (0.01mol) of Schiff bases (A3,A4, A7, A10) with (0.02 mol, 2.17g) of (3-Chloropropanoic acid) in (20mL) of dry benzene and two drops of (Ammonia), the mixture was refluxed for(6h). Then the solvent evaporated and then formed precipitate was re-crystallized from, absolute ethanol, physical properties are given in table 5 [29].

3- Synthesis of 2,4,4,6_Tetrabromo_2,5_Cyclohexadinone (2,4,4,6-TBCD): A mixture (0.02 mol, 1.88 g) of (phenol) and (0.06 mol, 6.714g) of (potassium bromide) with (0.03 mol, 4.797g) (potassium bromate) in (60 mL) of distilled water and then added to the mixture slowly (8.7 mL) of hydrochloric acid (36%) for 2 h after it was mix move and refluxed for two hours, then the precipitate was filtered and washed with distilled water , physical properties are given bellow. m.p. =121-124 , FT-IR:C=C 1581 cm^{-1}, =C-H 3050 cm^{-1}, C-Br 683-702 cm^{-1}, C=H 1381 cm^{-1}, C=0 1679 cm^{-1}.

Synthesis of N-bromo amines Derivatives (D3, D4, D7, D10): A Solution of (0.01 mol) of the compound 2,4,4,6- tetra bromo-2,5- Cyclohexadinone in (20 mL) of dry benzene and then added to a small amount of tri-aluminum chloride ($AlCl_3$) in (100 mL) round bottom flask equipped with magnetic stirrer and condenser and the mixture was refluxed for 15 min, equivalent moles of 1,3-Oxazine -6-one Derivatives (B3, B4, B7, B10) the same solvent were added to the mix and refluxed for 5 h. Then cold in the ice bath [29] the colored crystals of derivatives (D3, D4, D7, D10) filtered and washed with distilled water.

RESULTS AND DISCUSSION

Schiff bases prepared by the reaction of some aldehydes (aliphatic and aromatic) with diamine in absolute ethanol [30-33] and is shown in scheme 1.



Scheme 1

The prepared compounds were characterized by melting point, Physical properties are given in table 1.

Table 1. Physical properties of Schiff bases [A3,A4,A7,A10]

Comp. symb.	Molecular Formula	Colour	yield %	M.P ^{o}C	M. Wt.
A3	$C_{16}H_{16}O_2N_2$	yellow	81	124-126	268
A4	$C_{20}H_{16}O_2N_2$	Light Orange	83	210-213	316
A7	$C_{18}H_{20}O_2N_4$	White	70	154-157	324
A10	$C_{19}H_{18}O_4N_2$	Adoption	89	144-147	388

The FT-IR spectrum of Schiff bases showed the disappearance of bands at (3309 -3413 cm^{-1}) for amino group, and appear of bands at (3008 –3085) cm^{-1} for benzene ring, at (2909-2989 cm^{-1}) for methylene groups, at (1236-1284) cm^{-1} for (C-N), at (1496–1573) cm^{-1} for (C=C) aromatic ring, FT-IR wave numbers are given in the table 2 .

Table 2. FT-IR spectrum data of Schiff bases [A3,A4,A7,A10]cm^{-1}

Comp symb.	ν O-H	ν N-H Amine	ν N=C-H	ν C-H Aromatic	ν C-H Aliphatic	ν C=N Imine	ν C-N	ν C=C Aromatic
A3	3475	3413	3135	3054	2909	1627	1284	1496
A4	3475	3413	3108	3008	2931	1616	1280	1573
A7	3432	3309	3189	3085	2989	1696	1268	1505
A10	3471	-	3162	3062	2911	1573	1236	1484

1,3-Oxazine-6-one compound compounds [B3, B4, B7, B10] prepared by reaction of 3-Chloropropanoic Acid compound with Schiff bases (A3,A4, A7, A10) by using dry benzene as a solvent and ammonia. FT-IR spectrums showed bands at (3417-3471) cm^{-1} for((N-H), at (3008 –3143) cm^{-1} for benzene ring, at (1616-1677)cm^{-1} for (C=O) lactone and lactam compounds, at (1523) cm^{-1} for (C=N) aromatic ring besides other at (1184-1284) cm^{-1} for (C-N) and (1460–1612) cm^{-1} for (C=C) aromatic ring.
Physical properties are given in table 3.

Table3. Physical properties 1,3-oxazinan-6-one compounds [B3,B4,B7,B10]

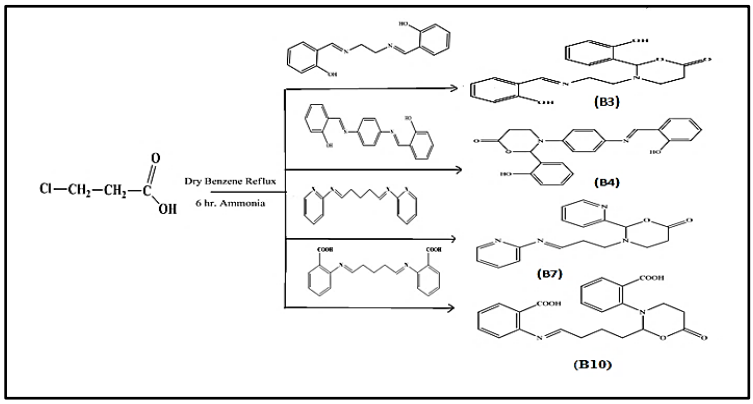
Comp. symb.	Molecular Formula	Colour	Yield %	M.P^{0}C	M.Wt
B3	$C_{19}H_{20}O_4N_2$	yellow	65	126-128	340
B4	$C_{23}H_{20}O_4N_2$	Dark Orange	84	142-144	388
B7	$C_{18}H_{20}O_2N_4$	yellow	87	112-114	324
B10	$C_{22}H_{22}O_4N_2$	greyish	95	112-115	378

FT-IR and UV-Vis spectrum data are given in table 4.

Table 4. FT-IR and UV-Vis spectrum data for 1,3-oxazinan-6-one compounds [B3,B4,B7,B10] cm^{-1}

Comp. symb.	ν N-H Amine	ν C-H Aromatic	ν C-H Aliphatic	ν C=O Lactone	ν C=C Arom.	ν C=N Arom.	ν C-N	λ_{max1} THF	λ_{max2} THF
B_3	3428	3008	2987	1627	1581	-	1284	405	200
B_4	3417	3062	2927	1616	1477	-	1272	330	205
B_7	3447	3074	2953	1646	1585	1523	1184	267	210
B_{10}	3471	3143	2933	1677	1492	-	1241	335	217

Scheme2 illustrates prepared B3, B4, B7, B10 compounds structures.



Scheme 2. Structure for prepared B3, B4, B7, B10 compounds

1,3-Oxazine 6,6- di one compound [C3,C4, C7, C10] prepared by reaction of two moles of 3-Chloropropanoic Acid with Schiff bases [A3,A4, A7, A10] by using dry benzene as a solvent and ammonia. Physical properties are given in table 5.

Table 5. Physical properties 1,3-oxazinan-6,6-Dione compounds[C3,C4,C7,C10]

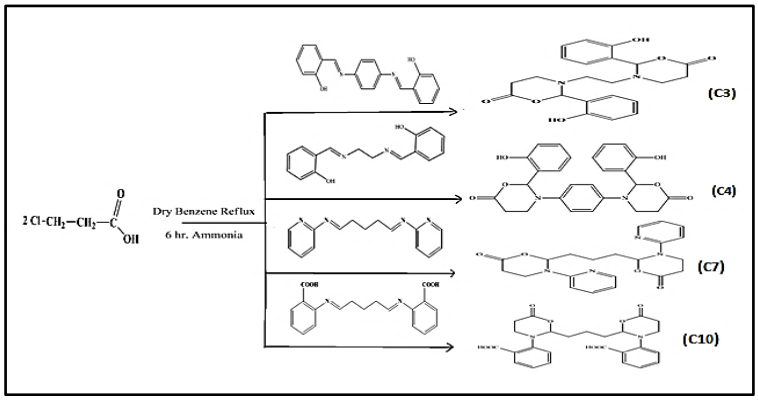
Comp. symb.	Molecular Formula	Colour	Yield %	M.P^{0}C	M.Wt
C3	$C_{22}H_{24}O_6N_2$	nutty	67	102-104	412
C4	$C_{26}H_{24}O_6N_2$	Dark Orange	61	194-196	460
C7	$C_{21}H_{24}O_4N_4$	nutty	81	104-107	396
C10	$C_{25}H_{26}O_6N_2$	Black	64	120-123	450

FT-IR spectrums showed bands at (1261-1287) cm^{-1} for (C-N), at (3413-3459) cm^{-1} for ((N-H)), at (3008 –3174) cm^{-1} for benzene ring, at (1581-1650) cm^{-1} for (C=O) for lactone and lactam compounds, at (2927–2987cm^{-1}) for (C-H) in methyl group, at (1527) cm^{-1} for (C=N) aromatic ring besides other at (1456–1511) cm^{-1} for (C=C) aromatic ring, FT-IR and UV-Vis spectrum data are given in the table 6.

Table 6. FT-IR and UV-Vis spectrum data for 1,3-oxazinan-6,6-Dione compounds [C3,C4,C7,C10] cm^{-1}

Comp. symb.	νs N-H Amine	ν_s C-H Aromatic	ν_s C-H Aliphatic	ν_s C=O Lacton	ν_s C=C Arom.	νs C=N Arom.	νs C-N	λ_{max1} THF	λ_{max2} THF
C_3	3413	3008	2927	1581	1500	-	1284	-	-
C_4	3432	3046	2923	1643	1508	-	1276	369	226
C_7	3459	3174	2945	1650	1456	1527	1287	315	214
C_{10}	3459	3128	2958	1646	1511	-	1261	355	216

Scheme3 illustrates prepared C3, C4, C7, C10 compounds structures



Scheme 3. Structure for prepared C3,C4, C7, C10compounds

N-bromoamine compounds [D3, D4, D7, D10] prepared by reaction of 2,4,4,6-Tetrabromo-2,5-cyclohexadienone with 1,3-Oxazine -6-one Derivatives using benzene as a solvent and $AlCl_3$ as a catalyst. Physical properties are given in table 7.

Table 7. Physical properties N-Bromoamine compounds[D3,D4,D7,D10]

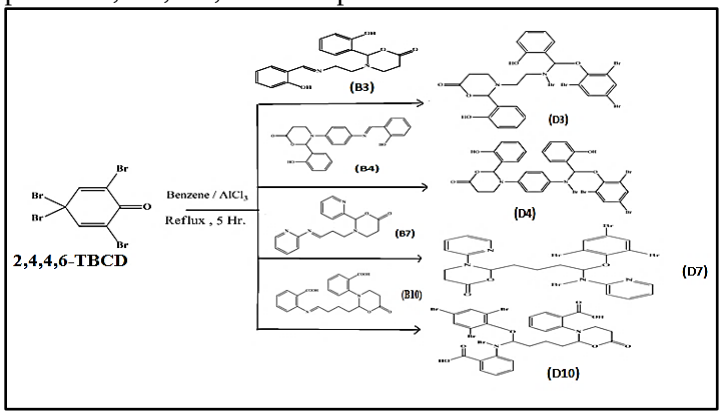
Comp. symb.	Molecular Formula	Colour	Yield %	M.P ^oC	M.Wt
D3	$C_{25}H_{22}O_5N_2Br_4$	nutty	67	118-121	721.616
D4	$C_{29}H_{22}O_5N_2Br_4$	Dark nutty	61	206-208	769.616
D7	$C_{24}H_{22}O_3N_4Br_4$	Light nutty	81	127-130	705.616
D10	$C_{26}H_{24}O_7N_2Br_4$	Leaden	64	102-105	791.616

FT-IR spectrums showed bands at (1118-1184)cm^{-1} for (C-O-C), at (3405-3421) cm^{-1}, at (3139-3158) cm^{-1} for benzene ring, at (1623-1747)cm^{-1} for (C=O)for lactone and lactam compounds, at (2927–2973cm^{-1}) for (C-H) in methyl group, at (1646) cm^{-1} for (C=N) aromatic ring, besides other at (1508-1569) cm^{-1} for (C=C) aromatic ring **[28,26]** . FT-IR and UV-Vis spectrum data are given in the table 8.

Table 8. FT-IR & UV-Vis spectrum data for N-Bromoamine compounds[D3,D4,D7,D10] cm^{-1}

Comp. symb.	vs N-H Amine	ν_s C-H Aromatic	ν_s C-H Aliphatic	ν_s C=O Lacton	ν_s C=C Arom.	vs C=N Arom.	ν_s C-O- C ethers	vs C-N	λ_{max1} THF	λ_{max2} THF
D_3	3405	3158	2938	1623	1508	-	1184	1272	323	215
D_4	3417	3158	2927	1639	1569	-	1184	1280	399	207
D_7	3413	3139	2938	1747	1529	1646	1137	1230	257	209
D_{10}	3421	3142	2973	1670	1523	-	1118	1238	343	215

Scheme4 illustrates prepared D3, D4, D7, D10 compounds structures.



Scheme4. Structure for prepared D3, D4, D7, D10 compounds

The following figures 1-4 show ^{1}H-NMR spectrums for A2, B4, C7 and D4 compounds

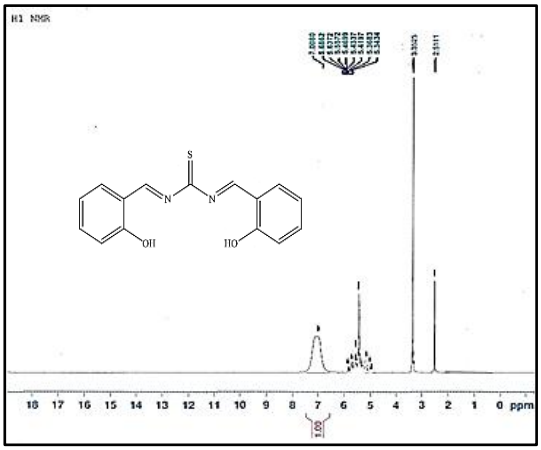


Figure (1): ^{1}H-NMR Spectrum for Compound A2

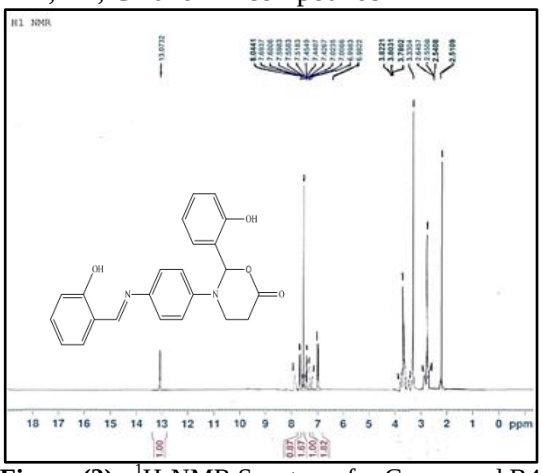


Figure (2): ^{1}H-NMR Spectrum for Compound B4

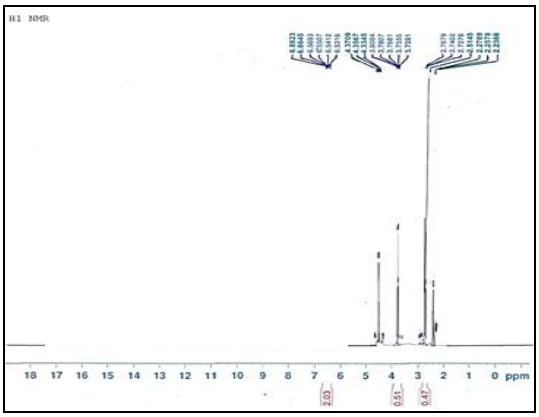


Figure (3): ^{1}H-NMR Spectrum for Compound C7

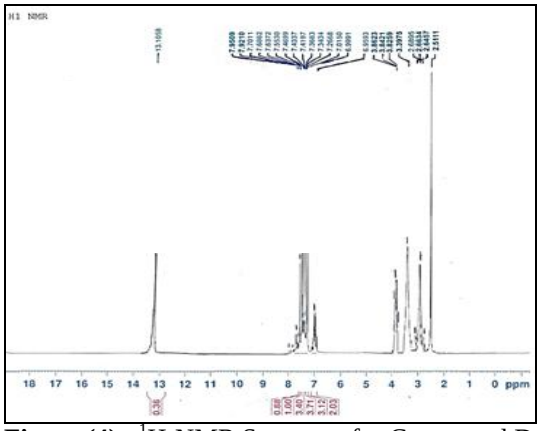


Figure (4): ^{1}H-NMR Spectrum for Compound D4

APPLICATIONS

We expect the prepared compounds have pharmaceutical applications in addition to the possibility of their use as anti-bacterial and antifungal.

CONCLUSIONS

A new some1,3-Oxazine -6-one, 1,3-Oxazine -6,6-dione and N-bromo amines derivatives were synthesized, purified and characterized by melting point, FT-IR, UV-Vis and ^{1}H- NMR spectra.

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