Selenium-containing heterocycles: Part 2. Reactions of 3-amino-4,6-dimethylselenolo[2,3-*b*]pyridine-2-carbonitrile and related fused tetracyclic systems

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A novel series of pyrido[3',2':4,5]selenolo[3,2-*d*]pyrimidine, 7,9-dimethylpyrido [3',2':4,5]selenolo[3,2-*d*]pyrimidine-2,4-(*1H,3H*)-dithione, 7,9-dimethylpyrido[3',2':4,5]selenolo[2,3-*e*]tetrazolo[1,5-*c*]pyrimidine-6(*5H*)-thione, 9,11-dimethylpyrido[3',2':4,5]selenolo[2,3-*e*][1,2,4]triazolo[1,5-*c*]pyrimidine, 7,9-dimethylpyrido[3',2':4,5]selenolo[2,3-*e*]imidazo[1,2-*c*]pyrimidine and 10,12-dimethylpyrido[3",2":4',5']selenolo[3',2':4,5]pyrimido[1,6-*a*]pyrimidine derivatives were prepared from 3-amino-4,6-dimethylselenolo[2,3-*b*]pyridine-2-carbonitrile.

Keywords: *o*-aminonitriles, fused selenophenes, imidazoles, pyridines, pyrimidines, tetrazoles, 1,2,4-triazoles

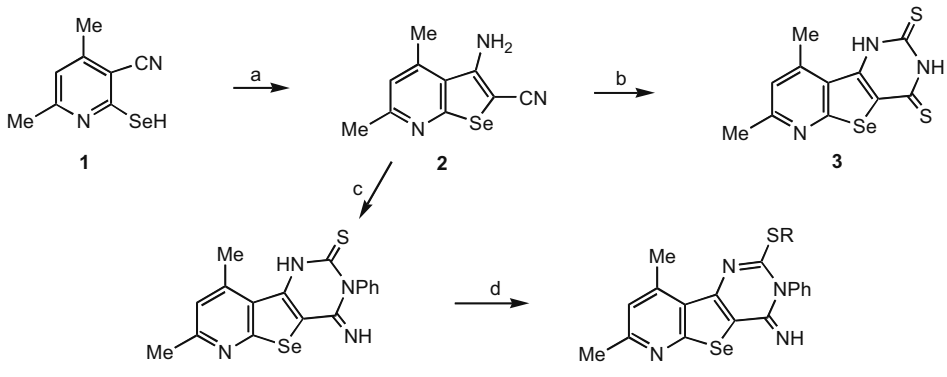
A literature survey indicates that only few publications are concerned with the incorporation of a selenium atom in the pyridine ring.[1-5] Consequently, the synthesis of new classes of heterocyclic systems containing the selenolopyridine moiety may be considered to be a virgin research area. Moreover, previous work in our laboratory describes the synthesis of pyrimidoselenolo[2,3-*b*]quinoline[6] and pyrimidoselenolo[2,3-*c*]pyridazine derivatives,[7] which indicate that certain compounds bearing the selenophene and quinoline or pyridazine nucleus possess significant anti-inflammatory and analgesic activities with strong fungicidal effects. Therefore, new efficient syntheses are an attractive goal of chemical research. In Part 1[8] we published the synthesis of selenolo[2,3-*b*]pyridine, pyrido[3',2':4,5]selenolo [3,2-*d*]pyrimidine, 7,8-dihydro-2,4-dimethylpyrrolo[1,2-*a*] pyrido[3',2':4,5]selenolo[3,2-*d*]pyrimidin-10(*6H*)-one and 7,9-dimethylpyrido[3',2':4,5]selenolo[3,2-*d*][1,2,4]triazolo [4,3-*c*]pyrimidine derivatives. The aforementioned properties of selenium organic compounds prompted further efforts in continuation of our work[6-10] on the quest for novel heterocyclic systems containing selenium exhibiting biological activity, and we report herein new classes of fused selenium-containing derivatives containing the selenolo[2,3-*b*]pyridine system.

Results and discussion

Our approach to the synthesis of the target compounds started from 3-amino-4,6-dimethylselenolo[2,3-*b*]pyridine-2-carbonitrile (**2**) which was prepared from the selenol **1** as previously described by Litvinov.[2] Compound **2** reacted with carbon disulfide and phenyl isothiocyanate to give the 7,9-dimethylpyrido[3',2':4,5]selenolo[3,2-*d*]pyrimidine-2,4-(*1H,3H*)-dithione (**3**) and 3,4-dihydro-7,9-dimethyl-3-phenyl-4-iminopyrido[3',2':4,5]selenolo[3,2-*d*]pyrimidine-2(*1H*)-thione (**4**), respectively (Scheme 1). The reactivity of the thione group of compound **4** was tested by alkylation with allyl bromide and methyl iodide which afforded derivatives (**5a,b)** respectively.

Heating of compound **2** with sodium azide and ammonium chloride in DMF followed by acidification of the reaction mixture led to the formation of the tetrazolyl compound **6**. The IR spectrum of compound **6** showed characteristic bands at 3300, 3400, 3500 cm^{-1} for NH_2 and NH, and the disappearance of the cyano group of compound **2**. The mass spectrum of **6** showed a molecular ion peak at *m/z* 294 (M^+, 100) which is in agreement with its molecular formula ($C_{10}H_{10}N_6Se$). When compound **6** was allowed to react with carbon disulfide and with aromatic aldehydes, the tetrazolopyridoselenolopyrimidine derivatives **7** and **8a–c** respectively were obtained in good yields.

In contrast, the reaction of tetrazolyl derivative **6** with triethyl orthoformate produced the azidopyrimidine derivative (**10**) (Scheme 2). The structure of compounds **7**, **8a–c** and **10** were assigned by elemental and spectral analyses. The IR spectrum of compound **10** showed a characteristic band at 2150 cm^{-1} for the azido group.



Reagents: a, $ClCH_2CN$; b, CS_2; c, PhNCS; d, Allyl-Br or Me-I

5a, R = $-CH_2-CH=CH_2$
5b, R = CH_3

Scheme 1

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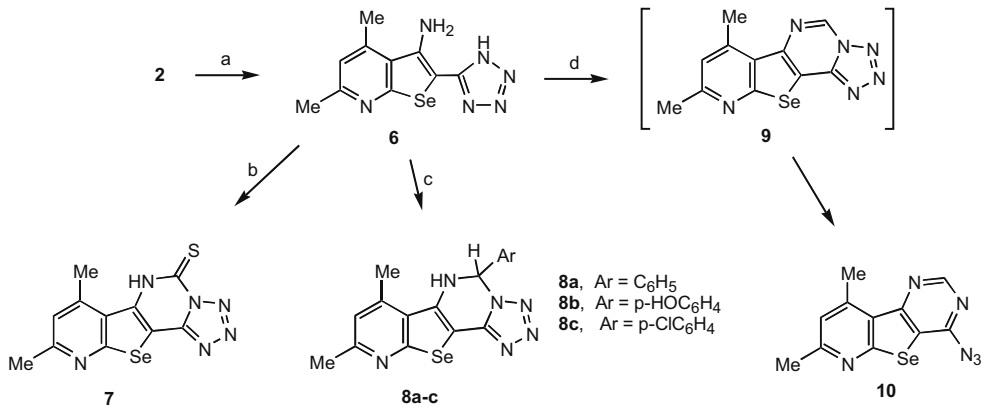


Reagents: a, NaN_3/NH_4Cl; b, CS_2; c, ArCHO; d, triethyl orthoformate

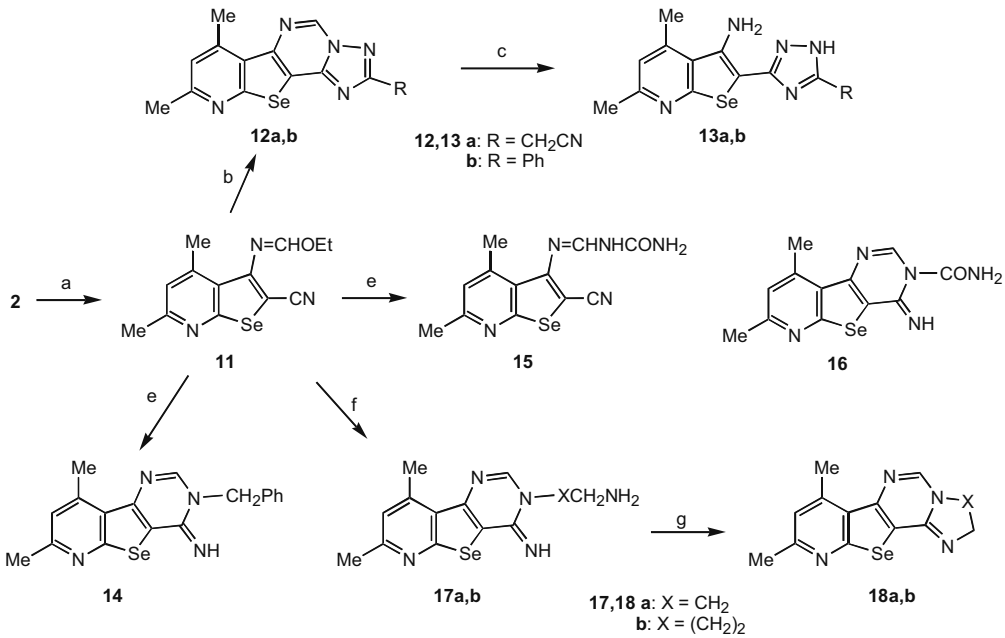
Scheme 2

Reaction of compound **2** with triethyl orthoformate afforded the methanimidate derivative **11**, which was prepared as previously described.[8] Heating of compound **11** with equimolar amounts of cyanoacetic acid or benzoic acid hydrazide furnished the 2-(substituted alkyl/phenyl)-7,9-dimethylpyrido [3',2':4,5]selenolo[2,3-*e*][1,2,4]triazolo[1,5-*c*]pyrimidine derivatives (**12a**,**b**). Treatment of **12a**,**b** with hydrochloric acid induced pyrimidine ring opening[11] to furnish the amines **13a**,**b** in excellent yield (Scheme 3).

Finally, stirring of **11** with benzylamine led to amino-deethoxylation accompanied by cyclisation to give 3-benzyl-7,9-dimethylpyrido[3',2':4,5]selenolo[3,2-*d*]pyrimidin-4(3*H*)-imine (**14**), while heating **11** with urea in acetic acid produced N'-carboxamido-*N*-(2-cyano-4,6-dimethylselenolo[2,3-*b*] pyridin-3-yl)methanimidate (**15**) rather than the expected fused pyrimidine (**16**) (Scheme 3). The IR spectrum of compound **15** showed characteristic bands at 3200–3400 cm^{-1} (NH, NH_2) and 2200 cm^{-1} for the CN group. The mass spectrum showed a molecular ion peak at *m/z* 321 (M^+, 0.5) in agreement with its molecular formula ($C_{12}H_{11}N_5OSe$). The ^1H NMR showed a characteristic peak at: δ 8.7 due to the CH=N group. Incorporation of the imidazoline and pyrimidine moieties into the selenolopyridine structure was successfully accomplished by reacting **11** with ethylenediamine and propylenediamine to give the intermediate derivatives **17a,b** which were boiled in ethanol to give 2,3-dihydro-7,9-dimethyl-pyrido[3',2':4,5]selenolo[2,3-*e*]imidazo[1,2-*c*]pyrimidine (**18a**) and 3,4-dihydro-8,10-dimethyl-2*H*-pyrido[3",2":4',5'] selenolo[3',2':4,5]pyrimido[1,6-*a*]pyrimidine (**18b**) respectively.

Experimental

Melting points were determined using a Kofler melting point apparatus. IR spectra were recorded on a Pye-Unicam SP3-100 instrument in KBr. The mass spectra (EI, 70 eV, ion source



Reagents: a, $HC(OEt)_3$; b, $RCONHNH_2$; c, 20% HCl/H_2O; d, $PhCH_2NH_2$; e, urea; f, H_2N-X-CH_2NH_2; g, EtOH, Δ

Scheme 3

temperature 210 °C) were recorded on a JEOL JMS600 instrument. ^{1}H NMR spectra were obtained on a Varian spectrometer (90 MHz) using tetramethylsilane as internal reference. ^{13}C NMR spectra were recorded on a Mercury-300BB NMR300 at Cairo University. Elemental analyses were obtained on an Elementar Vario EL 1150 C analyser. Purity of the compounds was checked by TLC.

Compounds **1**, **2**[1,2,5] and **11**[8] were prepared as previously described.

7,9-Dimethylpyrido[3',2':4,5]selenolo[3,2-d]pyrimidine-2,4(1H,3H)-dithione (**3**): Compound **2** (0.25 g, 1 mmol) and carbon disulfide (5 mL) in pyridine (10 mL) were gently heated on a water bath for 6 h. The solid product that formed while hot was collected and recrystallised from DMF/water mixture, forming yellow crystals (0.21 g, 65%), m.p. >300 °C. IR: ν_{max} 3100 cm^{-1} (NH). NMR (TFA): δ_H 3.0 (s, 3H, CH_3), 3.2 (s, 3H, CH_3), 7.7 (s, 1H, CH-pyridine). MS: *m/z* (%) 327 (M^+, 100). Anal: Calcd for $C_{11}H_9N_3S_2Se$ (326.33): C, 40.48; H, 2.78; N, 12.87; S, 19.65. Found: C, 40.33; H, 2.59; N, 12.58; S, 19.56%.

3,4-Dihydro-7,9-dimethyl-3-phenyl-4-iminopyrido[3',2':4,5]selenolo[3,2-d]pyrimidine 2(1H)-thione (**4**): Compound **2** (0.250 g, 1 mmol) and phenyl isothiocyanate (0.125 mL, 1 mmol) were gently heated under reflux for 6 h in pyridine (10 mL). The solid product that formed on cooling was collected and recrystallised from acetic acid to give orange crystals of **4** (0.32 g, 85%), m.p >300 °C. IR: ν_{max} 3200 cm^{-1} (NH). NMR (TFA): δ_H 3.0 (s, 3H, CH_3), 3.2 (s, 3H, CH_3), 7.5–7.8 (m, 6H, ArH, CH-pyridine). MS: *m/z* (%) 386 (M^+, 36). Anal: Calcd for $C_{17}H_{14}N_4SSe$ (385.38): C, 52.97; H, 3.66; N, 14.54; S, 8.32. Found: C, 52.55; H, 3.34; N, 14.24; S, 7.98%.

2-Alkylthio-7,9-dimethyl-3-phenylpyrido[3',2':4,5]selenolo[3,2-d]pyrimidin-4(3H)-imine (**5a,b**): *general procedure*

Allyl iodide or methyl iodide (4 mmol) was added to a mixture of **4** (1.54 g, 4 mmol) and sodium acetate trihydrate (1.36 g, 10 mmol) in ethanol (30 mL), and the reaction mixture was heated under reflux for 2 h. After cooling the reaction mixture was poured into ice-water and the precipitate that formed was collected and recrystallised from ethanol to give compounds **5a,b** respectively.

Allylthio compound **5a**: Yellow crystals (1.53 g, 90%), m.p. 192–194 °C. IR: ν_{max} 3200 (NH). NMR (TFA): δ_H 3.0 (s, 3H, CH_3), 3.3 (s, 3H, CH_3), 4.2 (d, 2H, SCH_2), 5.2–5.4 (m, 3H, CH=CH_2), 7.3–7.7 (m, 6H, 5H ArH, 1H CH-pyridine). MS: *m/z* (%) 426 (M^+, 44). Anal: Calcd for $C_{20}H_{18}N_4SSe$ (425.45): C, 56.45; H, 4.27; N, 13.17; S, 7.53. Found: C, 56.50; H, 4.28; N, 12.98; S, 7.19%.

Methylthio compound **5b**: Pale yellow crystals (1.42 g, 89%), m.p 212–214 °C: IR: ν_{max} 3200 cm^{-1} (NH). NMR (DMSO-d_6): δ_H 2.3 (s, 3H, SCH_3), 2.5 (s, 3H, CH_3), 2.8 (s, 3H, CH_3), 7.4–7.6 (m, 5H, ArH), 7.2 (s, 1H, CH-pyridine), 9.5 (s, 1H, NH); δ_H 19.8 (SCH_3), 23.8, 33.0 (2 CH_3-pyridine), 110.55, 117.2, 122.3, 123.8, 125.7, 128.4 (C-Aryl), 134.3, 138.7, 147.5 (C-selenophene), 156.5, 158.8, 159.3, 163.8 (C=N pyrimidine), 165.85 (C=NH pyrimidine). MS: *m/z* (%) 400 (M^+, 89). Anal: Calcd for $C_{18}H_{16}N_4SSe$ (399.40): C, 54.12; H, 4.04; N, 14.03; S, 8.02. Found: C, 54.50; H, 4.28; N, 13.98; S, 7.88%.

3-Amino-4,6-dimethyl-2-(tetrazol-5-yl)selenolo[2,3-b]pyridine (**6**): The aminonitrile **2** (1.25 g, 5 mmol), sodium azide (0.4 g, 6 mmol) and ammonium chloride (0.32 g, 6 mmol) in DMF (15 mL) were heated on a water bath for 5 h. The reaction mixture was cooled and acidified with dilute acetic acid. The solid product that formed was collected and recrystallised from ethanol to give yellow crystals of the tetrazole **6** (1.20 g, 82%), m.p. 273–275 °C. IR: ν_{max} 3300, 3400, 3500 cm^{-1} (NH_2, NH). NMR (TFA): δ_H 3.1 (s, 3H, CH_3), 3.2 (s, 3H, CH_3), 7.6 (s, 1H, CH-pyridine). MS: *m/z* (%) 294 (M^+, 100). Anal: Calcd for $C_{10}H_{10}N_6Se$ (293.22) C, 40.95; H, 3.44; N, 28.66. Found: C, 40.65; H, 3.36; N, 28.45%.

7,9-Dimethylpyrido[3',2':4,5]selenolo[2,3-e]tetrazolo[1,5-c]pyrimidine-5(6H)-thione (**7**): The tetrazole **6** (0.586 g, 2 mmol) was heated on a water bath with carbon disulfide (5 mL) in pyridine (10 mL) for 6 h. The solid product that formed was collected and recrystallised from DMF-water mixture to give yellow crystals of the thione **7** (0.48 g, 72%), m.p >300 °C. ^{1}H NMR (TFA): δ 3.0 (s, 3H, CH_3), 3.3 (s, 3H, CH_3) 7.8 (s, 1H, CH-pyridine). MS: *m/z* (%) 336 (M^+, 37). Anal: Calcd for $C_{11}H_8N_6SSe$ (335.28): C, 39.40; H, 2.40; N, 25.07; S, 9.56. Found: C, 39.59; H, 2.51; N, 24.98; S, 9.40%.

5-Aryl-5,6-dihydro-7,9-dimethylpyrido[3',2':4,5]selenolo[2,3-e]tetrazolo[1,5-c]pyrimidine (**8a–c**): *general procedure*

To a mixture of compound **6** (0.586 g, 2 mmol) and benzaldehyde, *p*-hydroxybenzaldehyde or *p*-chlorobenzaldehyde (2 mmol) in ethanol (15 mL), a few drops of piperidine were added. The reaction mixture was heated under reflux for 2 h. The solid product that formed on cooling was collected and recrystallised from ethanol to give compounds **8a–c** respectively.

5-Phenyl compound **8a**: Formed yellow crystals (0.64 g, 85%), m.p: 172–174 °C. IR: ν_{max} 3300 cm^{-1} (NH). NMR (DMSO-d_6): δ_H 2.5 (s, 3H, CH_3), 2.8 (s, 3H, CH_3), 7.3–7.5 (m, 7H, ArH, CH-pyridine, pyrimidine). MS: *m/z* (%) 382 (M^+, 20). Anal: Calcd for $C_{17}H_{14}N_6Se$ (381.33): C, 53.53; H, 3.70; N, 22.04. Found: C, 53.44; H, 3.56; N, 21.87%.

4-Hydroxyphenyl compound **8b**: Yellow crystals (0.68 g, 86%), m.p: 204–206 °C. IR: ν_{max} 3300 cm^{-1} (NH). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.3 (s, 3H, CH_3), 6.2 (s, 1H, CH pyrimidine), 7.6–7.8 (m, 5H, ArH, CH-pyridine). MS: *m/z* (%) 398 (M^+, 16). Anal: Calcd for $C_{17}H_{14}N_6OSe$ (397.20): C, 51.40; H, 3.55; N, 21.16. Found: C, 50.99; H, 3.24; N. 20.97%.

4-Chlorophenyl compound **8c**: Pale yellow crystals (0.74 g, 90%) from ethanol, m.p: 152–154 °C. IR: ν_{max} 3300 cm^{-1} (NH). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.3 (s, 3H, CH_3) 7.5–7.6 (m, 6H, 4H ArH, 1H CH-pyridine, 1H pyrimidine). MS: *m/z* (%) 417 (M^+, 17). Anal: Calcd for $C_{17}H_{13}ClN_6Se$ (415.77): C, 49.10; H, 3.15; Cl, 8.52; N, 20.21. Found: C, 48.92; H, 3.11; Cl, 8.32; N, 20.01%.

7,9-Dimethyl-4-azidopyrido[3',2':4,5]selenolo[3,2-d]pyrimidine (**10**): The tetrazole **6** (0.586 g, 2 mmol) and triethyl orthoformate (10 mL) were heated under reflux for 2 h. The precipitate which formed while hot was collected and recrystallised from ethanol giving yellow crystals of **10** (0.52 g, 87%), m.p: 182–184 °C. IR: ν_{max} 2150 cm^{-1} (N_3). NMR (DMSO-d_6): δ_H 2.6 (s, 3H, CH_3), 2.8 (s, 3H, CH_3), 7.2 (s, 1H, CH-pyridine), 10.2 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 304 (M^+, 44). Anal: Calcd for $C_{11}H_8N_6Se$ (303.21): C, 43.57; H, 2.66; N, 27.72. Found: C, 43.26; H, 2.21; N, 27.43%.

2-(Cyanomethyl/phenyl)-7,9-dimethylpyrido[3',2':4,5]selenolo[2,3-e][1,2,4]triazolo[1,5-c]pyrimidine (**12a,b**): *general procedure*

A mixture of the iminoether **11**[8] (1.53 g, 5 mmol) and cyanoacetic acid or benzoic acid hydrazides (5 mmol) in acetic acid (20 mL) was heated under reflux for 3 h. The precipitate that formed on cooling in the case of **12a** and that formed while hot in case of **12b** were collected and recrystallised from the indicated solvent to give compounds **12a,b**.

Cyanomethyl compound **12a**: Yellow crystals (1.45 g, 85%), m.p >300 °C from acetic acid. IR: ν_{max} 2200 cm^{-1} (CN). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.6 (s, 3H, CH_3), 4.7 (s, 2H, CH_2), 8.0 (s, 1H, CH-pyridine), 9.8 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 342 (M^+, 100). Anal: Calcd for $C_{14}H_{10}N_6Se$ (341.26): C, 49.27; H, 2.95; N, 24.63. Found: C, 49.07; H, 2.53; N, 24.95%.

Phenyl compound **12b**: White crystals (1.55 g, 82%)separated from DMF/water, m.p >300 °C. IR: ν_{max} 3050 cm^{-1} (CH-arom). NMR (TFA): δ_H 3.1 (s, 3H, CH_3), 3.5 (s, 3H, CH_3) 7.8–8.2 (m, 6H, 5H ArH, 1H CH-pyridine), 9.9 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 379 (M^+, 100). Anal: Calcd for $C_{18}H_{13}N_5Se$ (378.32): C, 57.14; H, 3.47; N, 18.51. Found: C, 56.88; H, 3.25; N, 18.22%.

3-Amino-2-[(5-(cyanomethyl/phenyl)-1,2,4-triazol-3-yl]-4,6-dimethylselenolo[2,3-b]pyridine (**13a,b**): *general procedure*

The tetracyclic compound **12a** or **12b** (5 mmol) in 20% aqueous hydrochloric acid (20 mL) was heated under reflux for 3 h. The solid product that formed while hot was collected and recrystallised from dioxan to give compounds **13a,b**.

Cyanomethyl compound **13a**: Golden crystals (1.42 g, 86%), m.p. 242–244 °C. IR: ν_{max} 2200 (CN), 3400–3500 cm^{-1} (NH, NH_2). NMR (TFA): δ_H 2.9 (s, 3H, CH_3), 3.2 (s, 3H, CH_3), 4.7 (s, 2H, CH_2CN), 7.6 (s, 1H, CH-pyridine). MS: *m/z* (%) 333 (M^+ + 1, 100). Anal: Calcd for $C_{13}H_{12}N_6Se$ (331.27): C, 47.13; H, 3.65; N, 25.37. Found: C, 46.91; H, 3.25; N. 25.18%.

Phenyl compound **13b** separated as yellow crystals (1.47 g, 80%). m.p >300 °C. IR: ν_{max} 3400–3500 cm^{-1} (NH, NH_2). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.5 (s, 3H, CH_3) 7.8–8.4 (m, 6H, 5H ArH, 1H CH-pyridine). MS: *m/z* (%) 369 (M+, 100). Anal: Calcd for $C_{17}H_{15}N_5Se$ (368.33): C, 55.43; H, 4.11; N, 19.01. Found: C, 55.15; H, 3.99; N 18.86%.

3-Benzylamino-7,9-dimethylpyrido[3',2':4,5]selenolo[3,2-d]pyrimidin-4(3H)-imine (**14**)

A mixture of compound **11** (1.53 g, 5 mmol) and benzylamine (0.54 mL, 5 mmol) in dioxan (10 mL) was stirred at room temperature for 4 h. the solid product that formed was collected and recrystallised from ethanol to give compounds **14** as white crystals (1.68 g, 92%), m.p: 173–175 °C. IR: ν_{max} 3200 (NH), 3050 (CH-arom). ^{1}H NMR (DMSO-d_6): δ 2.6 (s, 3H, CH_3), 2.9 (s, 3H, CH_3), 4.8 (s, 2H, CH_2Ph) 7.1–7.3 (m, 6H, 5H ArH, 1H CH-pyridine), 8.2 (s, 1H, NH), 8.5 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 368 (M^+, 100). Anal: Calcd for $C_{18}H_{16}N_4Se$ (367.34) Calcd. C, 58.85; H, 4.39; N, 15.25. Found: C, 58.43; H, 4.57; N, 14.96%.

N'-Carboxamido-N-(2-cyano-4,6-dimethylselenolo[2,3-b]pyridin-3-yl)methanimidate (**15**).
A mixture of compound **11** (1.53 g, 5 mmol) and urea (0.3 g, 5 mmol) in acetic acid (10 mL) was refluxed for 2 h. The solid product that formed on cooling was collected and recrystallised from acetic acid giving compound **15** as yellow crystals, (1.39 g, 87%), m.p: 242–244 °C. IR: ν_{max} 3200–3400 (NH, NH_2), 2200 cm^{-1} (CN). NMR (TFA): δ_H 3.0 (s, 3H, CH_3), 3.2 (s, 3H, CH_3), 7.8 (s, 1H, CH-pyridine), 8.7 (s, 1H, CH=N). MS: *m/z* (%) 321 (M^+, 0.5), 277 (M^+-$CONH_2$, 15). Anal: Calcd for $C_{12}H_{11}N_5OSe$ (320.24): C, 45.00; H, 3.46, N, 21.87. Found: C, 44.78; H, 3.92; N, 21.55%.

3-(2-Aminoethyl/3-aminopropyl)-3,4-dihydro-4-imino-7,9-dimethylpyrido[3',2':4,5]selenolo[3,2-d]pyrimidin-4(3H)-imine (**17a,b**): *general procedure*
A mixture of compound **11** (1.53 g, 5 mmol) and the appropriate diamine (5 mmol) in dioxan (10 mL) was stirred at room temperature for 4 h. The solid product (**17a,b**) that formed was collected and dried.

Aminoethyl compound **17a**: White powder (1.31 g, 82%), m.p undetermined (on heating cyclised to compound **18a**). IR: ν_{max} 3200–3400 cm^{-1} (NH, NH_2). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.5 (s, 3H, CH_3), 4.1 (m, 2H, CH_2), 5.2 (m, 2H, CH_2), 8.0 (s, 1H, CH-pyridine), 8.9 (s, 1H, CH-pyrimidine). MS (FAB): *m/z* (%) 321 (M^+, 10), 304 (M^+-NH_3, 60).

Aminopropyl compound **17b**: White powder (1.35 g, 81%), m.p undetermined (on heating cyclised to compound **18b**). IR: ν_{max} 3200–3400 cm^{-1} (NH, NH_2). NMR (TFA): δ_H δ 2.4 (m, 2H, CH_2), 3.3 (s, 3H, CH_3), 3.7 (s, 3H, CH_3), 4.1(m, 2H, CH_2), 4.9 (m, 2H, CH_2), 7.6 (s, 1H, CH-pyridine), 8.9 (s, 1H, CH-pyrimidine). MS (FAB): *m/z* (%) 335 (M^+, 18), 318 (M^+-NH_3, 30).

Cyclisation reactions; preparation of **18a**,**b**: *general procedure*
Compound **17a** (1.6 g, 5 mmol) or **17b** (1.67 g, 5 mmol) was boiled in ethanol for 5 min and the solution left to cool. The crystals that formed on cooling was collected and recrystallised from the proper solvent giving compounds **18a, b**.

7,9-Dimethylpyrido[3',2':4,5]selenolo[2,3-e]imidazo[1,2-c]pyrimidine (**18a**): Yellow crystals (1.36 g, 90%) from ethanol, m.p. 291–293 °C. IR: ν_{max} 3050 cm^{-1} (CH-arom). NMR (TFA): δ_H 3.2 (s, 3H, CH_3), 3.5 (s, 3H, CH_3), 4.5 (m, 2H, CH_2-imidazole), 5.2 (m, 2H, CH_2-imidazole), 7.2 (s, 1H, CH-pyridine), 8.1 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 304 (M^+, 86). Anal: Calcd for $C_{13}H_{12}N_4Se$ (303.25): C, 51.48; H, 3.99; N, 18.47. Found: C, 51.07; H, 3.98; N, 18.47%.

8,10-Dimethylpyrido[3",2":4',5']selenolo[3',2':4,5]pyrimido[1,6-a]pyrimidine (**18b**): Crystallised from ethanol as yellow crystals (1.42 g, 90%), m.p. 282–284 °C. IR: ν_{max} 3050 cm^{-1} (CH-arom). NMR (TFA): δ_H 2.6 (m, 2H, CH_2-pyrimidine), 3.1 (s, 3H, CH_3), 3.4 (s, 3H, CH_3), 4.0 (m, 2H, CH_2-pyrimidine), 4.6 (m, 2H, CH_2-pyrimidine), 7.8 (s, 1H, CH-pyridine), 8.7 (s, 1H, CH-pyrimidine). MS: *m/z* (%) 318 (M^+, 100). Anal: Calcd for $C_{14}H_{14}N_4Se$ (317.28): C, 52.99; H, 4.45; N, 17.66. Found: C, 52.50; H, 4.33; N, 17.72%.

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