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Tetramethylguanidinium triflate: An efficient catalyst solvent for the convergent synthesis of fused spiro[1,4-dihydropyridine-oxindole] compounds

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ABSTRACT

Synthesis of spiro[1H-indeno[1,2-b]benzoquinolin-13,3′-indoline]-7,13-dihydro-12,2′-dione derivatives was achieved expediently through three-component reactions between 2H-indene-1,3-dione, 1- or 2-naphthalenamine, and isatin derivatives under catalysis of the ionic liquid *N,N,N,N*-tetramethylguanidinium triflate. The ionic liquid appeared as a task-specific catalyst solvent being amenable to successive recycling without appreciable decrease in activity. This synthetic method similarly works well when acenaphthylene-1,2-dione or 1H-indene-1,2,3-trione is employed in place of isatin and likewise when 2-aminouracil is used instead of naphthalenamine to furnish the structurally related spirocyclic products. A convergent mechanism was suggested for the reactions to account for the selective formation of the spiro-compounds corresponding to the employed components.

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1. Introduction

Recent decades have witnessed a resurgence of interest in development of economical procedures, especially those having low chemical impact on environment [1]. Owing to this interest, a plethora of researches has been directed toward extension of diverse multi-component reactions (MCRs). The inherent convergence and high productive nature of MCRs along with their exploratory and complexity-generating power have popularized them as a context for exploring novel eco-friendly synthetic processes [2]. In this direction, designing MCRs without employing toxic catalysts and solvents, or at least using recoverable solvents such as ionic liquids, is particularly worthwhile; as such considerations should also complement significant characters of MCRs to satisfy the green chemistry's principles [3]. Although ionic liquids were initially introduced as alternative green reaction media, due to their nonvolatility, nonflamability, thermal stability, and controlled miscibility properties [4], today they have received the recognition that can control many reactions as catalysts [5]. Ionic liquids, by virtue of their organic and ionic nature, are potent solvents, exerting nearly all kinds of interactions on reacting species, including transition states, whereupon sometimes give rise to improved yields and rate enhancements [6]. Structural variation of ionic liquids gives more flexibility to their applications, as provides fine tunning of their miscibility to merit phase-separation from products [4]. Moreover, functionalized ionic liquids offer the task-specific elements for designed catalysis applications [7]. In this view and in line with our interest in performing reactions with the aid of ionic liquids [8,9], and also in continuation of our studies on synthesis of spiro-oxindole heterocycles [10,11], herein we report an expedient synthesis of some fused spiro[1,4-dihydropyridine-oxindole] compounds under catalysis of a task-specific ionic liquid. The heterocyclic spiro-oxindole ring system is a widely distributed structural framework present in a number of pharmaceuticals and natural products [12–14]. Spiro[pyrrolidine-oxindole] ring systems, for example, are found in a number of alkaloids such as horsifiline, spirotryprostatine A and B and elacomine [15]. This prevalence has led to interests in development of new methods for construction of various diversely functionalized spirocylic oxindoles [16–18].

2. Results and discussion

Our recent investigations on synthesis of spiro[1,4-dihydropyridine-oxindole] compounds using the three-component reaction between an isatin derivative, 2*H*-indene-1,3-dione, and 2-naphthalenamine, revealed a trend in which more solvent polarity led to a pronounced increase in yield [11]. However, water appeared a less useful solvent for this reaction, presumably due to its reduced solubility, requiring to be improved by mixing with co-solvents, which certainly would reduce its polarity. Beside the important role of solvent, the presence of an acid catalyst, i.e. *p*-TSA, was also found crucial for implementing the reaction. Bearing these findings in mind and considering the practical advantages of ionic liquids, we planned to verify the possible promotion of the reaction in ionic liquids, specially in those that can satisfy all the

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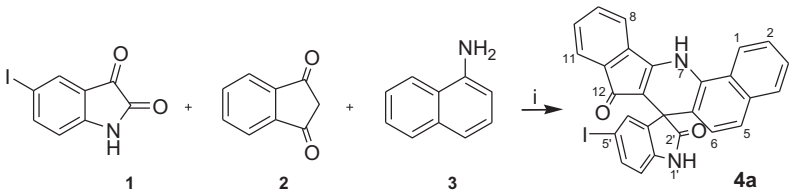
Table 1
Optimization of the reaction condition .



i = Ionic liquid, 80 °C

Entry	Catalyst	Time (min)	Yield (%)
1	–	300	Trace
2	[BMIm]BF_4	60	30
3	[BMIm]Br	60	24
4	$TMGT_f$	5	95
5	TMGT	15	88
6	[HMIm]/HSO_4	35	68
7	[BMIm]BF_4/*p*-TSA	30	62

requirements by themselves. In this regard, our attention turned toward application of two Brønsted acid–base ionic liquids, *N*,*N*,*N*,*N*-tetramethylguanidinium triflate ($TMGT_f$) and *N*,*N*,*N*,*N*-tetramethylguanidinium trifluoroacetate (TMGT), which are remarkably polar room temperature liquids composed of organic acid catalysts. Preliminary expeiments on screening the model reaction involving 5-iodoisatin **1**, 2*H*-indene-1,3-dione **2**, and 1-naphthalenamine **3** in a set of ionic liquids have confirmed our expectations. We were delighted to find that the reaction went to complete within a few minutes in both the ionic liquids, $TMGT_f$ and TMGT. A comparison between the ionic liquids which we applied to the model reaction, as shown in Table 1 in terms of reaction time and yield, clearly shows the efficiency of guanidinium ionic liquids and the supremacy of $TMGT_f$.

The neutral ionic liquid, [BMIm]BF_4, was not as effective as $TMGT_f$ or TMGT for catalysis of the reaction, however, it found comparable catalysis activity when contaminated with 20 mol% of *p*-TSA. Notably, nearly no product was detected when the trial reaction was heated at 100 °C for 8 h in the absence of any solvent (Table 1, entry 1).

The catalysis effect of $TMGT_f$ was explored by conducting the model reaction in 15 mol% solution of $TMGT_f$ in 1:5 ethanol:water. In this condition the reaction gave 82% yield after 20 min. Increasing the amount of $TMGT_f$ to 35 mol% has improved the yield (89%) and the reaction time (15 min), however, the best result was obtained in pure $TMGT_f$. Presumably, the efficient catalysis effect of $TMGT_f$ is related to its acid–base functions (Fig. 1) as well as to providing tuned ionic medium. After determination of $TMGT_f$ as a suitable catalyst solvent, a variety of isatin derivatives and naphthalenamines were employed under similar conditions to evaluate the substituent scope of this reaction.

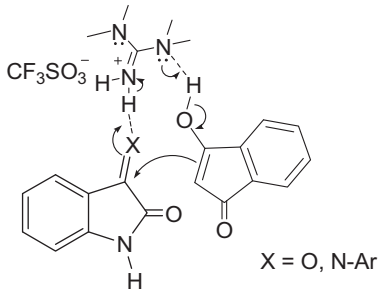


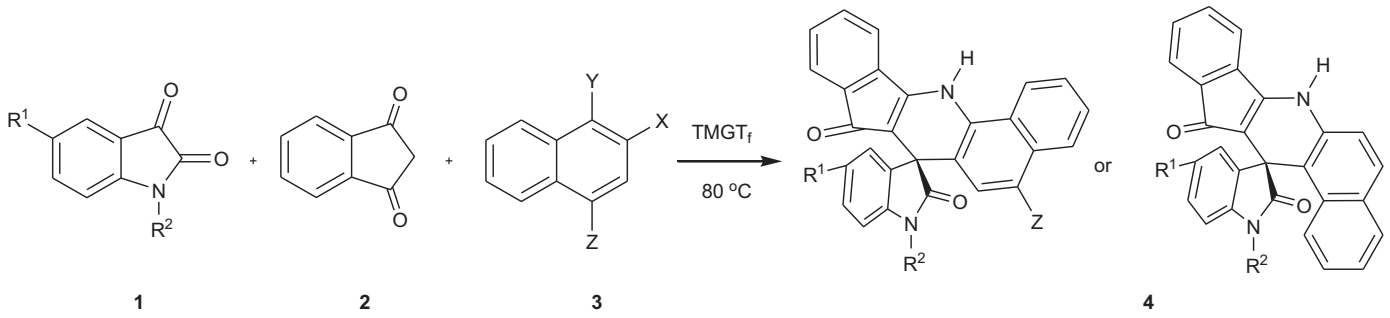
Fig. 1. Supposed acid–base functions of $TMGT_f$.

Generally, the cyclocondensation reactions proceeded well under catalysis of $TMGT_f$ and afforded the desired products **4a**–**j** in fairly high yields. As shown in Table 2, the reaction is compatible with a variety of electron-donating and electron-withdrawing substituents on the aromatic ring of isatin. It is also noteworthy that the reaction is likewise effective with *N*-benzyl isatin and equally goes to complete with both 1- and 2-naphthalenamines. Table 2 also compares the results of the present method with those obtained by our previously reported method using EtOH–H_2O as solvent in presence of *p*-TSA [11]. Interestingly, all the reactions appeared more effective in $TMGT_f$ with certain advantages in terms of reaction time and yields.

Encouraged by these fascinating results, we attempted to expand the scope of the method by using aniline derivatives in place of naphthalenamines. However, all the efforts we made to take aniline derivatives into a similar reaction remained entirely unsuccessful. This observation may be attributed to the more aromatic character of anilines in comparison with naphthalenamines which are prone to act as enamine reactants. Similar endeavours we made to employ 1- or 2-naphthols in place of naphthalenamines also failed to give the corresponding fused spiro[xanthene-oxindole] compounds. In all these abortive cases, the competitive formation of 3,3-di(2*H*-indene-1,3-dione-2-yl)indolin-2-ones, e.g. [19] **7**, outran the three-component synthesis of the desired spiro-product 4 (Scheme 1). Attempts to bring 5,5-dimethyl-1,3-cyclohexadione, ethyl acetoacetate, 1,3-cyclohexadione, or Meldrum's acid into alternative domino reactions with isatin and naphthalenamines were unsatisfactory, as they produced multiple of products polluted with unreacted substrates or otherwise remained completely unreactive.

A plausible mechanism for the formation of spiro-oxindoles **4a**–**j** from this three-component reaction is depicted in (Scheme 2). Compounds **4a**–**j** are likely produced from the initial condensation of 2*H*-indene-1,3-dione **2** with isatins **1** to yield the intermediates **5**, which upon Michael addition of naphthalenamine **3** followed by cyclocondensation of the resultant adducts **6a–j** give the corresponding products **4a**–**j** (pathway A). Alternatively, the key intermediates **5** may be produced by condensation of 2*H*-indene-1,3-dione **2** with the preformed imine derived from the reaction between isatin **1** and naphthalenamine **3** (pathway B). The fact that no byproducts, resulting from other combination of reactants, have been resolved from the reaction mixture may lend support to the proposal that the two pathways converge at the formation of intermediates **5**. In contrast to isatins, these intermediates are soft

Table 2
Synthesis of the fused spiro[1,4-dihydropyridine-oxindole] products **4a–j** .



Compound	R^1	R^2	X	Y	Z	Yield at 5 min	Yield at 15 min
						In $TMGT_f$	In $EtOH:H_2O$[a]
4a	I	H	H	NH_2	H	95	85
4b	Cl	H	H	NH_2	H	93	82
4c	NO_2	H	H	NH_2	Br	92	83
4d	Cl	H	H	NH_2	Br	93	80
4e	CH_3	H	H	NH_2	H	93	83
4f	H	H	NH_2	H	H	90	85[a]
4g	OCH_3	H	NH_2	H	H	90	78[a]
4h	NO_2	H	NH_2	H	H	95	81[a]
4i	Br	H	NH_2	H	H	97	83[a]
4j	H	Bn	NH_2	H	H	93	79[a]

[a] Ref. [11].

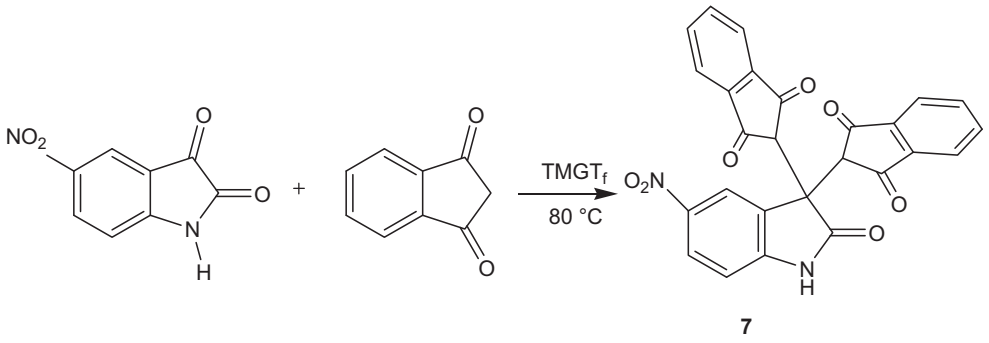
electrophiles which would be attacked in a Michael manner by the soft nucleophilic centre of, for example, 1-naphthalenamine **3** at β-carbon atom (Scheme 2).

Upon further exploration of the scope and limits of this method, two other α-dicarbonyl compounds, i.e. acenaphthylene-1,2-dione **8** and 1*H*-indene-1,2,3-trione **9** (ninhydrin) have shown similar reactivity when used instead of isatin and thereby the relative spirocyclic compounds **10** and **11** were obtained efficiently under the ionic liquid condition (Scheme 3). However, under the same conditions phenanthrene-9,10-dione did not go into the reaction when used in place of isatin component.

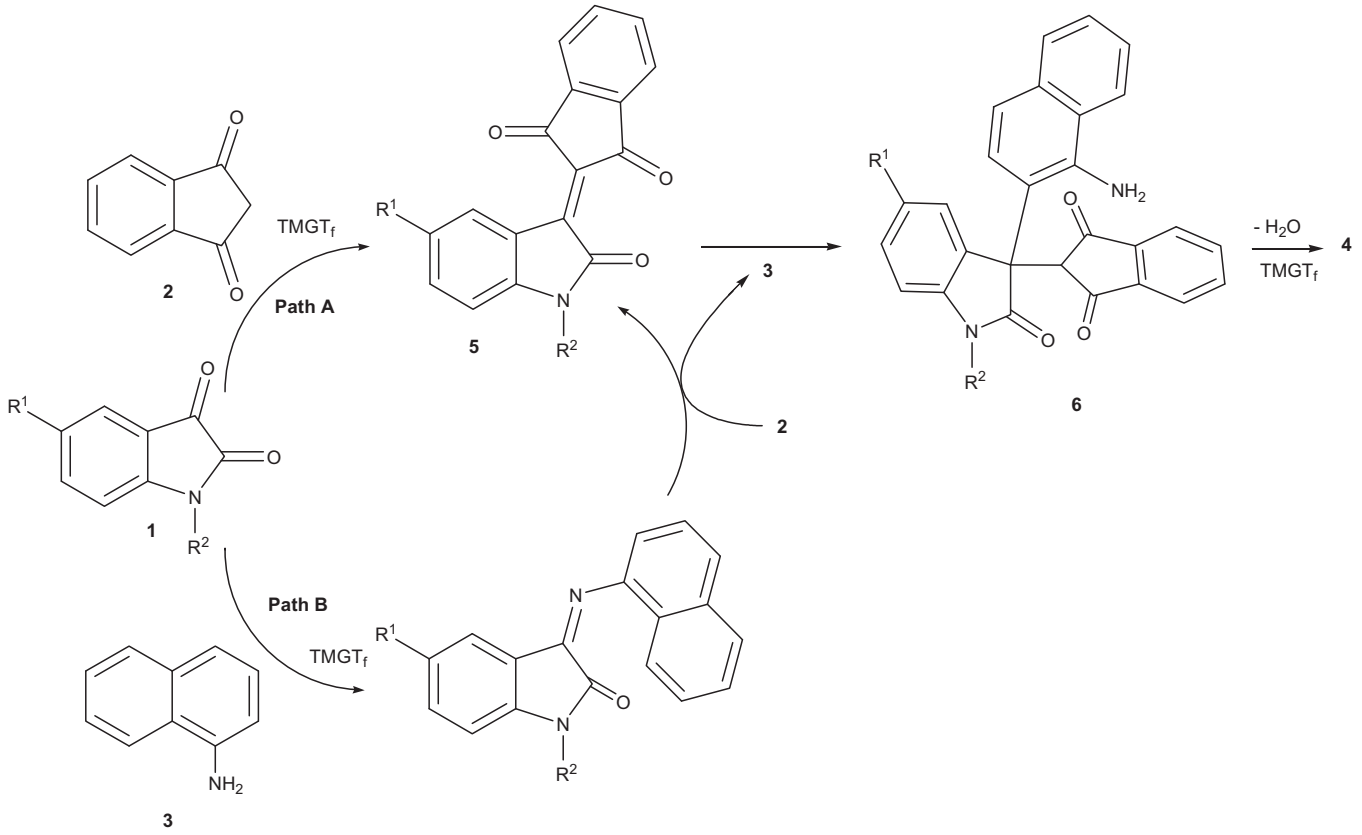
Our experiments also showed that the recently reported reaction [20] of 6-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione **12**, as an enamine partner, with 2*H*-indene-1,3-dione **2** and isatin derivatives is successfully promoted in $TMGT_f$ to give quickly the relative spiro-compounds **13a–c** in fairly high yields (Table 3).Formation of compounds **4a–j** was assessed from their elemental analyses, IR, ^{1}H- and ^{13}C NMR, as well as mass spectral data. The ^{1}H NMR spectra of these compounds follow a pattern of first order resonances precisely consistent with the spatial arrangement and different environments of protons. A more distinctive feature is associated with the signal of 1-H proton of compounds **4f–j** which experience the characteristic up-field shifts at about δ 6.58–6.67, in contrast to the previous structurally related 13-aryl-7,13-dihydro-1*H*-indeno[1,2-*b*]benzo[*f*]quinolin-12-ones [21]. This observation is attributable to anisotropic shielding effect of oxindole ring orienting perpendicular to the plane of indeno[1,2-*b*]benzoquinoline backbone. In compound **10** the shielding effect of acenaphthylene ring is even greater as the signal of 1-H proton shifts to higher field in aromatic region, appearing at δ 6.27 (Fig. 2).

The next phase of our studies dealt with investigating the recyclability of $TMGT_f$ for the model reaction synthesizing **4a**. After completion of reaction, the product was phase-separated from the reaction mixture by addition of water. The ionic liquid was recovered from the aqueous phase by evaporation of water under reduced pressure and then subjected to subsequent run of the reaction by charging with the same substrates. As shown in Table 4, the ionic liquid can be reused without any significant loss of its catalytic activity after five runs.

In conclusion, an expedient three-component reaction for synthesis of spiro[dihydropyridine-oxindole] systems using simple and readily available starting materials under catalysis of the ionic liquid, *N,N,N,N*-tetramethylguanidinium triflate, was introduced here. All the reactions, as a consequence of their conceivable convergent mechanism, solely afforded the corresponding spir-



Scheme 1. Formation of 3,3-di(2*H*-indene-1,3-dione-2-yl)indolin-5-nitro-2-one 7 from the side reaction of 5-nitroisatin and 2*H*-indene-1,3-dione.



Scheme 2. A proposed mechanism for the reaction.

ocyclic products. The ionic liquid plays the role of a catalyst solvent and can be recovered for reuse several times. Another advantage of the present method may be requiring no metal catalysts or additional solvent, whilst proceeding with an appropriate rate in comparison with other methods that give similar skeletones [20,21].

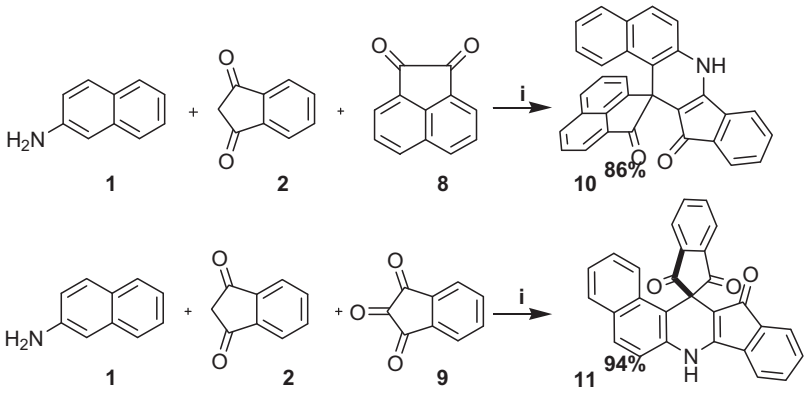
3. Experimental

3.1. Materials and methods

Melting points were measured on an Electrothermal apparatus and are uncorrected. Elemental analyses (C, H, N) were conducted using a Foss Heraeus CHN-O-Rapid analyzer. IR spectra were obtained in KBr wafers on Shimadzu IR-470 spectrometer. ^{1}H- and ^{13}C NMR spectra were measured in DMSO, with a Brucker DRX-400 AVANCE spectrometer at 400.1 and 100.6 MHz, respectively. Mass spectra were recorded on a Shimadzu QP1100EX mass spectrometer operating at an ionization potential of 70 eV. Chemicals were obtained from Merck & Fluka companies and were used without further purification.

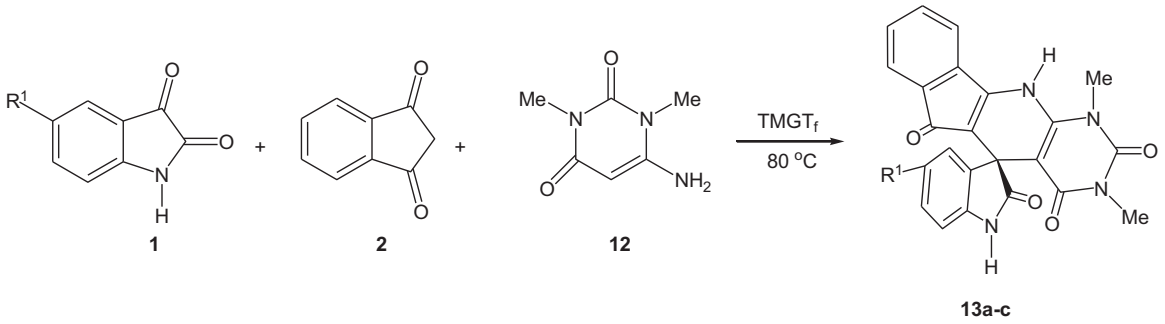
3.2. General procedure for preparation of compounds

A mixture of 5-iodo-isatin (0.273 g, 1 mmol), 2*H*-indene-1,3-dione (0.144 g, 1 mmol) and naphthalen-1-amine (0.144 g, 1 mmol) was added to a vial containing a magnetic stirring bar and the ionic liquid (TMGT$_f$, 1 mL). The reaction mixture was sealed and stirred at 80 °C until disappearance of the starting materials (about 5 min, as monitored by TLC on silica gel using a 5:1 mixture of ethyl acetate/n-hexane). After completion of the reaction, the residue was washed with 2 × 15 mL of water to



Scheme 3. Extension of the method by using acenaphthylene-1,2-dione 8 or ninhydrin 9 instead of isatin derivatives. Reagents and conditions: (i) TMGT$_f$, 80 °C.

Table 3
Extension of the method by using 6-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione 12, as enamine, and the results compared with those obtained from the reactions in EtOH/H_2O solution using *p*-TSA as catalyst .



Compound	R^1	Yield after 5 min	Yield after 15 min
		In $TMGT_f$	In EtOH:H_2O
13a	H	91	85 (89)[a]
13b	Cl	93	88
13c	I	92	84

[a] Ref. [20].

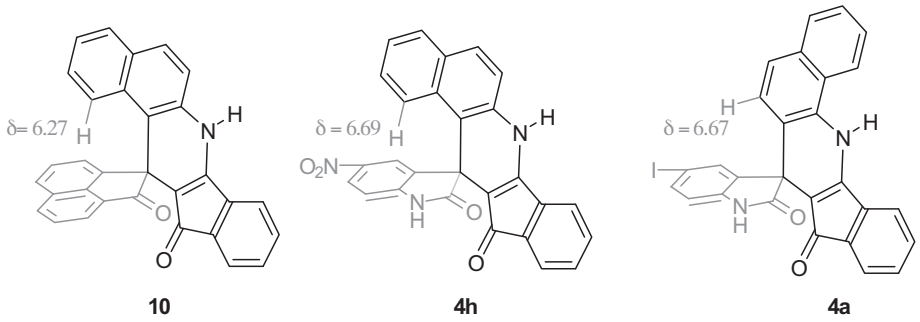


Fig. 2. Anisotropic shielding effects of the crossed rings on the incident protons.

extract the ionic liquid. The solid residue was washed with ethanol (95.5%) to obtain pure product **4a** (0.499 g, 95%). The ionic liquid was recovered from the aqueous extracts by evaporating in reduced pressure and reused in the next cycles.

3.2.1. Spiro[1H-indeno[1,2-b]benzo[h]quinolin-13,3′-indoline]-7,13-dihydro-5′-iodo-12,2′-dione ***(4a)***

Red powder: mp 303–305 °C; IR (KBr); ν 3230, 3180, 1697, 1614, 1583, 1524 cm^{-1}. ^1H NMR (400.13 MHz, DMSO-d_6): δ 6.67 (1H, d, *J* = 8.8 Hz, 6-H), 6.84 (1H, d, *J* = 8.4 Hz, 7′-H), 7.27 (1H, d, *J* = 1.2 Hz, 4′-H), 7.28 (1H, d, *J* = 7.2 Hz), 7.43 (1H, t, *J* = 7.2 Hz), 7.54–7.58 (m, 3H), 7.63 (1H, t, *J* = 7.2 Hz), 7.74 (1H, t, *J* = 7.2 Hz), 7.90 (1H, d, *J* = 8.0 Hz), 8.20 (1H, d, *J* = 7.6 Hz), 8.79 (1H, d, *J* = 8.8 Hz, 11-H), 10.41 (1H, s, NH), 10.79 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.5 (C_{spiro}), 85.6 (C–I), 103.4 (C-12a), 112.6 (C-11a), 119.8, 120.7, 121.3, 122.5, 124.0, 125.3, 125.4, 127.1, 127.4, 128.7, 131.0, 132.0, 132.1, 133.3, 133.4, 134.2, 136.8, 137.5, 140.8, 141.8, 156.7 (C-7a), 179.3 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, *m*/*z* (rel. int.): 526 $[M]^+$ (21), 498 $[M–CO]^+$ (35), 481 (19), 480 $[M–C=O + H_2O]^+$ (17), 369 (28), 341 (34), 43 (100). Anal. calcd. for $C_{27}H_{15}IN_2O_2$: C, 61.61; H, 2.87; N, 5.32. Found: C, 61.69; H, 2.93; N, 5.27.

Table 4
Recycling of $TMGT_f$ for the synthesis of **4a**.

Entry	Yield (%)	Cycle
1	95	fresh
2	93	first recycle
3	92	second recycle
4	90	third recycle
5	90	fourth recycle

3.2.2. Spiro[1H-indeno[1,2-b]benzo[h]quinolin-13,3′-indoline]-7,13-dihydro-5′-chloro-12,2′-dione ***(4b)***

Red powder: mp 305–307 °C; IR (KBr); ν 3200, 3100, 1700, 1680, 1620, 1570, 1520, 1480, 1395 cm^{-1}. ^1H NMR (400.13 MHz, DMSO-d_6): δ 6.67 (1H, d, *J* = 8.4 Hz, 6-H), 6.99 (1H, d, *J* = 8.4 Hz, 7′-H), 7.05 (1H, d, *J* = 2.0 Hz, 4′-H), 7.27 (1H, d, *J* = 7.2 Hz), 7.28 (1H, dd, *J* = 8.4 and 2.0 Hz, 6′-H), 7.42 (1H, t, *J* = 7.4 Hz), 7.55 (1H, dt, *J* = 8.4 and 0.8 Hz), 7.56 (1H, d, *J* = 8.8 Hz), 7.62 (1H, t, *J* = 7.6 Hz), 7.74 (1H, dt, *J* = 7.6 and 1.2 Hz), 7.89 (1H, d, *J* = 7.6 Hz), 8.19 (1H, d, *J* = 7.6 Hz), 8.78 (1H, d, *J* = 8.4 Hz, 11-H), 10.43 (1H, s, NH), 10.82 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.6 (Cspiro), 103.3 (C-12a), 111.5 (C-11a), 119.7, 120.7, 121.3, 122.4, 124.0, 125.1, 125.2, 125.4, 126.6, 127.1, 127.4, 128.7, 128.8, 131.0, 131.9, 132.0, 133.5, 134.2, 136.8, 140.1, 141.0, 156.7 (C-7a), 179.7 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, *m*/*z* (rel. int.): 436 $[M + 2]^+$ (1), 434 $[M]^+$ (3), 406 $[M–C=O]^+$ (6), 389 $[M–C=O + H_2O)]^+$ (4), 377 (5), 239 (4), 149 (9), 111 (11), 43 (100). Anal. calcd. for $C_{27}H_{15}ClN_2O_2$: C, 74.57; H, 3.48; N, 6.44. Found: C, 74.59; H, 3.52; N, 6.37.

3.2.3. Spiro[1H-indeno[1,2-b]benzo[h]quinolin-13,3′-indoline]-7,13-dihydro-5-bromo-5′-nitro-12,2′-dione ***(4c)***

Red powder: mp 288–290 °C; IR (KBr); ν 3250, 3190, 1706, 1680, 1620, 1600, 1522, 1332 cm^{-1}. ^1H NMR (400.13 MHz, DMSO-

d_6): δ 6.92 (1H, s, 6-H), 7.23 (1H, d, J = 8.4 Hz, 7′-H), 7.27 (1H, d, J = 6.8 Hz), 7.43 (1H, t, J = 7.2 Hz), 7.57 (1H, t, J = 7.4 Hz), 7.80–7.91 (m, 3H), 7.95 (1H, d, J = 1.8 Hz, 4′-H), 8.12 (1H, d, J = 8.0 Hz), 8.20 (1H, d, J = 7.2 Hz), 8.24 (1H, dd, J = 8.4 and 1.8 Hz, 6′-H), 8.88 (1H, d, J = 8.4 Hz, 11-H), 10.61 (1H, s, NH), 11.48 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.2 (C_{spiro}), 102.9 (C-12a), 110.7 (C-11a), 117.9 (C–Br), 119.5, 120.9, 121.6, 123.3, 123.4, 125.2, 126.6, 127.5, 128.3, 129.4, 131.2, 131.4, 132.3, 132.9, 133.9, 136.6, 136.9, 138.2, 143.4, 148.5 (C–NO_2), 156.8 (C-7a), 180.0 (amidic C=O), 189.5 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 527 [M + 2, ^{81}Br]$^+$ (0.5), 525 [M, ^{79}Br]$^+$ (0.5), 524 (1), 499 [527–C=O]$^+$ (0.5), 497 [525–C=O]$^+$ (0.5), 270 (14), 191 (76), 106 [HC≡C–^{81}Br]$^+$ (13), 104 [HC≡C–^{79}Br]$^+$ (13), 56 (100). Anal. calcd. for $C_{27}H_{14}$ BrN_3O_4: C, 61.85; H, 2.69; N, 8.01. Found: C, 61.89; H, 2.73; N, 7.93

3.2.4. Spiro[1H-indeno[1,2-b]benzo[h]quinolin-13,3′-indoline]-7,13-dihydro-5-bromo-5′-chloro-12,2′-dione (4d)

Red powder: mp 304–306 °C; IR (KBr); ν 3340, 1708, 1682, 1618 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.85 (1H, s, 6-H), 7.03 (1H, d, J = 8.0 Hz), 7.14 (1H, s, 4′-H), 7.21 (1H, d, J = 6.8 Hz), 7.31 (1H, d, J = 7.2 Hz, 6′-H), 7.39 (1H, t, J = 7.2 Hz), 7.53 (1H, t, J = 7.4 Hz), 7.77–7.84 (2H, m), 8.07 (1H, d, J = 8.0 Hz), 8.11 (1H, d, J = 7.2 Hz), 8.80 (1H, d, J = 8.0 Hz, 11-H), 10.53 (1H, s, NH), 10.94 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.4 (C_{spiro}), 103.4 (C-12a), 111.9 (C-11a), 117.7 (C–Br), 120.2, 120.8, 121.3, 123.2, 125.1, 125.4, 126.9, 127.4, 128.2, 128.4, 129.1, 129.2, 131.0, 131.2, 132.1, 132.7, 133.8, 136.6, 139.5, 140.8, 156.6 (C-7a), 179.5 (amidic C=O), 189.5 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 518 [M + 4, ^{37}Cl, ^{81}Br]$^+$ (5), 516 [M + 2, ^{35}Cl, ^{81}Br and ^{37}Cl, ^{79}Br]$^+$ (19), 514 [M, ^{35}Cl, ^{79}Br]$^+$ (13), 488 [516–C=O]$^+$ (3), 486 [514–C=O]$^+$ (6), 470 [488–H_2O]$^+$ (14), 468 [486–H_2O]$^+$ (20), 388 [470-^{81}Br]$^+$ or [468-^{79}Br]$^+$ (11), 311 (17), 276 (15), 221 (17), 43 (100). Anal. calcd. for $C_{27}H_{14}$ $BrClN_2O_2$: C, 63.12; H, 2.75; N, 5.45. Found: C, 63.08; H, 2.70; N, 5.52.

3.2.5. Spiro[1H-indeno[1,2-b]benzo[h]quinolin-13,3′-indoline]-7,13-dihydro-5′-methyl-12,2′-dione (4e)

Red powder: mp 294–296 °C; IR (KBr); ν 3220, 1700, 1680, 1620, 1583, 1524 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 2.10 (3H, s, CH_3), 6.67 (1H, d, J = 8.8 Hz, 6-H), 6.76 (1H, s, 4′-H), 6.86 (1H, d, J = 7.6 Hz), 7.00 (1H, dd, J = 8.8 and 1.2 Hz, 6′-H), 7.25 (1H, d, J = 6.8), 7.41 (1H, t, J = 7.2 Hz), 7.53 (1H, d, J = 9.6 Hz), 7.54 (1H, t, J = 8.0 Hz), 7.61 (1H, t, J = 7.2 Hz), 7.73 (1H, dt, J = 7.2 and 1.2 Hz), 7.88 (1H, d, J = 7.6 Hz), 8.17 (1H, d, J = 7.2 Hz), 8.76 (1H, d, J = 7.6 Hz, 11-H), 10.35 (1H, s, NH), 10.57 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 20.9 (CH_3), 51.3 (C_{spiro}), 104.1 (C-12a), 109.7 (C-11a), 120.6, 120.7, 121.1, 122.3, 123.9, 125.2, 125.5, 125.6, 127.0, 127.2, 128.7, 129.1, 130.9, 131.6, 131.8, 132.0, 133.3, 134.2, 136.9, 138.6, 139.4, 156.5 (C-7a), 180.0 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 414 [M]$^+$ (55), 398 (14), 386 [M–C=O]$^+$ (85), 385 (100), 369 [386–H_2O]$^+$ (76), 358 (60). Anal. calcd. for $C_{28}H_{18}N_2O_2$: C, 81.14; H, 4.38; N, 6.76. Found: C, 81.12; H, 4.43; N, 6.69.

3.2.6. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,3′-indoline]-7,13-dihydro-12,2′-dione (4f)

Red powder: mp 298–300 °C; IR (KBr); ν 3470, 3210, 3050, 1688, 1672, 1618 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.67 (1H, d, J = 8.4 Hz, 1-H), 6.86 (1H, t, J = 7.2 Hz, 4′-H), 6.96 (2H, t, J = 8 Hz, 5′-H and 6′-H), 7.21 (1H, t, J = 7.6 Hz, 7′-H), 7.26 (1H, d, J = 6.8 Hz), 7.41 (1H, t, J = 7.2 Hz), 7.54 (1H, d, J = 8.8 Hz), 7.55 (1H, t, J = 8.0 Hz), 7.62 (1H, t, J = 7.2 Hz), 7.73 (1H, t, J = 6.8 Hz), 7.88 (1H, d, J = 8 Hz), 8.18 (1H, d, J = 7.2 Hz), 8.76 (1H, d, J = 8.4 Hz), 10.36 (1H, s, NH), 10.65 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.3 (C_{spiro}), 104.0 (C-12a), 110.0 (C-11a), 120.5, 120.6, 121.1, 122.3, 122.7, 123.9, 125.1, 125.2, 125.4, 127.0, 127.3, 128.7, 128.8, 130.9, 131.8, 132.0, 133.3, 134.2, 136.8, 138.4, 141.9 (C-6a), 156.5 (C-7a), 180.0 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 400 [M]$^+$ (56), 384 (14), 371 (100), 355 (95). Anal. calcd. for $C_{27}H_{16}N_2O_2$: C, 80.99; H, 4.03; N, 7.00. Found: C, 81.03; H, 4.21; N, 6.89.

3.2.7. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,3′-indoline]-7,13-dihydro-5′-methoxy-12,2′-dione (4g)

Orange powder: mp 306–308 °C; IR (KBr); ν 3237, 1703, 1684, 1532, 1187, 1175 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 3.56 (3H, s, OCH_3), 6.56 (1H, d, J = 2.4 Hz, 4′-H), 6.67 (1H, d, J = 8.8 Hz, 1-H), 6.79 (1H, dd, J = 8.4 and 2.0 Hz, 6′-H), 6.89 (1H, d, J = 8.8 Hz, 7′-H), 7.26 (1H, d, J = 6.8 Hz), 7.41 (1H, t, J = 7.2 Hz), 7.54 (1H, d, J = 8.0 Hz), 7.55 (1H, t, J = 6.4 Hz), 7.61 (1H, t, J = 7.6 Hz), 7.72 (1H, t, J = 7.2 Hz), 7.88 (1H, d, J = 8.0 Hz), 8.17 (1H, d, J = 7.2 Hz), 8.76 (1H, d, J = 8.4 Hz), 10.35 (1H, s, NH), 10.48 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.8 (C_{spiro}), 55.8 (OCH_3), 102.8 (C-12a), 110.4 (C-11a), 111.7, 113.7, 120.5, 120.6, 121.1, 122.4, 123.9, 125.2, 125.4, 127.0, 127.2, 128.7, 130.9, 131.8, 132.0, 133.4, 134.3, 135.3, 136.8, 139.6, 155.8 (C-6a), 156.6 (C-7a), 179.9 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 430 [M]$^+$ (98), 402 [M–C=O]$^+$ (100), 385 (88). Anal. calcd. for $C_{28}H_{18}N_2O_3$: C, 78.13; H, 4.21; N, 6.51. Found: C, 78.22; H, 4.33; N, 6.46

3.2.8. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,3′-indoline]-7,13-dihydro-5′-nitro-12,2′-dione (4h)

Red powder: mp 304 °C (decomp.); IR (KBr); ν 3470, 3240, 1706, 1620, 1527, 1330 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.69 (1H, d, J = 8.8 Hz, 1-H), 7.21 (1H, d, J = 8.8 Hz, 7′-H), 7.28 (1H, d, J = 7.2 Hz), 7.43 (1H, t, J = 7.2 Hz), 7.57 (1H, t, J = 7.2 Hz), 7.58 (1H, d, J = 8.8 Hz), 7.65 (1H, t, J = 7.6 Hz), 7.76 (1H, t, J = 7.2 Hz), 7.86 (1H, d, J = 2.0 Hz, 4′-H), 7.91 (1H, d, J = 8.0 Hz), 8.22 (1H, dd, J = 8.8 and 2.4 Hz), 8.23 (1H, d, J = 8.4 Hz), 8.82 (1H, d, J = 8.4 Hz), 10.52 (1H, s, NH), 11.42 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 51.4 (C_{spiro}), 102.7 (C-12a), 110.4 (C-11a), 119.0, 120.7, 120.8, 121.5, 122.5, 124.0, 125.1, 125.7, 126.3, 127.2, 127.6, 128.8, 131.1, 132.1, 132.3, 133.6, 134.2, 136.7, 138.8, 143.2, 148.6 (C-6a), 157.0 (C-7a), 180.4 (amidic C=O), 189.6 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 445 [M]$^+$ (48), 443 [M-2]$^+$ (100), 417 [M–C=O]$^+$ (60), 400 (89), 399 [417–H_2O]$^+$ (75) 369 (76). Anal. calcd. for $C_{27}H_{15}N_3O_4$: C, 72.80; H, 3.39; N, 9.43. Found: C, 72.76; H, 3.42; N, 9.37.

3.2.9. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,3′-indoline]-7,13-dihydro-5′-bromo-12,2′-dione (4i)

Red powder: mp 300–302 °C; IR (KBr); ν 3245, 1700, 1617, 1584, 1523, 990 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.67 (1H, d, J = 8.4 Hz, 1-H), 6.95 (1H, d, J = 8.0 Hz), 7.16 (1H, s, 4′-H), 7.27 (1H, d, J = 6.0 Hz), 7.37–4.49 (2H, m, 6′-H and 7′-H), 7.56 (2H, d, J = 7.6 Hz), 7.58–7.67(1H, m), 7.69–7.78 (1H, m), 7.89 (1H, d, J = 7.6 Hz), 8.19 (1H, d, J = 6.8 Hz), 8.78 (1H, d, J = 8.0 Hz), 10.42 (1H, s, NH), 10.81 (1H, s, NH). EIMS (probe) 70 eV, m/z (rel. int.): 480 [M, ^{81}Br]$^+$ (61), 478 [M, ^{79}Br]$^+$ (63), 451 (100), 452 [480–C=O]$^+$ (87), 450 [478–C=O]$^+$ (88), 434 [452–H_2O]$^+$ (21), 432 [450–H_2O]$^+$ (41). Anal. calcd. for $C_{27}H_{15}BrN_2O_2$: C, 67.66; H, 3.15; N, 5.84. Found: C, 67.71; H, 3.09; N, 5.78.

3.2.10. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,3′-indoline]-7,13-dihydro-1′-benzyl-12,2′-dione (4j)

Orange powder: mp 310 °C (decomp.); IR (KBr); ν 3235, 1686, 1622, 1604, 1526, 1362 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 5.00 (1H, d, J = 16 Hz), 5.13 (1H, d, J = 16 Hz), 6.58 (1H, d, J = 8.8 Hz, 1-H), 6.91–6.96 (2H, m), 7.05 (1H, d, J = 7.2 Hz, 7′-H), 7.21 (1H, t, J = 7.6 Hz), 7.27–7.45 (6H, m), 7.51–7.59 (3H, m), 7.64 (1H, t, J = 7.6 Hz), 7.75 (1H, t, J = 7.6 Hz), 7.89 (1H, d, J = 8.4 Hz), 8.22 (1H, d, J = 7.2 Hz), 8.79 (1H, d, J = 8.4 Hz), 10.47 (1H, s, NH). EIMS (probe) 70 eV, m/z (rel. int.): 490 [M]$^+$ (34), 461 [M–HCO]$^+$ (4), 399 [M-

benzyl]$^+$ (100), 371 [399–C=O]$^+$ (86). Anal. calcd. for $C_{34}H_{22}N_2O_2$: C, 83.25; H, 4.52; N, 5.71. Found: C, 83.12; H, 4.59; N, 5.58.

Due to the very low solubility of compound **4i** and **4j** we were unable to obtain their ^{13}C NMR spectra

*3.2.11. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,1′(2′H)-acenaphthylene]-7,13-dihydro-12,2′-dione (**10**)*

Brick red powder: mp 284–286 °C; IR (KBr); ν 3295, 1702, 1684, 1620, 1586, 1525 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.27 (1H, d, J = 8.8 Hz, 1-H), 7.17 (1H, d, J = 6.8 Hz), 7.34 (1H, d, J = 6.8 Hz), 7.38–7.40 (2H, m), 7.57 (1H, t, J = 7.2 Hz), 7.62 (1H, t, J = 8.6 Hz), 7.64 (1H, t, J = 8.4 Hz), 7.74–7.91 (3H, m), 8.02 (1H, d, J = 8.4 Hz), 8.12 (1H, d, J = 6.8 Hz), 8.22 (1H, d, J = 7.6 Hz), 8.41 (1H, d, J = 8.4 Hz), 8.82 (1H, d, J = 8.8 Hz), 10.45 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 57.2 (C_{spiro}), 105.3 (C-12a), 120.6 (C-11a), 121.2, 121.9, 122.4, 123.18, 123.22, 124.0, 125.1, 125.2, 125.6, 127.1, 127.3, 128.7, 129.4, 129.9, 130.4, 131.0, 132.0, 132.1, 132.5, 132.7, 133.3, 134.1, 136.7, 136.8, 141.9 (C-6a), 158.7 (C-7a), 189.9 (C=O), 209.8 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 435 [M]$^+$ (82), 407 [M–C=O]$^+$ (78), 406 [M–HC=O]$^+$ (100). Anal. calcd. for $C_{31}H_{17}NO_2$: C, 85.50; H, 3.93; N, 3.22. Found: C, 85.61; H, 4.11; N, 3.14.

*3.2.12. Spiro[1H-indeno[1,2-b]benzo[f]quinolin-13,2′-indene]-7,13-dihydro-12,1′,3′-trione (**11**)*

Red powder: mp 286–288 °C; IR (KBr); ν 3303, 1740, 1697, 1624, 1587, 1524, 1250 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 6.63 (1H, d, J 8.4 Hz, 1-H), 7.28 (1H, d, J 7.2 Hz), 7.45 (1H, t, J 7.2 Hz), 7.58 (1H, t, J 8.4 Hz), 7.59 (1H, t, J 8.4 Hz), 7.66 (1H, t, J 7.6 Hz), 7.76 (1H, t, J 7.2 Hz), 7.93 (1H, d, J 8.4 Hz), 8.15–8.19 (4H, m), 8.20 (1H, d, J 7.2 Hz), 8.78 (1H, d, J 8.8 Hz), 10.58 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 57.5 (C_{spiro}), 118.1 (C-12a), 120.9 (C-11a), 121.6, 122.4, 123.4, 124.0, 124.4 (2CH), 124.6, 125.8, 127.5, 127.7, 128.9, 131.4, 132.3, 132.8, 133.5, 133.7, 136.5, 137.5 (2CH), 142.8 (2C), 157.9 (C-7a), 189.6 (C=O), 200.6 (2C=O). EIMS (probe) 70 eV, m/z (rel. int.): 413 [M]$^+$ (87), 397 (60), 385 [M–C=O]$^+$ (65), 384 [M–HC=]$^+$ (100), 369 (54), 356 [384–C=O]$^+$ (45). Anal. calcd. for $C_{28}H_{15}NO_3$: C, 81.35; H, 3.66; N, 3.39. Found: C, 81.39; H, 3.73; N, 3.32.

*3.2.13. Spiro[1H-indeno[1,2-b]pyrido[2,3-d]pyrimidine-5,3′-indoline]-1,3-dimethyl-2,2′,4,6(3′H,10′H)-tetraone **(13a)***

Red powder: mp 294–296 °C; IR (KBr); ν 3398, 1754, 1695, 1640, 1624, 1539, 1480 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 3.02 (3H, s, CH_3), 3.44 (3H, s, CH_3), 7.02 (1H, dt, J = 7.4 and 1.2 Hz), 7.08 (1H, dd, J = 7.6 and 1.2 Hz), 7.21 (1H, dd, J = 8.0 and 1.2 Hz), 7.24 (1H, d, J = 6.8 Hz), 7.29 (1H, dt, J = 7.4 and 1.4 Hz), 7.40 (1H, dt, J = 7.4 and 1.2 Hz), 7.49 (1H, dt, J = 7.4 and 1.2 Hz), 7.65 (1H, d, J = 7.2 Hz), 10.99 (1H, s, NH), 11.69 (1H, s, NH).

*3.2.14. Spiro[1H-indeno[1,2-b]pyrido[2,3-d]pyrimidine-5,3′-indoline]-5′-chloro-1,3-dimethyl-2,2′,4,6(3′H,10′H)-tetraone **(13b)***

Red powder: mp 296–298 °C; IR (KBr); ν 3419, 3220, 1758, 1683, 1720, 1634, 1535, 1477 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 3.04 (3H, s, CH_3), 3.43 (3H, s, CH_3), 7.20 (1H, d, J = 2.4 Hz, 4′-H), 7.23 (1H, d, J = 8.4 Hz, 7′-H), 7.25 (1H, d, J = 7.2 Hz), 7.35 (1H, dd, J = 8.4 and 2.4 Hz, 6′-H), 7.40 (1H, t, J = 7.4 Hz), 7.50 (1H, t, J = 7.6 Hz), 7.62 (1H, d, J = 7.2 Hz), 11.09 (1H, s, NH), 11.72 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 27.6, 31.9 (2CH_3), 50.1 (C_{spiro}), 95.8 (C-4a), 100.3 (C-5a), 119.8, 120.7, 125.5, 127.9, 128.7, 129.3, 131.2, 132.0, 134.8, 136.0, 136.1, 151.7, 153.2, 156.4 (C=O), 157.4 (C=O), 180.5 (C=O), 188.9 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 446 [M]$^+$ (1), 393 (4), 316 (20), 250 (26), 43 (100). Anal. calcd. for $C_{23}H_{15}ClN_4O_4$: C, 61.82; H, 3.38; N, 12.54. Found: C, 61.73; H, 3.43; N, 12.57.

*3.2.15. Spiro[1H-indeno[1,2-b]pyrido[2,3-d]pyrimidine-5,3′-indoline]-5′-iodo-1,3-dimethyl-2,2′,4,6(3′H,10′H)-tetraone **(13c)***

Red powder: mp 301–303 °C; IR (KBr); ν 3300, 3200, 1740, 1700, 1720, 1680, 1640 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 3.04 (3H, s, CH_3), 3.44 (3H, s, CH_3), 7.02 (1H, d, J = 8.4 Hz, 7′-H), 7.25 (1H, d, J = 7.2 Hz), 7.37 (1H, d, J = 2.0 Hz), 7.39 (1H, t, J = 7.2 Hz, 8-H), 7.49 (1H, t, J = 7.4 Hz, 9-H), 7.61 (1H, d, J = 7.2 Hz, 7-H), 7.62 (1H, dd, J = 8.4 and 2.0 Hz, 6′-H), 11.07 (1H, s, NH), 11.72 (1H, s, NH). ^{13}C NMR (100.6 MHz, DMSO-d_6): δ 27.6, 31.9 (2CH_3), 49.8 (C_{spiro}), 88.8 (C–I), 95.9 (C-4a), 100.6 (C-5a), 119.7, 120.3, 120.7, 125.9, 131.2, 132.0, 134.8, 136.1, 136.9, 138.0, 146.2, 151.6, 153.1 (C=O), 156.2 (C=O), 180.6 (C=O), 188.7 (C=O). EIMS (probe) 70 eV, m/z (rel. int.): 538 [M]$^+$ (21), 523 [M–CH_3]$^+$ (11), 510 [M–C=O]$^+$ (12), 494 (32), 411 [M–I]$^+$ (28), 382 (14), 354 (18), 43 (100). Anal. calcd. for $C_{23}H_{15}IN_4O_4$: C, 51.32; H, 2.81; N, 10.41. Found: C, 51.40; H, 2.87; N, 10.29.

*3.2.16. 3,3-Di(2H-indene-1,3-dione-2-yl)indolin-5-nitro-2-one **(7)***

White powder: mp 234–236 °C; IR (KBr); ν 3300, 1740, 1710, 1684, 1580, 1482, 1260, 1187 cm^{-1}. ^{1}H NMR (400.13 MHz, DMSO-d_6): δ 4.82 (2H, s, CH), 6.72 (1H, d, J = 8.4 Hz, 7-H), 6.87 (1H, d, J = 1.6 Hz, 4-H), 7.29 (1H, dd, J = 8.4 Hz, 1.6 Hz, 6-H), 7.86–7.97 (8H, m, Ar), 10.80 (1H, s, NH).

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