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New General Synthesis of Benzo[4,5]imidazo[1,2-*a*]pyrimidine and Benzo[4,5]imidazo[2,1-*b*]quinazoline Derivatives

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Abstract—The reactions of benzene-1,2-diamine with 5-substituted 2-(alkylsulfanyl)-4-methylpyrimidin-6(1*H*)-ones and 2-(propylsulfanyl)- and 5-iodo-2-(propylsulfanyl)-3,4-dihydroquinazolines at 175–185°C under solvent-free conditions unexpectedly afforded benzo[4,5]imidazo[1,2-*a*]pyrimidine, benzo[4,5]imidazo[2,1-*b*]-quinazoline, and 2,2′-(benzene-1,2-diyldiimino)bis[pyrimidin-4(3*H*)-ones]. The described reaction is the first example of synthesis of these heterocyclic systems by fusion of benzimidazole to pyrimidine or quinazoline and is likely to follow ANRORC mechanism.

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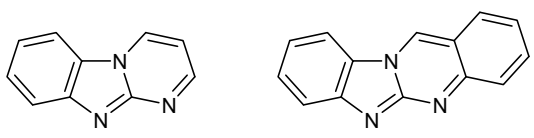
In recent years, an increased interest has been observed in the synthesis and properties of fused nitrogen heterocycles based on pyrimidine, quinazoline, and benzimidazole, in particular benzo[4,5]imidazo[1,2-*a*]-pyrimidine (**1**) and benzo[4,5]imidazo[2,1-*b*]quinazoline (**2**) derivatives, many of which exhibit a broad spectrum of biological activity [1–4].

All known synthetic approaches to benzo[4,5]-imidazo[1,2-*a*]pyrimidine derivatives are based on the condensation of benzimidazol-2-amine (**3a**) as 1,3-N,N-binucleophile with various 1,3-bielectrophiles and their synthetic equivalents [2, 3, 5], while benzo-[4,5]imidazo[2,1-*b*]quinazoline derivatives are obtained by condensation of 2-substituted benzimidazoles **3a**–**3d** with anthranilic or 2-chloro or 2-bromobenzoic acid, as well as by thermal rearrangement of substituted indazole or by reaction of isatoic anhydride with benzene-1,2-diamine [6]. Thus, all so-far-known



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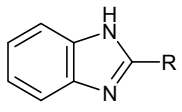


3a–3d

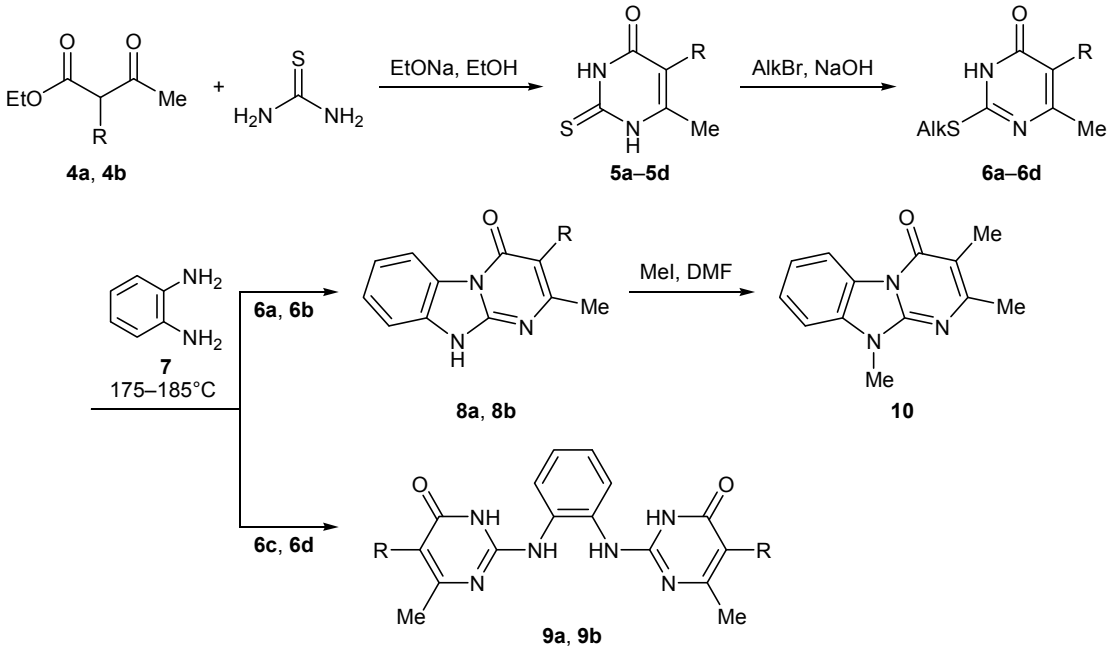
R = H_2N (**a**), Cl (**b**), MeS (**c**), $HOSO_2$ (**d**).

methods for the synthesis of heterocyclic systems **1** and **2** involve mainly closure of pyrimidine or quinazoline ring on benzimidazole derivatives, whereas an alternative approach implying fusion of benzimidazole to already existing pyrimidine or quinazoline ring has not been reported.

As an implementation of the latter approach, the present article describes a new general method for the synthesis of derivatives of heterocyclic systems **1** and **2** according to a cascade mechanism. The condensation of substituted ethyl acetoacetates **4a** and **4b** with thiourea in anhydrous ethanol in the presence of sodium ethoxide gave 2-thioxopyrimidines **5b** and **5d** which, as well as previously described analogs **5a** and **5c**, were alkylated with propyl bromide and butyl bromide to obtain key 2-(alkylsulfanyl)pyrimidines **6a**–**6d**. 2-Propylsulfanyl derivatives **6a** and **6b** reacted with benzene-1,2-diamine (**7**) at 175–185°C without a solvent to give targeted benzo[4,5]imidazo[1,2-*a*]pyrimidines **8a** and **8b**. Under analogous conditions, 5-arylmethyl-substituted compounds **6c** and **6d** gave rise to mixtures of products, from which bis-pyrimidines **9a** and **9b** were isolated in poor yield by crystallization from DMF (Scheme 1).

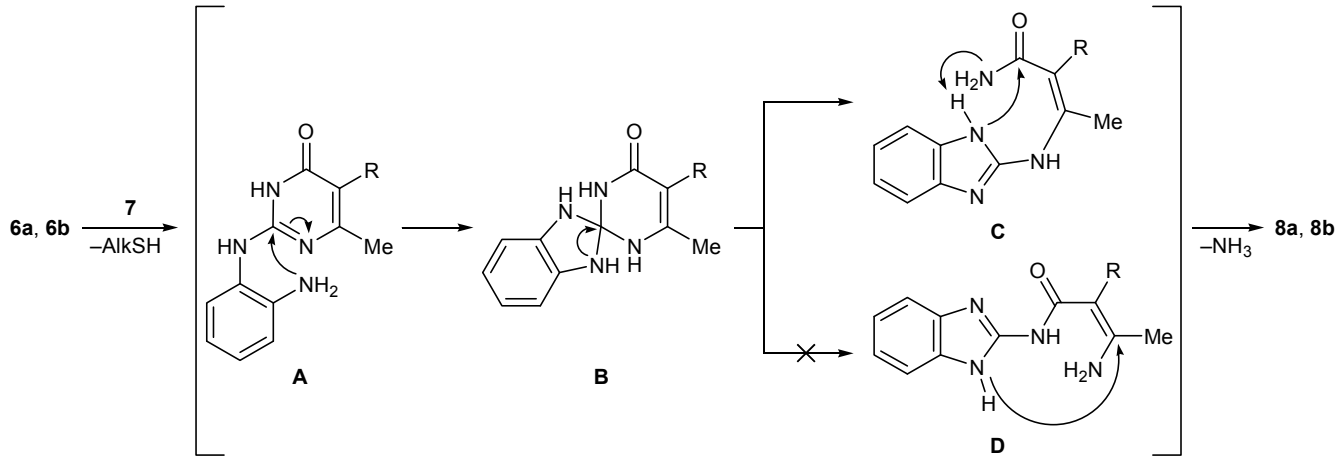
These unexpected results may be rationalized in terms of the ANRORC mechanism including several fast consecutive reactions: double nucleophilic attack on C^2 in **6a** and **6b** by the vicinal amino groups of benzene-1,2-diamine with elimination of propane-1-

Scheme 1.



4, R = 2,4-$Me_2C_6H_3$ CH_2 (**a**), CH_2=C(Me)CH_2 (**b**); **5**, R = Me (**a**), CH_2=C(Me)CH_2 (**b**), $PhCH_2$ (**c**), 2,4-$Me_2C_6H_3CH_2$ (**d**); **6**, Alk = Pr, R = Me (**a**), $PhCH_2$ (**c**), 2,4-$Me_2C_6H_3CH_2$ (**d**); Alk = Bu, R = CH_2=C(Me)CH_2 (**b**); **8**, R = Me (**a**), CH_2=C(Me)CH_2 (**b**); **9**, R = $PhCH_2$ (**a**), 2,4-$Me_2C_6H_3CH_2$ (**b**).

Scheme 2.

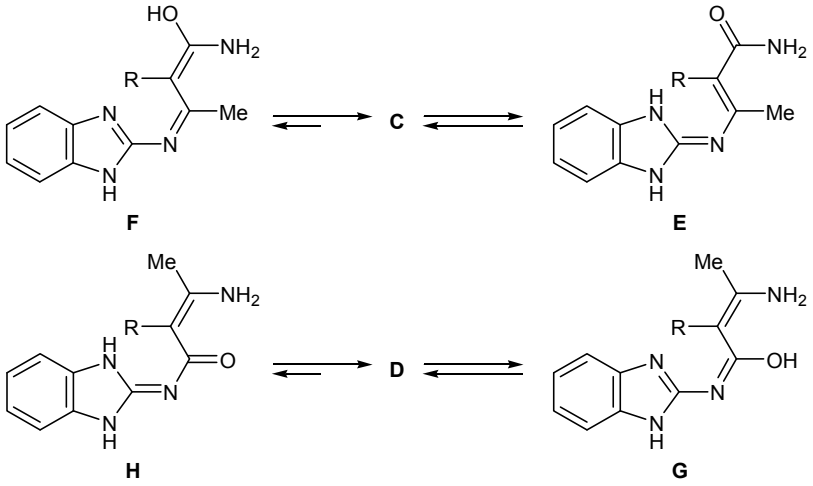


thiol and 1,3-proton migration (assumed intermediates **A** and **B**), selective opening of the pyrimidine ring at the C^2–N^3 bond, and final recyclization via nucleophilic substitution of the amino group in intermediate butenamide by the NH group of the benzimidazole fragment (assumed intermediate **C**) with elimination of ammonia (Scheme 2).

The formation of only one isomer, 4-oxo derivative **8a** or **8b**, is likely to be favored by the higher strength of the exocyclic C^2–N bond in intermediate **C** which may exist mainly as tautomer **E** where the above bond is double (cf. structure **F**). On the other hand, both N^1,N^3-prototropy (tautomer **H**) and amide–imidic acid tautomerism (tautomer **G**) are possible for assumed intermediate **D** with reduced contribution of the HNC(NH)=NC=O structure (Scheme 3).

The alkylation of 2,3-dimethyl-substituted benzo[4,5]imidazo[1,2-*a*]pyrimidine **8a** with methyl iodide in DMF in the presence of potassium carbonate regioselectively afforded 2,3,10-trimethyl derivative **10** (Scheme 1). The structure of **10** was confirmed by the 2D NOESY spectrum which revealed NOE be-

Scheme 3.



tween the 10-CH_3 and 9-H protons. The lack of NOE between the 10-CH_3 and 2-CH_3 protons also indicated methylation at the 10-position.

Likewise, the alkylation of 2-thioxoquinazolines **11a** and **11b** with propyl bromide in water in the presence of NaOH gave 2-(propylsulfanyl)quinazolines **12a** and **12b**. Quinazoline **12c** (R = R′ = H) was described previously [7]. Compounds **12a**–**12c** reacted with diamine **7** at 175–185°C under solvent-free conditions, yielding benzo[4,5]imidazo[2,1-*b*]quinazoline derivatives **13a** and **13b**. The formation of **13a** from both 3-(3-methylphenyl)quinazoline **12a** and 3-unsubstituted analog **12c** confirmed selective cleavage of the C^2–N^3 bond in the quinazoline ring system during the reaction (Scheme 4).

The structure of the isolated compounds was proved by spectral data and elemental analyses. Signals in the ^1H and ^{13}C NMR spectra were assigned using double resonance, DEPT, HMQC, and NOESY techniques.

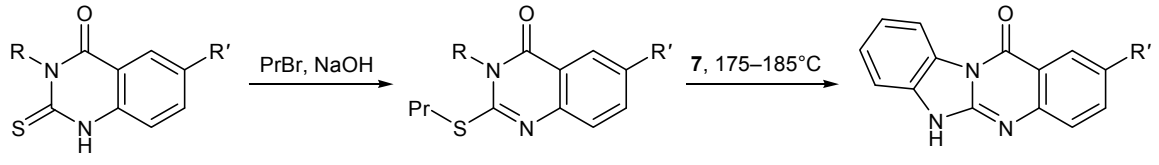
Thus, a new environmentally benign, economic, and experimentally convenient method has been developed for the synthesis of benzo[4,5]imidazo-[1,2-*a*]pyrimidine and benzo[4,5]imidazo[2,1-*b*]quinazoline derivatives from readily accessible 2-(alkylsulfanyl)pyrimidines and -quinazolines. The proposed method avoids the use of benzimidazol-2-amine which is obtained from highly toxic starting materials.

EXPERIMENTAL

The IR spectra were recorded on a Nicolet Avatar 330 spectrometer from samples dispersed in mineral oil. The ^1H and ^{13}C NMR spectra were measured on a Varian Mercury-300 VX instrument at 300.8 and 75.46 MHz, respectively, using tetramethylsilane as internal standard. Analytical thin-layer chromatography was performed on Silufol UV-254 plates using ethanol–dichloroethane (1 : 10) as eluent; spots were visualized by treatment with iodine vapor.

2-Sulfanylidenepyrimidines 5b and 5d (*general procedure***).** Sodium metal, 2.3 g (0.1 mol), was dissolved in 100 mL of anhydrous ethanol, 13.0 g (0.1 mol) of ethyl acetoacetate and 15.5 g (0.1 mol) of 2-(chloromethyl)-1,3-dimethylbenzene were added, and the mixture was refluxed for 6 h. Excess ethanol was distilled off, the residue was treated with 100 mL of benzene, the benzene extract was washed with water, dried, and evaporated, and the residue (ester **4a**, 24 g, 97%) was used in the next step without additional purification. Thiourea, 0.76 g (0.01 mol), and

Scheme 4.



11, **12**, R = 3-MeC_6H_4, R′ = H (**a**); R = Me, R′ = I (**b**); **13**, R = H (**a**), I (**b**).

ester **4a** or **4b** [8], 0.01 mol, were added to a solution of 0.46 g (0.02 mol) of sodium metal in 50 mL of anhydrous ethanol, and the mixture was refluxed for 7 h. The solvent was distilled off, the residue was dissolved in 50 mL of water, the solution was acidified with acetic acid to pH 6, the mixture was kept for 8–10 h in the cold, and the precipitate was filtered off and dried.

6-Methyl-5-(2-methylprop-2-en-1-yl)-2-sulfanylidene-2,3-dihydropyrimidin-4(1*H*)-one (5b) was synthesized from ester **4b**. Yield 1.0 g (51%), mp 226–228°C (from EtOAc), R_f 0.28. IR spectrum, ν, cm^{-1}: 3064 (N–H), 1648 (C=O), 1590 (C=C, C=N). Found, %: N 14.39. $C_9H_{12}N_2OS$. Calculated, %: N 14.27.

5-[(2,4-Dimethylphenyl)methyl]-6-methyl-2-sulfanylidene-2,3-dihydropyrimidin-4(1*H*)-one (5d) was synthesized from ester **4a**. Yield 1.2 g (46%), mp 307–308°C (from 60% AcOH), R_f 0.41. IR spectrum, ν, cm^{-1}: 3060 (N–H), 1676 (C=O), 1620 (C=C, C=N). Found, %: N 10.80. $C_{14}H_{16}N_2OS$. Calculated, %: N 10.76.

5-Substituted 2-(alkylsulfanyl)pyrimidinones 6a–6d (*general procedure*). Propyl or butyl bromide, 0.011 mol, was added to a solution of 0.01 mol of pyrimidinone **5a** [9], **5b**, **5c** [10], or **5d** and 0.4 g (0.01 mol) of sodium hydroxide in 50 mL of water. The mixture was heated for 15 h under reflux and left overnight, and the precipitate was filtered off, washed with water, and dried.

5,6-Dimethyl-2-(propylsulfanyl)pyrimidin-4(3*H*)-one (6a). Yield 79%, finely crystalline powder, mp 139–141°C (from EtOH), R_f 0.65. IR spectrum, ν, cm^{-1}: 1665 (C=O), 1620 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.02 t (3H, CH_2CH_3, J = 7.3 Hz), 1.70 q.t (2H, CH_2CH_3, J = 7.3, 7.1 Hz), 1.89 s (3H, CH_3), 2.19 s (3H, CH_3), 3.05 t (2H, SCH_2, J = 7.1 Hz), 12.17 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 10.1 (CH_3), 12.9 (CH_3), 21.1 (CH_3), 22.0 (CH_2), 31.2 (CH_2), 114.3, 156.3, 158.2, 162.6. Found, %: N 14.33. $C_9H_{14}N_2OS$. Calculated, %: N 14.13.

2-(Butylsulfanyl)-4-methyl-5-(2-methylprop-2-en-1-yl)pyrimidin-4(3*H*)-one (6b). Yield 76%, powder, mp 92–93°C (from 80% EtOH), R_f 0.57. IR spectrum, ν, cm^{-1}: 1638 (C=O), 1570 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 0.96 t (3H, CH_3CH_2, J = 7.3 Hz), 1.39–1.51 m (2H, CH_3CH_2), 1.61–1.71 m (2H, $CH_3CH_2CH_2$), 1.73 br.s (3H, $CH_3C=CH_2$), 2.15 s (3H, 4-CH_3), 3.07 br.s (2H, CH_2CCH_3), 3.08 t (2H, SCH_2, J = 7.2 Hz), 4.50–4.53 m and 4.65–4.68 m (1H each, $=CH_2$), 12.25 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 13.2 (CH_3CH_2), 20.9 (CH_3), 21.2 (CH_2), 22.2 (CH_3), 28.9 (CH_2), 30.7 (CH_2), 32.1 (CH_2), 109.5 ($=CH_2$), 116.2, 142.0, 157.2, 159.8, 162.4. Found, %: N 11.10. $C_{13}H_{20}N_2OS$. Calculated, %: N 11.10.

5-Benzyl-4-methyl-2-(propylsulfanyl)pyrimidin-4(3*H*)-one (6c). Yield 79%, powder, mp 148–150°C (from 60% EtOH), R_f 0.67. IR spectrum, ν, cm^{-1}: 1653 (C=O), 1573 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.03 t (3H, CH_3CH_2, J = 7.4 Hz), 1.71 q.t (2H, CH_3CH_2, J = 7.4, 7.1 Hz), 2.19 s (3H, 4-CH_3), 3.06 t (2H, SCH_2, J = 7.1 Hz), 3.73 s (2H, $CH_2C_6H_5$), 7.06–7.22 m (5H, C_6H_5), 12.38 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 12.9 (CH_3), 21.3 (CH_2), 22.0 (CH_3), 30.0 (CH_2), 31.2 (CH_2), 125.2 (C^p), 127.6 (C^o, C^m), 139.5 (C^i). Found, %: N 10.18. $C_{15}H_{18}N_2OS$. Calculated, %: N 10.21.

5-[(2,4-Dimethylphenyl)methyl]-4-methyl-2-(propylsulfanyl)pyrimidin-4(3*H*)-one (6d). Yield 63%, powder, mp 175–177°C (from 60% EtOH), R_f 0.62. IR spectrum, ν, cm^{-1}: 1654 (C=O), 1553 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.05 t (3H, CH_3CH_2, J = 7.3 Hz), 1.74 sext (2H, CH_3CH_2, J = 7.3 Hz), 2.09 s (3H, CH_3), 2.25 s (3H, CH_3), 2.32 s (3H, CH_3), 3.09 t (2H, SCH_2, J = 7.3 Hz), 3.61 s (2H, $CH_2C_6H_3$), 6.64 d (1H, 6′-H, J = 7.7 Hz), 6.78 d.d (1H, 5′-H, J = 7.7, 1.7 Hz), 6.90 d (1H, 3′-H, J = 1.7 Hz), 12.37 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 13.0 (CH_3CH_2), 19.1 (CH_3), 20.3 (CH_3), 21.2 (CH_2), 22.0 (CH_3), 26.8 (CH_2), 31.3 (SCH_2), 38.9, 39.2, 117.0, 125.8 (CH), 126.0 (CH), 130.1 (CH), 133.7, 133.9, 135.0, 157.4 br, 160.0, 162.5. Found, %: N 9.40. $C_{17}H_{22}N_2OS$. Calculated, %: N 9.26.

2-Methyl-3-R-pyrimido[1,2-*a*]benzimidazol-4(10*H*)-ones 8a–8d (*general procedure*). A mixture of 0.01 mol of compound **6a**–**6d** and 1.19 g (0.011 mol) of benzene-1,2-diamine was heated for 6 h at 175–185°C on a Wood's metal bath. After cooling, the melt was treated with 20 mL of ethanol, the mixture was kept for 2 h at 0°C, and the precipitate was filtered off and recrystallized.

2,3-Dimethylpyrimido[1,2-*a*]benzimidazol-4(10*H*)-one (8a) was synthesized from **6a**. Yield 1.46 g (69%), powder, mp 331–332°C (from 60% AcOH), R_f 0.73; published data [11]: mp 330°C. IR spectrum, ν, cm^{-1}: 1682 (C=O), 1643 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.02 s (3H,

CH_3), 2.33 s (3H, CH_3), 7.26 d.d.d (1H, H_{arom}, J = 7.9, 7.3, 1.2 Hz), 7.41 d.d.d (1H, H_{arom}, J = 8.0, 7.3, 1.2 Hz), 7.49 br.d (1H, H_{arom}, J = 7.9 Hz), 8.40 br.d (1H, H_{arom}, J = 8.0 Hz), 12.49 br.s (1H, NH). Found, %: N 19.76. $C_{12}H_{11}N_3O$. Calculated, %: N 19.71.

2-Methyl-3-(2-methylprop-2-en-1-yl)pyrimido-[1,2-*a*]benzimidazol-4(10*H*)-one (8b) was synthesized from **6b**. Yield 1.8 g (72%), powder, mp 292–294°C (from 60% AcOH), R_f 0.43. IR spectrum, ν, cm^{-1}: 1668 (C=O), 1653, 1620 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.74 br.s (3H, C**H**$_3$C=CH_2), 2.29 s (3H, 2-CH_3), 3.24 br.s (2H, CH_2C=), 4.55–4.58 m and 4.70–4.73 m (1H each, =CH_2), 7.27 d.d.d (1H, 8-H, J = 7.9, 7.3, 1.2 Hz), 7.42 d.d.d (1H, 7-H, J = 8.0, 7.3, 1.2 Hz), 7.51 d.d.d (1H, 9-H, J = 7.9, 1.2, 1.2 Hz), 8.39 d.d.d (1H, 6-H, J = 8.0, 1.2, 1.2 Hz), 12.60 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 19.7 (CH_3), 22.4 (CH_3), 32.2 (CH_2), 106.7, 109.7 (=CH_2), 113.42 br, 115.0 (CH), 121.0 (CH), 125.4 (CH), 126.9, 143.1, 146.8, 159.4. Found, %: N 16.72. $C_{15}H_{15}N_3O$. Calculated, %: N 16.59.

2,2′-(Benzene-1,2-diyldiimino)bis[5-benzyl-6-methylpyrimidin-4(3*H*)-one] (9a) was synthesized from **6c**. Yield 1.31 g (26%), mp 350–352°C (from DMF), R_f 0.59. IR spectrum, ν, cm^{-1}: 3460, 3375 (N–H), 1647 (C=O), 1615 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 2.08 s (6H, CH_3), 3.70 s (4H, CH_2), 7.04–7.12 m (4H, H_{arom}), 7.14–7.23 m (8H, H_{arom}), 7.70–7.79 m (2H, H_{arom}), 7.89 sh.s (2H, NH), 10.99 br.s (2H, NH). Found, %: N 16.47. $C_{30}H_{28}N_6O_2$. Calculated, %: N 16.66.

2,2′-(Benzene-1,2-diyldiimino)bis{5-[(2,4-dimethylphenyl)methyl]-6-methylpyrimidin-4(3*H*)-one} (9b) was synthesized from **6d**. Yield 2.4 g (43%), mp 326–328°C (from DMF), R_f 0.37. IR spectrum, ν, cm^{-1}: 3342, 3312 (N–H), 1689 (C=O), 1609 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.94 s (6H, CH_3), 2.20 s (6H, CH_3), 2.28 s (6H, CH_3), 3.57 s (4H, CH_2), 6.68 d (2H, 5′-H, J = 7.8 Hz), 6.83 d.d (2H, 6′-H, J = 7.8, 1.8 Hz), 6.95 d (2H, 3′-H, J = 1.8 Hz), 7.10–7.19 m and 7.70–7.80 m (2H each, C_6H_4), 8.08 br.s (2H, NH), 11.16 br.s (2H, NH). Found, %: N 14.69. $C_{34}H_{36}N_6O_2$. Calculated, %: N 14.99.

2,3,10-Trimethylpyrimido[1,2-*a*]benzimidazol-4(10*H*)-one (10). A mixture of 1.1 g (5 mmol) of **8a**, 0.69 g (5 mmol) of K_2CO_3, and 0.71 g (5 mmol) of methyl iodide in 7 mL of DMF was left overnight at room temperature. The mixture was then heated for 1 h on a boiling water bath, poured into 30 mL of water, and kept in the cold. The white solid was filtered off and dried. Yield 0.87 g (77%), mp 235–237°C (from DMF), R_f 0.61. IR spectrum, ν, cm^{-1}: 1670 (C=O), 1610 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 2.05 s (3H, CH_3), 2.35 s (3H, CH_3), 3.72 s (3H, NCH_3), 7.34 d.d.d (1H, 7-H, J = 8.1, 7.4, 1.2 Hz), 7.52 d.d.d (1H, 8-H, J = 8.1, 7.4, 1.0 Hz), 7.63 d.d (1H, 9-H, J = 8.1, 1.2 Hz), 8.47 d.d (1H, 6-H, J = 8.1, 1.0 Hz). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 10.6 (CH_3), 22.5 (CH_3), 28.0 (CH_3), 106.6, 109.4 (CH), 115.3 (CH), 121.7 (CH), 124.7, 125.8 (CH), 131.6, 146.2, 159.3, 159.5. Found, %: N 18.65. $C_{13}H_{13}N_3O$. Calculated, %: N 18.49.

3-(3-Methylphenyl)-2-(propylsulfanyl)quinazolin-4(3*H*)-one (12a) was synthesized by alkylation of quinazolinone **11a** [12] as described above for **6a**–**6d**. Yield 2.44 g (79%), powder, mp 91–93°C (from EtOAc–hexane), R_f 0.47. IR spectrum, ν, cm^{-1}: 1695 (C=O), 1610 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.03 t (3H, C**H**$_3$$CH_2$, J = 7.3 Hz), 1.72 sext (2H, CH_3C**H**$_2$, J = 7.3 Hz), 2.46 s (3H, CH_3), 3.04–3.18 m (2H, SCH_2), 7.05–7.09 m (2H, C_6**H**$_4$$CH_3$), 7.31–7.35 m (1H, C_6**H**$_4$$CH_3$), 7.38 d.d.d (1H, 6-H, J = 7.9, 7.1, 1.2 Hz), 7.39–7.45 m (1H, C_6**H**$_4$$CH_3$), 7.54 d.d (1H, 8-H, J = 8.2, 1.2 Hz), 7.72 d.d.d (1H, 7-H, J = 8.2, 7.1, 1.6 Hz), 8.09 d.d (1H, 5-H, J = 7.9, 1.6 Hz). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 13.0 (CH_3), 20.7 (CH_3), 21.5 (CH_2), 33.5 (SCH_2), 119.4, 124.9 (CH), 125.6 (CH), 125.8 (CH), 126.4 (CH), 128.6 (CH), 129.2 (CH), 129.8 (CH), 133.7 (CH), 135.5, 138.4, 147.1, 156.8, 160.2. Found, %: N 8.85. $C_{18}H_{18}N_2OS$. Calculated, %: N 9.02.

6-Iodo-3-methyl-2-(propylsulfanyl)quinazolin-4(3*H*)-one (12b) was synthesized in a similar way from quinazolinone **11b** [13]. Yield 63%, powder, mp 107–108°C (from EtOAc–hexane), R_f 0.64. IR spectrum, ν, cm^{-1}: 1665 (C=O), 1595 (C=C, C=N). ^1H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 1.09 t (3H, C**H**$_3$$CH_2$, J = 7.4 Hz), 1.80 q.t (2H, CH_3C**H**$_2$, J = 7.4, 7.2 Hz), 3.24 t (2H, SCH_2, J = 7.2 Hz), 3.53 s (3H, NCH_3), 7.24 d (1H, 8-H, J = 8.6 Hz), 7.92 d.d (1H, 7-H, J = 8.6, 2.1 Hz), 8.36 d (1H, 5-H, J = 2.1 Hz). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 13.0 (CH_3), 21.5 (CH_2), 29.6 (CH_3), 33.2 (CH_2), 88.5, 120.3, 127.5 (CH), 134.8 (CH), 141.9 (CH), 146.0, 157.5, 158.9. Found, %: N 8.29. $C_{11}H_{12}IN_2OS$. Calculated, %: N 8.07.

Compounds **13a** and **13b** were synthesized according to the procedure described above for **8a** and **8b**.

Benzimidazo[2,1-*b*]quinazolin-12(6*H*)-one (13a) was synthesized from **12a** (yield 69%) or **12c** (yield

86%). Powder, mp >380°C (from DMF), R_f 0.69; published data [14]: mp 395–400°C. IR spectrum, ν, cm^{-1}: 1691 (C=O), 1657, 1610 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 7.19–7.28 m (2H, H_{arom}), 7.35 t.d (1H, H_{arom}, J = 7.6, 1.2 Hz), 7.42 br.d (1H, H_{arom}, J = 7.9 Hz), 7.50 br.d (1H, H_{arom}, J = 8.3 Hz), 7.67 t.d (1H, H_{arom}, J = 8.3, 1.6 Hz), 8.25 d.d (1H, H_{arom}, J = 8.0, 1.6 Hz), 8.44 br.d (1H, H_{arom}, J = 7.8 Hz), 12.36 br.s (1H, NH).

2-Iodobenzimidazo[2,1-*b*]quinazolin-12(6*H*)-one (13b) was synthesized from **12b**. Yield 77%, powder, mp >340°C (from 60% AcOH), R_f 0.63. IR spectrum, ν, cm^{-1}: 1692 (C=O), 1664, 1608 (C=C, C=N). ^{1}H NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ, ppm: 7.23 d.d.d (1H, 8-H, J = 7.9, 6.6, 2.0 Hz), 7.30 d (1H, 4-H, J = 8.7 Hz), 7.34–7.42 m (2H, 7-H, 9-H), 7.88 d.d (1H, 3-H, J = 8.7, 2.1 Hz), 8.45 br.d (1H, 10-H, J = 8.0 Hz), 8.52 d (1H, 1-H, J = 2.1 Hz), 12.47 br.s (1H, NH). ^{13}C NMR spectrum (DMSO-d_6–CCl_4, 1:3), δ_C, ppm: 83.7, 111.4, 115.1, 117.2, 120.7, 125.0, 125.3, 126.5, 134.8, 141.6, 146.9, 146.9, 157.1. Found, %: N 11.30. $C_{14}H_8IN_3O$. Calculated, %: N 11.64.

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