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Physical Sciences

Reagents For New Heteroannelation Reactions: Aza Reagents

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Aza reagents were prepared from different hydrazides. Reactions of *ortho*-aminoesters and *ortho*-aminonitriles with aza reagents have been studied in different solvents. Aza reagents with *ortho*-aminoesters and *ortho*-aminonitriles in presence of *p*-toluene sulphonic acid in dry CH_3CN or acetic acid furnished the tricyclic derivatives N-[2-(methylthio)-4-oxo-5, 6, 7, 8-tetrahydro [1]benzothieno [2, 3-d]pyrimidine-3 (4H)-yl]carbamic acid ethyl ester (**8**), 5, 6, 7, 8-tetrahydro-2-methylthio-4-oxo [1] benzothieno [2, 3-d]pyrimidin-3 (4H) acetamide (**9**) and 5, 6, 7, 8-tetrahydro-2-methylthio-4-oxo [1] benzothieno [2, 3-d]pyrimidine-3 (4H)-benzamide (**10**), whereas, in pyridine medium these reactions afforded uncyclized urethane products (**11, 12** and **13**).

Key words: Aza reagents, Heteroannelation, Hydrazides.

Introduction

Fused pyrimidines are found in a broad variety of natural products [e.g. purines, pyrrolopyrimidines, pteridines], pharmaceuticals, agrochemicals and veterinary products (Taylor and Petel 1991). With the development of clinically useful anticancer, antihypertensive agents, antiviral, antibacterial, antiallergic, antimalarial, antianalgesic and anti-inflammatory drugs, there has recently been remarkable interest in the preparation of annelated pyrimidines (Ram *et al* 1981; Petrie *et al* 1985; Albert 1986; Chern *et al* 1993). For the last several years we are interested in the synthesis of heterocycles containing the thieno-pyrimidine systems with the aim of finding compounds with antihypertensive, antifungal and antibacterial activities (Sauter *et al* 1996; Chowdhury *et al* 1997).

The aromatic and heteroaromatic *Ortho*-aminoesters or *ortho*-aminonitriles undergo cyclization readily which allow convenient preparation of variety of condensed pyrimidines (Taylor 1987). The reaction of *ortho*-aminoesters or *Ortho*-aminonitriles with the N-[bis(methylthio) methylene] amino reagent, called BMMA, are versatile reagents for the preparation of various fused pyrimidine systems (Sauter *et al* Part-I & II 1996 and III 1997).

In this paper, the synthesis of benzothieno [2, 3-d] pyrimidine system from aza-regents and *ortho*-aminoesters and *ortho*-aminonitriles is reported.

Experimental

Melting points were determined on an electrothermal melting point apparatus and were uncorrected. ^{1}H- and ^{13}C-NMR spectra were recorded on a Bruker AC 200 (200 MHz) spectrometer (internal standard TMS, solvents $CDCl_3$ or DMSO-d_6, δ-values in ppm) at the Institute of Organic Chemistry, Technical University of Vienna. Elemental analyses were performed at the Microanalytical Laboratory, Institute of Physical Chemistry, University of Vienna, Austria. Thin layer chromatography (TLC) was performed on silica gel (Woelm, Germany) and spots were detected by heating the plates at 150-200°C. Column chromatography was carried out at room temperature with kieselgel (Merck 70, 230-400 mesh). All evaporations were conducted under reduced pressure with a bath temperature below 50°C.

Ethyl 2-amino-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (1). Title compound (**1**) was prepared from cyclohexanone by reacting with ethyl cyanoacetate and sulphur using the literature procedure (Gewald *et al* 1966) as white crystals. Yield: 76%; mp 113°C [lit.[10] 115°C].

Ethyl 2-amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (2). This compound (**2**) was prepared from 4-methylcyclohexanone as white needles. Yield: 74%; mp 107-109°C [lit.[10] 108-110°C].

2-Amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carbonitrile (3). This compound (**3**) was obtained from 4-methylcyclohexanone by reacting with malononitrile and sulphur as white crystals. Yield: 85%; mp 143-144°C [lit.[10] 143-144°C].

2-Amino-5,6,7,8-tetrahydro-4H-cyclohepta [b] thiophene-3-carbonitrile (4). Title compound (**4**) was prepared from cycloheptanone. Yield: 38%; mp 128-129°C [lit.[10] 128-129°C].

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Preparation of aza-reagent (general procedure). To a solution of hydrazide (95 mmol) and carbon disulphide (99 mmol) in chloroform (100 ml), triethylamine (105 mmol) was added dropwise maintaining the temperature below 40°C. Then methyl iodide (190 mmol) was added to this reaction mixture and refluxed for 2h. The reaction mixture was cooled and washed with water (2x40ml) and dried over anhydrous $MgSO_4$. The solvent was evaporated under reduced pressure to give the aza-reagents (**5**), (**6**) and (**7**).

N-[Bis(methylthio)-methylene] hydrazine carboxylic acid ethyl ester (5). Title compound (**5**) was prepared from hydrazine monocarboxylic acid ethyl ester as a syrup. Yield: 70%; bp. 113-115°C 0.02 mm Hg; Anal. Calc. for $C_6H_{12}N_2O_2S_2$(208.30); C 34.60; H 5.81; N 13.45. Found: C 34.66; H 5.89; N 13.51%, ^{1}H-NMR ($CDCl_3$); δ_H 8.20 (bs, ^{1}H, NH); 4.26 (q, 2H, OCH_2); 2.49 (s, 6H, two SMe); 1.30 (t, 3H, CH_3); ^{13}C-NMR ($CDCl_3$); δ_H 153 (s), 145.36 (s), 61.28 (t), 15.33 (q), 14.57 (q), 14.09 (q).

N-[Bis(methylthio)methylene]acetamide (6). This compound (**6**) was prepared from acid hydrazide (**11**) as yellow solid. Yield: 5%, mp 126-128°C; IR(KBr) 1680 (C=O): 1560, 1520, 1480, 1365, 1110, 1035 cm^{-1}; ^{1}H-NMR ($CDCl_3$); δ_H 8.7 (bs, 1H, NH), 2.33 (s, 3H, Me), 1.9 (s, 3H, SMe), 1.8 (s, 3H, SMe).

N-Bis(methylthio)methylene] benzamide (7). This compound (**7**) was obtained from benzahydrazide (Uddin 1988) as a syrup. Yield: 57%; bp. 116-118°C; IR (KBr) 3440, 3400, 3000, 2920, 2640, 1620, 1480, 1400 Cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ_H 7.93 (m, 2H, Ar-H), 7.5 (m, 3H, Ar-H), 4.73 (bs, 1H, NH), 2.86 (6H, two SMe).

N-[2-(Methylthio)-4-oxo-5,6,7,8-tetrahydro [1]benzothieno [2,3-d] pyrimidine-3 (4H)-yl]carbamic acid ethyl ester (8). A mixture of ethyl 2-amino-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (**1**) (0.33g, 1.5mmol), N-[bis(methylthio) methylene] hydrazine carb oxylic acid ethyl ester (**5**) (0.31g, 1.5 mmol) and *p*-toluene sulphonic acid (0.1g) in dry acetonitrile (8ml) was refluxed for 60 h. The reaction mixture was filtered, the filtrate, was evaporated *in vacuo* and the residue was crystallized from ether. After recrystallization from ether-ethyl acetate white crystals were obtained. Yield: 0.285g (57%); mp 145°C; Anal. Calc. for $C_{14}H_{17}N_3O_3S_2$ (339.44); C 49.54; H 5.04; N 12.37. Found: C 49.45; H 4.84; N 12.13%; H-NMR ($CDCl_3$): δ_H 7.40 (s, 1H, NH); 4.25 (q, 2H, CH); 2.90 (m, 2H, 8-H); 2.70 (m, 2H, 5-H); 2.50 (s, 3H, SMe); 1.80 (m, 4H, 6- and 7-H); 1.25 (t, 3H, Me); ^{13}C-NMR ($CDCl_3$): δ_c 162.38 (s, C=O); 159.64 (s, C=O); 156.90 (s, C-2); 155.27 (s, C-9a); 131.96 (s, C-8a); 131.14 (s, C-4b); 118.42 (s, C-4a); 62.88 (t, OCH_2); 24.98 (t, C-8); 24.88 (t, C-5); 27.74 (t, C-7); 22.02 (t, C-6); 14.33 (q, Me); 14.04 (q, SMe).

5,6,7,8-Tetrahydro-2-methylthio-4-axo[1] benzothieno [2, 3-d] pyrimidine-3 (4H)-acetamide (9). A mixture of *Ortho*-aminoester (**1**) (0.8g, 3.5 mmol) and N-[bis(methylthio) methylene]-acetamide (**6**) (0.6g, 3.5 mmol) in dry acetic acid (5 ml) was refluxed for 12 h. The reaction mixture was poured into ice-water. The precipitate was collected by filtration and obtained the compound (**9**) as crystals. Yield: 0.564g (57%); mp 70-71°C; IR (KBr): 3260, 2960, 2900, 1690; (C=O) 1600, 1560, 1440, 1400, 1275, 1140, 1075 cm^{-1}; ^{1}H-NMR ($CDCl_3$); δ_H 7.50 (s, 1H, NH); 2.80 (m, 2H, 8-H); 2.65 (m, 2H, 5-H); 2.54 (s, 3H, Me); 2.45 (s, 3H, SMe); 1.85 (m, 4H, 6-and 7H).

5,6,7,8-Tetrahydro-2-methylthio-4-oxo [1] benzothieno [2,3-d] pyrimidine-3 (4H)-benzamide (10). A solution of *Ortho*-aminoester (**1**) (0.5g, 2.22 mmol) in dry acetic acid (5 ml) was treated with N-[bis(methylthio)methylene] benzamide (**7**) (0.53g, 2.22 mmol). The reaction mixture was heated under reflux for 8 h and poured into ice-water. The precipitate was collected by filtration and the title compound (**10**) obtained as brown gummy-mass compound, which resisted recrystallization. Yield: 0.42g (52%); IR (KBr) 3300, 2920, 1660, 1480, 1120 cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ_H 8.19 (m, 2H, Ar-H), 7.7 (m, 3H, Ar-H); 4.66 (s, 1H, NH); 2.59 (s, 3H, SMe); 2.16 (m, 2H, 8-H); 1.90 (m, 2H, 5-H); 1.73 (m, 4H, 6- and 7-H).

2-[Bis (methylthio) methylene] hydrazinocarbonylamino]-6-methyl-4,5,6,7-tetrahydrobenzo[b]-thiophene-3-carboxylic acid ethyl ester (11). A mixture of ethyl 2-amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (**2**) (0.48g, 2 mmol) and N-[bis(methylthio) methylene] hydrazine carboxylic acid ethyl ester (**5**) (0.41g 2 mmol) in pyridine (5 ml) was refluxed for 7 h. The solid was collected by filtration and recrystallized from ethanol to give (**11**) as white crystals. Yield: 0.40g (49.6%); mp 225°C; Anal. Calc. for $C_{16}H_{23}N_3O_3S_4$ (401.57); C 47.85; H 5.77; N 10.46. Found: C 47.92; H 5.63; N 10.28%; ^{1}H-NMR ($CDCl_3$):δ_H 11.65 (s, 1H, NH); 8.40 (s, 1H, NH); 4.25 (q, 2H, OCH_2); 2.95-1.80 (m, 7H, 4, 5, 6 and 7-H); 2.70 (s, 3H, SMe); 2.50 (s, 3H, SMe); 1.40 (t, 3H, Me); 1.10 (d, 3H, 6-Me); ^{13}C-NMR ($CDCl_3$): δ_c 166.01 (s, C=O); 151.74 (s, C=O); 148.81 (s, N=C-S); 145.24 (s, C-2); 130.28 (s, C-7a); 124.98 (s, C-3a); 110.59 (s, C-3); 59.80 (t, CH_2); 32.30 (t, C-7); 31.03 (t, C-4); 29.09 (d, C-6); 26.09 (t, C-5); 21.28 (q, 6-Me); 15.18 (q, SMe); 14.16 (q, Me).

2-[Bis (methylthio) methylene] hydrazinocarbonylamino]-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carbonitrile (12). A mixture of 2-amino-6-methyl-4, 5, 6, 7-tetrahydrobenzo [b] thiophene-3-carbonitrile (**3**) (0.29g, 1.5 mmol) and N-[bis(methylthio) methylene] hydrazine

carboxylic acid ethyl ester (**5**) (0.31g, 1.5 mmol) in 5 ml of pyridine was refluxed for 10 h. The solid was collected by filtration and recrystallized from ethanol to give (**12**) as white crystals. Yield: 0.23g (44.8%); mp 195°C; Anal. Calc. for $C_{14}H_{18}N_4OS_3$ (345.52); C 47.43; H 5.12; N 15.80; Found: C47.17; H 4.88; N 15.52%; ^{1}H-NMR ($CDCl_3$); δ_H 9.00 (s, 1H, NH); 8.25(s, 1H, NH) 2.75-1.80 (m, 7H, 4,5,6 and 7H); 2.50 (s, 6H, two SMe); 1.05 (d, 3H, 6-Me); ^{13}C-NMR ($CDCl_3$); δ_c 150.80 (s, C=O); 148.10 (s, N=C-S); 147.48 (s, C-2); 130.22 (s, C 7a); 126.73 (s, C-3a); 114.39 (s, CN); 92.96 (s, C-3); 31.91 (t, C-7); 30.19 (t, C-4); 29.48 (d, C-6); 23.68 (t, C-5); 21.19 (q, 6-Me); 15.39 (q, SMe); 15.28 (q, SMe).

2-[Bis(methylthio) methylene] hydrazinocarbonylamino]-5,6,7,8-tetrahydro-4H-cyclohepta [b]-thiophene-3-carbonitrile(13). A mixture of 2-amino-5,6,7,8-tetrahydro-4H-cyclohepta [b]thiophene-3-carbonitrile (**4**) (0.29g, 1.5 mmol) and N-[bis(methylthio) methylene] hydrazine carboxylic acid ethyl ester (**5**) (0.31g, 1.5 mmol) in 3 ml of pyridine was refluxed for 16 h. The reaction mixture was cooled and the solid was collected by filtration and recrystallized from ethanol to give (**13**) as white crystals. Yield: 0.21g (39.5%); mp 205°C; Anal. Calc. for $C_{14}H_{18}N_4OS_3$ (354.52); C 47.43; H 5.12; N 15.80. Found: C 47.70; H 5.00; N 15.68%; IR (KBr): 3360, 3178, 3070, 2912, 2847, 2282, 1688, 1564, 1531, 1471, 1425, 1350, 1280, 1224, 1187, 1111, 1028 Cm^{-1}; ^{1}H-NMR ($CDCl_3$): δ_H 8.95 (s, 1H, NH); 8.25 (s, 1H, NH); 2.70 (m, 4H, 4 and 8-H); 2.65 (s, 6H, two SMe); 1.85 (m, 2H, 7-H); 1.65 (m, 4H, 5 and 6-H); 13C-NMR ($CDCl_3$): δ_c 150.54 (s, C=O); 146.79 (s, N=C-S); 134.65 (s, C-2 and C-8a); 129.21 (s, C-3a); 114.86 (s, CN); 95.00 (s, C-3); 31.37 (t, C-8); 28.34 (t, C-4) and C-7); 27.64 (t, C-6); 26.86 (t, C-5); 14.79 (q, two SMe).

Results and Discussion

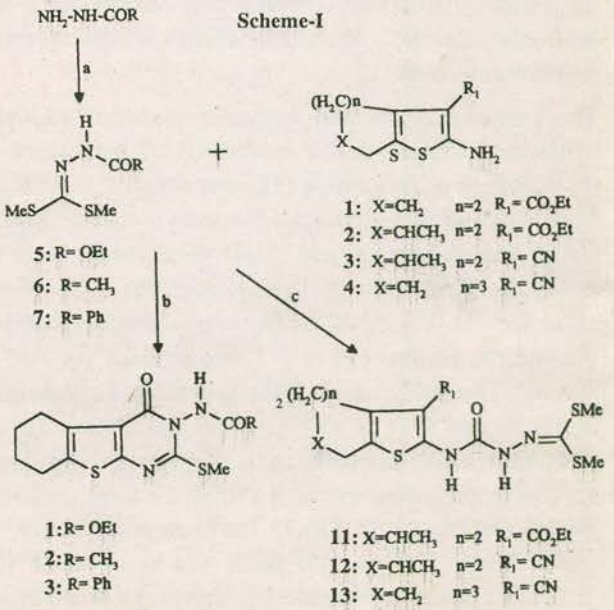
Ortho-aminoesters, ethyl 2-amino-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (**1**), ethyl 2-amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (**2**) and *ortho*-aminonitriles, 2-amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carbonitrile (**4**) were prepared using Gewald method (Gewald *et al* 1966).

Aza reagents (**5, 6** and **7**) were prepared by one-step reaction of hydrazide with CS_2, Et_3 N and MeI. The molar ratio of acid hydrazide, CS_2, Et_3 N and MeI was 1:1::2:2. The structural assignment of N-[bis (methylthio) methylene] hydrazine carboxylic acid ethyl ester (**5**) was done by Sauter *et al* (1995). In the IR spectrum of commpound (**6**) the signals at 1680 and 1520 cm^{-1} were indicated for C=O (amide carbonyl) and C=N bands existing in the molecule. Appearance of a three-proton singlet at δ 2.33 in the ^{1}H-NMR spectrum showed the presence of -CH_3 proton and a one-proton broad singlet at δ 8.7 for NH proton suggested the presence of acetamide group in the molecule. Two three-proton singlets at δ 1.9 and 1.8 indicated for two -SMe groups in the compound. The IR spectrum of compound (**7**) showed absorption bands at 3440 and 1620 cm^{-1}confirmed the presence of -NH and C=O stretchings. In ^{1}H-NMR spectrum of compound (**7**) showed a broad singlet at δ 7.39-7.5 due to the presence of aromatic ring and a broad singlet at δ 4.73 for -NH proton.

The tricyclic derivative (**8**) with a carbamate substituent was synthesized from ethyl 2-amino-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carboxylate (**1**) and reagent (**5**) with *p*-toluene sulphonic acid in dry CH_3CN refluxing for 60 h in 57% yield as white crystal (Scheme-1). In the ^{1}H-NMR, it showed a one-proton singlet at δ 7.40, a three-proton singlet at δ 2.50 due to -NH and SMe groups in the molecule and also a two-proton quartet at δ 4.25 and a three-proton triplet at δ 1.25 suggested the presence of ethyl group in the molecule. In ^{13}C-NMR, the singlets appeared at δ 162.38 and 159.64 for two carbonyl carbons and δ 156.90 for C-2 confirmed the formation of pyrimidine ring. The spectrum also exhibited the presence of fourteen carbon atoms corresponding to the molecular formula $C_{14}H_{17}N_3O_3S_2$ of the compound (**8**), which is also supported by its elemental analysis.

The *ortho*-amminoester (**1**), on reaction with aza reagent (**6**) in dry acetic acid afforded compound (**9**) in 50% as crystals,

Scheme-I



Reagents: (a) CS_2, EtN_3, MeI, (b) (i) p-TsOH, CH_3CN, reflux (ii) AcOH, reflux (c) Pyridine, reflux.

mp 70-71°C. In its IR spectrum, the absorption bands at 1960 and 3260 cm^{-1} corresponded to C=O and -NH stretching respectively. In ^{1}H-NMR spectrum of the compound (**9**), two three-proton singlets at δ 2.54 and 2.45 demonstrated the presence of Me and SMe groups in the molecule. The rest of the ^{1}H-NMR spectrum consisted the structure established as benzothieno [2, 3-d] pyrimidine derivative (**9**).

The compound 5,6,7,8-tetrahydro-2-methylthio-4-oxo (**1**) benzothieno [2, 3-d] pyrimidine-3 (4H)-benzamide (**10**) was also obtained in a same manner as of (**9**) from *ortho*-aminoester (**1**) and aza reagent (**7**). In ^{1}H-NMR spectrum of the compound (**10**), the main peaks were recognizable for the absorption at δ 8.19-7.7 due to the presence of phenyl group and also gave two singlets at δ 4.66 and 2.59 which indicated the presence of -NH and -SMe groups respectively in the molecule.

Ortho-aminoester (**2**) reacted with aza-reagent (**5**) in pyridine medium afforded uncyclized urethane (**11**) in 49.6% yield as white crystal, mp 225°C. In its ^{1}H-NMR spectrum, a one-proton singlet at δ 11.65 and 8.40 indicated two-NH protons in the urethane molecule. The resonances at δ 2.70 and 2.50 for two - SMe groups and at δ 4.25 and 1.40 for ester group and absence of $-NH_2$ group suggested that the compound (**11**) was uncyclized urthane.

Uncyclized urethane compound was also supported by ^{13}C-NMR, the spectrum displayed two singlets at δ 166.01 and 151.74 for two singlets at δ 166.01 and 151.74 for two C=O groups and two quarters at δ 15.48 and 15.18 for two - SMe groups in the molecule. The spectrum also displayed the presence of sixteen carbon atoms corresponding to its molecular formula $C_{16}H_{23}N_3O_3S$, which was also suported by its elemental analysis.

The urethane compound 2-[Bis(methylthio) methylene hydrazinocarbonylamino]-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3-carbonitrile (**12**) was obtained in 45% yield from 2-amino-6-methyl-4,5,6,7-tetrahydrobenzo [b] thiophene-3carbonitrile (**3**) and N-[bis (methylthio) methylene hydra-zine carboxylic acid ethyl ester (**5**) in pyridine at reflux for 10 h. The ^{1}H-NMR spectrum of the compound (**12**) showed the resonances at δ 9.00 and 8.25 for two NH protons. The substitutents SMe and 6-Me were observed at δ 2.50 and 1.50 as singlets. The rest of the ^{1}H-NMR spectrum was also accorded to its structure. ^{13}C-NMR spectrum gave the resonances at δ 150.80 for C=O, 148.10 for N=C-S, 130.22 C-7a, 126.73 for C-3a respectively. The presence of -CN(δ 114.39) group and two SMe (δ 15.39 and 15.28) groups in the molecule proved that the compound was uncyclized urethane (**12**) and it was also evident from its microanalytical data. Treatment of *ortho*-aminonitrile (**4**) and aza reagent (**5**) in pyridine led 2-[Bis (methylthio) methylene] hydrazino-carbonylamino]-5,6,7,8-tetrahydro-4H-cyclohepta [b] thiophene-3-carbonitrile (**13**) in 39.5% yield as white crystals. The structure of this compound was confirmed by spectroscopical and microanalytical data.

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