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SYNTHESIS AND REACTIONS OF SOME NEW SPIRO {INDENO[1,2-*b*] PYRAN-4,3'-INDOLINES}

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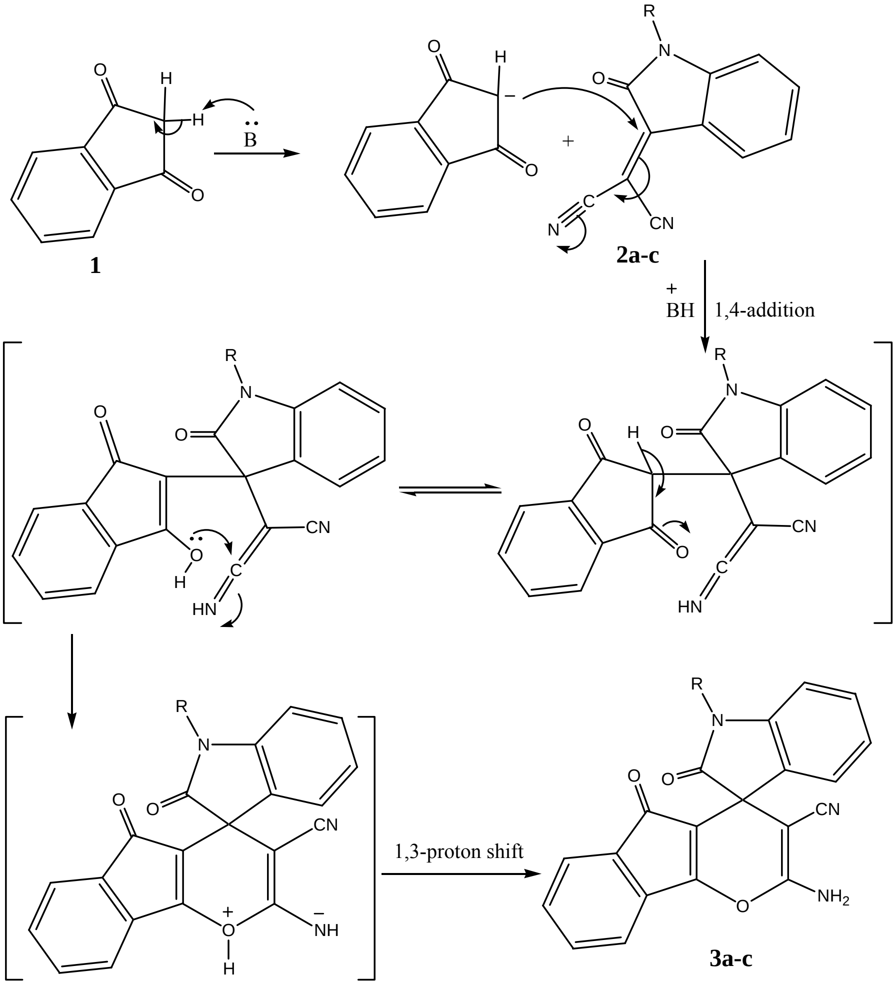
Abstract- Indan-1,3-dione **(1)** reacted with 3-dicyanomethylidene-2-oxoindolines **(2a-c)** in refluxed ethanol to afford 2-amino-3-cyanospiro{5H-indeno[1,2-*b*] pyrane-4,3'-(1'-substitutedindoline)}-2',5-diones **(3a-c)** which underwent different reactions to afford new spiro{indeno[2',1':5,6] pyrano[2,3-*d*]pyrimidine-5,3'-(1'-substitutedindoline)} **(4a,b)-(12a,b)** which are analogues of some reported biologically active spiropolycyclic compounds.

Several authors[1-11] reported the synthesis and application of spiroheterocyclic derivatives. From this point of view and in continuation to our previous work[12-20] we report herein the synthesis of some new spiroheterocycles of pyrans and pyranopyrimidines containing indoline moiety. Our syntheses started with the reaction of indan-2,3-dione **(1)** with 3-dicyanomethylidene-2-oxoindolines **(2a-c)** in absolute ethanol and catalytic amount of triethyl amine to give 2-amino-3-cyanospiro{5H-indeno [1,2-*b*]pyran-4,3'-indoline} derivatives **(3a-c)** via a type of Michael addition shown in **(Scheme 1)**.

The structures of the prepared compounds **(3a-c)** were established from their elemental analyses and spectral data. The IR spectrum of compound **(3a)** showed characteristic absorption bands at ύ 3350-3300 for NH_2, 3200 for NH, 2200, for CN, 1725, and 1700 cm^{-1} corresponding to C=O in indene and indoline rings respectively. The ^{1}H-NMR spectrum of **(3b)** showed multiplet signal at δ 7.81-6.38 for aromatic protons and singlet signals at 4.80, 3.20, for NH_2, and methyl protons, respectively.

Compounds **(3a,b)** were subject to further reactions to give fused spiroheterocyclic systems incorporate pyrimidine nucleus in addition to indeno[1,2-*b*]pyran and indoline moieties. Thus the reaction of **(3a,b)** with formic acid gave spiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} derivatives **(4a,b)**. Reaction of **(3b)** with acetic anhydride/pyridine mixture (2/1, v/v) afforded 1',2-dimethylspiro-

{6H-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',4,6(3H)-trione **(5)**. Compounds **(4a,b)** and **5** were formed via Dimroth rearrangement illustrated in **(Scheme 2)**.



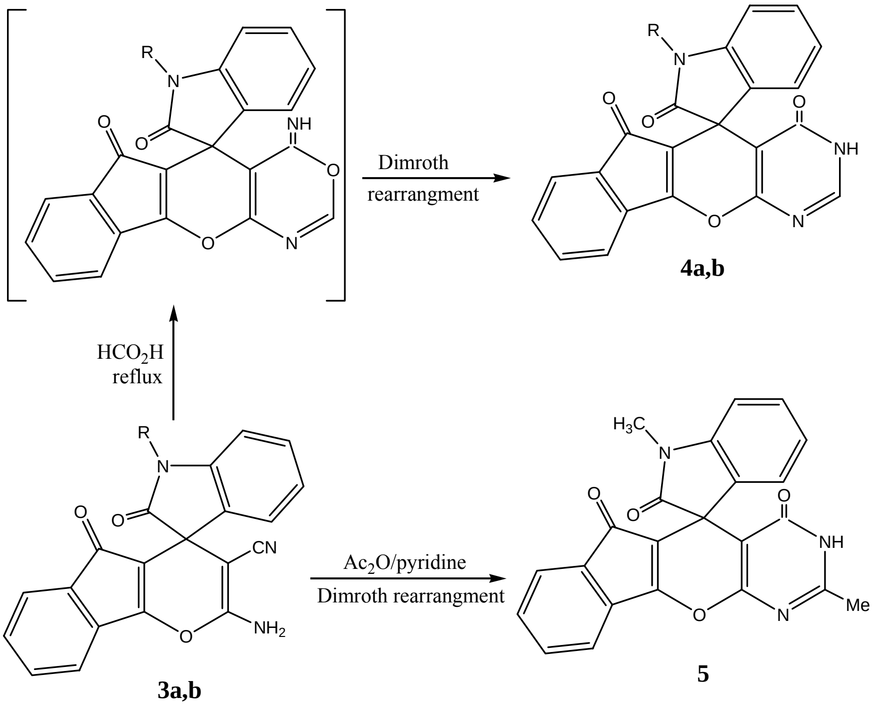
a, R= H; b, R=Me; c, R= Et

Scheme 1

The IR spectrum of compounds **(4a)**, (**4b)**, and (**5**) revealed absorption bands at ύ 3300, 3350, 3400 respectively, are corresponding to NH group with disappearance of the absorption bands corresponding to NH group with disappearance of the absorption bands corresponding to the cyano and amino groups. The ^{1}H-NMR spectrum of compounds **(4a**) and **(4b)** revealed a singlet signal for one proton at δ 11.64, 11.12 respectively, are exchanged with D_2O indicated to NH proton with absence of the signal corresponding to NH_2 protons. However, the ^{1}H-NMR spectrum of compound **(5)** revealed a singlet signal at δ 11.60 exchanged with D_2O for NH proton, and two singlet signals at 3.46 and 2.50 for two methyl protons.

Interaction of **(3a,b)** with formamide afforded 4-aminospiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*] pyrimidine-5,3'-indoline} derivatives **(6a,b)**. Reaction of **(3a,b)** with malononitrile in pyridine or dry dioxane/Et_3N afforded the condensation product 2-amino-3-cyano-5-dicyanomethylidenespiro{5*H*-indeno

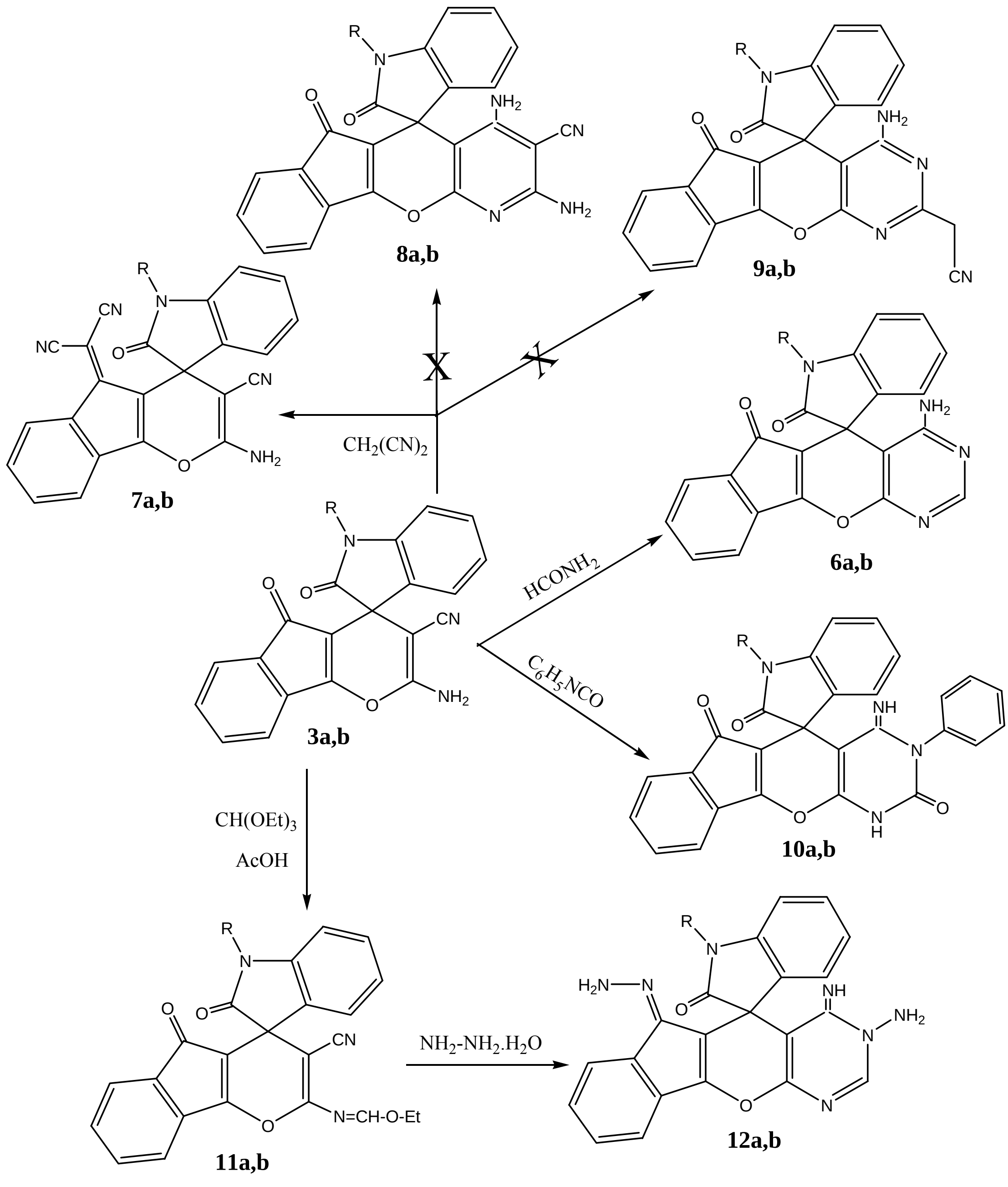
[1,2-*b*]pyran-4,3'-indoline} derivatives **(7a,b)** rather than spiro{indeno[2',1':5,6]pyrano[2,3-*b*] pyridine-5,3'-indoline} derivatives **(8a,b)** or spiro{indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} derivatives **(9a,b)**. Reaction of **(3a,b)** with phenyl isocyanate in refluxed pyridine afforded 4-iminospiro {1,2,3,4-tetrahydro-6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} derivatives **(10a,b)**. The reaction of **(3a,b)** with triethyl orthoformate in the presence of a few drops of glacial acetic acid gave ethoxymethyleniminospiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline} derivatives **(11a,b)**. Finally, compounds **(11a,b)** reacted with hydrazine hydrate in benzene to afford 6-hydrazono-4-iminospiro{3,4-dihydro-6*H*-indeno-[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} derivatives **(12a,b) (Scheme 3)**.



Scheme 2

The chemical structures of all compounds **(6a,b)-(12a,b)** were established based on their elemental and spectral analyses. The IR spectrum of compounds **(6a)** and **(6b)** revealed absorption band at ύ 3390-3300 corresponding to NH_2 group with disappearance of the absorption band corresponding to the cyano group. The structures of 2-amino-3-cyano-5-dicyanomethylidenespiro{5*H*-indeno[1,2-*b*]pyrane-4,3'-indoline} derivatives **(7a,b)** were tentatively preferred for these products based on its ^{1}H-NMR spectrum of compound **(7a)** for example, shown the following signals: two singlet signals at δ 10.76, and 4.71 exchanged with D_2O for NH, and NH_2 protons respectively and the absence of any protons attached to sp^3 carbons. The IR spectrum of compounds **(10a,b)** shown the absorption bands at ύ 3320, and 3290 for two NH groups with absence of the absorption band corresponding to cyano group. The IR spectrum of compounds **(11a,b)** shown the disappearance of the band for NH_2 group, and the ^{1}H-NMR spectrum of compound **(11b)** shown the following signals: multiplet signals at δ 7.69-6.81 for aromatic protons, singlet signal at 5.16 for the methine proton, quartet and triplet signals at 4.05 and 1.12 respectively for ethyl protons, and singlet signal at 3.21 for the methyl protons. The IR spectrum of compounds **(12b)** revealed the absorption bands ύ 3360-3210 for NH_2, 3150 for NH group with absence of the band for cyano group and its ^{1}H-NMR spectrum shown the absence of signals corresponding to ethyl protons.

The previously discussed reactions described a simple synthetic procedure to prepare new spiroheterocyclic derivatives of indenopyrans and indenopyranopyrimidines which might have important biological applications.



Scheme 3

EXPERIMENTAL

The time required for completion of each reaction was monitored by TLC. Melting points are uncorrected and were measured on a Gallen Kamp apparatus. The IR spectra were recorded on a Shimadz 470 IR spectrometer (KBr) ύ cm^{-1}. The ^{1}H-NMR spectra were measured on Varian EM-200 MHz spectrometer with TMS as internal standard. Mass spectra were determined on a Jeol-600 spectrometer. Elemental analyses were performed on an elemental analyses system GmbH varioel $V_{2.3}$.

Synthesis of 2-amino-3-cyanospiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione and its 1'-alkyl derivatives (3a-c):

General procedure[19]: A mixture of indan-1,3-dione **(1)** (1.46 g, 0.01 mol) and 3-dicyanomethylidine-2-oxoindolines **(2a-c)** (0.01 mol) in absolute EtOH (20 mL) in the presence of Et_3N (1 mL) was stirred at rt for 3 h, and/or refluxed for 2 h. Then the product was precipitated, collected by filtration, dried, and recrystallized from AcOH.

2-Amino-3-cyanospiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione (3a)

Brown crystals; yield (81%); mp 171-174°C; IR: ύ cm^{-1} 3350, 3300 (NH_2), 3200 (NH), 2200 (CN), 1725 (C=O), 1700 (C=O); ^{1}H-NMR (DMSO-*d*$_6$): δ 9.45 (s, 1H, NH), 7.75-6.56 (m, 8H, Ar-H), 4.75 (s, 2H, NH_2); Anal. Calcd for $C_{20}H_{11}N_3O_3$ (341.17): C, 70.38; H, 3.25; N, 12.31. Found: C, 70.35; H, 3.30; N, 12.24.

2-Amino-3-cyano-1'-methylspiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione (3b)

Bright brown crystals; yield (60%); mp 230-233°C; IR: ύ cm^{-1} 3310-3200 (NH_2), 2200 (CN), 1718 (C=O), 1705 (C=O); ^{1}H-NMR (DMSO-*d*$_6$): δ 7.81-6.38 (m, 8H, Ar-H), 4.81 (s, 2H, NH_2), 3.20 (s, 3H, CH_3); Anal. Calcd for $C_{21}H_{13}N_3O_3$ (355.18): C, 70.98; H, 3.69; N, 11.83. Found: C, 70.92; H, 3.64; N, 11.78.

2-Amino-3-cyano-1'-ethylspiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione (3c)

Bright orange crystals; yield (45%); mp 208-210°C; IR: ύ cm^{-1} 3300-3200 (NH_2), 2200 (CN), 1710 (C=O); ^{1}H-NMR (DMSO-*d*$_6$): δ 7.51-6.60 (m, 8H, Ar-H), 3.50 (s, 2H, NH_2), 2.75 (q, 2H, CH_2), 0.95 (s,3H, CH_3); Anal. Calcd for $C_{22}H_{15}N_3O_3$ (369.19): C, 71.54; H, 4.09; N, 11.38. Found: C, 71.48; H, 4.05; N, 11.35.

Synthesis of spiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',4,6(3*H*)-trione and its 1'-methyl derivative (4a,b):

General procedure: A mixture of **3a** or **3b** (0.01 mol) and formic acid (5 mL, 85%) was heated under reflux for 5 h. After cooling the reaction mixture was poured into an ice/water mixture and the formed solid product thus formed was filtered off, dried and recrystallized from AcOH.

Spiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',4,6(3*H*)-trione (4a)

Scarlet red crystals; yield (50%); mp 161-165°C; IR: ύ cm^{-1} 3300 (NH), 1710 (C=O), 1700 (C=O), 1620

(C=N); ^{1}H-NMR (DMSO-d_6): δ 11.64 (s, 1H, NH indoline), 10.59 (s, 1H, NH pyrimidine), 7.90-6.91 (m, 9H, Ar-H and CH pyrimidine); Anal. Calcd for $C_{21}H_{11}N_3O_4$ (369.17): C, 68.29; H, 3.00; N, 11.38. Found: C, 68.25; H, 2.89; N, 11.16.

1'-Methylspiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',4,6(3*H*)-trione (4b)

Brown crystals; yield (50%); mp 155-157°C; IR: ύ cm^{-1} 3350 (NH), 1710 (C=O), 1700 (C=O), 1620 (C=N); ^{1}H-NMR (DMSO-d_6): δ 11.21 (s, 1H, NH pyrimidine), 7.90-6.94 (m, 9H, Ar-H and CH pyrimidine), 3.50 (s, 3H, CH_3); Anal. Calcd for $C_{22}H_{13}N_3O_4$ (383.18): C, 68.93; H, 3.42; N, 10.96. Found: C, 68.80; H, 3.31; N, 10.76.

Synthesis of 1',2-dimethylspiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',4,6-(3*H*)-trione (5):

A solution of **3b** (3.55 g, 0.01 mol) in Ac_2O/pyridine mixture (15 mL, 2/1 v/v) was heated in water bath for 5h. Then cooled to rt, poured into an ice/water mixture, the formed solid product was collected by filtration, washed several times with cold water, dried and recrystallized from EtOH/AcOH (3/1) to give brown crystals; yield (66%); mp 241-245°C; IR: ύ cm^{-1} 3400 (NH), 1715 (C=O), 1700 (C=O), 1650 (C=N); ^{1}H-NMR (DMSO-d_6): δ 11.61 (s, 1H, NH), 7.95-6.90 (m, 8H, Ar-H), 3.46 (s, 3H, CH_3), 2.50 (s, 3H, CH_3); Anal. Calcd for $C_{23}H_{15}N_3O_4$ (397.19): C, 69.52; H, 3.80; N, 10.57. Found: C, 69.42; H, 3.58; N, 10.56.

Synthesis of 4-aminospiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',6-dione and its 1'-methyl derivative (6a,b):

General procedure: A mixture of **3a** or **3b** (0.05 mol), formamide (10 mL), and DMF (1 mL) was heated under reflux for 4 h. The reaction mixture was cooled, then poured into an ice/water mixture and the formed product was filtered off, washed several times by cold water, dried and recrystallized from EtOH.

4-Aminospiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',6-dione (6a)

Green crystals; yield (40%); mp 220-224°C; IR: ύ cm^{-1} 3390-3300 (NH_2), 3290 (NH), 1700 (C=O), 1690 (C=O); ^{1}H-NMR (DMSO-d_6): δ 10.65 (s, 1H, NH), 7.90-6.94 (m, 9H, Ar-H and CH pyrimidine), 5.61 (s, 2H, NH_2); Anal. Calcd for $C_{21}H_{12}N_4O_3$ (368.18): C, 68.48; H, 3.28; N, 15.210. Found: C, 68.38; H, 3.11; N, 15.21.

4-Amino-1'-methylspiro{6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline}-2',6-dione (6b)

Green crystals; yield (46%); mp 240-243°C; IR: ύ cm^{-1} 3340-3200 (NH_2), 1710 (C=O), 1690 (C=O), 1625 (C=N); ^{1}H-NMR (DMSO-d_6): δ 7.91-6.95 (m, 9H, Ar-H and CH pyrimidine), 5.52 (s, 2H, NH_2), 3.46 (s, 3H, CH_3); Anal. Calcd for $C_{22}H_{14}N_4O_3$ (382.19): C, 69.10; H, 3.69; N, 14.65. Found: C, 68.02; H, 3.59; N, 14.21.

Synthesis of 2-amino-3-cyano-5-dicyanomethylidenespiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indolin}-2'-one and its 1'-methyl derivative (7a,b):

General procedure *A*: A mixture of **3a** or **3b** (0.05 mol) and malononitrile (0.33 g, 0.05 mol) in pyridine (10 mL) was heated under reflux for 3 h. The reaction mixture was cooled to rt, then poured into an ice/water mixture containing a few drops of AcOH. The formed solid product was filtered off, washed by cold water, dried and recrystallized from dioxane.

General procedure *B*: A mixture of **3a** or **3b** (0.05 mol) and malononitrile (0.33 g, 0.05 mol) in dry dioxane (10 mL) in the presence of catalytic amount of Et_3N was stirred at rt for 2 h. The formed solid product was collected by filtration, dried and recrystallized from dioxane.

2-Amino-3-cyano-5-dicyanomethylidenespiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indolin}-2'-one (7a)

Violet crystals; yield (68%); mp 178-180°C; IR: ύ cm^{-1} 3320, 3210 (NH_2), 3190 (NH), 2200 (CN), 2190 (CN), 1700 (C=O); ^{1}H-NMR (DMSO-d_6): δ 10.76 (s, 1H, NH), 8.00-6.50 (m, 8H, Ar-H), 4.71 (s, 2H, NH_2); Anal. Calcd for $C_{23}H_{11}N_5O_2$ (389.21): C, 70.95; H, 2.85; N, 17.99. Found: C, 70.81; H, 2.73; N, 17.89.

2-Amino-3-cyano-5-dicyanomethylidene-1'-methylspiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indolin}-2'-one (7b)

Violet crystals; yield (70%); mp 190-193°C; IR: ύ cm^{-1} 3300-3200 (NH_2), 2200 (CN), 2190 (CN), 1705 (C=O); ^{1}H-NMR (DMSO-d_6): δ 8.05-6.60 (m, 8H, Ar-H), 4.70 (s, 2H, NH_2), 3.60 (s, 3H, CH_3); Anal. Calcd for $C_{24}H_{13}N_5O_2$ (403.22): C, 71.46; H, 3.25; N, 17.36. Found: C, 71.39; H, 3.06; N, 17.35.

Synthesis of 4-imino-2,2',6-trioxospiro{1,2,3,4-tetrahydro-6*H*-indeno[2',1':5,6]pyrano[2,3-*d*] pyrimidine-5,3'-indoline} and its 1'-methyl derivative (10a,b):

General procedure: A solution of **3a** or **3b** (0.01 mol) in pyridine (15 mL) and phenylisocyanate (0.01 mol) was heated under reflux for 5 h. The reaction mixture was cooled to rt, and then poured into an ice/water mixture; the formed product was collected by filtration, washed several times by cold water, dried and recrystallized from EtOH.

4-Imino-2,2',6-trioxospiro{1,2,3,4-tetrahydro-6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} (10a)

Green crystals; yield (40%); mp 182-185°C; IR: ύ cm^{-1} 3320 (NH), 3290 (NH), 3190 (NH), 1710 (C=O), 1700 (C=O), 1645 (C=N); ^{1}H-NMR (DMSO-d_6): δ 10.45 (s, 1H, NH), 8.60 (s, 1H, NH), 7.81 (s, 1H, NH), 7.65-6.60 (m, 13H, Ar-H); Anal. Calcd for $C_{27}H_{16}N_4O_4$ (460.23): C, 70.43; H, 3.50; N, 12.17. Found: C, 70.40; H, 3.32; N, 12.10.

4-Imino-1'-methyl-2,2',6-trioxospiro{1,2,3,4-tetrahydro-6*H*-indeno[2',1':5,6]pyrano[2,3-*d*]pyrimidine-5,3'-indoline} (10b)

Brown crystals; yield (45%); mp 190-192°C; IR: ύ cm^{-1} 3320 (NH), 3290 (NH), 3190 (NH), 1710 (C=O), 1700 (C=O), 1640 (C=N); ^{1}H-NMR (DMSO-d_6): δ 8.80 (s, 1H, NH), 7.85 (s, 1H, NH), 7.60-6.80 (m, 13H, Ar-H), 3.42 (s, 3H, CH_3); Anal. Calcd for $C_{28}H_{18}N_4O_4$ (474.24): C, 70.88; H, 3.82; N, 11.81. Found:

C, 70.78; H, 3.78; N, 11.75.

Synthesis of 3-cyano-2-ethoxymethyleniminospiro{5*H*-indeno[1,2-*b*]pyrane-4,3'-indoline}-2',5-dione and its 1'-methyl derivative (11a,b):

General procedure: A mixture of **3a** or **3b** (0.01 mol) and triethyl orthoformate (0.5 mol)/glacial AcOH (1 mL) was heated under reflux for 5 h. The reaction mixture was cooled then the solvent was removed under vacuum, the formed product was collected by filtration, washed several times by cold water, dried and recrystallized from EtOH.

3-Cyano-2-ethoxymethyleniminospiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione (11a)

Brown crystals; yield (65%); mp 150-154°C; IR: ύ cm^{-1} 3250 (NH), 2210 (CN), 1715 (C=O), 1705 (C=O), 1665 (C=N); ^{1}H-NMR (DMSO-d_6): δ 10.40 (s, 1H, NH), 7.67-6.80 (m, 8H, Ar-H), 5.05 (s, 1H, CH), 4.10 (q, 2H, CH_2), 1.35 (t, 3H, CH_3); Anal. Calcd for $C_{23}H_{15}N_3O_4$ (397.19): C, 69.52; H, 3.80; N, 10.57. Found: C, 69.46; H, 3.70; N, 10.49.

3-Cyano-2-ethoxymethylenimino-1'-methylospiro{5*H*-indeno[1,2-*b*]pyran-4,3'-indoline}-2',5-dione (11b)

Brown crystals; yield (67%); mp 165-167°C; IR: ύ cm^{-1} 2200 (CN), 1715 (C=O), 1705 (C=O), 1660 (C=N); ^{1}H-NMR (DMSO-d_6): δ 7.69-6.81 (m, 8H, Ar-H), 5.16 (s, 1H, CH), 4.05 (q, 2H, CH_2), 3.21 (s, 3H, CH_3), 1.12 (t, 3H, CH_3); Anal. Calcd for $C_{24}H_{17}N_3O_4$ (411.20): C, 70.07; H, 4.16; N, 10.21. Found: C, 69.91; H, 4.10; N, 10.09.

Synthesis of 3-amino-6-hydrazono-4-iminospiro{3,4-dihydro-6*H*-indeno[2',1':5,6]pyran[2,3-*d*] pyrimidine-5,3'-indolin}-2'-one and its 1'-methyl derivative (12a,b):

General procedure: A mixture of **11a** or **11b** (0.01 mol) and hydrazine hydrate (80%) (0.015 mol) in benzene (15 mL) was stirred at rt for 1 h. The formed product was collected by filtration, dried and recrystallized from benzene.

3-Amino-6-hydrazono-4-iminospiro{3,4-dihydro-6*H*-indeno[2',1':5,6]pyran[2,3-*d*]pyrimidine-5,3'-indolin}-2'-one (12a)

Scarlet red crystals; yield (65%); mp 200-204°C; IR: ύ cm^{-1} 3370, 3210 (NH_2), 3160 (NH), 1700 (C=O), 1680 (C=N); ^{1}H-NMR (DMSO-d_6): δ 10.54 (s, 1H, NH), 10.47 (s, 1H, NH), 9.56 (s, 2H, NH_2), 9.49 (s, 2H, NH_2), 8.74-6.49 (m, 9H, Ar-H); Anal. Calcd for $C_{21}H_{15}N_7O_2$ (397.19): C, 63.47; H, 3.80; N, 24.67. Found: C, 63.39; H, 3.68; N, 24.57.

3-amino-6-hydrazono-4-imino-1'-methylspiro{3,4-dihydro-6*H*-indeno[2',1':5,6]pyran[2,3-*d*]pyrimidine-5,3'-indolin}-2'-one (12b)

Red crystals; yield (60%); mp 185-188°C; IR: ύ cm^{-1} 3360, 3210 (NH_2), 3150 (NH), 1705 (C=O), 1680 (C=N); ^{1}H-NMR (DMSO-d_6): δ 10.54 (s, 1H, NH), 9.61 (s, 2H, NH_2), 9.45 (s, 2H, NH_2), 8.33-6.50 (m, 9H, Ar-H), 3.11 (s, 3H, CH_3); Anal. Calcd for $C_{22}H_{17}N_7O_2$ (411.20): C, 64.23; H, 4.16; N, 23.83. Found:

C, 64.10; H, 4.00; N, 23.75.

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