New approaches to pyrimido[4,3-*b*][1,3,5]selenadiazines

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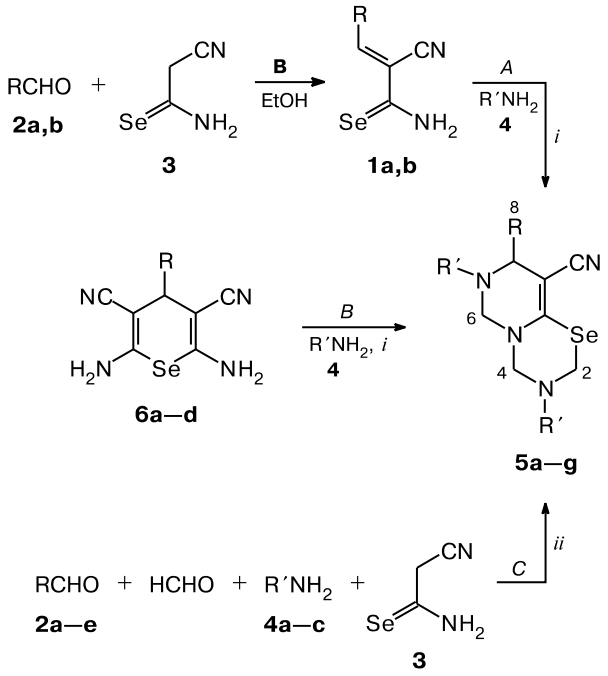
The Mannich reactions of (arylmethylidene)(cyano)selenoacetamides with primary amines and formaldehyde gave 3,7-disubstituted 8-R-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*]-[1,3,5]selenadiazine-9-carbonitriles. Alternatively, these compounds can be obtained by reactions of 4-R-2,6-diamino-4*H*-selenopyran-3,5-dicarbonitriles with primary amines and formaldehyde or by multicomponent condensation of an appropriate aldehyde, cyanoselenoacetamide, a primary amine, and formaldehyde.

Key words: pyrimido[4,3-*b*][1,3,5]selenadiazines, the Mannich reaction, recyclization, 4-R-2,6-diamino-4*H*-selenopyran-3,5-dicarbonitriles, aminomethylation.

Interest in the chemistry of selenium-containing heterocycles is largely due to a broad spectrum of their biological activity.[1,2] 1,3,5-Selenadiazines are relatively poorly studied. Only a few ways are currently available for constructing the 1,3,5-selenadiazine ring. They include (1) multicomponent reactions between benzylamine, sodium hydroselenide, formaldehyde, and aryl isoselenocyanates in acidic media,[3] (2) reactions of 1,3-dichloro-1,3-bis-(dimethylamino)-2-azapropenylium chlorides with selenamides or selenoureas,[4,5] (3) reactions of *N*,*N*-dialkyl-3-aryl-3-chloro-2-azaprop-1-enylideneammonium perchlorate with *N*-acylselenoureas,[6] and (4) recyclization of 1,3,5-oxaselenazines[7,8] and a 1,2,4-selenadiazolium salt.[9] Nevertheless, the literature provides only one example[10] of the synthesis of fused 1,3,5-selenadiazines, while 1,3,5-thiadiazine derivatives are studied much better.[11—13] (Arylmethylidene)(cyano)selenoacetamides **1**, which are easily prepared from aromatic aldehydes **2** and cyanoselenoacetamide (**3**),[14,15] can serve as convenient starting materials for the synthesis of fused 1,3,5-selenadiazines (Scheme 1). Earlier, we have found that brief heating of (cyano)(2-thienylmethylidene)selenoacetamide (**1a**) with an excess of aqueous formaldehyde and two equivalents of primary amine **4** under argon gives the corresponding pyrimido[4,3-*b*][1,3,5]selenadiazines **5** in 38—50% yields (see Scheme 1, pathway *A*).[10] The goal of the present work was to obtain novel pyrimido[4,3-*b*][1,3,5]selenadiazines according to this method, as well as to study (1) recyclization of selenopyrans **6** in their Mannich aminomethylation and (2) multicomponent cyclocondensation of aldehydes, cyanoselenoacetamide **3**, primary amines, and HCHO as possible routes to compounds of the type **5**.

Apart from (cyano)(2-thienylmethylidene)selenoacetamide (**1a**), we successfully used its analog, namely, (cyano)(4-dimethylaminophenylmethylidene)selenoacetamide (**1b**), in the Mannich reaction with amines **4** and HCHO to give pyrimido[4,3-*b*][1,3,5]selenadiazines **5a**,**d** in 46 and 47% yields, respectively (see Scheme 1, pathway *A*).

Scheme 1



i. HCHO, EtOH, *t*°; *ii*. EtOH, **B**, *t*° (**B** is *N*-methylmorpholine).

1: R = C_4H_3S (**a**), 4-$Me_2NC_6H_4$ (**b**)
2: R = C_4H_3S (**a**), 4-$Me_2NC_6H_4$ (**b**), Ph (**c**), CH_2Me (**d**), $CHMe_2$ (**e**)
4: R′ = 4-MeC_6H_4 (**a**), CH_2Ph (**b**), 4-FC_6H_4 (**c**)
6: R = $CHMe_2$ (**a**), Ph (**b**), CH_2Me (**c**), CH_2CH_2Me (**d**)

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Table 1. The yields of compounds **5** obtained by methods *A*—*C*

Compound	R	R^1	Yield (%) (method)
5a	2-Thienyl	4-FC_6H_4	46 (*A*)
5b	2-Thienyl	4-MeC_6H_4	50 (*A*)
			32 (*C*)
5c	2-Thienyl	CH_2Ph	38 (*A*)
			29 (*C*)
5d	4-$Me_2NC_6H_4$	4-MeC_6H_4	47 (*A*)
5e	$CHMe_2$	4-MeC_6H_4	16 (*B*)
			22 (*C*)
5f	Ph	4-MeC_6H_4	20 (*B*)
			28 (*C*)
5g	Et	4-MeC_6H_4	0 (*B*)
			14 (*C*)

Alternatively, pyrimido[4,3-*b*][1,3,5]selenadiazines **5** can be obtained by recyclization of easily accessible[16—18] 2,6-diamino-4*H*-selenopyran-3,5-dicarbonitriles **6**, which are "masked" synthetic analogs of unsaturated selenoacetamides **1**. For instance, reactions of selenopyrans **6a**,**b** with an excess of formaldehyde and a twofold amount of *p*-toluidine in boiling EtOH gave compounds **5e**,**f** in 15 and 20% yields, respectively (see Scheme 1, pathway *B*). Aminomethylation of selenopyrans **6c**,**d** was accompanied by strong resinification and the formation of inseparable complex mixtures of unidentified products. At the same time, method *B* allows the synthesis of compound **5e** (R = $CHMe_2$, R′ = 4-MeC_6H_4), which cannot be obtained by method *A* because (alkylidene)(cyano)selenoacetamides of the type **1** (R is alkyl) are inaccessible. A possible mechanism of the recyclization of selenopyrans **6** can involve elimination of malononitrile and the formation of unsaturated selenamides **1** followed by their aminomethylation leading to compounds **5** (Scheme 2). The low yields of the final products can be attributed to the aminomethylation of malononitrile as a side process.

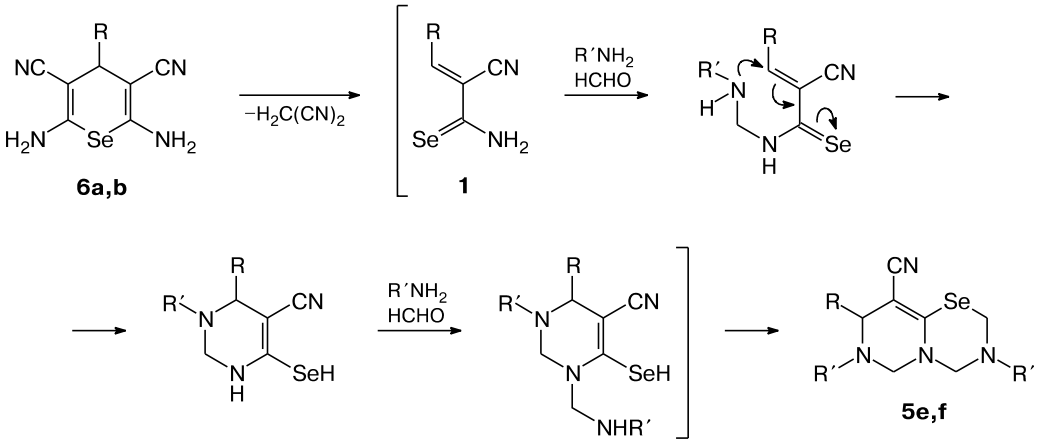
Pyrimido[4,3-*b*][1,3,5]selenadiazines **5** can be obtained by "one pot" condensation of aldehydes, cyanoselenoacetamide **3**, primary amines, and HCHO (pathway *C*) without isolation or purification of unsaturated selenamides **1**, which are generated *in situ* and undergo further transformations. Another advantage of this approach is that the range of documented[14] unsaturated selenamides of the type **1** is very limited. We found that benzaldehyde and aliphatic aldehydes react with cyanoselenoacetamide **3**, primary amines **4**, and an excess of formaldehyde in boiling EtOH to give selenadiazines **5** in 14—32% yields (see Scheme 1). The structural data and yields of compounds **5** are given in Table 1.

Method *C* (see Scheme 1) also allows the synthesis of pyrimido[4,3-*b*][1,3,5]selenadiazines **5** containing either alkyl or aryl substituent at the C(8) atom in higher yields compared to method *B*. It should be noted that the yields of compounds **5** from aromatic aldehydes are low and comparable with those achieved by method *A*. The compounds obtained are fine crystalline powders varying in color from light beige to claret; they are well soluble in acetone, DMF, and DMSO and moderately soluble in EtOH.

The structures of the compounds obtained were confirmed by spectroscopic and elemental analysis data. Among the most characteristic signals in the ^1H NMR spectra of pyrimido[4,3-*b*][1,3,5]selenadiazines **5a**—**d**,**f**, note a singlet at δ 4.35—5.13 for the C(8)H proton. For selenadiazines **5e**,**g** containing aliphatic substituents at the C(8) atom, the signal for the C(8)H proton appears as a doublet shifted upfield (δ 3.23—3.62, ^3J = 6.6—8.6 Hz). The protons of the fragment N—CH_2—Se are manifested as a doublet of doublets at δ 3.82—4.48 (^2J = 12.0—12.8 Hz), while the protons of the fragment N—CH_2—N—CH_2—N, as two doublets of doublets at δ 4.13—5.28 (^2J = = 10.2—13.6 Hz).

Thus, we proposed three approaches to the synthesis of pyrimido[4,3-*b*][1,3,5]selenadiazines: aminomethylation

Scheme 2



of (arylmethylidene)(cyano)selenoacetamides (method *A*), cross recyclization of 4-R-2,6-diamino-4*H*-selenopyran-3,5-dicarbonitriles in the presence of primary amines and excess formaldehyde (*B*), and "one pot" condensation of appropriate aldehydes, cyanoselenoacetamide, primary amines, and formaldehyde (*C*).

When comparing these three approaches, one can conclude that the highest yields of the target products are achieved by method *A*. However, because (alkylidene)-(cyano)selenoacetamides (**1**, R = alkyl) are inaccessible, this approach is suitable only for the synthesis of 8-arylpyrimido[4,3-*b*][1,3,5]selenadiazines **5**. Method *B* provides a route to 8-alkylpyrimidoselenadiazines **5**. However, the yields of the target products are much lower because this method involves an additional step (preparation of selenopyran **6**) and is not economical. On the whole, the above drawbacks makes the recyclization of selenopyrans **6** not very suitable for preparative synthesis of compounds **5**. Method *C* ("one pot" multicomponent condensation) combines the advantages of methods *A* and *B* and seems to be most versatile and convenient, though not providing the highest yields of the target products.

Experimental

^{1}H NMR spectra were recorded on a Bruker Avance II 400 instrument (399.97 MHz) in DMSO-d_6 with $SiMe_4$ as the internal standard. IR spectra (Nujol) were recorded on an IKS-29 spectrophotometer. Elemental analysis was carried out on a Perkin—Elmer C,H,N-analyzer. The purity of the compounds obtained was checked by TLC on Silufol UV-254 plates with acetone—heptane (1 : 1) as an eluent; spots were visualized in the iodine vapor or under UV light. Melting points were determined on a Kofler hot stage and are given uncorrected. All manipulations were carried out under argon.

(Cyano)(2-thienylmethylidene)selenoacetamide (1a) was prepared according to a general procedure.[14]

(Cyano)(4-dimethylaminophenylmethylidene)selenoacetamide (1b) was prepared in a similar way from 4-dimethylaminobenzaldehyde **2b** (2.03 g, 1.36 mmol) and cyanoselenoacetamide **3** (2.0 g, 1.36 mmol). The yield of compound **1b** was 59%, dark violet fine crystalline powder, decomp. > 150 °C. Found (%): C, 51.91; H, 4.78; N, 14.99. $C_{12}H_{13}N_3Se$ (M = 279.23). Calculated (%): C, 51.81; H, 4.71; N, 15.10. IR (Nujol), ν/cm^{-1}: 3275 (NH_2); 2210 (C≡N); 1610 (C=C). ^{1}H NMR (DMSO-d_6), δ: 3.16 (s, 6 H, 2 Me); 6.75, 7.85 (both d, 2 H each, Ar, ^3J = 9.1 Hz); 8.12 (s, 1 H, CH=); 8.26, 9.63 (both br.s, 1 H each, C(Se)NH_2).

Pyrimido[4,3-*b*][1,3,5]selenadiazines (5). Method *A*. For the detailed synthesis of compounds **5b,c** by this method, see Ref. 10. Compounds **5a,d** were obtained according to a modified procedure as follows. A mixture of 2-cyano-3-(4-dimethylaminophenyl)prop-2-eneselenamide (**1b**) (0.7 g, 2.5 mmol), an appropriate primary amine **4a**—**c** (5.2 mmol), and 37% formalin (1.0 mL, 12.5 mmol) in DMF (5 mL) was refluxed to complete homogenization (~1—2 min), immediately filtered through a paper filter, and left at ~20 °C for 24 h. The precipitate of compound **5** that formed was filtered off and washed with EtOH and hexane.

***B*.** A mixture of an appropriate selenopyran **6a**,**b** (1 mmol) prepared according to a known procedure,[16,18] *p*-toluidine **4a** (0.23 g, 2.1 mmol), and an excess of 37% formalin (2.5 mL, 33.6 mmol) in EtOH (30 mL) was refluxed for 3 min, filtered through a paper filter, and left at ~20 °C for 72 h. The crystals that formed were filtered off and washed with EtOH and hexane.

***C*.** A drop of *N*-methylmorpholine was added to a mixture of cyanoselenoacetamide **2** (0.2 g, 1.4 mmol) and an appropriate aldehyde **1a**,**c**,**d**,**e** (1.4 mmol) in EtOH (15 mL). The mixture was stirred for 0.5 h to homogenization. Then an appropriate primary amine (**4a**,**b**) (3 mmol) and an excess of 37% formalin (2.0 mL, 10 mmol) were added. The reaction mixture was refluxed for 3 min, filtered through a paper filter, and left at ~20 °C for 24 h. Products **5b**,**c**,**e**,**f**,**g** were filtered off and washed with EtOH and hexane. The yields of compounds **5** obtained by methods *A*—*C* are given in Table 1.

3,7-Bis(4-fluorophenyl)-8-(2-thienyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5a), m.p. 158—160 °C (Me_2CO), beige fine crystalline powder. Found (%): C, 53.72; H, 4.06; N, 12.53. $C_{23}H_{18}F_2N_4SSe$ (M = 499.45). Calculated (%): C, 54.06; H, 4.02; N, 12.61. IR (Nujol), ν/cm^{-1}: 2158 (C≡N); 1590 (C=C). ^{1}H NMR (DMSO-d_6), δ: 4.45 (dd, AB system, 2 H, $SeCH_2N$, ^2J = 12.8 Hz); 4.83 (dd, AB system, 2 H, NCH_2N, ^2J = 11.5 Hz); 5.10 (s, 1 H, C(8)H); 5.27 (dd, AB system, 2 H, NCH_2N, ^2J = 11.0 Hz); 6.91—6.97 (m, 8 H, 2 Ar); 7.05—7.09 (m, 2 H, $C(3)H_{thienyl}$, $C(4)H_{thienyl}$); 7.32 (m, 1 H, $C(5)H_{thienyl}$).

3,7-Bis(4-methylphenyl)-8-(2-thienyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5b), m.p. 173—175 °C (Me_2CO), light beige fine crystalline powder. The spectroscopic characteristics of compound **5b** are identical with the literature data.[10]

3,7-Dibenzyl-8-(2-thienyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5c), m.p. 127—128 °C (Me_2CO) (*cf.* Ref. 10: 127—130 °C). The spectroscopic characteristics of compound **5c** are identical with the literature data.[10]

8-(4-Dimethylaminophenyl)-3,7-bis(4-methylphenyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5d), m.p. 187—189 °C (DMF), dark claret fine crystalline powder. Found (%): C, 66.24; H, 5.93; N, 13.18. $C_{29}H_{31}N_5Se$ (M = 528.56). Calculated (%): C, 65.90; H, 5.91; N, 13.25. IR (Nujol), ν/cm^{-1}: 2170 (C≡N); 1615 (C=C). ^{1}H NMR (DMSO-d_6), δ: 2.28, 2.31 (both s, 3 H each, 2 $\underline{H}_3CC_6H_4$); 2.92, 2.98 (both s, 3 H each, Me_2N); 4.27 (dd, AB system, 2 H, $SeCH_2N$, ^2J = 12.5 Hz); 4.80 (dd, AB system, 2 H, NCH_2N, ^2J = 13.6 Hz); 4.85 (s, 1 H, C(8)H); 5.27 (dd, AB system, 2 H, NCH_2N, ^2J = 10.9 Hz); 6.55—7.04 (m, 12 H, 3 Ar).

8-Isopropyl-3,7-bis(4-methylphenyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5e), m.p. 164—166 °C (EtOH), beige fine crystalline powder. Found (%): C, 64.13; H, 6.29; N, 12.28. $C_{24}H_{28}N_4Se$ (M = 451.48). Calculated (%): C, 63.85; H, 6.25; N, 12.41. IR (Nujol), ν/cm^{-1}: 2177 (C≡N); 1605 (C=C). ^{1}H NMR (DMSO-d_6), δ: 0.98, 1.07 (both d, 3 H each, $(C\underline{H}_3)_2CH$, ^3J = 6.6 Hz); 1.82 (m, 1 H, $Me_2C\underline{H}$); 2.25, 2.28 (both s, 3 H each, 2 $\underline{H}_3CC_6H_4$); 3.25 (d, 1 H, C(8)H, ^3J = 8.6 Hz); 4.47 (dd, AB system, 2 H, $SeCH_2N$, ^2J = 12.0 Hz); 4.77 (pseudo s, 2 H, NCH_2N); 5.22 (dd, AB system, 2 H, NCH_2N, ^2J = 11.0 Hz); 6.66—6.98 (m, 8 H, 2 $MeC_6\underline{H}_4$).

3,7-Bis(4-methylphenyl)-8-phenyl-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5f), m.p.

176—178 °C (DMF), yellow-green fine crystalline powder. Found (%): C, 67.54; H, 5.46; N, 11.41. $C_{27}H_{26}N_4Se$ (M = 485.50). Calculated (%): C, 66.80; H, 5.40; N, 11.54. IR (Nujol), ν/cm^{-1}: 2185 (C≡N); 1610 (C=C). ^{1}H NMR (DMSO-d_6), δ: 2.30, 2.31 (both s, 3 H each, 2 $\underline{H}_3CC_6H_4$); 4.31 (dd, AB system, 2 H, $SeCH_2N$, ^2J = 12.7 Hz); 4.83 (dd, AB system, 2 H, NCH_2N, ^2J = 13.6 Hz); 4.98 (s, 1 H, C(8)H); 5.28 (dd, AB system, 2 H, NCH_2N, ^2J = 10.8 Hz); 6.91—7.31 (m, 13 H, 3 Ar).

8-Ethyl-3,7-bis(4-methylphenyl)-3,4,7,8-tetrahydro-2*H*,6*H*-pyrimido[4,3-*b*][1,3,5]selenadiazine-9-carbonitrile (5g), m.p. 168—170 °C (EtOH), beige fine crystalline powder. Found (%): C, 63.24; H, 6.03; N, 12.91. $C_{23}H_{26}N_4Se$ (M = 437.45). Calculated (%): C, 63.15; H, 5.99; N, 12.81. IR (Nujol), ν/cm^{-1}: 2155 (C≡N); 1610 (C=C). ^{1}H NMR (DMSO-d_6), δ: 0.99 (t, 3 H, $CH_2C\underline{H}_3$, ^3J = 7.3 Hz); 1.52—1.76 (m, 2 H, $C\underline{H}_2Me$); 2.25, 2.27 (both s, 3 H each, 2 $\underline{H}_3CC_6H_4$); 3.60 (br.d, 1 H, C(8)H, ^3J = 6.6 Hz); 4.48 (dd, AB system, 2 H, $SeCH_2N$, ^2J = 12.7 Hz); 4.80 (pseudo s, 2 H, NCH_2N); 5.23 (dd, AB system, 2 H, NCH_2N, ^2J = 11.0 Hz); 6.72—6.99 (m, 8 H, 2 $MeC_6\underline{H}_4$).

4-R-2,6-Diamino-4*H*-selenopyran-3,5-dicarbonitriles (6a,b) were prepared as described earlier.[16,18]

4-R-2,6-Diamino-4*H*-selenopyran-3,5-dicarbonitriles (6c,d) were prepared according to a modified procedure[18] as follows. A mixture of an appropriate aldehyde (10 mmol), malononitrile (0.66 g, 10 mmol), cyanoselenoacetamide (1.47 g, 10 mmol), and *N*-methylmorpholine (0.1 mL) in EtOH (20 mL) was stirred at 20 °C for 3 h. The precipitate that formed was filtered off and washed with EtOH and hexane.

2,6-Diamino-4-ethyl-4*H*-selenopyran-3,5-dicarbonitrile (6c). Yield 17%, m.p. 253—255 °C (EtOH—Me_2CO, 3 : 1), orange fine crystalline powder. Found (%): C, 42.13; H, 4.01; N, 21.95. $C_9H_{10}N_4Se$ (M = 253.17). Calculated (%): C, 42.70; H, 3.98; N, 22.13. IR (Nujol), ν/cm^{-1}: 3285 (2 NH_2); 2215, 2235 (2 C≡N); 1640 (2 C=C). ^{1}H NMR (DMSO-d_6), δ: 1.30 (t, 3 H, Me, ^3J = 7.6 Hz); 2.79 (m, 2 H, CH_2); 2.91 (br.d, 1 H, C(4)H, ^3J = = 6.6 Hz); 7.74 (br.s, 4 H, 2 NH_2).

2,6-Diamino-4-propyl-4*H*-selenopyran-3,5-dicarbonitrile (6d). Yield 29%, m.p. 220—222 °C (EtOH—Me_2CO, 3 : 1), light yellow fine crystalline powder. Found (%): C, 45.37; H, 4.57; N, 20.62. $C_{10}H_{12}N_4Se$ (M = 267.19). Calculated (%): C, 44.95; H, 4.53; N, 20.97. IR (Nujol), ν/cm^{-1}: 3345 (2 NH_2); 2215 (2 C≡N); 1650 (2 C=C). ^{1}H NMR (DMSO-d_6), δ: 1.30 (t, 3 H, Me, ^3J = 7.3 Hz); 1.72 (m, 2 H, $MeC\underline{H}_2CH_2$); 2.76 (m, 2 H, $MeCH_2C\underline{H}_2$); 2.96 (br.d, 1 H, C(4)H, ^3J = 6.7 Hz); 7.76 (br.s, 4 H, 2 NH_2).

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