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Synthesis of novel indenoquinoxaline derivatives as potent α-glucosidase inhibitors

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1,2-Di(11*H*-indeno[2,1-*b*]quioxalin-11-ylidene)hydrazine

ABSTRACT

A series of new *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide derivatives (**3a**–**3p**) were synthesized and evaluated for their α-glucosidase inhibitory activity. The synthesized compounds **3d**, **3f**, **3g**, **3k**, **3n**, **3p** and **4** showed significant α-glucosidase inhibitory activity as compared to acrabose, a standard drug used to treat type II diabetes. Structures of the synthesized compounds were determined by using FT-IR, ^{1}H NMR, ^{13}C NMR, mass spectrometry and elemental analysis techniques.

1. Introduction

α-Glucosidase (α-D-glucoside glucohydrolase, EC 3.2.1.20) is an enzyme that lies on the brush border surface membrane of the small intestinal cells and breaks down starch and disaccharides to liberate free glucose in the blood.[1] Compounds with α-glucosidase inhibition activity can lower the rate of glucose absorption and suppression of postprandial hyperglycemia thus is helpful in treating Type-II diabetes mellitus.[2] Acrabose,[3] miglitol[4] and voglibose[5] are clinically important α-glucosidase inhibitors and are glucosidic in nature. Similarly, iminosugars like nojirimycin[6] and 1-deoxynojirimycin[7] are well known α-glucosidase inhibitors. There are also many reports on the non-glycosidic α-glucosidase inhibitors like phenolic compounds[8,9] and tetrachlorophthalimide analogues.[10]

Quinoxaline derivatives are well known for their diverse biological activities including antimicrobial,[11] anti-hypertensive,[12] anti-tubercular,[13] anti-depressant,[14] anti-malarial,[15] anti-inflammatory,[16] anti-convulsant,[17] anti-HIV,[18] anti-diabetic[19] and anti-cancer.[20] Previously our group has reported the synthesis of novel quinoxaline derivatives and their anti-bacterial, insecticidal and phytotoxic activities.[21] There are few recent reports on the enzyme inhibition activities of quinoxalines[22,23] which prompted us to synthesize a new series of indenoquinoxaline derivatives (**3a**–**3q**, **4**) and evaluate their α-glucosidase, tyrosinase, and α-chymotrypsin inhibition activities.

2. Results and discussion

2.1. Chemistry

11*H*-Indeno[1,2-*b*]quinoxalin-11-one (**1**) was synthesized by refluxing an equimolar mixture of *o*-phenylenediamine and ninhydrin using ethanol/acetic acid (1:1) as a solvent. The benzohydrazides (**2a**–**2r**) (prepared from a series of benzoic acids) were made to react with **1** to yield a series of *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazides (**3a**–**3q**) (Scheme 1) (Table 1) and an unexpected product 1,2-Di(11*H*-indeno[2,1-*b*]quioxalin-11-ylidene)hydrazine (**4**) in case of **2r**.

The Formation of 1,2-bis(11*H*-indeno[2,1-*b*]quioxalin-11-ylidene)hydrazine (**4**) probably occurred due to the decomposition of an expected indenobenzohydrazide (**3q**) to **5** followed by reaction with **1** to yield **4** (Scheme 2).

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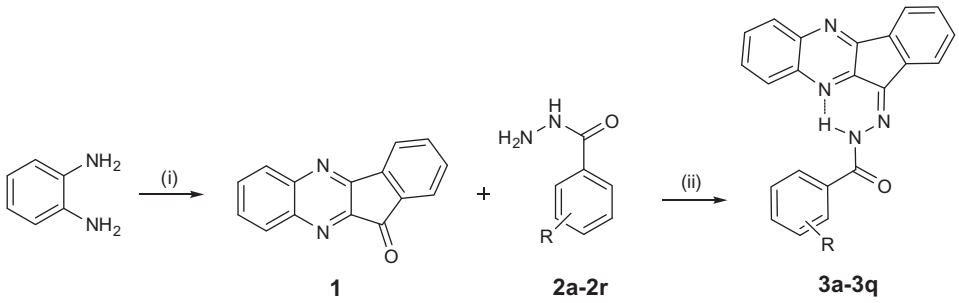
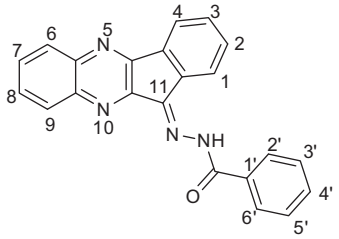


Scheme 1. Reagents and conditions: (i) Ninhydrin, ethanol/acetic acid (5:1), reflux, 3 h. (ii) Ethanol/acetic acid (1:1), reflux, 3–4 h.

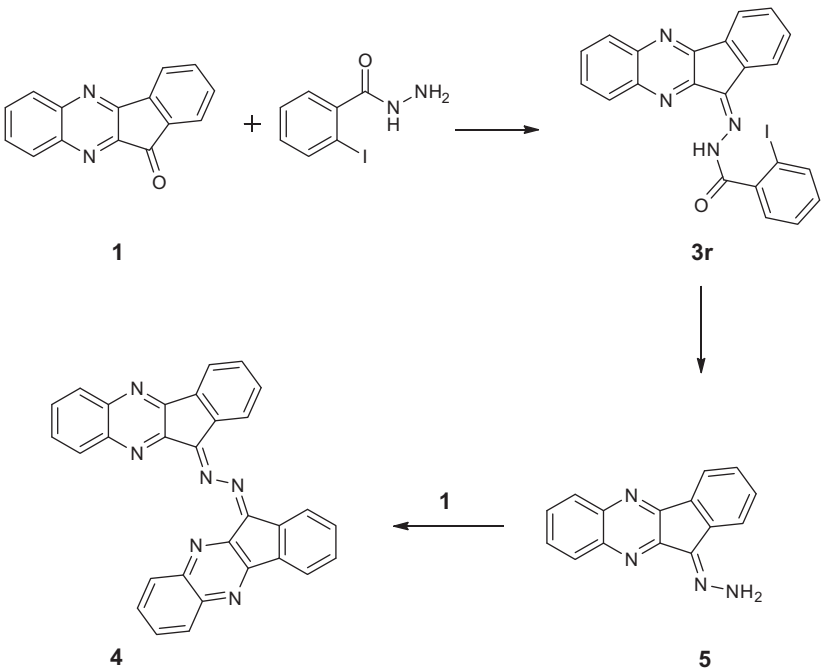
Table 1
N-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazides produced via Scheme 1

S. no.	R	2a–2q	3a–3q
1	H	**2a**	**3a**
2	3-F	**2b**	**3b**
3	4-F	**2c**	**3c**
4	2-Br	**2d**	**3d**
5	3-Br	**2e**	**3e**
6	4-Br	**2f**	**3f**
7	2-Cl	**2g**	**3g**
8	3-Cl	**2h**	**3h**
9	4-Cl	**2i**	**3i**
10	3-I	**2j**	**3j**
11	4-I	**2k**	**3k**
12	4-CH_3	**2l**	**3l**
13	2-NH_2	**2m**	**3m**
14	2-NO_2	**2n**	**3n**
15	4-NO_2	**2o**	**3o**
16	3-OH, 4-OCH_3	**2p**	**3p**
17	4-OH	**2q**	**3q**

The structures of the synthesized compounds **3a**–**3q** and **4** were established on the basis of spectral data. The mass spectra of all compounds showed a specific splitting pattern. The common peaks were at *m*/*z* 245, 217 and 190. The M^+ peaks were corresponding to the molecular weights of the compounds **3a**–**3q**. M^+ and M^++2 peaks were observed with the ratio of 3:1 and 1:1 for chloro and bromo derivatives, respectively. Other important peaks were M^+-28 and M^+-245 for all the compounds.



Similarly, the ^{1}H NMR spectra of *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazides (**3a**–**3q**) showed a singlet in the range of δ 13.00–δ 14.50 for hydrazide NH. This down field shift is probably due to the hydrogen bonding of NH proton with one of the nitrogen of quinoxaline moiety. The eight protons of indenoquinoxaline moiety appeared in the region of δ 7.00–δ 8.90 as expected. The protons of benzohydrazide moiety showed splitting patterns



Scheme 2. Synthetic route for the formation of compound **4**.

according to the position of the substituents attached. In ^{13}C NMR spectra the C=O carbon appeared in the range of δ 163.9–δ 169.9 while C=N appeared in the range of δ 153.9–δ 154.1. In compound **3b** the ^{1}H NMR spectrum showed a broad doublet for H-4′ (J = 9 Hz) and in ^{13}C NMR spectrum C-3′ showed doublet at δ 163.8 and δ 162.1 (J = 247 Hz) due to the effect of fluorine resonance. The methyl carbon can be seen at δ 21.7 in the spectrum of compound **3l**. FT-IR and elemental analyses were also found to be helpful in confirmation.

The mass spectrum of compound **4** showed molecular ion peak at m/z 460.1 corresponding to its molecular weight while the base peak was observed at m/z 431.2 probably due to the removal of nitrogen molecule. The ^{1}H NMR and ^{13}C NMR spectra were found in full agreement with the reported structure.

2.2. α-Glucosidase inhibition assay

The slightly modified method of Pierre et al. (1978)[24] was adopted for α-glucosidase inhibition assay. A 100 µl assay mixture containing 70 µl phosphate buffer saline (50 mM at pH 6.8), 10 µl corresponding test compounds (0.5 mM) and 10 µl (0.0234 units) α-glucosidase enzyme were incubated for 10 min at 37 °C and the absorbance was recorded 400 nm using Synergy HT BioTek (USA) 96-well plate reader. The reaction was initiated by the addition of 10 µl *p*-nitrophenyl-α-D-glucopyranoside (substrate, 0.5 mM, code No. N1377 from Sigma Inc). The change in absorbance of *p*-nitrophenol formed was recorded after 30 min 400 nm. Both positive and negative controls were run. Acarbose was used as positive control. All experiments were carried out in triplicate and results are mean ± SEM. For the determination of IC_{50} values, suitable dilutions of active compounds were carried out for the assay. Data was computed for the determination of IC_{50} values using EZ-Fit Enzyme kinetics software from Perrella Scientific Inc. Amherst, USA.

The inhibition percentages were calculated as follows

$$Inhibition\ (\%) = \frac{abs\ of\ control - abs\ of\ test}{abs\ of\ control} \times 100$$

α-Glucosidase (Cat No. 5003-1KU Type I) from *Saccharomyces cereviciae* has been used in the assay because of the structural and functional similarities between the yeast (eukaryote) and mammalian enzyme (eukaryote). The highest enzyme inhibition was shown by **4** with IC_{50} value 18.23 ± 0.37 µmoles/L which was even better than the standard acarbose (38.25 ± 0.12 µmoles/L). The high inhibiting activity of the **4** may be due to the presence of ylidene type structure as compared to the parent compound **1** with IC_{50} value 62.51 ± 0.35 µmoles/L (Table 2).

The presence of deactiviating groups at *ortho* and *para*-positions in the series **3a**–**3q** seem to be necessary for the potent enzyme inhibiting activity, as the corresponding un-substituted derivative **3a** was inactive. Therefore, six derivatives were found to be more active including **3f** (4-Br) > **3d** (2-Br) > **3n** (2-NO_2) > **3g** (2-Cl) > **3o** (4-NO_2) > **3k** (4-I) with IC_{50} values better than that of acarbose, **3f** being the most potent compound with IC_{50} value of 22.67 ± 0.14 µmoles/L. However, **3b** (3-F) > **3j** (3-I) > **3h** (3-Cl) > **3e** (3-Br) > **3j** (3-I) were found to be moderately active with IC_{50} values ranging between 77.45 ± 0.15 and 112.53 ± 1.45 µmoles/L. The compounds with electron donating substituents attached exhibited moderate to poor inhibitory activity among these **3m** (2-NH_2) was found to be more active with IC_{50} value 62.34 ± 1.12 µmoles/L followed by **3q** (4-OH) with IC_{50} value 102.13 ± 1.25 µmoles/L. However, **3l** (4-CH_3) and **3p** (3-OH-4-OMe) were inactive against α-glucosidase enzyme. The electron withdrawing inductive effect of chlorine seemed to be the dominating factor in case of **3g** (2-Cl) derivative with IC_{50} value 25.91 ± 0.19 µmoles/L as compared to the **3i** (4-Cl) with IC_{50} value 99.85 ± 0.32.

Table 2
α-Glucosidase inhibiting activity of *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazides (**3a**–**3q**), compounds **1** and **4**

Code no.	Substitution	Percentage inhibition at 0.5 mM	IC_{50} (µmoles/L)
1	—	97. 85 ± 0.85	62.51 ± 0.35
3a	H	7.17 ± 1.15	—
3b	3F	96.09 ± 0.55	90.92 ± 0.25
3c	4F	99.12 ± 0.95	53.42 ± 0.45
3d	2Br	99.34 ± 0.95	23.12 ± 0.75
3e	3Br	75.42 ± 1.68	112.53 ± 1.45
3f	4Br	98.37 ± 0.18	22.67 ± 0.14
3g	2Cl	99.45 ± 0.32	25.91 ± 0.19
3h	3Cl	94.48 ± 0.34	105.41 ± 0.22
3i	4Cl	88.51 ± 0.45	99.85 ± 0.32
3j	3I	94.13 ± 0.18	77.45 ± 0.15
3k	4I	93.22 ± 0.75	37.72 ± 0.45
3l	4CH_3	47.92 ± 1.25	>500
3m	2NH_2	90.78 ± 1.22	62.34 ± 1.12
3n	2NO_2	98.72 ± 2.15	23.54 ± 1.25
3o	4NO_2	99.34 ± 0.96	29.47 ± 0.99
3p	3OH-4OMe	18.04 ± 0.82	—
3q	4OH	95.42 ± 1.55	102.13 ± 1.25
4	—	96.09 ± 0.55	18.23 ± 0.37
Acarbose		92.23 ± 0.14	38.25 ± 0.12

It was also noted that the size or molecular weight of the substituents also contributed in increasing or decreasing the activity. Thus, **3c** (4-F), **3b** (3-F), **3i** (4-Cl), **3l** (4-CH_3), **3m** (2-NH_2), **3p** (3-OH-4-OMe), **3q** (4-OH) being lesser in molecular weight showed moderate to poor or no inhibition as compared to the higher molecular weight derivatives. However, **3k** (4-I) being the bulkiest showed lesser inhibition as compared to the **3f** (4-Br). These results suggested that the moderately deactivating groups at *ortho* and *para* positions enhanced the activity of the compounds while same groups at *meta* positions moderately contributed in the enzyme inhibiting activity. Also the particular molecular weight and size of the compounds contributed in increasing or decreasing the inhibition activity.

The synthesized compounds **3a**–**3q** & **4** were also evaluated for tyrosinase and α-chymotrypsin inhibition assays which showed less to moderate activity for α-chymotrypsin inhibition assay while all compounds were almost inactive against tyrosinase (data not shown).

3. Conclusion

Novel indenoquinoxaline derivatives (**3a**–**3q** and **4**) were prepared by a canonical method using easily available starting materials. The synthesized compounds were evaluated as inhibitors for the enzymatic activity of α-glucosidase which manifested a strong competitive inhibition against normal enzyme action. Amazingly, data shows that moderately deactivating substituents like Br appear to act as strong inhibitors than strongly deactivating substituents that is NO_2. From this data it might be speculated that bromo derivative **3f** fit better in the cavity of enzyme and block the active site. Noticeably, compounds **4**, **3d**, **3f**, **3g**, **3k**, **3n** and **3o** were highly active and showed better results than the standard drug acarbose so these compounds may be used as potential candidates in search for antidiabetic drugs.

4. Experimental

4.1. General methods

^{1}H NMR and ^{13}C NMR spectra of the compounds were recorded in $CDCl_3$/DMSO on a Bruker AVANCE spectrophotometer 600 MHz

and 150 MHz respectively (Otherwise stated). Chemical shifts are reported in parts per million (δ) using internal TMS standard. The IR spectra have been recorded, in film form, on a SHIMADZU FTIR 8400, between 4000 and 600 cm^{-1} with a resolution of 4 cm^{-1}. Mass spectra were measured on JEOL MS Route. Elemental analyses were carried on Perkin Elmer 2400-CHN Analyzer. Melting points were determined on a Gallenkamp melting point apparatus and are uncorrected. Thin layer chromatography was carried out on Merck silica gel $60F_{254}$ plates while column chromatography was carried out using Merck silica gel 60.

Commercial ethanol was dried over calcium carbide and distilled to get absolute ethanol. Glacial acetic acid was obtained from Merck. All substituted benzoic acids to be converted into different substituted benohydrazides were also obtained from Merck. Ninhydrin and *o*-phenylenediamine were obtained from Sigma–Aldrich.

4.2. Synthesis of 11*H*-Indeno[1,2-*b*]quinoxalin-11-one (1)

A mixture of *o*-phenylenediamine (2.16 g, 0.02 mol) and ninhydrin (3.56 g, 0.02 mol) in ethanol/acetic acid (5:1, 30 ml) was heated under reflux for 3 h. The reaction contents were cooled and the resulting yellow precipitates were filtered, washed with ethanol and dried to yield 11*H*-indeno[1,2-*b*]quinoxalin-11-one (**1**). Yield: 4.39 g, 95%, mp 195–200 °C (lit. mp: 217–219 °C).[25] IR (KBr): ν_{max}, cm^{-1}: 1730 (C=O),^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.24 (1H, d, *J* = 7.8 Hz, H-1), 8.13–8.11 (2H, m, H-9 & H-6), 7.93 (1H, d, *J* = 7.8 Hz, H-4), 7.83 (1H, t, *J* = 7.2 Hz, H-2), 7.79-7.74 (2H, m, H-7 & H-8), 7.61 (1H, t, *J* = 7.2, Hz H-3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ 189.9 (C-11), 156.6 (C-10a), 149.3 (C-9a), 143.1 (C-4b), 142.6 (C-5a), 141.5 (C-4a), 136.8 (C-11a), 136.6 (C-1), 132.6 (C-9), 132.5 (C-6), 131.6 (C-3), 130.3(C-8), 129.6 (C-7), 124.8 (C-2), 122.5 (C-4). EI-MS (*m*/*z*, %): 231.9 (M^+, 100). 204.0 (M^+−28, 94.9), 177.0 (204-HCN, 59.2). Anal. Calcd for $C_{15}H_8N_2O$ (232.24): C, 77.58; H, 3.47; N, 12.06. Found: C, 77.41; H, 3.64; N, 12.18.

4.3. General method for the synthesis of *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazides (3a–3q)

An equimolar mixture of 11*H*-indeno[1,2-*b*]quinoxalin-11-one (**1**) (0.232 g, 1 mmol) and a benzohydrazide (**2a**–**2q**) (1 mmol) was refluxed in ethanol/acetic acid (1:1, 20 ml) for 3–4 h. The resulting precipitates were filtered while hot, washed with hot ethanol and dried. The solid obtained was further purified by preparative TLC using chloroform as a solvent.

4.3.1. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3a)

Off white solid, yield: 63%, mp 198–200 °C, IR (KBr): ν_{max}, cm^{-1}: 3215 (NH, amide), 3061 (Ar-H), 1680 (C=O), 1560 (C=N).^{1}H NMR($CDCl_3$, 600 MHz): δ_H: 14.29 (1H, s, NH), 8.14–8.23 (5H, m, H-2′, H-6′, H-1, H-9, H-6), 8.08 (1H, d, *J* = 7.8 Hz, H-4), 7.86 (1H, dt, *J* = 7.5 Hz, 1.2 Hz, H-8), 7.81 (1H, dt, *J* = 7.5 Hz, 1.2 Hz, H-7), 7.59-δ7.69 (5H, m, H-4′, H3′, H5′, H-3, H-2). ^{13}C NMR($CDCl_3$, 150 MHz): δ_C: 164.3 (C=O), 154.0 (C=N), 147.9 (C-4b), 142.9 (C-10a), 142.0 (C-9a), 140.0 (C-5a), 138.1(C-4a), 135.8 (C-1′), 132.8 (C-4′), 132.7 (C-3), 132.4 (C-1), 131.5 (C-8 & C-7), 130.0 (C-3′ & C-5′), 129.8 (C-11a), 129.2 (C-9), 129.0 (C-6), 127.9 (C-2), 122.8 (C-4), 122.5 (C-2′, C-6′). EI-MS (*m*/*z*, %): 350.0 (M^+, 8.2), 322.1 (M^+−28, 3.4), 245.0 (M^+−105, 82.5), 217.0 (M^+−133, 100), 190.1 (217-HCN, 16.8) 105.1 (M^+−245, 30.8). Anal. Calcd for $C_{22}H_{14}N_4O$ (350.37): C, 75.42; H, 4.03; N, 15.03. Found: C, 75.78; H, 3.87; N, 15.29.

4.3.2. 3-Fluoro-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3b)

Yellow solid, yield 81%. Mp 250–252 °C.IR (KBr): ν_{max}, cm^{-1}: 3233.6 (NH, amide), 3076.46 (Ar-H), 1718.58 (C=O), 1564.2 (C=N), 1251.8 (C-F). ^{1}H NMR($CDCl_3$, 600 MHz):δ_H: 14.32 (IH, s, NH), 8.12–8.21 (3H, m, H-9, H-6, H-2′), 8.07 (1H, d, *J* = 7.8 Hz, H-6′), 7.98 (1H, d, *J* = 7.2 Hz, H-1), 7.91 (1H, broad d *J* = 9 Hz, H-4), 7.85 (1H, t, *J* = 7.2 Hz, H-8), 7.81 (1H, t, *J* = 7.2 Hz, H-7), 7.58-7.67 (3H, m, H-4′, H-5′, H-2), 7.38 (1H, m, H-3). ^{13}C NMR ($CDCl_3$, 150 MHz): δ_C: 163.8 & 162.1 (d, C-F), 162.8 (C=O), 154.1 (C=N), 147.8 (C-4b), 143.4 (C-10a), 142.2 (C-9a), 139.9 (C-5a), 137.9 (C-4a), 135.9 (C-3), 132.4 (C-1′), 131.7 (C-8), 131.6 (C-7), 131.7 (d, C-5′), 130.2 (C-1), 129.9 (C-11a), 129.1 (C-2), 123.5 (C-4), 122.9 (C-6′), 122.5 (C-9 & C-6), 119.8 (d, C-4′), 115.1 (d, C-2′). EI-MS (*m*/*z*, %): 368.4 (M^+, 6.9), 369.4 (M^++1, 2.1), 245.3 (M^+−123, 80.6), 217.2 (M^+−152, 100), 190.2 (217-HCN, 15.1), 123.1 (M^+−245, 10.5). Anal. Calcd for $C_{22}H_{14}FN_4O$ (368.36): C, 71.73; H, 3.56; N, 15.21. Found: C, 71.68, H, 3.65, N, 15.07.

4.3.3. 4-Fluoro-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3c)

Yellow solid; yield: 83%. Mp 208–210 °C.IR (KBr): ν_{max}, cm^{-1}: 3215.34 (NH, amide), 3053.32 (Ar-H), 1710.86 (C=O), 1602.85 (C=N), 1255.66 (C-F). ^{1}H NMR($CDCl_3$ 600 MHz): δ_H: 14.30 (1H, s, NH), 8.18–8.25 (5H, m, H-9, H-6, H-2′, H-6′, H-1), 8.08 (1H, d, *J* = 8.4 Hz, H-4), 7.88(1H, dt, *J* = 7.2 Hz, 1.2 Hz, H-8), 7.83 (1H, t, *J* = 7.2 Hz, 1.2 Hz, H-7), 7.63 (2H, m, H-3′ & H-5′), 7.32 (2H, m, H-2 & H-3). EI-MS (*m*/*z*, %): 368.3 (M^+, 25.6), 369.3 (M^++1, 17.3), 340.2 (M^+−28, 5.5), 245.0 (M^+−123, 100), 217.0 (M^+−152, 100), 190.1 (217-HCN, 59.3), 123.1 (M^+−245, 93.9). Anal. Calcd for $C_{22}H_{13}FN_4O$ (368.36): C, 71.73; H, 3.56; N, 15.21. Found: C, 71.97; H, 3.69; N, 14.99.

4.3.4. 2-Bromo-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3d)

Yellow solid. Yield: 69%. Mp 186–188 °C.IR (KBr): ν_{max}, cm^{-1}: 3192.19 (NH, amide), 3055.24 (Ar-H), 1691.57 (C=O), 1552.70 (C=N), 759.95 (C-Br). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 13.26 (1H, s, NH), 8.16-8.25 (3H, m, H-9, H-6 & H-3′), 7.81-7.86 (3H, m, H-6′, H-1 & H-4), 7.69-7.74 (2H, m, H-8 & H-7), 7.53-7.60 (2H, m, H-4′ & H-5′), 7.43-7.49 (2H, m, H-2 & H-3). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 164.8 (C=O), 154.0 (C=N), 147.7 (C-4b), 143.4 (C-10a), 142.2 (C-5a), 140.0 (C-9a), 137.9 (C-4a), 136.1 (C-1′), 133.9 (C-4′), 132.4 (C-8), 132.3 (C-7), 131.7 (C-3′), 131.4 (C-3), 130.9 (C-11a), 130.6 (C-6′), 129.8 (C-1), 129.7 (C-9), 129.5 (C-6), 127.8 (C-2), 122.9 (C-5′), 122.4 (C-4), 119.9 (C-2′). EI-MS (*m*/*z*, %): 427.9 (M^+, 5.3), 430.0 (M^++2, 5.5), 399.9 (M^+−28, 1.7), 402.1 (M^++2−28, 1.4), 245 (M^+−182.9, 100), 217.0 (M^+−211, 100), 190.0 (217-HCN, 33.5), 184.9 (M^++2−245, 18.2), 183.0 (M^+−245, 18.6). Anal. Calcd for $C_{22}H_{13}BrN_4O$ (429.27): C, 61.55; H, 3.05; N, 13.05. Found: C, 61.89; H, 2.81; N, 13.32.

4.3.5. 3-Bromo-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3e)

Yellow solid. Yield. 29%, mp 188–190 °C.IR (KBr): ν_{max}, cm^{-1}: 3223.05 (NH, amide), 3057.17 (Ar-H), 1705.07 (C=O), 1562.34 (C=N), 682.80 (C-Br). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.39 (1H, s, NH), 8.37 (1H, s, H-2′), 8.14–8.21 (5H, m, H-9, H-6, H-6′, H-4′ & H-1), 7.80–7.88 (3H, m, H-5′, H-8 & H-7), 7.62–7.63 (2H, m, H-4 & H-3), 7.52 (1H, t, *J* = 7.5 Hz, H-2). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 162.6 (C=O), 154.1 (C=N), 147.8 (C-4b), 143.6 (C-10a), 142.3 (C-5a), 139.9 (C-9a), 137.7 (C-1′), 136.1 (C-4′), 135.5 (C-3), 134.6 (C-4a), 132.4 (C-5′), 131.7 (C-11a), 131.6 (C-1), 130.7 (C-8), 130.6 (C-7), 130.2 (C-9), 129.9 (C-6), 129.3 (C-2′), 126.8 (C-6′), 123.1 (C-3′), 122.4 (C-2 & C-4). EI-MS (*m*/*z*, %): 428.0 (M^+, 7.8), 430.1 (M^++2, 8.2), 402 (M^++2−28, 1.7), 400 (M^+−28, 1.7), 245 (M^+−183, 92.5), 217 (M^+−211, 100), 190.1 (217-HCN, 33.5), 184.9 (M^++2−245, 13.1), 183 (M^+−245, 13.8). Anal. Calcd for $C_{22}H_{13}BrN_4O$ (429.27): C, 61.55; H, 3.05; N, 13.05. Found: C, 61.34; H, 3.23; N, 13.31.

4.3.6. 4-Bromo-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3f)

Yellow solid. Yield: 74%. Mp 245–248 °C. ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.29 (1H, s, NH), 8.19-8.23 (2H, m, H-9 & H-6), 8.12-8.14 (1H, m, H-2′& H-6′), 8.05-8.06 (3H, m, H3′ & H5′), 7.78-7.87 (4H, m, H-7, H-8, H-1, H-4), 7.61 (2H, m, H-2 & H-3).^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 163.4 (C=O), 154.1 (C=N), 147.8 (C-4b), 143.3 (C-10a), 142.2 (C-5a), 139.8 (C-9a), 137.8 (C-1′), 135.9 (C-3′, C-5′), 133.1 (C-4a), 132.4 (C-11a), 132.3 (C-3), 131.6 (C-8), 131.5 (C-7), 130.1 (C-1), 129.8 (C-9), 129.3 (C-6), 129.1 (C-2), 127.6 (C-4′), 122.8 (C-4), 122.5 (C2′& C6′).EI-MS (*m*/*z*, %): 429.9 (M^+, 3.5) 431.1 (M^++1, 6.3), 432.0 (M^++2, 5.6) 400.1 (M^+−28, 0.8), 245.0 (M^+−184.9, 88.8), 217.0 (M^+−212.9, 100), 190.1 (217-HCN, 16.8) 184.9 (M^++2−245, 10.5) 182.9 (M^+−245, 11.2). Anal. Calcd for $C_{22}H_{13}BrN_4O$ (429.27): C, 61.55; H, 3.05; N, 13.05. Found: C, 61.72; H, 3.14; N, 12.89.

4.3.7. 2-Chloro-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3g)

Yellow solid. Yield: 76%. Mp 180–183 °C. IR (KBr): ν_{max}, cm^{-1}: 3415.93 (NH, amide), 1774.51 (C=O), 1681.93 (C=N), 715.59 (C-Cl). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.00 (1H, s, NH), 8.23–8.26 (3H, m, H-9, H-6, H-1), 7.75–8.01 (3H, m, H-7, H-8, H-4), 7.41–7.64 (6H, m, H-3′, H-4′, H-5′, H-6′, H-2, H-3). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 163.8 (C=O), 153.9 (C=N), 143.2 (C-4b), 141.2 (C-10a), 140.6 (C-9a), 140.3 (C-5a), 138.3 (C-4a), 135.2 (C-4′), 134.1 (C-1′), 132.7 (C-2′), 132.7 (C-3), 131.8 (C-8), 131.7 (C-7), 131.3 (C-1), 131.0 (C-11a), 130.7 (C-9), 130.1 (C-6), 129.6 (C-3′), 129.4 (C-6′), 127.4 (C-2), 123.0 (C-4), 121.8 (C-5′). EI-MS (*m*/*z* %): 384.0 (M^+, 6.3), 385.0 (M^++1, 3.0), 386.0 (M^++2, 2.5), 356.0 (M^+−28, 1.2), 245.0 (M^+−139, 97.3), 217.0 (M^+−167, 100), 190.1 (217-HCN, 20.3), 140.9 (M^++2−245, 6.8), 138.9 (M^+−245, 20.9). Anal. Calcd for $C_{22}H_{13}ClN_4O$ (384.82): C, 68.67; H, 3.41; N, 14.56. Found: C, 68.89; H, 3.47; N, 14.44.

4.3.8. 3-Chloro-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3h)

Yellow solid. Yield: 72%. Mp 240–242 °C.IR (KBr): ν_{max} cm^{-1}: 3236.55 (NH, amide), 3074.53 (Ar-H), 1718.58 (C=O), 1566.20 (C=N), 759.95 (C-Cl). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.38 (1H, s, NH), 8.21–8.22 (3H, m, H-9, H-6, H-2′), 8.15–8.17 (2H, m, H-6′, H-1), 8.11 (1H, d, *J* = 7.2 Hz, H-4′), 7.87 (1H, dt, *J* = 7.5 Hz, 1.2 Hz, H-8), 7.83 (1H, dt, *J* = 7.5 Hz, 1.2 Hz, H-7), 7.57–7.66 (4H, m, H-5′, H-4, H-2 & H-3).^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 162.7 (C=O), 154.1 (C=N), 147.9 (C-4b), 143.5 (C-10a), 142.2 (C-5a), 139.9 (C-9a), 137.8 (C-4a), 135.9 (C-1′), 135.1 (C-3′), 134.5 (C-11a), 132.7 (C-4′), 132.4 (C-3), 131.7 (C-8), 131.6 (C-7), 130.4 (C-5′), 130.2 (C-1), 129.8 (C-9), 129.2 (C-6), 127.8 (C-2), 126.3 (C-4), 122.9 (C-2′), 122.6 (C-6′). EI-MS (*m*/*z*):384.0 (M^+, 3.3), 385.1 (M^++1, 1.3), 386.0 (M^++2, 1.3), 245.2 (M^+−139, 57.5), 217.2 (M^+−167, 100), 190.3 (217-HCN, 13.6), 141.2 (M^++2−245, 2.7), 139.2 (M^+−245, 8.4). Anal. Calcd for $C_{22}H_{13}ClN_4O$ (384.82): C, 68.67; H, 3.41; N, 14.56. Found: C, 68.73; H, 3.26; N, 14.81.

4.3.9. 4-Chloro-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3i)

Yellow solid. Yield: 78%. Mp: 234–238 °C.IR (KBr): ν_{max} cm^{-1}: 3199.91 (NH, amide), 3059.10 (Ar-H), 1703.14 (C=O), 1637.56 (C=N), 756.10 (C-Cl). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.30 (1H, s, NH), 8.20 (2H, m, H-9 & H-6), 8.14 (3H, m, H-2′, H-6′, H-1), 8.05 (1H, d, *J*=7.2 Hz, H-4), 7.87 (1H, t, *J* = 7.2 Hz, H-8), 7.82 (1H, t, *J* = 7.2 Hz, H-7), 7.62 (4H, m, H-3′, H-5′, H-2, H-3).^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 163.3 (C=O), 154.2 (C=N), 147.9 (C-4b), 143.2 (C-10a), 142.2 (C-9a), 139.9 (C-5a), 139.1 (C-4a), 137.9 (C-1′), 135.9 (C-4′), 132.4 (C-3), 131.7 (C-8), 131.6 (C-7), 131.2 (C-11a), 130.1 (C-2′ & C-6′), 129.9 (C-1), 129.3 (C-9), 129.2 (C-6) 129.1 (C-2), 122.9 (C-4′), 122.5 (C3′ & C5′). EI-MS (*m*/*z* %):384.0 (M^+, 3.8), 385.0 (M^++1, 1.6), 386.0 (M^++2, 1.4), 356.0 (M^+−28, 1), 245.2 (M^+−139, 72.2), 217.2 (M^+−167, 100), 190.2 (217-HCN, 14.8), 141.1 (M^++2−245, 5.8), 139.1 (M^+−245, 18.9). Anal. Calcd for $C_{22}H_{13}ClN_4O$ (384.82): C, 68.67; H, 3.41; N, 14.56. Found: C, 68.78; H, 3.23; N, 14.61.

4.3.10. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)-3-iodobenzohydrazide (3j)

Yellow solid. Yield: 80%. Mp 246–248 °C.IR (KBr): ν_{max} cm^{-1}: 3217.27 (NH, amide), 3120.00 (Ar-H), 1701.22 (C=O), 1589.34 (C=N), 629.44 (C-I).^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.36 (1H, s, NH), 8.55 (1H, s, H-2′), 8.27 (1H, d, *J* = 7.5 Hz, H-6′), 8.21–8.22 (3H, m, H-9, H-6, H4′), 8.16 (1H, d, *J* = 8.4 Hz, H-1), 8.00 (1H, d, *J* = 7.2 Hz, H-4), 7.87 (1H, t, *J* = 7.2 Hz, H-8), 7.83 (1H, t, *J* = 7.2 Hz, H-7), 7.62 (2H, m, H-2 & H-3), 7.38 (1H, t, *J* = 7.5 Hz, H-5′). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 162.5 (C=O), 154.0 (C=N), 147.9 (C-4b), 143.5 (C-10a), 142.1 (C-9a), 141.5 (C-4′), 139.9 (C-5a), 137.8 (C-4a), 136.3 (C-2′), 135.9 (C-1′), 134.7 (C-11a), 132.4 (C-3), 131.7 (C-8), 131.6 (C-7), 130.8 (C-5′), 130.2 (C-1), 129.8 (C-9), 129.5 (C-6), 127.6 (C-2), 122.9 (C-4), 122.6 (C-6′), 94.5 (C-3′). EI-MS (*m*/*z*): 476.3 (M^+, 25.5), 477.3 (M^++1, 13.6), 478.3 (M^++2, 2.7), 448.2 (M^+−28, 10.1), 245.1 (M^+−231, 95.7), 231.0 (M^+−245, 37.1), 217 (M^+−259.3, 100), 190.1 (217-HCN, 51.9). Anal. Calcd for $C_{22}H_{13}IN_4O$ (476.27): C, 55.48; H, 2.75; N, 11.67. Found: C, 55.21; H, 3.01; N, 11.59.

4.3.11. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)-4-iodobenzohydrazide (3k)

Yellow solid. Yield: 70%. Mp 230–232 °C. IR (KBr): ν_{max} cm^{-1}: 3221.12 (NH, amide), 3047.43 (Ar-H), 1707.00 (C=O), 1585.49 (C=N), 765.74 (C-I). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.33 (1H, s, NH), 8.29–8.13 (3H, m, H-9, H-6, H-1), 7.91-8.10 (5H, m, H-3 , H-5′, H-2′, H-6′, H-4), 7.80–7.90 (2H, m, H-8, H-7), 7.59-7.69 (2H, m, H-2, H-3). EI-MS (*m*/*z*): 476.4 (M^+, 6.1), 477.3 (M^++1), 448.3 (M^+−28, 2.4), 245.2 (M^+−231, 91.4), 231.1 (M^+−245, 17.2), 217.2 (M^+−259.3, 100), 190.2 (217-HCN, 12.1). Anal. Calcd for $C_{22}H_{13}IN_4O$ (476.27): C, 55.48; H, 2.75; N, 11.67. Found: C, 55.67; H, 2.98; N, 11.43.

4.3.12. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)-4-methylbenzohydrazide (3l)

Yellow solid. Yield: 69%. Mp 198–200 °C. IR (KBr): ν_{max} cm^{-1}: 3224.98 (NH, amide), 3043.67 (Ar-H), 1699.29 (C=O), 1608.63 (C=N). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.27 (1H, s, NH), 8.09–8.22 (6H, m, H-9, H-6, H-2′, H-6′, H-1 & H-4), 7.86 (1H, t, *J* = 7.5 Hz, H-8), 7.80 (1H, t, *J* = 7.5 Hz, H-7), 7.61 (2H, m, H-2 & H-3), 7.44 (2H, d, *J* = 7.2 Hz, H-3′ & H-5′), 2.51 (3H, s, H-CH_3). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 164.2 (C=O), 154.0 (C=N), 148.1 (C-4b), 143.4 (C-10a), 142.6 (C-9a), 141.9 (C-5a), 140.0 (C-4′), 138.2 (C-4a), 135.7 (C-3′ & C-5′), 132.4 (C-3), 131.5 (C-2′ & C-6′), 131.4 (C-1′), 130.0 (C-8 & C-7), 129.8 (C-9), 129.7 (C-6), δ129.2 (C-11a), 127.9 (C-1), 122.8 (C-2), 122.5 (C-4), 21.7 (C-CH_3). EI-MS (*m*/*z*, %): 364.3 (M^+, 11.6), 336.2 (M−28, 7), 245.2 (M−119.1, 83), 217.2 (M−147, 100), 190.1 (217-HCN, 9), 119.1 (M−245.2, 46.6). Anal. Calcd for $C_{23}H_{16}N_4O$ (364.40): C, 75.81; H, 4.43; N, 15.38. Found: C, 76.01; H, 4.29; N, 15.52.

4.3.13. 2-Amino-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3m)

Red solid. Yield: 85%. Mp 228–230 °C. IR (KBr): ν_{max} cm^{-1}: 3560.59 (NH, amide), 3417.86 & 3309.85 (NH_2), 3055.24 (Ar-H), 1680.00 (C=O), 1554.63 (C=N), 1242.16 (C-N).^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.29 (1H, s, NH),8.13–8.26 (4H, m, H-9, H-6, H-1, H-6′), 7.92–7.97 (2H, m, H-7 & H-8), 7.75–7.86 (3H, m, H-4, H-3, H-2), 7.61-7.63 (3H, m, H-4′, H-5′, H-3′). EI-MS (*m*/*z* %): 365.1 (M^+, 57.1), 337.0 (M^+−28, 1.6), 245.0 (M^+−120, 83.2), 217.0

(M^+−149, 100), 190.0 (217-CHN, 13), 120.0 (M^+−245, 100). Anal. Calcd for $C_{22}H_{15}N_5O$ (365.39): C, 72.32; H, 4.14; N, 19.17. Found: C, 72.01; H, 3.97; N, 19.34.

4.3.14. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)-2-nitrobenzohydrazide (3n)

Yellowish green solid. Yield 50%. Mp 268–270 °C. IR (KBr): ν_{max} cm^{-1}: 3190.26 (NH, amide), 3068.75 (Ar-H), 1685.79 (C=O), 1610.56 (C=N), 1525.69 (N=O, nitro).^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 13.23 (1H, s, NH), 8.24 (1H, d, J = 8.4 Hz, H-3′), 8.21 (1H, d, J = 7.8 Hz, H-6′), 8.16 (1H, d, J = 7.8 Hz, H-9), 8.09 (1H, d, J = 7.8 Hz, H-6), 7.62–7.85 (4H, m, H-4′, H-5′, H-1, H-8), 7.72 (1H, t, J = 7.8 Hz, H-7), 7.53 (1H, dt, J = 7.2 Hz,1.2 Hz, H-3), 7.45–7.50 (2H, m, H-2 & H-4). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 169.6 (C=O), 153.8 (C=N), 147.7 (C-2′), 147.2 (C-4b), 142.1 (C-10a), 140.3 (C-9a), 140.2 (C-5a), 137.6 (C-4a), 136.1 (C-5′), 132.0 (C-4′), 131.5 (C-8), 131.4 (C-7), 130.8 (C-3), 130.3 (C-1), 130.2 (C-9), 130.1 (C-6), 129.8 (C-2), 129.5 (C-6′), 123.6 (C-4), 122.5 (C-3′), 121.8 (C-1′). EI-MS (m/z): 395.0 (M^+, 4.9), 245.0 (M^+−150, 66.7), 217.0 (M^+−178, 100), 190.1 (217-HCN, 34.2), 150.1 (M^+−245, 2.0). Anal. Calcd for $C_{22}H_{13}N_5O_3$ (395.37): C, 66.83; H, 3.31; N, 17.71. Found: C, 66.67; H, 3.59; N, 17.52.

4.3.15. *N*-(11*H*-Indeno[1,2-*b*]quinoxalin-11-ylidene)-4-nitrobenzohydrazide (3o)

Yellow solid. Yield: 66%. Mp 265–270 °C. IR (KBr): ν_{max} cm^{-1}: 3196.05 (NH, amide), 3039.81 (Ar-H), 1707.00 (C=O), 1583.56 (C=N), 1514.12 (N=O, nitro).^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.44 (1H, s, NH), 8.09–8.51 (7H, m, H-3′, H-5′, H-2′, H-6′, H-9, H-6 & H-1), 7.86–8.01 (3H, m, H-4, H-8 & H-7), 7.66-7.76 (2H, m, H-2 & H-3). EI-MS (m/z, %):395.2 (M^+, 4.1), 245.2 (M^+−150, 59), 217.2 (M^+−178, 100), 190.1 (217-HCN, 13.4), 150.1 (M^+−245, 1.7). Anal. Calcd for $C_{22}H_{13}N_5O_3$ (395.37): C, 66.83; H, 3.31; N, 17.71. Found: C, 66.99; H, 3.02; N, 17.90.

4.3.16. 3-Hydroxy-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)-4-methoxybenzohydrazide (3p)

Yellow solid. Yield:78%. Mp 274–277 °C. IR (KBr): ν_{max} cm^{-1}: 3234.62 (broad peak, NH, amide & OH), 3074.53 (Ar-H), 1691.57 (C-O), 1570.06 (C=N), 1284.59 (C-O).^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 14.30 (1H, s, NH), 8.23–8.30 (3H, m, H-9, H-6, H-1), 8.12 (1H, d, J = 7.8 Hz, H-6′), 7.83- 7.90 (2H, m, H-7, H-8), 7.77 (1H, s, H-2′), 7.75 (1H, d, J = 8.4 Hz, H-4), 7.64 (2H, m, H-2&H-3), 7.16 (1H, d, J = 7.8 Hz, H5′), 4.07 (3H, s, OCH_3). ^{13}C NMR ($CDCl_3$ 150 MHz): δ 56.3 (C-OMe). EI-MS (m/z, %): 396.0 (M^+, 11.8), 367.9 (M-28, 12.2), 245.0 (M-151, 69.7), 217.0 (M-179, 100), 190.0 (217-HCN, 12.3) 151.0 (M-245, 87.4). Anal. Calcd for $C_{23}H_{16}N_4O_3$ (396.40): C, 69.69; H, 4.07; N, 14.13. Found: C, 69.45; H, 3.98; N, 13.99.

4.3.17. 4-Hydroxy-*N*-(11*H*-indeno[1,2-*b*]quinoxalin-11-ylidene)benzohydrazide (3q)

Pale yellow solid. Yield: 77%. Mp 315–320 °C. IR (KBr): ν_{max} cm^{-1}: 3298.28 (OH), 3200.00 (NH, amide), 3068.75 (Ar-H), 1666.50 (C=O), 1602.85 (C=N), 1267.23 (C-O). ^{1}H NMR (DMSO 300 MHz): δ_H: 13.96 (1H, s, NH), 10.44 (1H, s, OH), 8.20–8.23 (2H, m, H-9 & H-6), 8.11–8.13 (1H, m, H-1), 7.93–8.00 (5H, m, H-2′, H-6′, H4, H-7 & H-8), 7.68–7.70 (2H, m, H-2 & H-3), 7.07 (2H, d, J = 8.7 Hz, H-5′ & H-3′). ^{13}C NMR (DMSO100 MHz): δ_C: 161.6 (C=O & C-4′), 153.3 (C=N), 141.3 (C-4b & C-10a), 139.4 (C-5a & C-9a), 137.8 (C-4a), 135.4 (C-11a), 132.5 (C-3), 131.6 (C-8), 131.3 (C-7), 130.4 (C-1), 129.6 (C-2′ & C-6′), 129.3 (C-9 & C-6), 122.7 (C-1′), 122.3 (C-2), 121.6 (C-4), 115.9 (C-5′ & C-3′). EI-MS (m/z %):366.1 (M^+, 9), 338.1 (M^+−28, 3.7), 245.1 (M^+−121, 73.8), 217.1 (M^+−149, 100), 190.1 (217-CHN, 12.1), 121.0 (M^+−245, 56.3). Anal. Calcd for $C_{22}H_{14}N_4O_2$ (366.37): C, 72.12; H, 3.85; N, 15.29. Found: C, 71.99; H, 3.67; N, 15.43.

4.3.18. 1,2-Di(11*H*-indeno[2,1-*b*]quioxalin-11-ylidene)hydrazine (4)

Yellow solid. Yield: 25%. Mp 288–290 °C. IR (KBr): ν_{max} cm^{-1}: 1666.50 (C=N), 1544.98-1402.25 (aromatic region). ^{1}H NMR ($CDCl_3$ 600 MHz): δ_H: 8.39 (2H, d, J = 7.8 Hz, H-9 & H-9′), 8.33 (2H, d, J = 7.8 Hz, H-6 & H-6′), 8.23 (2H, d, J = 7.8 Hz, H-1 & H-1′), 8.21 (2H, d, J = 8.4 Hz, H-4 & H-4′), 7.83 (4H, p, J = 7.8 Hz, H-8, H-8′, H-7 & H-7′), 7.64 (2H, t, J = 7.5 Hz, H-2 & H-2′), 7.54 (2H, t, J = 7.5 Hz, H-3 & H-3′). ^{13}C NMR ($CDCl_3$ 150 MHz): δ_C: 154.3 (C-11 & C-11′), 150.8 (C-4b & C-4′b), 149.2 (C-10a & C-10′a), 142.9 (C-9a & C-9′a), 142.5 (C-5a & C-5′a), 138.4 (C-4a & C-4′a), 133.6 (C-11a & C-11′a), 133.1 (C-3 & C-3′), 132.1 (C-1 & C-1′), 130.9 (C-8 & C-8′), 130.7 (C-7 & C-7′), 130.0 (C-9 & C-9′), 129.8 (C-6 & C-6′), 129.5 (C-2 & C-2′), 122.5 (C-4 & C-4′). EI-MS (m/z, %): 460.1 (M^+, 29), 431.2 (M^+−28, 100), 231.1 (M^+−229, 22.1), 216.1 (M^+−244, 22.2). Anal. Calcd for $C_{30}H_{16}N_6$ (460.49): C, 78.25; H, 3.50; N, 18.25. Found: C, 78.09; H, 3.46; N, 18.49.

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Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.bmc.2013.12.024.

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