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Synthesis ,characterization and study thermal properties of some new .benzidine derivatives

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Abstract:

New 1,3-oxazepine derivatives containing azo group have been prepared. The first step, (benzidine) was converted to 4,4'-bis(3-amino phenol)Azobenzidine [A]. by coupling reaction with 3-amino phenol, Then amino group of the azo derivative [A] was condensed with different aromatic aldehydes [4-chlrobenzaldehyde,3-hydroxybenzaldehyde,p-dimethyleamin-obenzaldehyde,4 bromobenzaldehyde,4-hydroxy-3-methoxybenzaldehyde,4-chloro-3-hydroxy-benzaldehyde, 3,4-dimethylbenzaldehyde, and 4-hydroxybenzaldehyde] in absolute ethanol to give new azo Schiff's bases derivatives [B_1-B_8] respectively. The resulting imines derivatives [B_1-B_8] were reacted with maleic anhydride and phathalic anhydride in dry benzene to give new 1,3-oxazepine-4,7-dione ring derivatives[C_1a-C_8a]and[C_1b-C_8b] respectively. For the last derivatives thermal properties were determind by using Thermo gravimetric analysis (TGA),All these compounds were characterized by melting points and FT.IR spectroscopy, some of them were characterized by^{1}H-NMR spectroscopy ,C.H.N analysis and TGA analysis.

Keywords: Benzidine ,Azo compound , Schiff's bases ,1,3-Oxazepine, thermal properties.

الخلاصه:

حضرت مشتقات 1،3-اوكسازيبين جديدة تحتوي في تركيبها على مجموعة الأزو. الخطوة الأولى تتضمن تحول (البنزدين) إلى 4, 4- بس (3- امينوفينول) ازو بنزدين [A].عن طريق تفاعل الازدواج مع (3- امينو فينول) الخطوة الثانية تتضمن إدخال مجموعة امين مشتق الأزو المحضرفي تفاعل تكاثف مع الديهايدات اروماتية مختلفة(4- كلورو بنزلدهايد , 3- هايدروكسي بنزلدهايد , بارا- داي مثل امينو بنزلدهايد, 4-برومو بنزلدهايد , 4-هايدروكسي – 3- ميثوكسي بنزلدهايد , 4- كلورو- 3- هايدوكسي بنزلهايد , 4,3 داي مثل بنزلهايد , 4-هايدوكسي بنزلهايد) بوجود الأيثانول المطلق فتم الحصول على مشتقات ازو قواعد شف الجديدة [B_1-B_8] على التوالي .بعد ذلك تم مفاعلة مشتقات إلايمين الناتجة [B_1-B_8]مع كل من إنهدريد المالك وإنهدريد الفثاليك في البنزين الجاف فتم الحصول على مشتقات حلقية جديدة 3,1-اوكسازيبين-7,4-دايون[C_1a-C_8a] و [C_1b-C_8b] على التوالي وان المخطط [I] يوضح خطوات تحضير كافة المركبات, كما تم دراسة الخواص

الحرارية لبعض هذه المشتقات باستخدام التحليل الحراري الوزني (TGA). إن جميع المركبات المحضرة شخصت بوساطة درجات الأنصهار ومطيافية الأشعة تحت الحمراء والبعض منها شخصت بوساطة مطيافية الرنين النووي المغناطيسي ألبروتوني وتحليل العناصر الدقيق (كاربون, هيدروجين, نيتروجين).

Introdection

Azo dyes are a class of compounds containing N=N bond.It is easily of synthesis so that it used in the textile, fiber, leather, paint and printing industries for more than a century (1).Azodyes,like five-membered heterocyclic azo dyes such as azothiazole, azothiophene, azopyrrole, and azofuran, are important since they have pronounced bathochromic absorptions compareds to azobenzene dyes and are environmentally friendly(2).Benzidine is a biphenyl amine that exists at room temperature as a white to slightly reddish crystalline powder(3).So that we can utilized as a source of Schiff base,thenenterd in cycloaddition reaction to prepare hetrocyclic compound(4).Azomethine group (-C=N-) are known as Schiff bases ,They are usually formed by condensation of a primary amine with a carbonyl compounds(5). Schiff bases are the imprtant compound owing to their wide range of biological activities and industrial application(6).Oxazepine-diones is a seven-membered ring containing nitrogen, oxygen and two carbonyl group(7).Oxazepine and their derivatives have some important biological pharmacological activities(8),psychoactive drugs (9).such as enzyme inhibitors(10).Analgesic(11).and antidepressant(12). Thermal analysis is the analysis of a change in a property of a sample, which is related to an imposed change in the temperature, To determine the thermo-physical properties several methods are commonly used differential scanning calorimetric (DSC),thermo gravimetric analysis (TGA)(13). These measurements provide quantitative andqualitative information about physical and chemicalchanges and determining the temperatures of the phase transitions like melting point, solidification onset, re-crystallization onset, evaporation temperature,Heats of Fusion and Reactions, Reaction Kinetics, and Purity(14).

Experimental

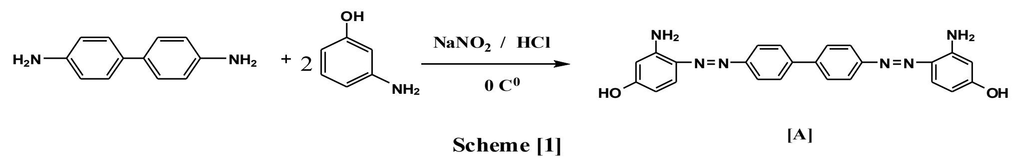
All chemicals were used supplied from Merck , BDH and Fluka chemicals company . The melting points were recorded using Electrothermal melting point apparatus, UK. The elemental analyses were measured on E.A.G.E.R.-100, Carlo Erba, Italy. FT.IR spectra were taken on Fourier transform infrared SHIMADZU FT.IR-8400S infrared spectrophotometer by

KBrdisc .^{1}H-NMR were recorded on Fourier transform Varian spectrometer, operating at 400 MHz.TGA were recorded by thermal gravimetric analyzer ,Company TA , Model TQ50, USA.

Synthesis Methods

Synthesis of4,4'-bis(3-amino phenol)Azobenzidine [A](15).

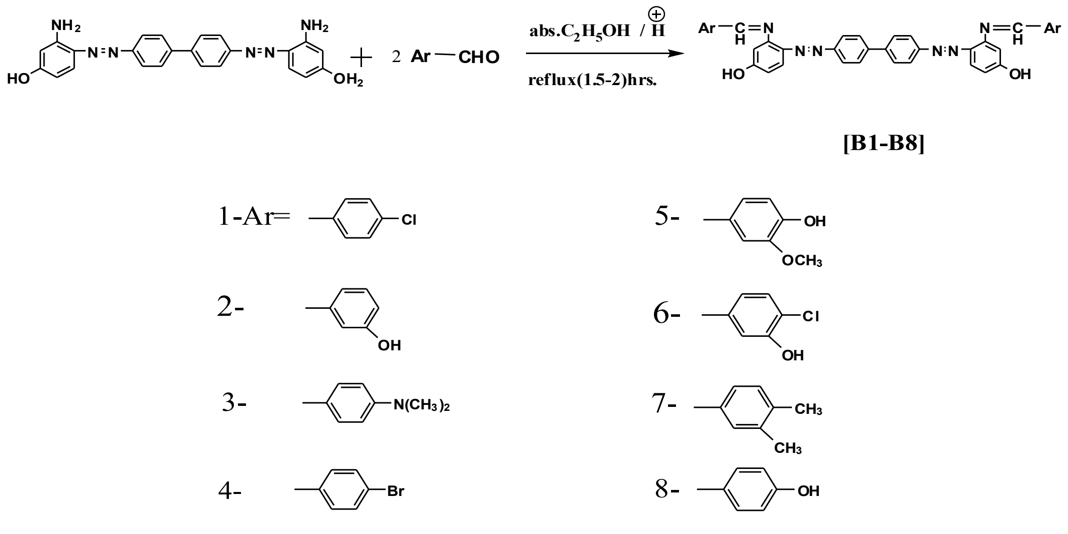
Benzidine (1.84gm, 0.01 mole) was dissolved in (3.5 ml) of concentrated hydrochloric acid and (40 ml) of distilled water. The mixture was cooled at (0C^0) in ice-water bath. a solution of sodium nitrite (1.72 gm, 0.02 mole) dissolved in (6 ml) of distilled water. Then was added a dropwise to the mixture with stirring. In the other beaker 3-amino phenol (2.18 gm, 0.02 mole) was dissolved in (3 gm) of sodium hydroxide and (200 ml) of distilled water and place this beaker in ice-water bath at (0C^0). The cold diazonium chloride was added to the coupling agent in small portions and stirred after each addition, after the addition was completed, the reaction mixture was stirred at (0C^0) for 15 minites. The black product was precipitated and filtered recrystallized from ethanol, yield (4.32 gm, 86%), (m.p. above 200) C and R_f=(0.74) (toluene : ethanol , 7 : 3).**Scheme(1).**



Scheme [1]

Synthesis of Schiff bases derivatives [B_1-B_8](16).

The Azo compound [A] (0.01 mole) was added to a solution of the different benzaldehyde derivatives (0.02 mole) in 40 ml of absolute ethanol and two drops of glacial acetic acid were also added to the above mixture. The mixture was refluxed for (1.5-2) hrs. TLC showed that the reaction was completed by using (ethyl acetate : toluene , 1 : 1). The precipitates were formed collected by filtration , dried and recrystallized from the appropriate solvent. Scheme(2).



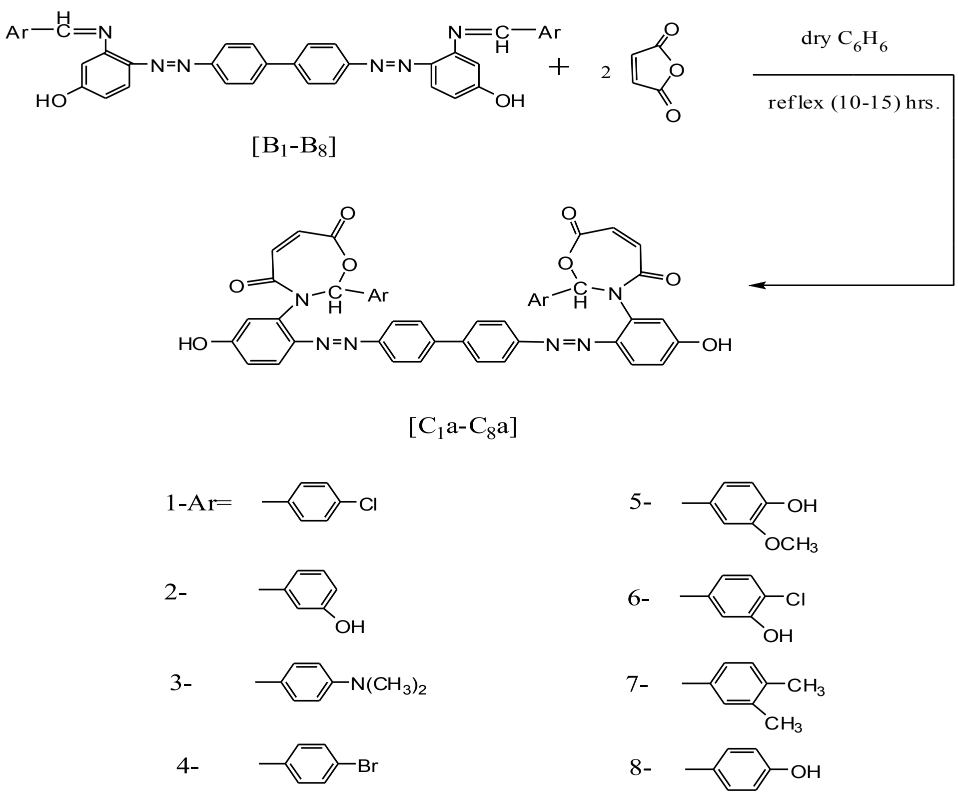
Scheme[2]

Table(1):some physical properties of compounds $[B_1-B_8]$

Comp. No.	Molecular Formula	M.P. °C	Yield %	R_f
B_1	$(C_{38}H_{26}Cl_2N_2O_2)$	above 200	82.85	0.43
B_2	$(C_{38}H_{28}N_6O_4)$	185	86	0.7
B_3	$(C_{42}H_{38}N_8O_2)$	196	86.85	0.76
B_4	$(C_{38}H_{26}Br_2N_8O_2)$	above 200	80.75	0.51
B_5	$(C_{40}H_{32}N_6O_6)$	193	89.06	0.72
B_6	$(C_{38}H_{26}Cl_2N_6O_4)$	180	81.91	0.76
B_7	$(C_{42}H_{36}N_6O_2)$	169	85.5	0.75
B_8	$(C_{38}H_{28}N_6O_4)$	above 200	83.6	0.64

General procedure for Synthesisof 4,4'-[bis(3-hydroxy-6,1-phenylene)-2-(aryl)-2,3-dihydro-1,3-oxazepine-4,7-dione]Azo benzidine $[C_1a-C_8a]$ (17).

To the mixture of Schiff bases [B_1-B_8](0.01 mol) in dry benzene (250ml), maleic anhydride (0.02mol,1.96 gm), The reaction mixture was stirred for (10-15 hrs.) at(55C^0),the TLC showed that the reaction was complete by using (ethyl acetate : toluene , 1 : 1) The precipitates were collected by filtration.), **Scheme(3).**



Scheme [3]

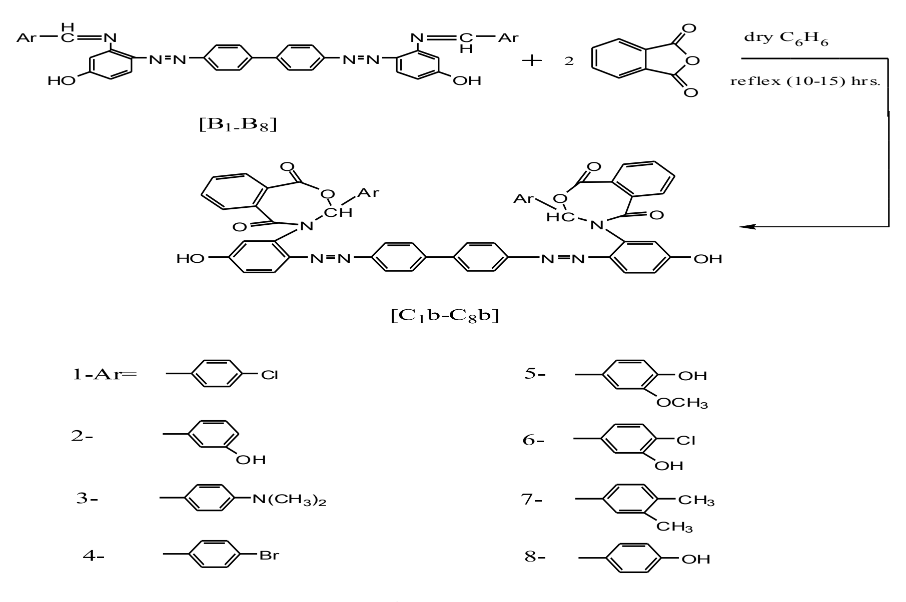
Table(2): Some physical properties of compounds [C_1a-C_8a]

Comp. No.	**Molecular Formula**	**M.P. °C**	**Yield %**	**R_f**		**C.H.N.**		
						C%	**H%**	**N%**
C_1a	($C_{46}H_{30}Cl_2N_6O_8$)	Above200	73.29	0.61	_			
C_2a	($C_{46}H_{32}N_6O_{10}$)	198	76.9	0.69	Calc.	66.66	3.89	10.14
					Found.	(66.44)	(3.37)	(10.07)
C_3a	($C_{52}H_{46}N_8O_8$)	Above200	72.5	0.75	_			
C_4a	($C_{46}H_{30}Br_2N_6O_8$)	Above200	77.4	0.94	Calc.	57.88	3.17	8.80
					Found.	(57.69)	(3.12)	(8.26)
C_5a	($C_{48}H_{36}N_6O_{18}$)	172	79.54	0.68	_			
C_6a	($C_{46}H_{30}Cl_2N_6O_{10}$)	183 Above200	74	0.73	_			
C_7a	($C_{50}H_{40}N_6O_8$)	Above200	78	0.7	_			

C_8a	$(C_{46}H_{32}N_6O_{10})$	175	75.2	0.64	_

General procedure for Syenthesis of4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(aryl)-2,3-dihydro- benzo[e]-1,3-oxazepine-4,7-dione]Azobenzidine [C_1b-C_8b][18]

To the mixture of Schiff bases [B_1-B_8](0.01 mol) in dry benzene(250 ml), phthalic anhydride (0.02mol, 2.96 gm), The reaction mixture was stirred for (10-15 hrs.) at(55C^0) . the TLC showed that the reaction was complete by using (ethyl acetate : toluene , 1 : 1).The precipitates were collected by filtration. **Scheme(4).**



Scheme [4]

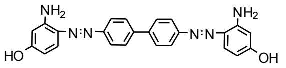
Table(3): Some physical properties of compounds [C_1b-C_8b].

Comp. No.	Molecular Formula	M.P. ^0C	Yield %	R_f		C.H.N.		
						C%	H%	N%
C_1b	$(C_{54}H_{34}Cl_2N_6O_8)$	Above 200	78.7	0.38	Calc.	67.16	3.55	8.70
					Found.	(67.06)	(3.45)	(8.56)
C_2b	$(C_{54}H_{36}N_6O_{10})$	191	74.4	0.73	_			
C_3b	$(C_{58}H_{46}N_8O_8)$	135	82	0.72	Calc.	70.86	4.72	11.40
					Found.	(70.32)	(4.49)	(11.23)
C_4b	$(C_{54}H_{34}Br_2N_6O_8)$	Above 200	73.43	0.89	_			
C_5b	$(C_{56}H_{46}N_6O_{12})$	Above200	72.75	0.66	_			
C_6b	$(C_{54}H_{34}Cl_2N_6O_{10})$	Above 200	70.27	0.41	_			

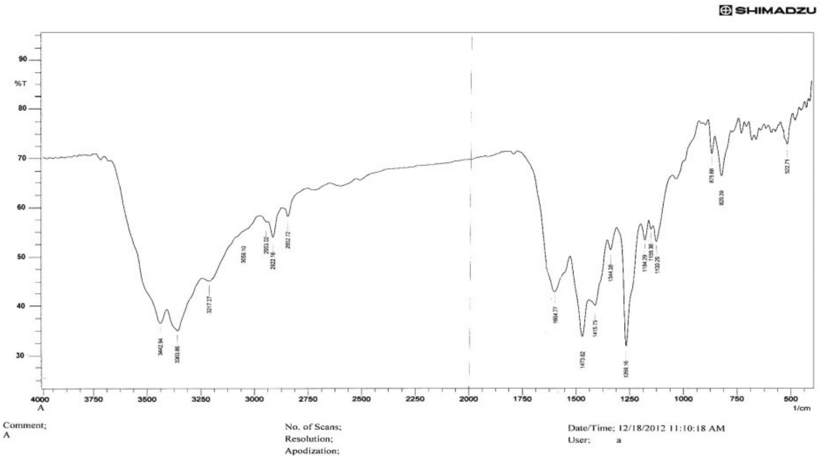
C_7b	($C_{50}H_{40}N_6O_8$)	161	80.42	0.74	_
C_8b	($C_{54}H_{36}N_6O_{10}$)	Above200	78.8	0.51	_

Results and discussion

The Key intermediate compound [A] by converted benzidine to the benzidinediazonium chloride by reaction with concentrated hydrochloric acid and sodium nitrite. Diazonium salt was directly introduced in a coupling reaction with 3-amino phenol to produce 4,4'-bis(3-amino phenol)Azobenzidine [A] The synthesis azo compound [A] was identified with FT.IR showed disappearance of the two absorption bands at (3383 cm^{-1}) and (3307 cm^{-1}) were due to the stretching vibrations of (-NH_2) group of benzidine and appearance of two absorption bands at (3442) cm^{-1} and (3363) cm^{-1} of the asymmetric and symmetric stretching vibration of the two(-NH_2)groups of azo compound . Which overlaps with the stretching vibration of the two (- OH) groups. FT.IR spectrum also showed the appearance of absorption band at (1473cm^{-1}) for (N=N) group,stretching vibration of the (C-H) aromatic appear at (2922) cm^{-1} and (829 cm^{-1}) were due to the bending vibration of (C-H) aromatic out of plane.



4,4'- bis(3-amino phenol)Azo benzidine

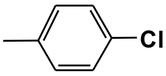
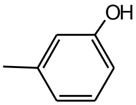
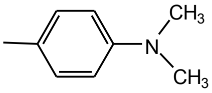
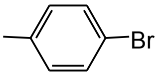
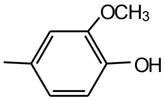
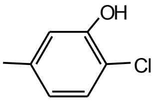
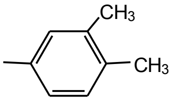
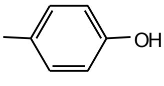


Figure(1)FT.IR spectrum of compound [A].

Azobenzidine derivative reaction with different carbonyl derivatives so as to form new Schiff bases compounds [B_1-B_8] these compounds were characterized by FT.IR which showed disappearance of two absorption bands at (3442) cm^{-1} and (3363) cm^{-1} of the asymmetric and symmetric streching vibration of the two(-NH_2)groups . Which overlaps with the stretching vibration of the (-OH) groups of azobenzidinederavaitive.and appearance band at(1624-1670)$cm^{-1}$of streachingvibraition of two (C=N) imine group**.**

Table (4): FT .IR data of Schiff bases compounds [B_1-B_8].

Comp. No.	**Ar**	**υ (C-H)Str.** **Aromatic** **Aliphatic**	**υ (C=N) Str. Imine**	**υ (O-H) Str. cm⁻**	**υ (N=N) cm^{-1}**	**δ(C-H) Bending**	**Others cm^{-1}**

		cm^{-1}	cm^{-1}			cm^{-1}	
B_1		2922 2852	1624	3406 3394	1541	825	υ (C-Cl) Str. : 1097
B_2		2922 2848	1670 1630	3433 3402	1452	785	_
B_3		2916	1666	3429 3205	1542	817	υ(N-CH_3)Str.: 2866asym. 2812sym
B_4		2923 2852	1635	3390 3217	1558	827	υ (Ar-Br)Str. : 1068
B_5		2927	1670 1623	3190	1512	821	υ (Ar-OCH_3)Str. : 2848
B_6		2880	1670	3305	1550	831	υ (Ar-Cl) Str. : 1091
B7		2918	1660	3421	1550 1533	817	_
B8		2916	1668	3325	1514 1454	833	_

Then, the imines results derivatives [B_1-B_8] were reacted with maleic anhydride andphathalic anhydride in dry benzene to give new 1,3-oxazepine-4,7-dione ring derivatives [C_1a-C_8a] and [C_1b-C_8b] respectively. these compounds were characterized by FT.IR intable 5,6 as flow:-

Table(5): FT .IR data of 4,4'-[bis(3-hydroxy-6,1-phenylene)-2-(aryl)-2,3-dihydro-1,oxazepine -4,7-dione]Azo benzidine [C_1a-C_8a]

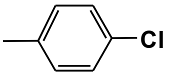
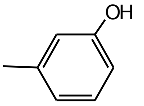
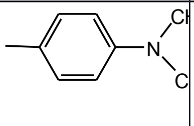
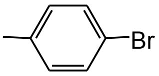
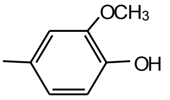
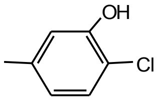
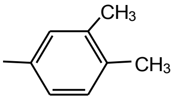
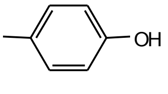
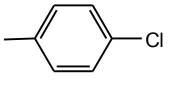
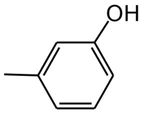
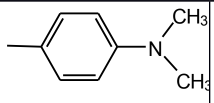
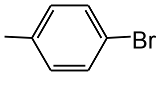
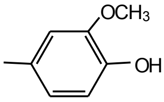
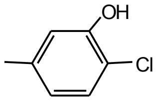
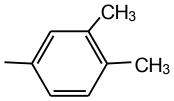
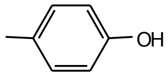
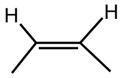
Comp. No.	**Ar**	**υ (C-H)Str. Aromatic Aliphatic cm^{-1}**	**υ (C-H) Str. Benzylic cm^{-1}**	**υ (C=O)str. Lactone Lactam cm^{-1}**	**υ (C-N) Str. cm^{-1}**	**υ (N=N) cm^{-1}**	**υ (O-H) Str. cm^{-}**	**υ (C-O) Str. Lactone cm^{-1}**	**Others cm^{-1}**
C_1 a		2925 2854	3224	1701 1693	1159 1135	1558 1523	3366 3250	1263 1230	υ (C-Cl) Str. : 827
C_2 a		2923	3109	1680	1172 1151	1492	3217	1284 1251	-
C_3 a		2921	3190	1733 1683 1662	1166 1124	1548 1533	3402	1232 1245	υ(N-CH_3)Str.: 2796asym. 2713sym.
C_4 a		2923 2854	3241	1701 1680	1164 1134	1558 1541	3390 3369	1236	υ (Ar-Br)Str. : 1070
C_5 a		2920	3026	1678	1172 1155	1537 1512	3186	1201	υ($ArOCH_3$) Str: 2850
C_6 a		2925 2854	3193	1670 1645	1172 1155	1550 1525	3276 3263	1238	υ (Ar-Cl) Str. : 1089
C_7 a		2920 2854	3259 3244	1714 1662	1166 1124	1550 1535	3413 3402	1236	_
C_8 a		2962 2920	3185	1708 1666	1161	1516 1462	3446	1278 1219	_

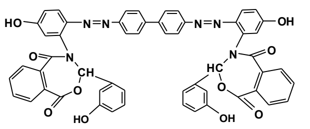
Table (6):FT .IR data of of4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(aryl)-2,3-dihydrobe- azo[e]-1,3-oxazepine-4,7-dione]Azobenzidine [C$_1$b-C$_8$b]

Comp. No.	Ar	υ (C-H)Str. Aromatic Aliphatic cm^{-1}	υ (C-H) Str. Benzylic cm^{-1}	υ (C=O)str. Lactone Lactam cm^{-1}	υ (C-N) Str. cm^{-1}	υ (N=N) cm^{-1}	υ (O-H) Str. cm^{-1}	υ (C-O) Str. Lactone cm^{-1}	Others cm^{-1}
C$_1$b		2921	3230	1766 1701	1145	1542	3402	1250	υ (C-Cl) Str. : 1074
C$_2$b		3028 2964	3095	1759 1674	1165	1494 1465	3213	1255	-
C$_3$b		2920	3111	1766 1710 1680	1166	1537	3344	1259	υ(N-CH$_3$)Str.: 2856asym 2819sym
C$_4$b		2923 2854	3215	1760 1701	1168	1490 1471	3361	1259	υ (Ar-Br)Str. : 1072
C$_5$b		2925	3176	1770 1743	1172 1155	1512	3380	1256	υ (Ar-OCH$_3$)Str. : 2863
C$_6$b		3070 2926	3194	1766 1670	1166	1541	3304 3265	1259	υ (Ar-Cl) Str. : 1097
C$_7$b		2921	3210	1758 1662	1185	1550 1535	3442 3363	1243	-
C$_8$b		2964 2927	3170	1762 1668	1163	1516 1467	3201	1259	-

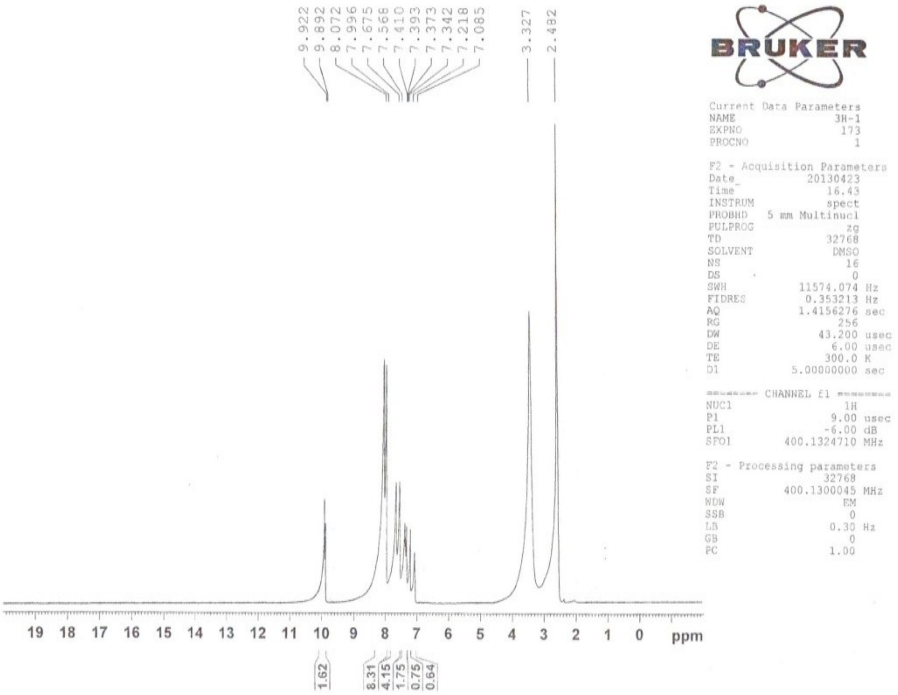
^{1}H-NMR data for (C$_1$a, C$_2$b, C$_3$a ,C$_4$b, C$_5$a, C$_6$b, C$_7$b and C$_8$a) showing in table (7).

Table (7):^{1}H-NMR data of some oxazepine compounds in (*DMSO-d*$_6$) solvent.

Comp. No.	**δ(C-H) Aromatic ppm**		**CH of oxazepine ring**	**OH**	**Others ppm**
C_1a	7.08-7.62	6.45-6.95	9.67	8.61	–
C_2b	7.08-7.99	–	9.89-9.92	8.07	–
C_3a	7.65-7.67	6.75-6.77	9.64	8.43	(s , 6H , N-CH_3): δ2.47
C_4b	7.31-7.99	–	9.64	8.43-8.56	–
C_5a	7.36-7.73	6.93	9.87	8.71	(s , 3H , O-CH_3): δ3.31
C_6b	7.30-7.99	–	9.53	8.27	–
C_7b	6.75-7.98	–	9.64	8.05	(s–CH3): δ 2.48
C_8a	7.06-7.74	6.17-6.91	9.76	8.44-8.56	–



4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(3-hydroxyphenyl)
-2,3-dihydrobenzo[e]-1,3-oxazepine-4,7-dione]Azo benzidine



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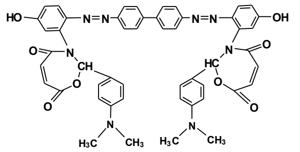
Current Data Parameters
NAME                 3H-1
EXPNO                 173
PROCNO                  1

F2 - Acquisition Parameters
Date_            20130423
Time                16.43
INSTRUM             spect
PROBHD   5 mm Multinucl
PULPROG                zg
TD                  32768
SOLVENT              DMSO
NS                     16
DS                      0
SWH             11574.074 Hz
FIDRES           0.353213 Hz
AQ              1.4156276 sec
RG                    256
DW                 43.200 usec
DE                   6.00 usec
TE                  300.0 K
D1             5.00000000 sec

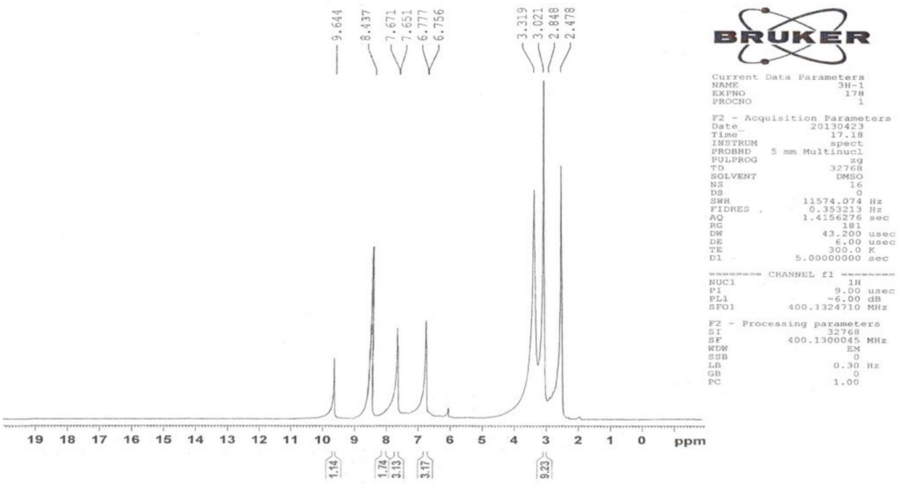
======== CHANNEL f1 ========
NUC1                   1H
P1                   9.00 usec
PL1                 -6.00 dB
SFO1          400.1324710 MHz

F2 - Processing parameters
SI                  32768
SF            400.1300045 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   1.00
```

Figure(2)^{1}H-NMRspectrum of compound [C_2b].



4,4'-[(bis(3-hydroxy-6,1-phenylene)-2-(4-(dimethylaminophenyl)
-2,3-dihydro-1,3-oxazepine-4,7-dione]Azo benzidine



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Current Data Parameters
NAME              3H-1
EXPNO              178
PROCNO               1

F2 - Acquisition Parameters
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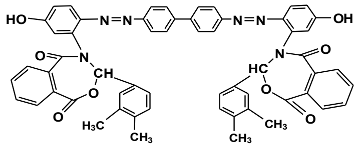
Time             17.18
INSTRUM          spect
PROBHD   5 mm Multinucl
PULPROG             zg
TD               32768
SOLVENT           DMSO
NS                  16
DS                   0
SWH          11574.074 Hz
FIDRES .      0.353213 Hz
AQ           1.4156276 sec
RG                 181
DW              43.200 usec
DE                6.00 usec
TE               300.0 K
D1          5.00000000 sec

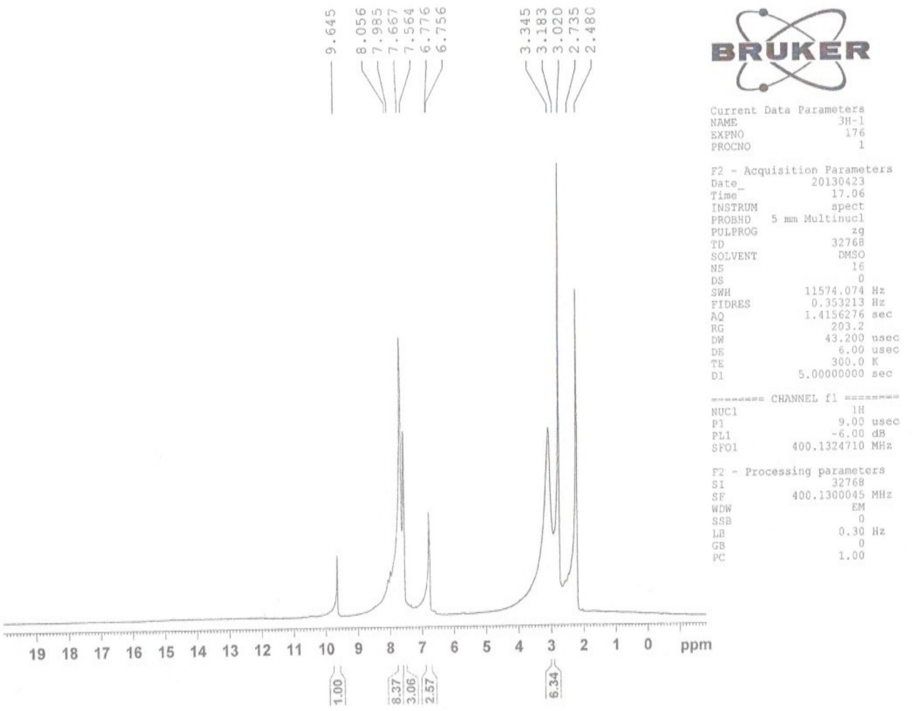
======== CHANNEL f1 ========
NUC1                1H
P1                9.00 usec
PL1              -6.00 dB
SFO1       400.1324710 MHz

F2 - Processing parameters
SI               32768
SF         400.1300045 MHz
WDW                 EM
SSB                  0
LB                0.30 Hz
GB                   0
PC                1.00
```

Figure(3)^{1}H-NMRspectrum of compound [C_3a].



4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(3,4-dimethylphenyl)
-2,3-dihydro- benzo[e]-1,3-oxazepine-4,7-dione]Azo benzidine



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Current Data Parameters
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EXPNO                176
PROCNO                 1

F2 - Acquisition Parameters
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Time               17.06
INSTRUM            spect
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PULPROG               zg
TD                 32768
SOLVENT             DMSO
NS                    16
DS                     0
SWH            11574.074 Hz
FIDRES          0.353213 Hz
AQ             1.4156276 sec
RG                 203.2
DW                43.200 usec
DE                  6.00 usec
TE                 300.0 K
D1            5.00000000 sec

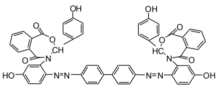
======== CHANNEL f1 ========
NUC1                  1H
P1                  9.00 usec
PL1                -6.00 dB
SFO1         400.1324710 MHz

F2 - Processing parameters
SI                 32768
SF           400.1300045 MHz
WDW                   EM
SSB                    0
LB                  0.30 Hz
GB                     0
PC                  1.00
```

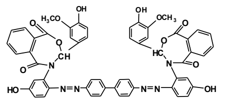
Figure(4)^{1}H-NMRspectrum of compound [C_7b].

As well as ,TGA analysis was done for some compounds and the results indicated that stability to compound (C_5b) was at (217 C^0), (C_6a) at (299 C^0), (C_7a) at (390C^0) and (C_8b) at (253C^0).

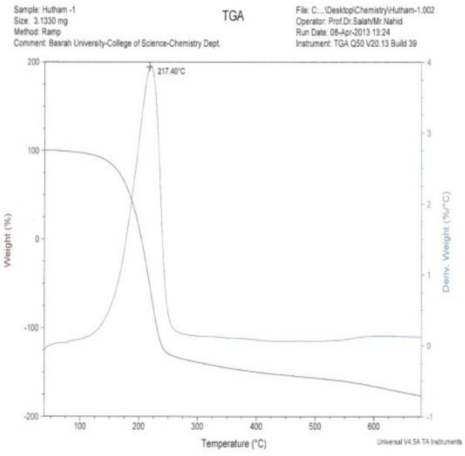
According of TGA spectrum:-

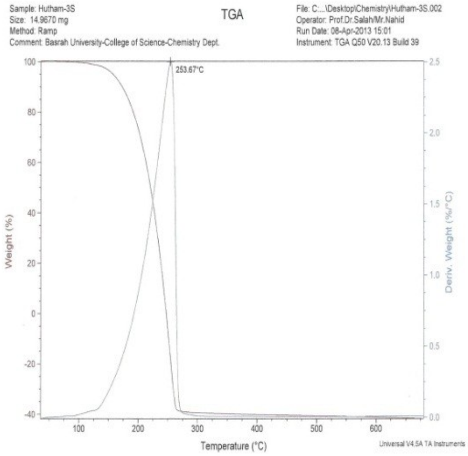


4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(4-hydroxyphenyl)
-2,3-dihydro- benzo[e]-1,3-oxazepine-4,7-dione]Azo benzidine

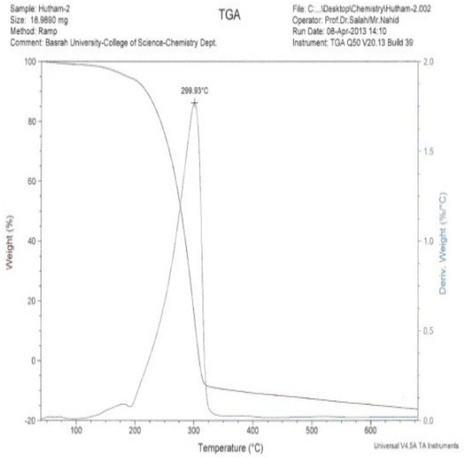


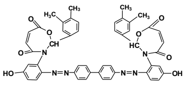
4,4'-[(bis(3-hydroxy-6,1-phenylene)-3-(4-hydroxy-3-methoxyphenyl)
-2,3-dihydrobenzo[e]-1,3-oxazepine-4,7-dione]Azo benzidine



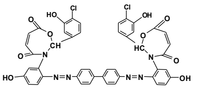


Figure(5)TGA spectrum of compound [C_5b]**Figure(6)**TGA spectrum of compound [C_8b]

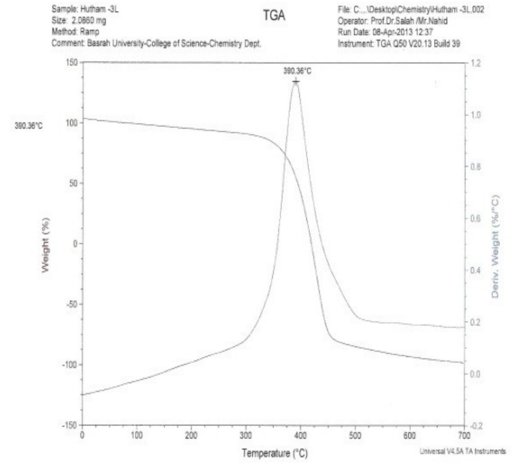




4,4'-[(bis(3-hydroxy-6,1-phenylene)-2-(3,4-dimethylphenyl)
-2,3-dihydro-1,3-oxazepine-4,7-dione]Azo benzidine



4,4'-[(bis(3-hydroxy-6,1-phenylene)-2-(4-chloro-3-hydroxyphenyl)
-2,3-dihydro-1,3-oxazepine-4,7-dione]Azo benzidine



Figure(7)TGA spectrum of compound [C_6a]**Figure(8)**TGA spectrum of compound [C_7a]

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