Highly Convergent One-Pot Four-Component Regioselective Synthesis of Spiro-pyranopyrazoles and Oxa-aza-[3.3.3]propellanes

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A concise and efficient route for the synthesis of spiro-pyranopyrazoles and oxa-aza-[3.3.3]propellanes by simple regioselective multicomponent reaction of ninhydrin, malononitrile, hydrazine derivatives, and β-keto esters or dimethyl acetylenedicarboxylate was developed. This protocol provides an alternative method for combinatorial and parallel syntheses in drug discovery. The value of this method lies in its simplicity, regioselectivity, and good yields. The structures of **3** and **4** were corroborated spectroscopically (IR, ^{1}H- and ^{13}C-NMR, and EI-MS). A plausible mechanism for this type of reaction is proposed (*Schemes 2* and *3*).

Introduction. – We have previously developed some synthetic methods for the preparation of propellanes [1] and spiro compounds [2] *via* multicomponent domino reactions. Encouraged by our past results, we became interested in producing propellanes and spiro-pyranopyrazoles by the multicomponent reaction of ninhydrin, malononitrile, β-keto esters or dimethyl acetylenedicarboxylate (DMAD), and hydrazine derivatives.

Results and Discussion. – Very recently, we have reported novel multicomponent methods for the preparation of propellanes [1]. These methods rely on the reaction of an electron-deficient *Knoevenagel* adduct, resulting from ninhydrin and malononitrile, with diverse nucleophiles such as enamines, ketene aminals, and thioureas. At the outset of our study, we examined the multicomponent reaction of ninhydrin, malononitrile, phenylhydrazine (**1a**) with methyl acetoacetate (**2a**), using piperidine as catalyst. First, **1a** and **2a** were refluxed for 2 h in EtOH, then the mixture was cooled to ambient temperature, and a solution of ninhydrin, malononitrile, and piperidine in EtOH was added. The reaction afforded the desired spiro-pyranopyrazole **3a** in moderate yield. To explore the scope of this transformation, a variety of hydrazines, β-keto esters, and DMAD were applied (*Scheme 1*). Surprisingly, for hydrazine derivatives with an electron-withdrawing group close to the $NHNH_2$ moiety, instead of the anticipated spiro-pyranopyrazole products, we observed an unexpected process leading to heterocyclic propellanes, **4a**–**4c**, in moderate yields. The results are compiled in the *Table*. The reaction of $H_2NNH_2 \cdot H_2O$, DMAD, ninhydrin, and malononitrile also led to desired spiro-pyranopyrazole **5**.

Scheme 1. *Synthesis of Spiro-pyranopyrazoles and Oxa-aza-[3.3.3]propellanes*

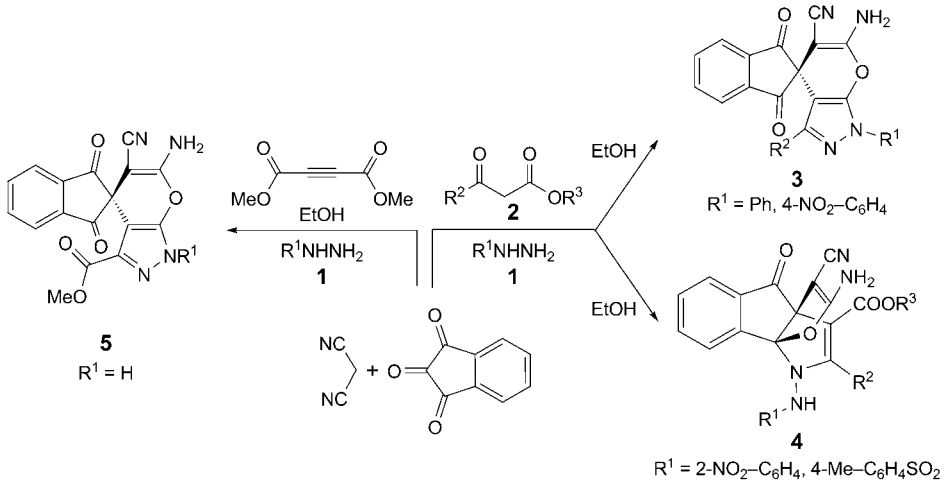


Table. *Spiro-pyranopyrazole and Oxa-aza-[3.3.3]propellane Derivatives Prepared*

Entry	R^1	R^2	R^3	Product	Yield [%]
1	Ph	Me	Me	**3a**	90
2	4-NO_2–C_6H_4	Me	Et	**3b**	85
3	Ph	Pr	Et	**3c**	81
4	4-NO_2–C_6H_4	Pr	Et	**3d**	87
5	2-NO_2–C_6H_4	Me	Me	**4a**	75
6	2-NO_2–C_6H_4	Me	Et	**4b**	70
7	4-Me–C_6H_4–SO_2	Me	Me	**4c**	81

NMR, IR, MS, and elemental-analysis data support the spiro and propellane structures. In contrast to the unsymmetrical structure of propellanes, spiro-pyranopyrazoles have a plane of symmetry. These findings are reflected in corresponding ^{13}C-NMR spectra. The mass spectra of **3a** displayed the molecular-ion peak at *m*/*z* 382. The IR spectrum of **3a** clearly indicated the presence of NH_2 (3375 and 3308 cm^{-1}), CN (2202 cm^{-1}), C=O (1716 cm^{-1}), and NC=C (1657 cm^{-1}) as the most significant functional groups of the product. The ^{1}H-NMR spectrum of **3a** showed two sharp *singlet*s at δ(H) 1.5 and 7.86 arising from the Me and NH_2 group, respectively. The signals of aromatic H-atoms appeared as two *triplet*s at δ(H) 7.34 and 7.49, a *doublet* at δ(H) 7.73, and a *singlet* at δ(H) 8.15. Due to the plane of symmetry of **3a**, 16 signals were observed in the ^{1}H-decoupled ^{13}C-NMR spectrum.

The mass spectra of **4b** displayed the molecular-ion peak at *m*/*z* 473. The IR spectrum of **4b** clearly indicated the presence of NH_2 and NH (at 3390 and 3201 cm^{-1}), CN (2189 cm^{-1}), CO_2 (1730 cm^{-1}), C=O (1670 cm^{-1}), NC=C (1632 cm^{-1}), and NO_2 (1460 and 1325 cm^{-1}) as the most significant functional groups of the product. The ^{1}H-NMR spectrum of **4b** exhibited a *triplet* at δ(H) 1.35 arising from the Me group, three sharp *singlet*s at δ(H) 2.04, 7.48, and 9.67, attributed to Me, NH_2, and NH groups,

respectively. The signal of the CH_2 group appeared as a *doublet* of *quartet* at δ(H) 4.19. The aromatic H-atom signals were four *triplet*s at δ(H) 7.02, 7.72, 7.88, and 7.89, and four *doublet*s at δ(H) 7.51, 7.66, 7.77, and 8.19. Observation of 24 distinct signals in the ^{1}H-decoupled ^{13}C-NMR spectrum of **4b** was in agreement with the proposed structure.

Wardell et al. investigated the reactions of methyl acetoacetate with a range of simple arylhydrazines. According to their report, polarized quinonoid form of (2-nitrophenyl)hydrazine is a significant contributor to the overall electronic structure of compound **6**, and it causes a reduction in the basicity and nucleophilicity of N(1) in (2-nitrophenyl)hydrazine as compared with phenylhydrazine itself (*Fig. 1*).

The aryl ring in (2-nitrophenyl)hydrazine is almost coplanar with the chain-extended fragment between C(1) and C(13) and the NO_2 groups are nearly coplanar with the ring. This nearly planar conformation may be influenced by the intramolecular N–H$\cdots$O H-bond, and the observed distance for N(1)–H$\cdots$O(21) in the X-ray structure is 1.97 Å (*Fig. 2*).

Therefore, from the reaction of methyl acetoacetate with (2-nitrophenyl)hydrazine or phenylhydrazine acyclic and cyclic products are obtained [3].

Based on this finding, we assume that heating of NH_2NH_2, $PhNHNH_2$, or (4-nitrophenyl)hydrazine with β-keto esters **2** or DMAD at reflux initially produces the pyrazole intermediate **7**, which undergoes tautomerization to enol form **8**. A *Knoevenagel* condensation of ninhydrin and malononitrile is also proposed to give adduct **9**, which acts as *Michael* acceptor. *Michael* addition of pyrazole intermediate **8** to *Knoevenagel* adduct **9** should provide intermediate **10**. Then, the intermediate **11**, which is the tautomeric form of intermediate **10**, undergoes intramolecular cyclization to give intermediate **12**. In the last step, intermediate **12** tautomerizes to product **3** and **5** (*Scheme 2*).

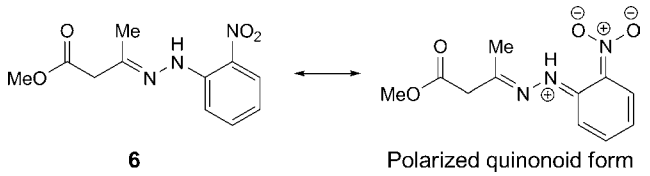


Fig. 1. *The polarized quinonoid form of intermediate* **6** (according to [3])

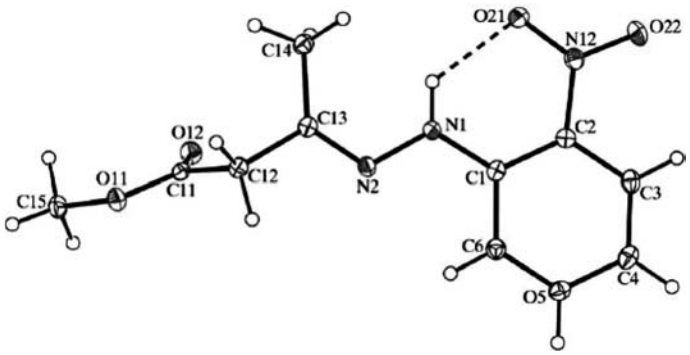
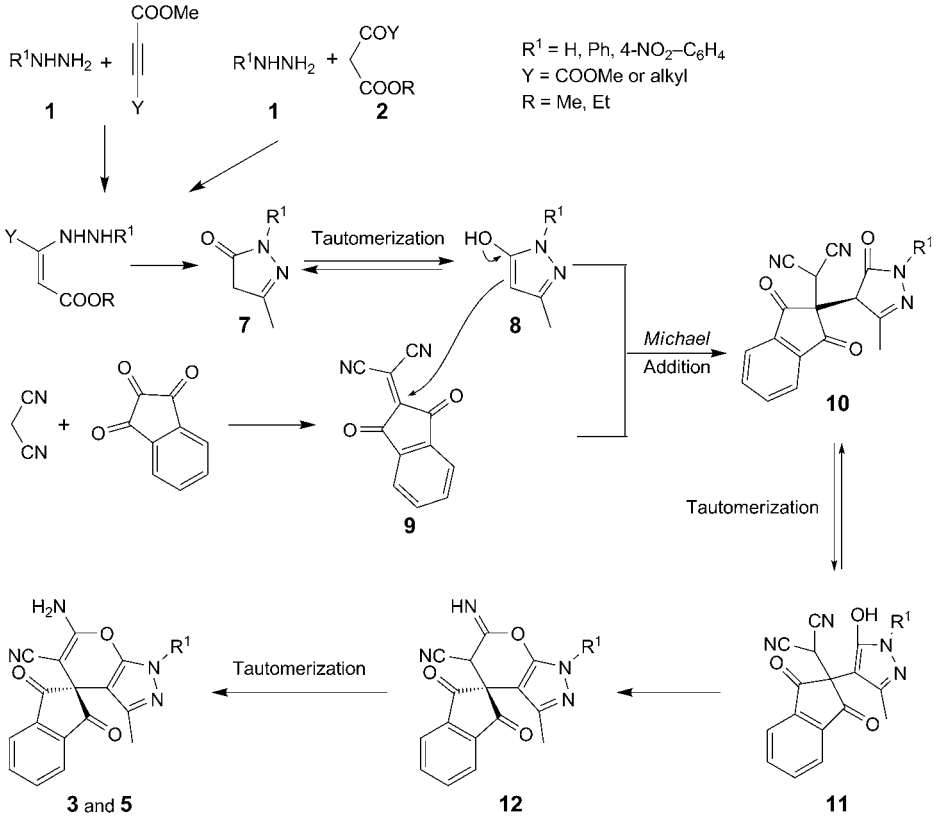


Fig. 2. *Intramolecular N–H$\cdots$O H-bond* (dashed line) *in compound* **6** (according to [3])

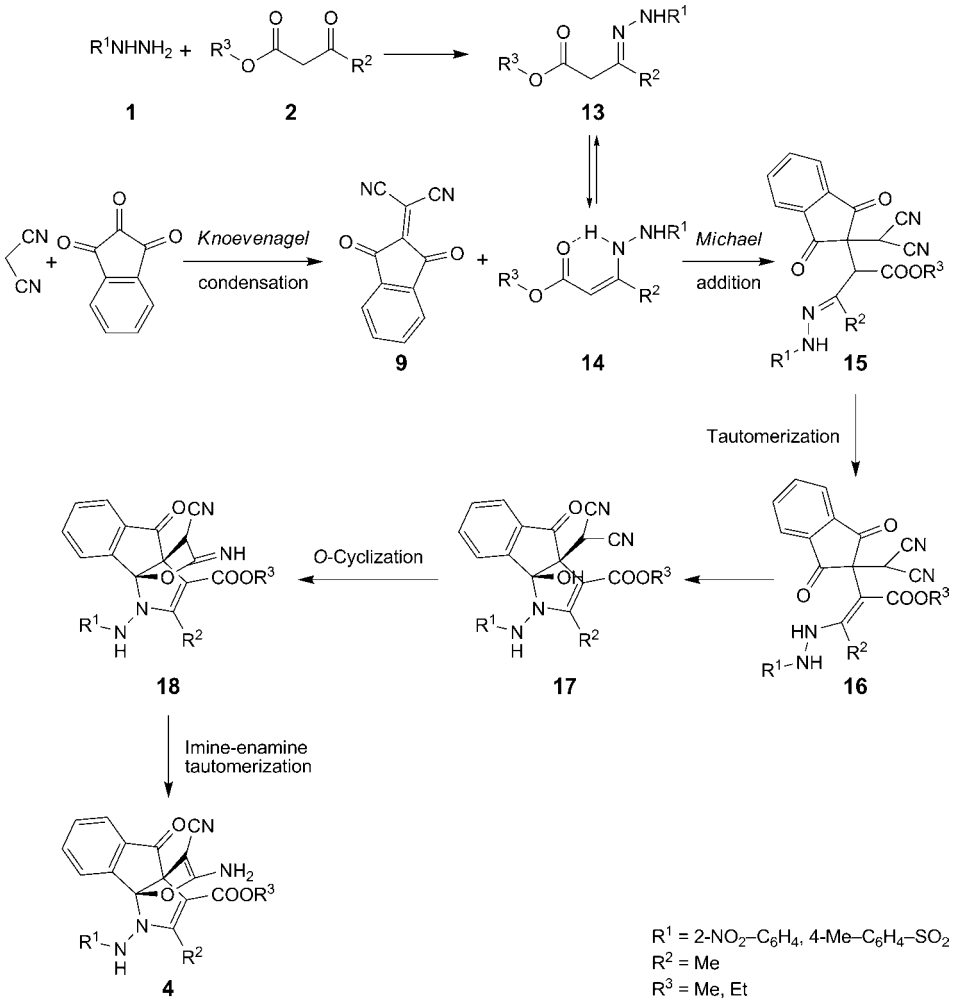
Scheme 2. *Proposed Mechanism of the Synthesis of* **3** *and* **5**



For the preparation of propellane, it is conceivable that initially the ninhydrin undergoes piperidine-promoted *Knoevenagel* condensation with malononitrile to give adduct **9**, which acts as *Michael* acceptor. The formation of intermediate **13** occurs through condensation of (2-nitrophenyl)hydrazine or tosylhydrazine with the C=O group of β-keto ester **2**. Then, the intermediate **14**, which is the tautomeric form of **13**, attacks the *Knoevenagel* adduct **9** in a *Michael*-type addition to produce an open-chain intermediate **15**, which is transformed to intermediate **16** through the migration of the H-atom. At this stage, the regioselective nucleophilic addition of the amino group to the C=O bond affords intermediate **17**, which is transformed to intermediate **18** through the *O*-cyclization. Then tautomerization of the imino group to the amino group could lead to product **4** (*Scheme 3*).

In conclusion, we have described a chemoselective four-component reaction for the synthesis of spiro-pyranopyrazoles and oxa-aza-[3.3.3]propellanes by taking advantage of the effect of an electron-withdrawing group on the reactivity of hydrazine derivatives. These reactions employ piperidine as catalyst. The advantages of this protocol include high yields, operational simplicity, regioselectivity, simple filtration,

Scheme 3. *Proposed Mechanism of the Synthesis of* **4**



and needing no metal catalyst. No extraction or separation by column chromatography is necessary.

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Experimental Part

General. The β-keto esters, hydrazine derivatives, ninhydrin, and malononitrile were obtained from *Merck* (Germany) and *Fluka* (Switzerland) and were used without further purification. M.p.: *Electrothermal 9100*. IR Spectra: as KBr pellets on a *NICOLET FT-IR 100* spectrometer; $\tilde{\nu}$ in cm^{-1}. ^{1}H- (300, 400, and 500 MHz) and ^{13}C-NMR (75, 100, and 125 MHz) spectra: *Bruker DRX-400 AVANCE* and *Bruker DRX-500 AVANCE* spectrometers. EI-MS: *FINNIGAN-MAT 8430* mass

spectrometer; at an ionization potential of 70 eV. Elemental analyses for C, H, and N: *Heraeus CHN–O–Rapid* analyzer.

General Procedure (exemplified for **3a**). A soln. of *methyl acetoacetate* (**2a**; 1 mmol) and *phenylhydrazine* (**1a**; 1 mmol) in EtOH (4 ml) was heated to reflux for 30 min. The soln. was cooled to ambient temp., and then a mixture of ninhydrin (1 mmol), malononitrile (1 mmol), and piperidine (one drop) in EtOH (4 ml) was added to the mixture. The resulting mixture was stirred at r.t. Upon completion (1 h), monitored by TLC, the mixture was filtered, and the precipitate was washed with EtOH (4 ml) to afford the pure product **3a**.

General Procedure (exemplified for **4b**). A soln. of *ethyl acetoacetate* (**2b**; 1 mmol) and *(2-nitrophenyl)hydrazine* (**1b**; 1 mmol) in EtOH (4 ml) was heated to reflux for 30 min. The soln. was cooled to r.t., and then a mixture of ninhydrin (1 mmol), malononitrile (1 mmol), and piperidine (one drop) in EtOH (4 ml) was added. The resulting mixture was stirred at r.t. Upon completion (48 h), monitored by TLC, the mixture was filtered, and the precipitate was washed with EtOH (4 ml) to afford the pure product **4b**.

General Procedure (exemplified for **5**). A soln. of *dimethyl acetylenedicarboxylate* (DMAD; 1 mmol) and $NH_2NH_2 \cdot H_2O$ (0.32 g, 1 mmol) in EtOH (4 ml) was stirred for 15 min at r.t. The soln. was cooled to r.t., and then a mixture of ninhydrin (1 mmol), malononitrile (1 mmol), and piperidine (one drop) in EtOH (4 ml) was added. The resulting mixture was stirred at r.t. Upon completion (1 h), monitored by TLC, the mixture was filtered, and the precipitate was washed with EtOH (4 ml) to afford the pure product **5**.

*6′-Amino-1,3-dihydro-3′-methyl-1,3-dioxo-1′-phenyl-1′*H*-spiro[indene-2,4′-pyrano[2,3-*c*]pyrazole]-5′-carbonitrile* (**3a**). Yield: 0.34 g (90%). Yellow powder. M.p. 205–207°. IR: 3375, 3308 (NH_2), 2202 (CN), 1716 (C=O), 1657 (NC=C), 1590, 1518 (Ar), 1455 (Me), 1265 (C–O). ^{1}H-NMR (500.1 MHz, (D_6)DMSO): 1.50 (*s*, 3 H); 7.34 (*t*, *J* = 7.4, 1 H); 7.49 (*t*, *J* = 7.9, 2 H); 7.73 (*d*, *J* = 7.9, 2 H); 7.86 (*s*, 2 H); 8.15 (*s*, 4 H). ^{13}C-NMR (125 MHz, (D_6)DMSO): 13.6; 53.4; 54.0; 94.5; 118.3; 121.4; 124.9; 127.9; 130.4; 137.7; 139.0; 141.5; 144.1; 146.3; 162.7; 199.7. EI-MS: 382 (100, M^+), 249 (60), 193 (18), 77 (44). Anal. calc. for $C_{22}H_{14}N_4O_3$ (382.38): C 69.11, H 3.69, N 14.65; found: C 69.18, H 3.62, N 14.58.

*6′-Amino-1,3-dihydro-3′-methyl-1′-(4-nitrophenyl)-1,3-dioxo-1′*H*-spiro[indene-2,4′-pyrano[2,3-*c*]pyrazole]-5′-carbonitrile* (**3b**). Yield: 0.36 g (85%). Green powder. M.p. 240° (dec.). IR: 3440, 3345 (NH_2), 2197 (CN), 1711 (C=O), 1657 (NC=C), 1594, 1519 (Ar), 1518, 1339 (NO_2), 1265 (C–O). ^{1}H-NMR (500.1 MHz, (D_6)DMSO): 1.54 (*s*, 3 H); 7.99 (*s*, 2 H); 8.05 (*d*, *J* = 8.8, 2 H); 8.15 (*s*, 4 H); 8.29 (*d*, *J* = 8.8, 2 H). ^{13}C-NMR (125 MHz, (D_6)DMSO): 13.7; 53.4; 53.8; 95.7; 118.0; 120.8; 125.0; 125.9; 137.0; 139.0; 141.4; 142.6; 146.0; 147.2; 162.5; 199.3. EI-MS: 427 (4, M^+), 354 (25), 208 (100), 152 (95), 104 (93), 76 (74), 50 (40). Anal. calc. for $C_{22}H_{13}N_5O_5$ (427.37): C 61.83, H 3.07, N 16.39; found: C 61.75, H 3.15, N, 16.43.

*6′-Amino-1,3-dihydro-1,3-dioxo-1′-phenyl-3′-propyl-1′*H*-spiro[indene-2,4′-pyrano[2,3-*c*]pyrazole]-5′-carbonitrile* (**3c**). Yield: 0.33 g (81%). Yellow powder. M.p. 206–208°. IR: 3383, 3320 (NH_2), 2201 (CN), 1712 (C=O), 1655 (NC=C), 1593, 1518 (Ar), 1260 (C–O). ^{1}H-NMR (500.1 MHz, (D_6)DMSO): 0.45 (*t*, *J* = 8.0, 3 H); 1.11 (*sext.*, *J* = 7.3, 2 H); 1.75 (*t*, *J* = 7.6, 2 H); 7.34 (*t*, *J* = 7.4, 1 H); 7.49 (*t*, *J* = 7.9, 2 H); 7.73 (*d*, *J* = 7.9, 2 H), 7.86 (*s*, 2 H), 8.16 (*s*, 4 H). ^{13}C-NMR (125 MHz, (D_6)DMSO): 14.2; 22.1; 30.1; 53.5; 54.2; 89.2; 118.4; 121.5; 124.9; 127.9; 130.4; 138.1; 139.0; 141.5; 143.3; 146.1; 162.6; 200.1. EI-MS: 410 (58, M^+), 217 (33), 167 (37), 149 (57), 129 (39), 105 (37), 93 (38), 81 (57), 69 (78), 55 (100). Anal. calc. for $C_{24}H_{18}N_4O_3$ (410.43): C 70.23, H 4.42, N 13.65; found: C 70.15, H 4.49, N 13.55.

*6′-Amino-1,3-dihydro-1′-(4-nitrophenyl)-1,3-dioxo-3′-propyl-1′*H*-spiro[indene-2,4′-pyrano[2,3-*c*]-pyrazole]-5′-carbonitrile* (**3d**). Yield: 0.40 g (87%). Yellow powder. M.p. 208° (dec.). IR: 3350, 3300 (NH_2), 2195 (CN), 1708 (C=O), 1657 (NC=C), 1593, 1520 (Ar), 1519, 1394 (NO_2), 1262 (C–N), 1109 (C–O). ^{1}H-NMR (400.1 MHz, (D_6)DMSO): 0.51 (*t*, *J* = 8.0, 3 H); 1.18 (*sext.*, *J* = 7.3, 2 H); 1.83 (*t*, *J* = 7.6, 2 H); 8.07 (*s*, 2 H); 8.12 (*d*, *J* = 8.8, 2 H); 8.23 (*s*, 4 H); 8.35 (*d*, *J* = 8.8, 2 H). ^{13}C-NMR (100 MHz, (D_6)DMSO): 13.3; 21.0; 29.2; 53.8; 54.6; 94.5; 118.0; 120.1; 124.1; 125.1; 137.7; 138.3; 140.5; 141.6; 145.0; 149.0; 161.5; 198.9. EI-MS: 455 (15, M^+), 354 (29), 247 (42), 185 (100), 105 (92), 76 (57). Anal. calc. for $C_{24}H_{17}N_5O_5$ (455.43): C 63.30, H 3.76, N 15.38; found: C 63.36, H 3.71, N 15.44.

*Methyl 10-Amino-11-cyano-2-methyl-1-[(2-nitrophenyl)amino]-4-oxo-1*H*,4*H*-8b,3a-(epoxyetheno)indeno[1,2-*b*]pyrrole-3-carboxylate* (**4a**). Yield: 0.34 g (75%). Green powder. M.p. 241–244°. IR:

3361, 3255 (NH_2), 2186 (CN), 1729 (CO_2), 1690 (C=O), 1640 (NC=C), 1565, 1325 (NO_2), 1272, 1210 (C–O of ester) 1148 (C–N), 1069 (C–O). ^{13}H-NMR (300.1 MHz, (D_6)DMSO): 2.03 (*s*, 3 H); 3.68 (*s*, 3 H); 7.00 (*t*, *J* = 7.6, 1 H); 7.48 (*s*, 2 H); 7.49 (*d*, *J* = 8.4, 1 H); 7.64 (*d*, *J* = 8.0, 1 H); 7.69 (*d*, *J* = 8.4, 1 H); 7.74 (*t*, *J* = 7.8, 1 H); 7.87 (*t*, *J* = 7.7, 2 H); 8.17 (*d*, *J* = 8.3, 2 H); 9.65 (*s*, 1 H). ^{13}C-NMR (75 MHz, (D_6)DMSO): 11.6; 49.9; 53.8; 66.9; 99.6; 108.8; 115.1; 117.8; 119.0; 124.7; 125.9; 126.1; 131.5; 132.1; 135.9; 136.4; 136.8; 142.6; 144.5; 160.6; 164.8; 167.1; 195.1. EI-MS: 459 (4, M^+), 416 (38), 279 (32), 247 (100), 138 (35). Anal. calc. for $C_{23}H_{17}N_5O_6$ (459.41): C 60.13, H 3.73, N 15.24; found: C 60.21, H 3.81, N 15.15.

*Ethyl 10-Amino-11-cyano-2-methyl-1-[(2-nitrophenyl)amino]-4-oxo-1*H,*4*H-*8b,3a-(epoxyetheno)indeno[1,2-*b*]pyrrole-3-carboxylate* (**4b**). Yield: 0.33 g (70%).Green powder. M.p. 237–240°. IR: 3390, 3201 (NH and NH_2), 2189 (CN), 1730 (CO_2), 1670 (CO), 1632 (NC=C), 1616, 1523, 1489 (Ar), 1460, 1325 (NO_2), 1273 (C–N), 1210, 1150 (C–O). ^{13}H-NMR (400.1 MHz, (D_6)DMSO): 1.35 (*t*, *J* = 7.0, 3 H); 2.04 (*s*, 3 H); 4.19 (*dq*, ^2J = 26.8, ^3J = 7.0, 2 H); 7.02 (*t*, *J* = 7.8, 1 H); 7.48 (*s*, 2 H), 7.51 (*d*, *J* = 8.8, 1 H); 7.66 (*d*, *J* = 7.6, 1 H); 7.72 (*t*, *J* = 7.2, 1 H); 7.77 (*d*, *J* = 7.6, 1 H); 7.88 (*t*, *J* = 7.2, 1 H); 7.89 (*t*, *J* = 7.6, 1 H); 8.19 (*d*, *J* = 8.4, 1 H); 9.67 (*s*, 1 H). ^{13}C-NMR (100 MHz, (D_6)DMSO): 11.6; 14.3; 53.8; 58.9; 70.0; 99.9; 108.7; 115.1; 117.9; 119.0; 124.7; 125.9; 126.0; 131.4; 132.1; 135.8; 136.5; 136.8; 142.5; 144.5; 160.5; 164.3; 167.1; 195.0. EI-MS: 473 (1, M^+), 430 (21), 310 (33), 264 (37), 249 (67), 138 (96), 65 (100). Anal. calc. for $C_{24}H_{19}N_5O_6$ (473.44): C 60.89, H 4.04, N 14.79; found: C 60.93, H 3.97, N 14.73.

*Methyl 10-Amino-11-cyano-2-methyl-1-[(4-methylphenyl)sulfonyl]-4-oxo-1*H,*4*H-*8b,3a-(epoxyetheno)indeno[1,2-*b*]pyrrole-3-carboxylate* (**4c**). Yield: 0.40 g (81%). Yellow powder. M.p. 185–186°. IR: 3570, 3441, 3337 (NH and NH_2), 2195 (CN), 1718 (C=O), 1653 (NC=C), 1337, 1157 (S=O). ^{1}H-NMR (400.1 MHz, (D_6)DMSO): 1.68 (*s*, 3 H); 2.45 (*s*, 3 H); 3.66 (*s*, 3 H); 7.49 (*s*, 2 H); 7.54 (*d*, *J* = 7.6, 2 H); 7.56 (*d*, *J* = 7.6, 1 H); 7.74 (*t*, *J* = 6.4, 1 H); 7.84 (*d*, *J* = 6.8, 3 H); 7.90 (*t*, *J* = 7.2, 1 H); 10.22 (*s*, 1 H). ^{13}C-NMR (100 MHz, (D_6)DMSO): 11.2; 21.1; 51.1; 53.1; 53.1; 70.4; 101.4; 108.5; 117.5; 124.7; 126.6; 127.6; 130.1; 131.7; 135.7; 135.8; 135.9; 142.2; 144.4; 158.4; 164.3; 170.1; 194.9. EI-MS: 492 (2, M^+), 449 (13), 294 (98), 262 (38), 139 (46), 91 (100), 65 (58). Anal. calc. for $C_{24}H_{20}N_4O_6S$ (492.5): C 58.53, H 4.09, N 11.38; found: C 58.11, H 3.79, N 11.80.

*Methyl 6′-Amino-5′-cyano-1,3-dihydro-1,3-dioxo-1′*H-*spiro[indene-2,4′-pyrano[2,3-*c*]pyrazole]-3′-carboxylate* (**5a**). Yield: 0.28 g (78%). Yellow powder. M.p. 248° (dec.). IR: 3436, 3298, 3185 (NH and NH_2), 2189 (CN), 1718 (C=O), 1636 (NC=C), 1583, 1487 (Ar), 1244 (C–N), 1066 (C–O). ^{1}H-NMR (400.1 MHz, (D_6)DMSO): 3.22 (*s*, 3 H); 7.71 (*s*, 2 H); 8.12 (*s*, 4 H); 14.22 (*s*, 1 H). ^{13}C-NMR (100 MHz, (D_6)DMSO): 51.8; 52.4; 54.0; 97.6; 117.6; 123.6; 128.3; 137.2; 140.8; 156.5; 157.5; 162.2; 198.6. EI-MS: 350 (83, M^+), 318 (67), 178 (48), 151 (38), 84 (67), 73 (100). Anal. calc. for $C_{17}H_{10}N_4O_5$ (350.29): C 58.29, H 2.88, N 15.99; found: C 58.33, H 2.94, N 15.91.

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