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Synthesis of some 1,3-diazepine -4,7-diones derivatives (part 2)

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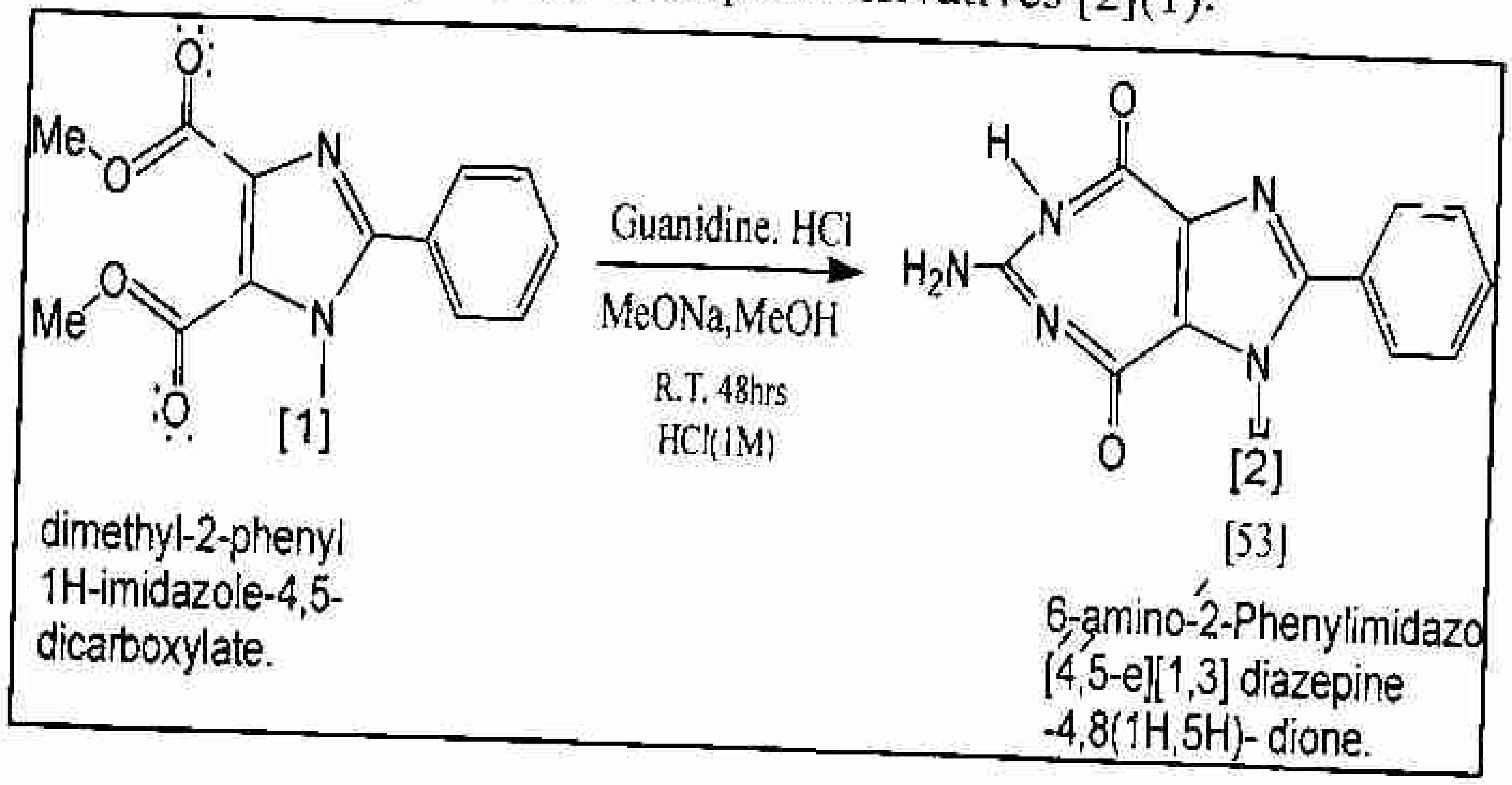
Abstract

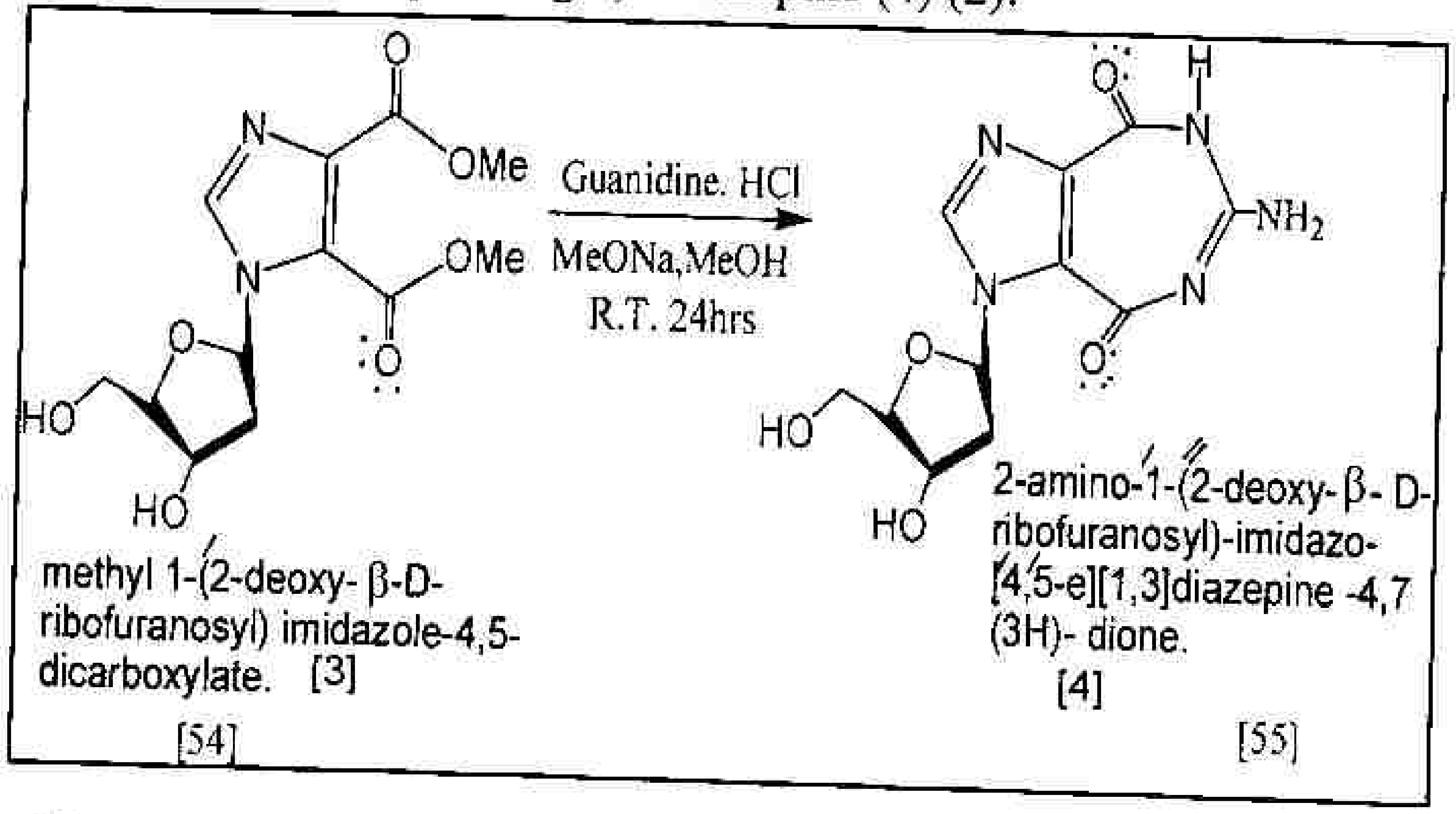
1,3-Oxazepine-4,7-diones were prepared by condensation of N-cinnamylideneareneamines with maleic anhydride, phthalic anhydride ,and 3-nitrophthalic anhydride. The oxazepines were reacted with primary aromatic amine to give the corresponding 1,3-diazepine-4,7-diones.

Introduction

The starting imidazole diester(1) reacts with guanidine hydrochloride to yield 1,3-diazepine derivatives [2](1):



While the starting methyl-1-($\bar{2}$-deoxy-β-D-ribofuranosyl) imidazole -4,5-dicarboxylate(3) reacts with guanidine hydrochloride to yield the corresponding 1,3-diazepine (4) (2):



methyl 1-(2-deoxy- β-D-ribofuranosyl) imidazole-4,5-dicarboxylate. [3]

2-amino-1-(2-deoxy-β- D-ribofuranosyl)-imidazo-[4,5-e][1,3]diazepine -4,7 (3H)- dione. [4]

Aim of the research

To synthesis new diazepines derivatives which are expected to have biological activityies.

Experimental Part

Materials and Solvents

1. Oxazepine- 4,7- diones (II) (prepared as part 1).
2. P- toluidine
3. Dry benzene
4. Ethanol

The instruments:

1. Melting points were determined with: Strut Melting point Apparatus and were uncorrected.
2. IR. Spectra were recorded with: PYE UNICAM SP3-300 Infrared Spectrophotometer (KBr disc) in the range (4000-200)cm^{-1}
3. FTIR Spectra were recorded with: SHIMADZU FTIR-8400S Infrared Spectrophotometer (KBr disc).

4. Elemental Analysis (C.H.N.) was carried with: Perkin Elmer B-240 Elemental Analyzer.

Synthesis of:

A. 1-(p-methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro[1,3]-diazepine-4,7-diones(V).

B. 1-(p-methylphenyl)-2-styryl-3-aryl-1,2,3-trihydrobenzo[1,2-e][1,3]-diazepine-4,7-diones(VI) .

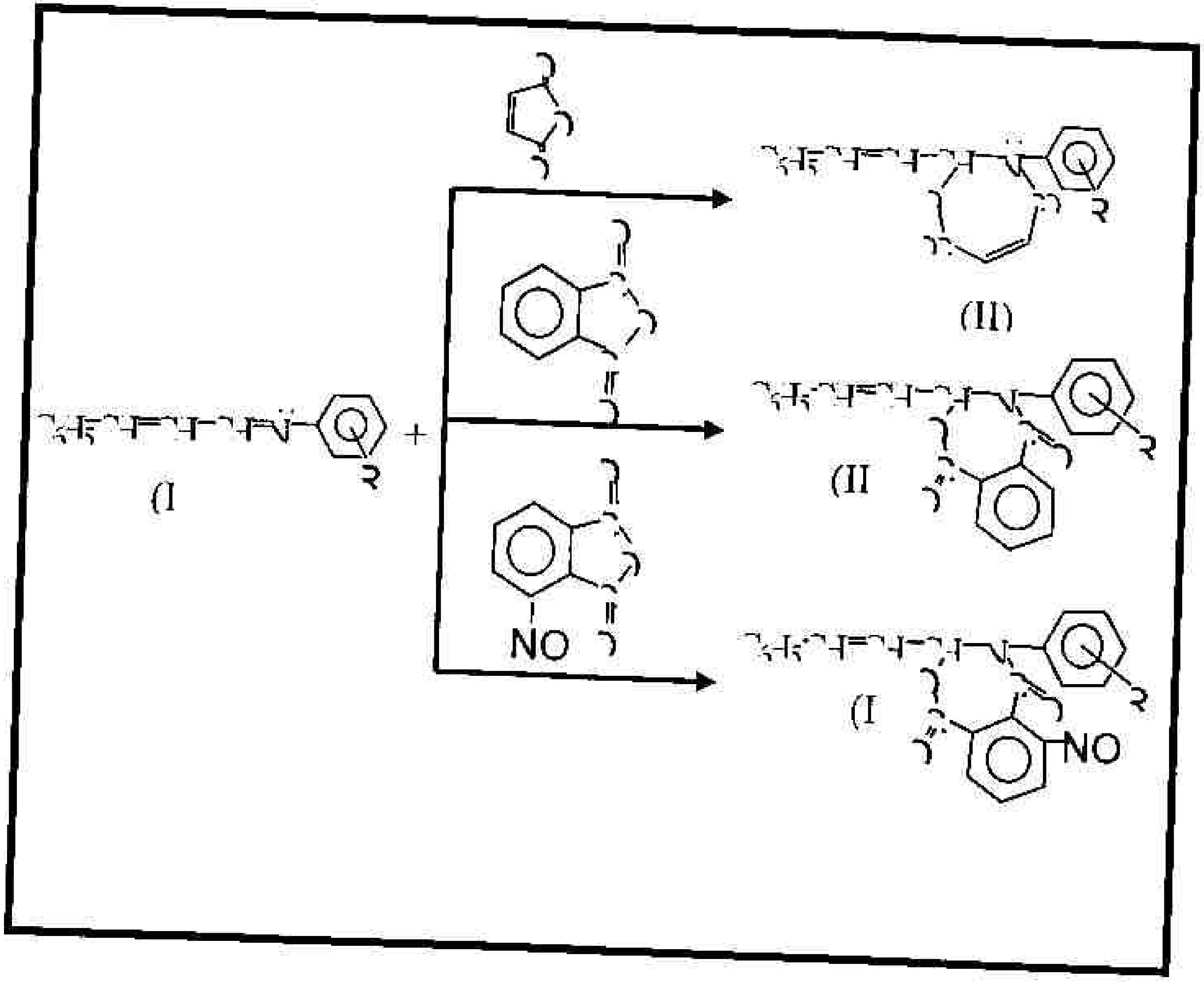
C. 1-(p-methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro-3-nitrobenzo[1,2-e][1,3]-diazepine-4,7-diones(VII).

A mixture of (0.03mole) of 2-styryl-3-aryl-2,3-dihydro[1,3]-oxazepine -4,7-dione (II) and (0.03mole) of p-toluidine suspended in (30mL) dry benzene was refluxed in water bath for 3hrs.The mixture then allowed to cool to room temperature and the separated crystalline solid was filtered and re-crystallized from ethanol to yield 1-(p-methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro[1,3]-diazepine-4,7-diones (V).

The corresponding derivatives of phthalic or 3-nitrophthalicdiazepines were obtained following the same procedure and using the same moles of oxazepine derivatives and p-toluidine.

Discussion

2-Styryl-3-aryl-2,3-dihydro-1,3-oxazepine-4,7-diones,benzo [1,2-e][1,3]-oxazepine-4,7-diones and 3-nitrobenzo [1,2-e][1,3]-oxazepine-4,7-diones were prepared by reaction of N-cinnamylideneareneamines with maleic anhydride,phthalic anhydride ,and 3-nitrophthalic anhydride (3), scheme 1.

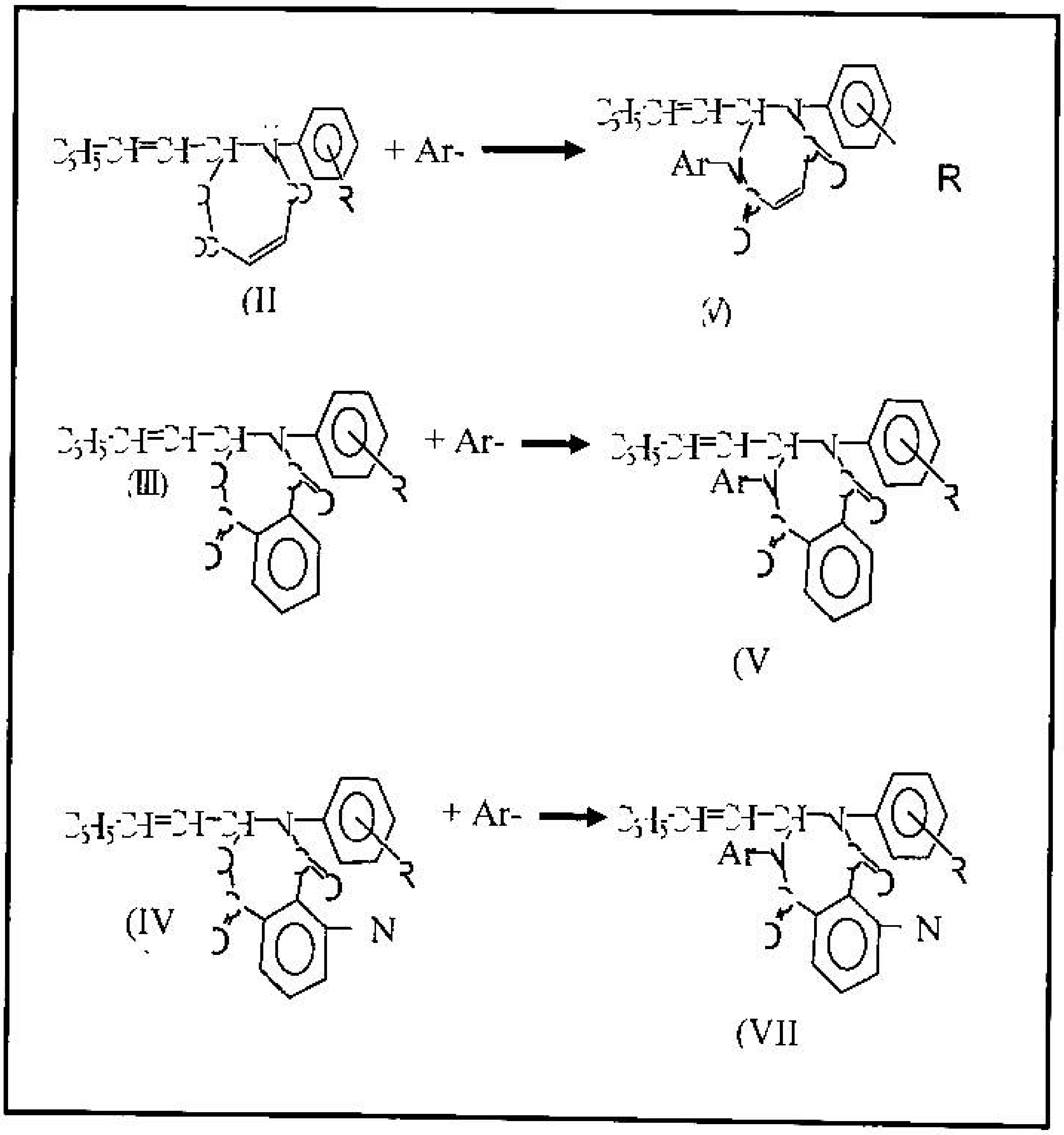


Where R= a) *m*-NO_2; b) *p*-NO_2; c) 2, 4, 6-trichloro; d) *p*-CH_3; e) *p*-OCH_3

Scheme (1)

1,3-Oxazepine-4,7-diones, II, III, and IV were identified by m.ps, and IR,FT.IR spectra(3) and were found to be good starting materials for the synthesis of another class of heterocyclics by their reaction with aromatic primary amines, namely the corresponding 1,3-diazepine-4,7-diones.

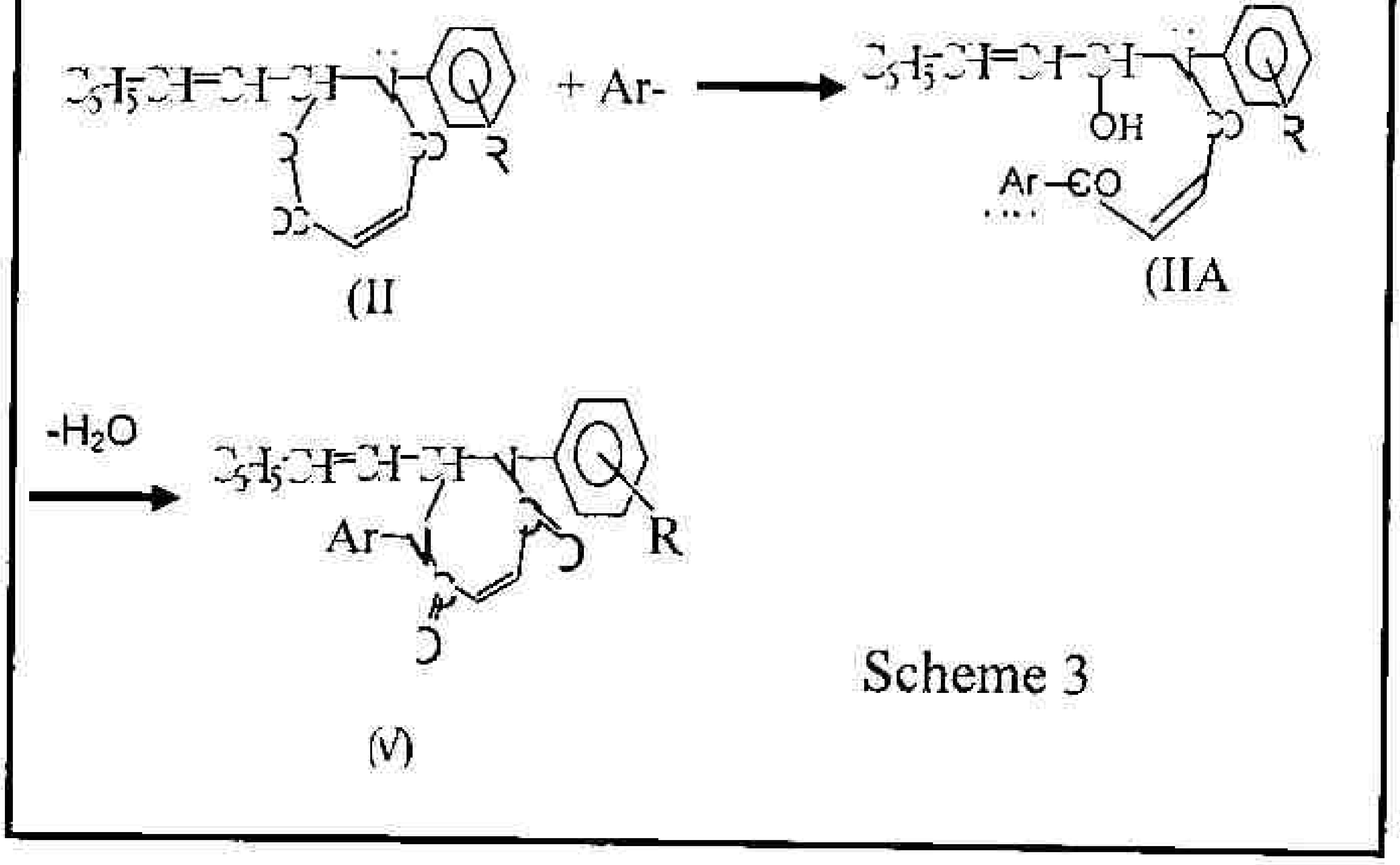
It is obvious that the structure of 1,3-oxazepine-4,7-dione is a combination of lactone and lactam in a 7-membered heterocyclic ring(4). The lactone (cyclic ester) can be converted into 1,3-diazepine-4,7-diones ring via tetrahedral mechanism(5) as shown by the following equation, scheme 2.



Ar= P-CH_3-C_6H_4 Scheme 2

The resulting 2-styryl-1,3-diaryl-1,2,3-trihydro-1,3-diazepine-4,7-diones (V), 2-styryl-1,3-diaryl-1,2,3-trihydrobenzo[1,2-e][1,3]-diazepine-4,7-diones,(VI) , and 2-styryl-1,3-diaryl-1,2,3-trihydro-3-nitrobenzo[1,2-e][1,3]-diazepine-4,7-diones (VII), were identified by their m.ps ,elemental analyse (tables-1,4,6), and IR, FT.IR spectra (tables-2,3,5,7).

It is noticeable that the IR spectra of the expected products show the appearance of (NH, OH) absorption bands at(3500, 3300) cm^{-1}(6). In fact, this is due to an open –chain hydroxyamide intermediate (IIA), formed by cleavage of the 1,3-oxazepine ring by the aromatic amine which upon cyclisation gave the diazepine (V) on further heating. This indicates that the alcoholic part of the lactone maintains its configuration throughout the course of the reaction of the amine with 1,3-oxazepine, scheme 3.



Scheme 3

References

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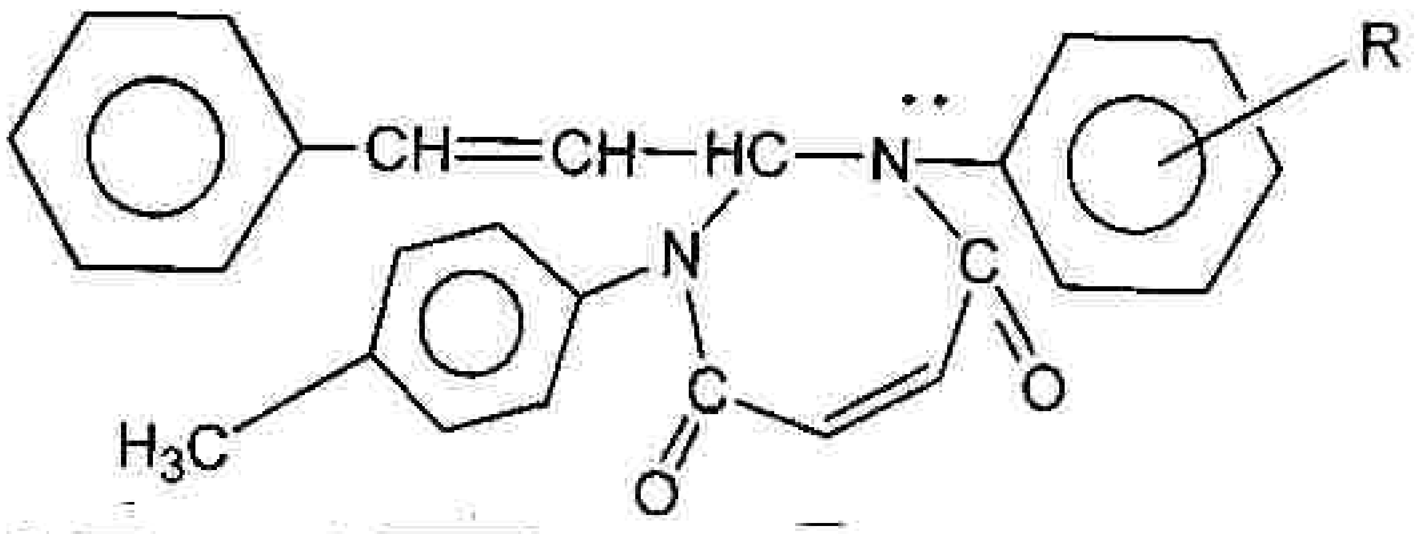
3. Hussein, F. A. and Muhanned J. Mahmoud., (2006) Al-Muatanairya J.Scim 1 (17) p.42.
4. Hussein, F.A. and Obaid H. Abid., (2001) Iraqi Journal of Chemistry 27(3) P.943.
5. March, J. (1992) "Advanced Organic Chemistry", Reactions, Mechanisms, and Structures, 4th ed., Megraw-Hill, International Book Co., John Wiley and Sons, New York, PP (896-918).
6. John R. Dyer(1965) "Application of Absorption Spectroscopy of Organic Compounds", Prentice- Hill, Inc., London p. 35.

Table (1):Molewlar formula M.F., m.ps, Yields %, and Elemental Analysis of 1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro[1,3]-diazepine4,7-diones(V).

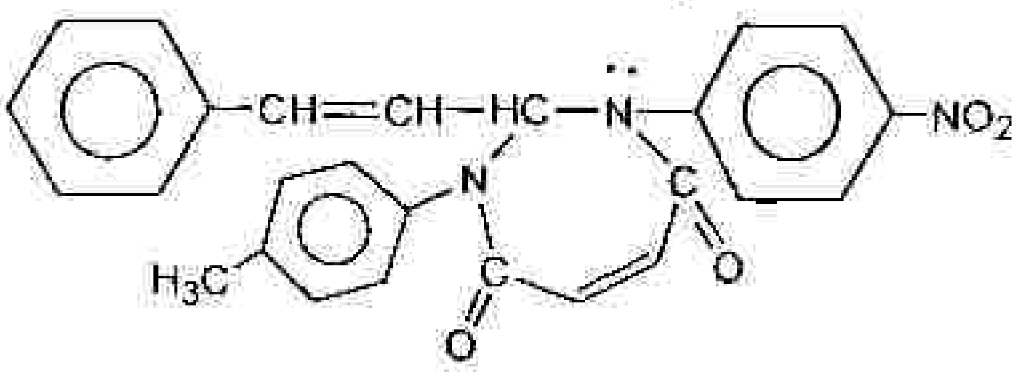


Comp	R	M.F.	m.p.C°	Yield %	Calc.			found		
					C%	H%	N%	C%	H%	N%
Va	3-NO2	$C_{26}H_{21}O_4N_3$	143-145	56	71.07	4.78	9.56	70.91	4.66	9.74
Vb	4-NO2	$C_{26}H_{21}O_4N_3$	162-164	54	71.07	4.78	9.56	70.51	4.51	9.87
Vc	2,4,6-trichloro	$C_{26}H_{19}O2N_2Cl_3$	216-218	66	62.71	3.81	5.62	62.98	3.49	5.93
Vd	4-Me	$C_{27}H_{24}O_2N_2$	178-179	56	79.41	5.88	6.86	78.97	5.53	6.35
Ve	4-OMe	$C_{27}H_{24}O_3N_2$	156-158	68	76.41	5.66	6.60	75.94	5.42	6.48

Table (2) IR absorption bands (cm^{-1}) of 1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro[1,3] –diazepine- 4,7-diones(V).

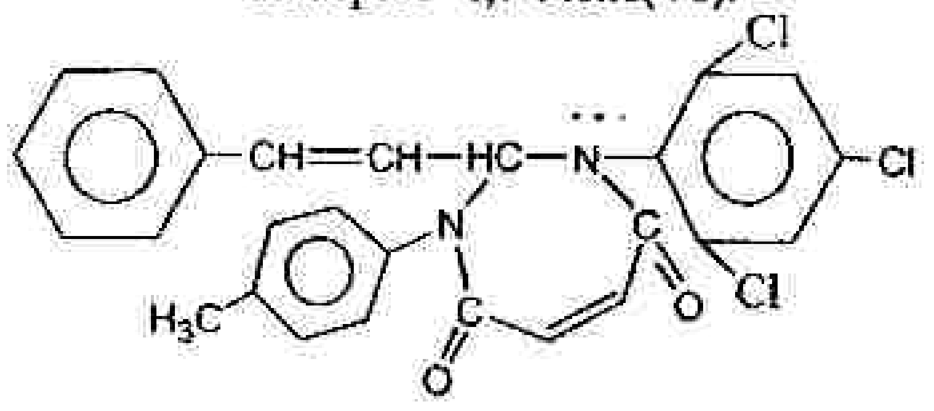
Comp.	C-H str. Allylic	=CH str. Vinylic	C-H str Aromatic	C-H str. Aliphatic	C=O str. Lactam	C=C str. Vinylic	C=C str. Aromatic	C-N str.	C-H bend. Aromatic	C-H bend.Allylic	Cis=CH bend. vinylic	Others
Va	3195	3155	3050	2895	1720	1455	1625	1170	1430	1270	840	C-NO2: 1540, 1350
Vh	3195	3155	3050	2890	1695	1455	1625	1170	1430	1270	840	C-NO2 1550, 1330
Vc	3195	3155	3050	2910	1720	1455	1645	1170	1410	1320	840	C-Cl: 720
Vd	3190	3145	3040	2885	1695	1510	1545	1170	1410	1320	830	
Ve	3190	3140	3060	2900	1730	1495	1630	1170	1400	1320	835	C-O-C 1030

Table (3) FTIR absorption bands (cm-1) of 1-(p--Methylphenyl)-2-styryl-3-(p- nitrophenyl)-1,2,3-trihydro[1,3]-diazepine-4,7-dione (Vb).



(Vb)

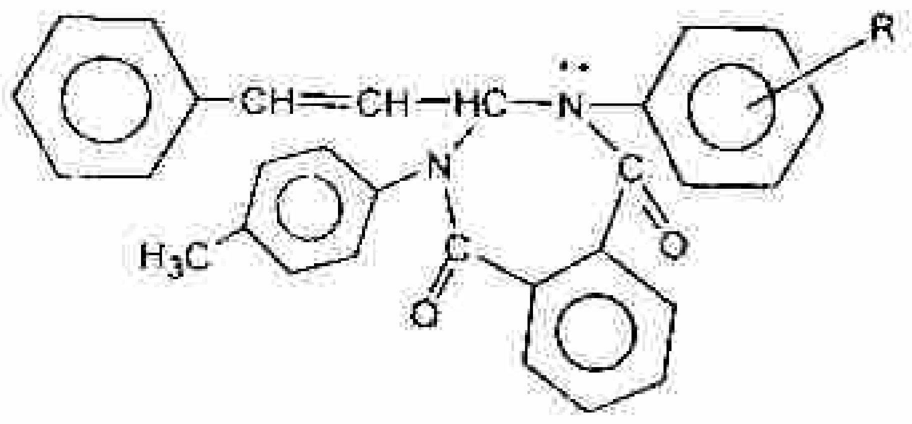
& 1-(*p*-Methylphenyl)-2-styryl-3-(2,4,6,)-trichlorophenyl)-1,2,3-trihydro[1,3]-diazepine -4,7-dione(Vc).



(Vc)

Comp.	C-H str. Allylic	=CH str Vinylic	C-H str. Aromatic	C-H str. Aliphatic	C=O str. Lactam	C=C str. Vinylic	C=C str. Aromatic	C-N str.	C-H bend. Aromatic	Cis=CH bend. Vinylic	C-H bend Allylic	C-NO2 Aromatic	C-Cl Aromatic
Vb	3238	3087	3031	2910	1707	1458	1636	1182	1458	837	1271	1530,1338	
Vc	3226	3072	3016	2921	1724	1450	1637	1176	1406	819	1326	—	700

Table (4) Molewlar formula M.F., m.ps, Yields %, and Elemental Analysisof1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydrobenzo[1,2-e][1,3]-diazepine-4,7-diones(VI).



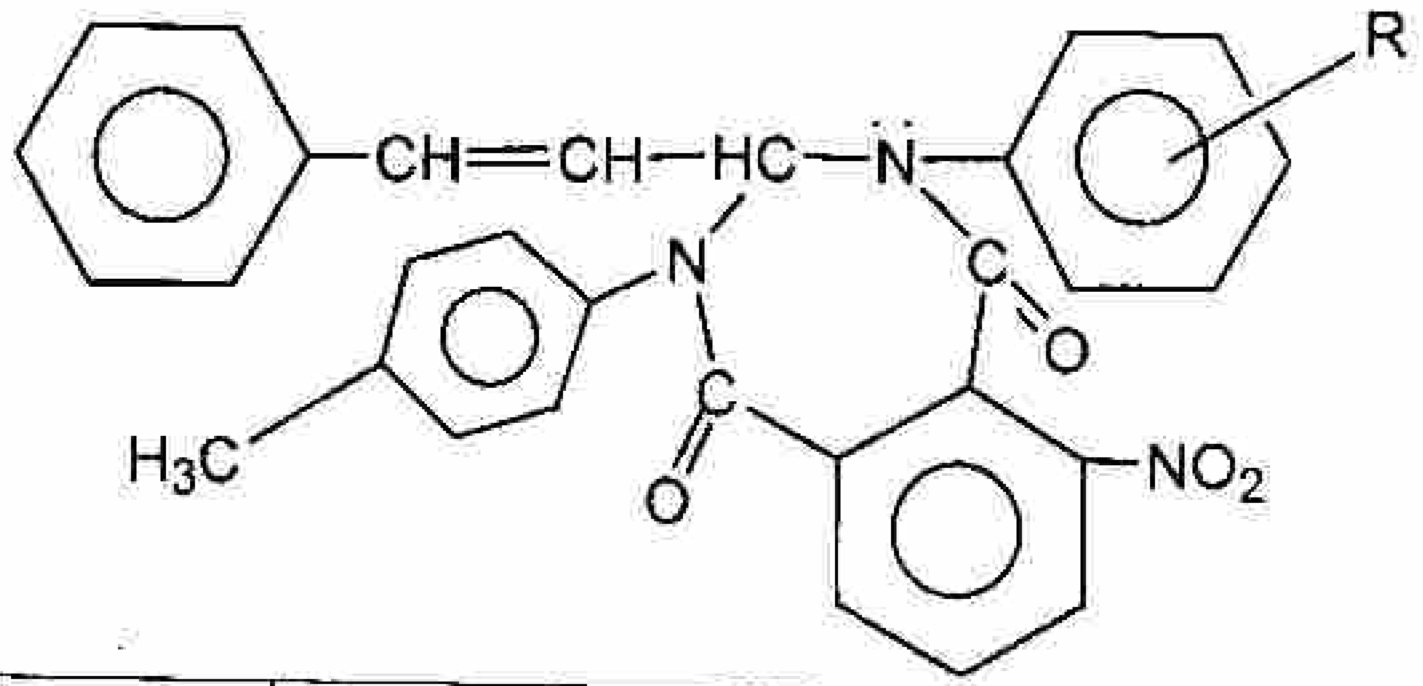
(VI)

Comp.	R	M.F.	m.p.C°	Yield%	Calc.			found		
					C%	H%	N%	C%	H%	N%
VIa	3-NO2	$C_{30}H_{23}O_4N_3$	117-119	59	73.61	4.70	8.58	73.11	4.16	8.73
VIb	4-NO2	$C_{30}H_{23}O_4N_3$	191(de.) 196-197(melt)	51	73.61	4.70	8.58	73.13	4.24	8.79
VIc	2,4,6-trichloro	$C_30H_{21}O_2N_2Cl_3$	196-198	53	65.75	3.83	5.11	65.19	3.24	5.47
VId	4-Me	$C_{31}H_{26}O_2N_2$	168-170	57	81.22	5.67	6.11	81.39	5.42	6.39
VIe	4-OMe	$C_{31}H_{26}O_3N_2$	177-179	65	78.48	5.48	5.90	78.66	5.17	5.98

Table (5) IR absorption bands (cm-1)of 1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydrobenzo[1,2-e][1,3]-diazepine-4,7-diones(VI).

Comp.	C-H str. Allylic	=CH str Vinylic	C-H str. Aromatic	C-H str. Aliphatic	C=O str. Lactam	C=C str. Vinylic	C=C str. Aromatic	C-N str.	C-H bend. Aromatic	C-H bend. Allylic	Cis =CH bend. Vinlylic	Others
VIa	3185	3145	3060	2895	1710	1510	1675	1170	1410	1325	830	C-NO2: 1560, 1375
VIb	3180	3140	3050	2890	1710	1485	1655	1170	1390	1315	825	C-NO2 1545, 1335
VIc	3185	3145	3065	2885	1690	1510	1645	1170	1390	1285	825	C-Cl: 795
VId	3185	3155	3060	2880	1710	1515	1665	1170	1390	1295	815	
VIe	3185	3150	3045	2880	1710	1510	1670	1170	1390	1315	830	C-O-C 1025

Table (6) Molewlar formula M.F., m.ps, Yields % and Elemental Analysis of 1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro-3-nitrobenzo[1,2-e][1,3]-diazepine-4,7-diones(VII).



Comp	R	M.F.	m.p.C°	Yield%	Calc.			found		
					C%	H%	N%	C%	H%	N%
VIIa	3-NO2	$C_{30}H_{22}O_6N_4$	163-165	66	67.41	4.11	10.48	66.93	3.91	10.82
VIIb	4-NO2	$C_{30}H_{12}O_6N_4$	149-151	61	67.41	4.11	10.48	67.05	3.96	10.97
VIIc	2,4,6-trichloro	$C_{30}H_{20}O_4N_3Cl_3$	177-179	63	60.75	3.37	7.08	60.39	3.12	7.23
VIId	4-Me	$C_{31}H_{25}O_4N_3$	180-182	64	73.95	4.97	8.34	74.11	4.63	8.06
VIIe	4-OMe	$C_{31}H_{25}O_5N_3$	189-191	69	71.67	4.81	8.09	71.85	4.58	8.27

Table (7) IR absorption bands (cm-1)of 1-(p-Methylphenyl)-2-styryl-3-aryl-1,2,3-trihydro-3-nitrobenzo [1,2,e][1,3]-diazepine-4,7-diones(VII).

Comp.	C-H str. Allylic	=CH st. Vinylic	C-H str. Aromatic	C-H str. Aliphatic	C=O str. Lactam	C=C str. Vinylic	C=C str. Aromatic	C-N str.	C-NO2 Aromatic	C-H bend. Aromatic	C-H bend. Allylic	Cis =CH bend. Vinylic	Others
VIIa	3175	3140	3040	2880	1720	1455	1615	1150	1530,1345	1330	1285	795	
VIIb	3180	3140	3040	2880	1720	1445	1625	1150	1520,1365	1330	1290	840	
VIIc	3180	3145	3040	2875	1725	1455	1590	1135	1510,1335	1290	1285	840	C-Cl: 810
VIId	3190	3155	3045	2875	1715	1465	1620	1130	1530,1355	1330	1285	835	
VIIe	3185	3150	3040	2870	1710	1470	1625	1130	1535,1370	1335	1285	835	C-O-C 1025

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تحضير بعض مشتقات 3,1-دايازيبين -7,4- دايونات

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الخلاصة

حضر عدد من مشتقات الـ 1و3-اوكسازيبين-4و7-دايون بتكاثف N-سيناميليدين ارين أمين مع انهدريد الماليك، انهدريد الفثاليك وانهدريد 3-نترو فثاليك.تفاعلت الأخيرة مع الأمينات الاروماتية الأولية فأعطت مشتقات 1و3-دايازيبين-4و7-دايون المقابلة.