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SUPPLEMENTARY MATERIAL TO

Synthesis and biological activity of some triazole-bearing benzimidazole derivatives

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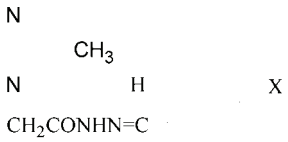
TABLE I-S. Physical and analytical properties of compounds **4a**–**k**



Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
4a	–	78	170–72	$C_{17}H_{16}N_4O$	292.33	Methanol	69.85; 69.83	5.52; 5.50	19.17; 19.15
4b	4-CH_3	75	188–90	$C_{18}H_{18}N_4O$	306.36	Methanol	70.57; 70.54	5.92; 5.90	18.29; 18.27
4c	4-CH_2CH_3	69	195–97	$C_{19}H_{20}N_4O$	320.38	Methanol	71.23; 71.23	6.29; 6.27	17.49; 17.48
4d	4-$CH_2CH_2CH_3$	72	196–98	$C_{20}H_{22}N_4O$	334.41	Methanol	71.83; 71.80	6.63; 6.60	16.75; 16.74
4e	2-OH	82	182–84	$C_{17}H_{16}N_4O_2$	308.33	Methanol–water	66.22; 66.20	5.23; 5.21	18.17; 18.15
4f	4-OH	74	185–87	$C_{17}H_{16}N_4O_2$	308.33	Methanol–water	66.22; 66.21	5.23; 5.20	18.17; 18.15
4g	2-OCH_3	75	160–62	$C_{18}H_{18}N_4O_2$	322.36	Acetone	67.07; 67.05	5.63; 5.60	17.38; 17.36
4h	4-OCH_3	70	169–71	$C_{18}H_{18}N_4O_2$	322.36	Acetone	67.07; 67.04	5.63; 5.60	17.38; 17.36
4i	2-Cl	81	196–98	$C_{17}H_{15}N_4OCl$	326.78	Methanol	62.48; 62.46	4.63; 4.61	17.15; 17.14
4j	4-Cl	80	178–80	$C_{17}H_{15}N_4OCl$	326.78	Methanol–water	62.48; 62.46	4.63; 4.60	17.15; 17.14
4k	4-NH_2	73	150–52	$C_{17}H_{17}N_5O$	307.35	Acetone	66.43; 66.41	5.58; 5.56	22.79; 22.77

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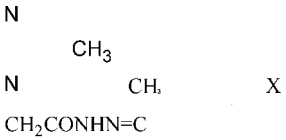
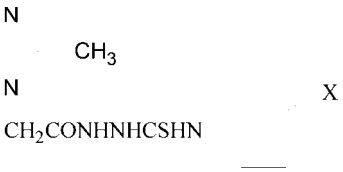
TABLE II-S. Physical and analytical properties of compounds **4l–v**



Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
4l	–	80	160–62	$C_{18}H_{18}N_4O$	306.36	Glacial acetic acid	70.57; 70.55	5.92; 5.90	18.29; 18.27
4m	4-CH_3	79	180–82	$C_{19}H_{20}N_4O$	320.38	Ethanol	71.23; 71.20	6.29; 6.27	17.49; 17.46
4n	4-CH_2CH_3	78	188–90	$C_{20}H_{22}N_4O$	334.41	Ethanol	71.83; 71.81	6.63; 6.60	16.75; 16.73
4o	4-$CH_2CH_2CH_3$	75	195–97	$C_{21}H_{24}N_4O$	348.44	Ethanol	72.39; 72.36	6.94; 6.92	16.08; 16.06
4p	2-OH	82	178–80	$C_{18}H_{18}N_4O_2$	322.36	Glacial acetic acid	67.07; 60.05	5.63; 5.61	17.38; 17.33
4q	4-OH	74	184–86	$C_{18}H_{18}N_4O_2$	322.36	Glacial acetic acid	67.07; 60.05	5.63; 5.60	17.38; 17.38
4r	2-OCH_3	75	145–47	$C_{19}H_{20}N_4O_2$	336.38	Chloroform–water	67.84; 67.82	5.99; 5.97	16.66; 16.64
4s	4-OCH_3	72	153–55	$C_{19}H_{20}N_4O_2$	336.38	Chloroform–water	67.84; 67.82	5.99; 5.97	16.66; 16.63
4t	2-Cl	81	178–80	$C_{18}H_{17}N_4OCl$	340.80	Chloroform–water	63.44; 63.41	5.03; 5.01	16.44; 16.42
4u	4-Cl	80	171–73	$C_{18}H_{17}N_4OCl$	340.80	Chloroform–water	63.44; 63.42	5.03; 5.01	16.44; 16.41
4v	4-NH_2	73	180–82	$C_{18}H_{19}N_5O$	321.34	Acetic acid	67.27; 67.25	5.96; 5.94	21.79; 21.76

TABLE III-S. Physical and analytical properties of compounds **5a–k**

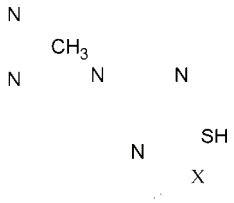


Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
5a	–	76	170–72	$C_{17}H_{17}N_5OS$	339.4g1	Methanol	60.16; 60.14	5.05; 5.03	20.63; 20.61
5b	4-CH_3	79	189–91	$C_{18}H_{19}N_5OS$	353.44	Methanol	61.17; 61.15	5.42; 5.40	19.81; 19.80
5c	4-CH_2CH_3	76	200–02	$C_{19}H_{21}N_5OS$	367.46	Methanol	62.10; 62.08	5.76; 5.74	19.06; 19.03

TABLE III-S. Continued

Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
5d	4-$CH_2CH_2CH_3$	71	197–99	$C_{20}H_{23}N_5OS$	381.49	Methanol	62.97; 62.95	6.08; 6.06	18.36; 18.33
5e	2-OH	71	182–84	$C_{17}H_{17}N_5O_2S$	355.41	Methanol–water	57.45; 57.42	4.82; 4.80	19.70; 19.68
5f	4-OH	80	185–87	$C_{17}H_{17}N_5O_2S$	355.41	Methanol–water	57.45; 57.43	4.82; 4.80	19.70; 19.65
5g	2-OCH_3	78	160–62	$C_{18}H_{19}N_5O_2S$	369.44	Acetone	58.52; 58.50	5.18; 5.16	18.96; 18.94
5h	4-OCH_3	82	169–71	$C_{18}H_{19}N_5O_2S$	369.44	Acetone	58.52; 58.50	5.18; 5.15	18.96; 18.93
5i	2-Cl	85	196–98	$C_{17}H_{16}N_5OSCl$	373.85	Methanol	54.61; 54.59	4.31; 4.29	18.73; 18.70
5j	4-Cl	74	178–80	$C_{17}H_{16}N_5OSCl$	373.85	Methanol–water	54.61; 54.60	4.31; 4.28	18.73; 18.70
5k	4-NH_2	82	150–52	$C_{17}H_{18}N_6OS$	354.42	Acetone	57.61; 57.59	5.12; 5.10	23.71; 23.70

TABLE IV-S. Physical and analytical properties of compounds **6a–k**



Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
6a	–	78	184–86	$C_{17}H_{15}N_5S$	321.39	Ethanol	63.53; 63.51	4.70; 4.68	21.79; 21.76
6b	4-CH_3	74	192–94	$C_{18}H_{17}N_5S$	335.42	Ethanol	64.45; 64.43	5.11; 5.10	20.88; 20.86
6c	4-CH_2CH_3	72	198–00	$C_{19}H_{19}N_5S$	349.45	Ethanol	65.30; 65.27	5.48; 4.47	20.04; 20.02
6d	4-$CH_2CH_2CH_3$	70	196–98	$C_{20}H_{21}N_5S$	363.46	Ethanol	66.09; 66.06	5.82; 5.79	19.27; 19.25
6e	2-OH	79	197–99	$C_{17}H_{15}N_5OS$	337.39	Ethanol	60.52; 60.50	4.48; 4.46	20.76; 20.74
6f	4-OH	70	181–83	$C_{17}H_{15}N_5OS$	337.39	Ethanol	60.52; 60.50	4.48; 4.45	20.76; 20.73
6g	2-OCH_3	65	165–67	$C_{18}H_{17}N_5OS$	351.42	Ethanol–water	61.52; 61.51	4.88; 4.85	19.93; 19.90

TABLE IV-S. Continued

Compd.	X	Yield %	M.p. °C	Formula	MW g mol^{-1}	Recrystalization solvent	Calcd.; Found, % C	H	N
6h	4-OCH_3	67	178–80	$C_{18}H_{17}N_5OS$	351.42	Ethanol–water	61.52; 61.50	4.88; 4.85	19.93; 19.90
6i	2-Cl	77	181–83	$C_{17}H_{14}N_5SCl$	355.84	Acetone–water	57.38; 57.36	3.97; 3.95	19.68; 19.65
6j	4-Cl	82	177–79	$C_{17}H_{14}N_5SCl$	355.84	Acetone–water	57.38; 57.36	3.97; 3.95	19.68; 19.65
6k	4-NH_2	75	148–50	$C_{17}H_{18}N_6S$	336.41	Ethanol	60.69; 60.67	4.79; 4.77	24.98; 24.96

SPECTRAL DATA OF THE SYNTHESIZED COMPOUNDS

*2-(2-Methyl-1*H*-benzimidazol-1-yl)-N'-(phenylmethylidene)acetohydrazide* ***(4a)***. IR (KBr, cm^{-1}): 1659 (–C=O amide). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.60 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.77 (*s*, 2H, $–N–CH_2$), 6.39-7.61 (*m*, 9H, *J* = 4.2 Hz, 5.5 Hz, Ar–H), 9.08 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.35 ($–CH_3$ at C-2 of benzimidazole), 50.80 ($–CH_2$), 175.42 (amide carbon), 145.50 (–N=CH), 135.19, 130.52, 111.31, 107.79, 122.24, 122.10, 137.43, 130.46, 114.35, 146.52, 125.47 (aromatic carbons). MS (*m*/*z*): 292 (M^+), 277, 266, 264, 250.

*2-(2-Methyl-1*H*-benzimidazol-1-yl)-*N*'-[(*E*)-(2-methylphenyl)methylidene]-acetohydrazide* ***(4b)***. IR (KBr, cm^{-1}): (–C=O amide). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.32 (*s*, 3H, $–CH_3$), 2.51 (*s*, 3H, $–CH_3$ at C-2 of Bz), 4.62 (*s*, 2H, $–N–CH_2$), 7.0 (*s*, 1H, –CONH), 7.22-7.79 (*m*, 8H, *J* = 4.5, 5.1, 3.9 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 39.7 ($–CH_2$), 171.0 (amide carbon), 147.7 (–N=CH), 16.7 ($–CH_3$), 149.3, 142.2, 134.2, 137.5, 131.0, 128.8. 128.2, 123.0, 119.8, 110.0 (aromatic carbons). MS (*m*/*z*): 306 (M^+).

*N'-[(*E*)-(2-Ethylphenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)ace-tohydrazide* ***(4c)***. IR (KBr, cm^{-1}): 1665 (–C=O amide). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 1.25 (*t*, 3H, *J* = 3.5, 1.2 Hz, $–CH_2CH_3$), 2.51 (*s*, 3H, $–CH_3$ at C-2 Bz), 2.60 (*q*, 2H, *J* = 4.7 and 1.2 Hz, $–CH_2CH_3$), 4.62 (*s*,2H, $–N–CH_2$), 7.0 (*s*, 1H, –CONH), 7.22–7.72 (*m*, 8H, *J* = 6.2, 4.9 Hz, Ar-H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 39.7 ($–CH_2$), 171.0 (amide carbon), 168.8 (–N=C), 17.0 ($–CH_3$), 27.9 ($–CH_2$), 14.5 ($–CH_3$), 149.3, 142.2, 134.2, 130.9, 128.3, 124.4, 126.0, 123.0, 119.8, 110.0 (aromatic carbons). MS (*m*/*z*): 320 (M^+).

*2-(2-Methyl-1*H*-benzimidazol-1-yl)-*N*'-[(*E*)-(2-propylphenyl)methylidene]-acetohydrazide* ***(4d)***. IR (KBr, cm^{-1}): 1670 (–C=O amide); ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.32 (*s*, 3H, $–CH_3$), 2.51 (*s*, 3H, $–CH_3$ at C-2 of Bz), 4.62 (*s*, 2H, $–N–CH_2$), 7.0 (*s*, 1H, –CONH), 7.22–7.52 (*m*, 8H, *J* = 6.2 and 4.7 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 ($–CH_3$ at C-2 of benzimida-

zole), 39.7 (–CH_2), 171.0 (amide carbon), 147.7 (–N=CH), 16.7 (–CH_3), 149.3, 142.2, 134.2, 137.5, 131.0, 128.8. 128.2, 123.0, 119.8, 110.0 (aromatic carbons). MS (*m*/*z*):334 (M^+).

N'-*[(2-Hydroxyphenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4e**)*. IR (KBr, cm^{-1}): 1672 (–C=O amide), 907 (Ar–OH) cm^{-1}. ^{1}H--NMR (300 MHz, $CDCl_3$, *δ* / ppm): δ 2.61 (*s*, 3H, –CH_3 at C-2 Bz), 3.81 (*s*, 2H, –N–CH_2), 6.53–7.68 (*m*, 8H, *J* = 6.2 and 4.7 Hz, Ar–H), 9.12 (*s*, 1H, –CONH), 12.25 (*s*, 1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 12.31 (–CH_3 at C-2 of benzimidazole), 50.82 (–CH_2), 175.73 (amide carbon), 147.79 (–N=CH), 135.19, 130.52, 111.31, 107.79, 122.24, 122.10, 137.43, 131.23, 115.32, 141.65, 124.23 (aromatic carbons). MS (*m*/*z*): 308 (M^+), 293, 282, 280, 266.

N'-*[(4-Hydroxyphenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1yl)acetohydrazide (**4f**)*. IR (KBr, cm^{-1}): 1677 (–C=O amide), 890 (Ar–OH). ^{1}H-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 2.65 (*s*, 3H, –CH_3 at C-2 Bz), 3.75 (*s*, 2H, –N–CH_2), 6.48–8.10 (*m*, 8H, *J* = 6.2 and 4.7 Hz, Ar–H), 9.10 (*s*, 1H, –CONH), 9.68 (*s*, 1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 12.30 (–CH_3 at C-2 of benzimidazole), 50.83 (–CH_2), 172.60 (amide carbon), 145.87 (–N=CH), 141.90, 135.98, 118.45, 102.45, 123.14, 123.30, 146.41, 160.65, 131.88, 128.56, 117.97, 116.23 (aromatic carbons). MS (*m*/*z*): 308 (M^+), 293, 282, 280, 266.

N'-*[(2-Methoxyphenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4g**)*. IR (KBr, cm^{-1}): 1679 (–C=O amide), 2810 (O–CH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, *δ* / ppm): δ 2.60 (s, 3H, –CH_3 at C-2 Bz), 3.78 (s, 2H, –N–CH_2), 4.80 (*s*, 3H, O–CH_3), 6.49–7.65 (*m*, 8H, *J* = 7.8 and 5.1 Hz, Ar–H), 9.08 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 12.34 (–CH_3 at C-2 of benzimidazole), 50.81 (–CH_2), 175.65 (amide carbon), 145.83 (–N=CH), 57.32 (O–CH_3), 141.90, 135.98, 118.45, 102.45, 123.14, 123.30, 146.41, 160.85, 131.45, 128.99, 117.34, 116.55 (aromatic carbons). MS (*m*/*z*): 322 (M^+), 307, 296, 294, 291.

N'-*[(4-Methoxyphenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4h**)*. IR (KBr, cm^{-1}): 1660 (–C=O amide), 2808 (O–CH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, *δ* / ppm): δ 2.59 (*s*, 3H, –CH_3 at C-2 Bz), 3.69 (*s*, 3H, O–CH_3), 3.72 (*s*, 2H, –N–CH_2), 5.52–7.70 (*m*, 8H, *J* = 7.8, 5.2 Hz, Ar–H), 9.00 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 12.35 (–CH_3 at C-2 of benzimidazole), 50.79 (–CH_2), 174.11 (amide carbon), 145.85 (–N=CH), 56.68 (O–CH_3), 143.10, 136.38, 116.15, 103.35, 124.14, 124.90, 144.81, 162.35, 132.15, 128.29, 113.14, 116.65 (aromatic carbons). MS (*m*/*z*): 322 (M^+), 307, 296, 294, 291.

N'-*[(2-Chlorophenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4i**)*. IR (KBr, cm^{-1}): 1668 (–C=O amide), 709 (C–Cl) cm^{-1}. ^{1}H-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 2.63 (*s*, 3H, –CH_3 at C-2 Bz), 3.70 (*s*, 2H, –N–CH_2), 6.34–7.51 (*m*, 8H, *J* = 8.0 and 7.3 Hz, Ar–H), 9.27 (*s*, 1H, -CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, *δ* / ppm): 12.36 (–CH_3 at C-2 of benzimidazole), 50.81 (–CH_2),

175.65 (amide carbon), 145.76 (–N=CH), 142.46, 132.50, 118.13, 108.12, 124.21, 123.78, 146.98, 118.24, 118.11, 161.72, 126.22, 121.56, 126.23, 121.77 (aromatic carbons). MS (*m*/*z*): 326 (M^+), 311, 300, 298, 290, 284.

N'-*[(4-Chlorophenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide* (***4j***). IR (KBr, cm^{-1}): 1674 (–C=O amide), 745 (C–Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.62 (*s*, 3H, $–CH_3$ at C-2 Bz), 3.74 (*s*, 2H, $–N–CH_2$), 6.53–7.68 (*m*, 8H, *J* = 8.0 and 7.3 Hz, Ar–H), 9.18 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.30 ($–CH_3$ at C-2 of benzimidazole), 50.82 ($–CH_2$), 175.62 (amide carbon), 145.12 (–N=CH), 142.22, 132.12, 117.87, 107.64, 124.23, 123.87, 146.56, 118.34, 118.25, 161.82, 126.44, 121.57, 126.55, 121.29 (aromatic carbons). MS (*m*/*z*): 326 (M^+), 311, 300, 298, 290, 284.

N'-*[(4-Aminophenyl)methylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide* (***4k***). IR (KBr, cm^{-1}): 1672 (–C=O amide), 3348 ($–NH_2$). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.50 (*s*, 3H, $–CH_3$ at C-2 Bz), 3.69 (*s*, 2H, $–N–CH_2$), 5.58 ($–C–NH_2$), 6.55 (*s*, 1H, –CONH), 6.50–7.88 (*m*, 8H, *J* = 5.4, 3.7 Hz, Ar–H). ^{13}C--NMR (300 MHz, $CDCl_3$, δ / ppm): 12.30 ($–CH_3$ at C-2 of benzimidazole), 50.82 ($–CH_2$), 175.62 (amide carbon), 145.71 (–N=CH), 14.35 ($–CH_3$), 135.19, 130.52, 111.31, 107.79, 122.24, 122.10, 137.43, 130.09, 114.12, 146.32, 125.37 (aromatic carbons); MS (*m*/*z*): 307 (M^+), 292, 281, 279, 263, 256.

*2-(2-Methyl-1*H*-benzimidazol-1-yl)*-N'-*[1-phenylethylidene]acetohydrazide* (***4l***). IR (KBr, cm^{-1}): 1685 (–C=O amide). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.17 (*s*, 3H, $–CH_3$), 2.43 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.73 (*s*, 2H, $–N–CH_2$), 7.10––7.80 (*m*, 9H, *J* = 6.4 and 6.9 Hz, Ar–H), 9.04 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.64 ($–CH_3$ at C-2 of benzimidazole), 51.07 ($–CH_2$), 176.88 (amide carbon), 148.96 (–N=CH), 15.14 ($–CH_3$), 140.29, 135.52, 118.97, 108.85, 123.24, 123.44, 145.41, 136.97, 127.69, 127.79, 125.23, 127.69 (aromatic carbons); MS (*m*/*z*). 306 (M^+), 291, 280, 278, 264, 262, 230.

*2-(2-Methyl-1*H*-benzimidazol-1-yl)*-N'-*[(1*E*)-1-(2-methylphenyl)ethylidene]-acetohydrazide* (***4m***). IR (KBr, cm^{-1}): 1689 (–C=O amide); ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 1.81 (*s*, 3H, $–CH_3$), 2.48 (*s*, 3H, $–CH_3$ at benzene ring), 2.54 (*s*, 3H, $–CH_3$ at C-2 of Bz), 4.65 (*s*, 2H, $–N–CH_2$), 7.00 (*s*, 1H, –CONH), 7.28–7.95 (*m*, 9H, *J* = 6.9, 6.3 and 6.3 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 39.7 ($–CH_2$), 171.0 (amide carbon), 168.8 (–N=C), 19.2 ($–CH_3$), 149.3, 142.2, 135.1, 134.2, 130.9, 129.6, 128.0, 125.8, 123.0, 119.8, 110.0 (aromatic carbons). MS (*m*/*z*): 320 (M^+).

N'-*[(1*E*)-1-(2-Ethylphenyl)ethylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)-acetohydrazide* (***4n***). IR (KBr, cm^{-1}): 1687 (–C=O amide); ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.02 (*q*, 2H, *J* = 4.7, 3.2, 0.9 Hz, CH_2CH_3), 2.48 (*s*, 3H, $–CH_3$ at benzene ring), 2.54 (*s*, 3H, $–CH_3$ at C-2 of Bz), 2.95 (*t*, 3H, *J* = 4.5 and 3.9 Hz, $–CH_2CH_3$), 4.65 (*s*, 2H, $–N–CH_2$), 7.02 (*s*, 1H, –CONH), 7.28–7.95 (*m*, 9H, *J* = 6.7 and 3.8 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4

(–CH_3 at C-2 of benzimidazole), 39.7 (–CH_2), 171.0 (amide carbon), 168.8 (–N=C), 19.2 (–CH_3), 149.3, 142.2, 135.1, 134.2, 130.9, 129.6, 128.0, 124.9, 123.2, 119.7, 110.3 (aromatic carbons). MS (*m*/*z*): 334 (M^+).

*2-(2-Methyl-1*H*-benzimidazol-1-yl)-*N*'-[(1*E*)-1-(2-propylphenyl)ethylidene]-acetohydrazide (**4o**)*. IR (KBr, cm^{-1}): 1686 (–C=O amide). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 0.90 (*t*, 3H, *J* = 6.5 Hz, –$CH_2CH_2CH_3$), 1.65 (*m*, 2H, *J* = = 6.8 and 7.5 Hz, –$CH_2CH_2CH_3$), 2.48 (*s*, 3H, –CH_3), 2.62 (*m*, 2H, *J* = 6.5 and 1.2 Hz, –$CH_2CH_2CH_3$), 2.54 (*s*, 3H, –CH_3 at C-2 of Bz), 4.65 (*s*, 2H, N–CH_2), 7.02 (*s*, 1H, CONH), 7.28–7.95(*m*, 9H, *J* = 6.7 and 3.8 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 (–CH_3 at C-2 of benzimidazole), 39.7 (–CH_2), 171.0 (amide carbon), 168.8 (–N=C), 19.2 (–CH_3), 148.2, 142.0, 134.1, 134.2, 130.9, 127.6, 128.4, 124.5, 123.0, 119.1, 109.5 (aromatic carbons). MS (*m*/*z*): 348 (M^+).

N'-*[1-(2-Hydroxyphenyl)ethylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4p**)*. IR (KBr, cm^{-1}): 1682 (–C=O amide), 982 (Ar–OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.21 (*s*, 3H, –CH_3), 2.40 (*s*, 3H, CH_3 at C-2 of Bz), 3.71 (*s*, 2H, NCH_2), 6.67–7.98 (*m*, 8H, *J* = 8.1 and 7.5 Hz, Ar–H), 9.12 (*s*, 1H, CONH), 12.03 (*s*, 1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.68 (–CH_3 at C-2 of benzimidazole), 51.05 (–CH_2), 176.73 (amide carbon), 152.69 (–N=CH), 15.72 (–CH_3), 140.24, 135.50, 118.91, 108.79, 123.14, 123.30, 146.41, 118.45, 118.89, 132.52, 159.13, 119.79, 127.33 (aromatic carbons). MS (*m*/*z*): 322 (M^+), 307, 296, 294, 278, 271.

N'-*[1-(4-Hydroxyphenyl)ethylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4q**)*. IR (KBr, cm^{-1}): 1687 (–C=O amide), 982 (Ar-OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.18 (*s*, 3H, –CH_3), 2.41 (*s*, 3H, –CH_3 at C-2 of Bz), 3.75 (*s*, 2H, –N–CH_2), 6.86–8.00 (*m*, 8H, *J* = 8.1 and 7.2 Hz, Ar–H), 9.10 (*s*, 1H, CONH), 9.49 (*s*, 1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.62 (–CH_3 at C-2 of benzimidazole), 51.03 (–CH_2), 176.76 (amide carbon), 150.49 (–N=CH), 15.21 (–CH_3), 140.24, 135.50, 118.91, 108.79, 123.14, 123.30, 146.41, 160.35, 130.88, 127.46, 118.97, 117.34 (aromatic carbons). MS (*m*/*z*): 322 (M^+), 307, 296, 294, 280, 278, 271.

N'-*[1-(2-Methoxyphenyl)ethylidene]-2-(2-methyl-1*H*-benzimidazol-1-yl)acetohydrazide (**4r**)*. IR (KBr, cm^{-1}): 1689 (–C=O amide), 2815 (O–CH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.19 (*s*, 3H, –CH_3), 2.45 (*s*, 3H, –CH_3 at C-2 of Bz), 3.75 (*s*, 2H, –N–CH_2), 3.89 (*s*, 3H, O–CH_3), 6.58–7.78 (*m*, 8H, *J* = 7.8 and 6.2 Hz, Ar–H), 9.08 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.60 (–CH_3 at C-2 of benzimidazole), 51.09 (–CH_2), 176.49 (amide carbon), 147.73 (–N=CH), 15.94 (–CH_3), 56.16 (O–CH_3), 140.29, 135.52, 118.97, 108.85, 123.24, 123.44, 146.41, 118.19, 118.88, 132.87, 159.76, 121.08, 129.24 (aromatic carbons). MS (*m*/*z*): 336 (M^+), 321, 310, 308.

N'-*[1-(4-Methoxyphenyl)ethylidene]-2-(2-methyl-1*H-*benzimidazol-1-yl)acetohydrazide (**4s**)*. IR (KBr, cm^{-1}): 1680 (–C=O amide), 2811 (O–CH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.12 (*s*, 3H, –CH_3), 2.48 (*s*, 3H, CH_3 at C-2 Bz), 3.78 (*s*, 2H, –NCH_2), 3.84 (*s*, 3H, OCH_3), 6.61–7.88 (*m*, 8H, *J* = 7.8 and 6.2 Hz, Ar–H), 9.00 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.62 (–CH_3 at C-2 of benzimidazole), 51.11 (–CH_2), 176.40 (amide carbon), 147.78 (–N=CH), 15.91 (–CH_3), 55.33 (O–CH_3), 140.29, 135.52, 118.97, 108.85, 123.24, 123.44, 146.41, 118.19, 118.88, 160.75, 126.97, 121.08, 126.97, 121.08 (aromatic carbons). MS (*m*/*z*): 336 (M^+), 321, 310, 308, 294, 260.

N'-*[1-(2-Chlorophenyl)ethylidene]-2-(2-methyl-1*H-*benzimidazol-1-yl)acetohydrazide (**4t**)*. IR (KBr, cm^{-1}): 1668 (–C=O amide), 775 (C–Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.10 (*s*, 3H, –CH_3), 2.37 (*s*, 3H, –CH_3 at C-2 of Bz), 3.61 (*s*, 2H, –N–CH_2), 6.54–7.63 (*m*, 8H, *J* = 7.1 and 5.5 Hz, Ar–H), 8.97 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.59 (–CH_3 at C-2 of benzimidazole), 51.08 (–CH_2), 176.38 (amide carbon), 147.75 (–N=CH), 15.85 (–CH_3), 140.26, 135.50, 118.93, 108.82, 123.21, 123.42, 146.38, 118.14, 118.85, 160.72, 126.97, 121.06, 126.94, 121.07 (aromatic carbons). MS (*m*/*z*): 340 (M^+), 325, 314, 312, 304, 296.

N'-*[-1-(4-Chlorophenyl)ethylidene]-2-(2-methyl-1*H-*benzimidazol-1-yl)acetohydrazide (**4u**)*. IR (KBr, cm^{-1}): 1674 (–C=O amide), 780 (C–Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.15 (*s*, 3H, –CH_3), 2.39 (*s*, 3H, –CH_3 at C-2 of Bz), 3.64 (*s*, 2H, –N–CH_2), 6.66–7.79 (*m*, 8H, *J* = 7.1, 5.4 Hz, Ar–H), 9.00 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.52 (–CH_3 at C-2 of benzimidazole), 51.04 (–CH_2), 176.36 (amide carbon), 147.71 (–N=CH), 15.80 (–CH_3), 140.23, 135.45, 118.89, 108.80, 123.15, 123.34, 146.36, 118.10, 118.81, 160.71, 126.94, 121.02, 126.90, 121.03 (aromatic carbons). MS (*m*/*z*): 340 (M^+), 325, 314, 312, 304, 296.

N'-*[1-(4-Aminophenyl)ethylidene]-2-(2-methyl-1*H-*benzimidazol-1-yl)acetohydrazide (**4v**)*. IR (KBr, cm^{-1}): 1672 (–C=O amide), 3348 (–NH_2). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.11 (*s*, 3H, –CH_3), 2.50 (*s*, 3H, –CH_3 at C-2 of Bz), 3.69 (*s*, 2H, –N–CH_2), 5.58 (*s*, 2H, C–NH_2), 6.55 (*s*, 1H, –CONH), 6.50–7.88 (*m*, 8H, *J* = 6.2 and 4.4 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.64 (–CH_3 at C-2 of benzimidazole), 51.07 (–CH_2), 176.88 (amide carbon), 146.84 (–N=CH), 14.97 (–CH_3), 55.26 (O–CH_3), 140.29, 135.52, 118.97, 108.85, 123.24, 123.44, 146.41, 131.13, 115.32, 147.93, 126.66 (aromatic carbons). MS (*m*/*z*): 321 (M^+), 306, 295, 293, 279, 245.

*2-[(2-Methyl-1*H-*benzimidazol-1-yl)acetyl]-*N-*phenylhydrazinecarbothioamide (**5a**)*. IR (KBr, cm^{-1}): 1650 (–C=O amide), 1220 (–C=S). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.38 (*s*, 3H, –CH_3 at C-2 of Bz), 3.68 (*s*, 2H, –N–CH_2), 7.08–7.68 (*m*, 9H, *J* = 6.6 and 6.3 Hz, Ar–H), 7.42 (*s*, 1H, CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.83 (–CH_3 at C-2 of benzimidazole), 50.16 (–CH_2),

171.43 (amide carbon), 178.04 (–C=S), 142.98, 137.27, 118.97, 108.26, 123.24, 123.44, 148.16, 13.00, 125.80, 128.25, 125.85 (aromatic carbons). MS (*m*/*z*): 339 [M^+], 324, 313, 311, 297, 263.

*2-[(2-Methyl-1*H*-benzimidazol-1-yl)acetyl]*-N-*(2-methylphenyl)hydrazine-carbothioamide (**5b**)*. IR (KBr, cm^{-1}): 1658 (–C=O amide), 1225 (–C=S). ^{1}H--NMR (300 MHz, $CDCl_3$, δ / ppm): 1.25 (*t*, 3H, *J* = 3.6 Hz, $–CH_3$), 2.00 (*s*, 1H, –NH), 2.51 (*s*, 3H, $–CH_3$ at C-2 of Bz), 2.60 (*q*, 2H, *J* = 3.3 Hz, $–CH_2$), 4.62 (*s*, 2H, $–N–CH_2$), 6.02–7.59 (*m*, 8H, *J* = 6.3, 6.6 Hz, Ar–H), 8.20 (*s*, 1H, –CONH), 10.08 (*s*, 1H, –NH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 40.0 ($–CH_2$), 164.4 (amide carbon), 181.1 (–C=S), 17.9 ($–CH_3$ at benzene) 142.20, 136.217, 135.02, 134.12, 130.24, 129.24, 127.16, 126.12, 125.80, 119.8, 110.02 (aromatic carbons). MS (*m*/*z*): 353 [M^+].

N-*(2-Ethylphenyl)-2-[(2-methyl-1*H*-benzimidazol-1-yl)acetyl]hydrazinecar-bothioamide (**5c**)*. IR (KBr, cm^{-1}): 1654 (–C=O amide), 1222 (–C=S). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 1.25 (*t*, 3H, *J* = 3.9 Hz, $–CH_3$), 2.51 (*s*, 3H, $–CH_3$ at C-2 of Bz), 2.60 (*q*, 2H, *J* = 3.2 Hz, $–CH_2$), 4.62 (*s*, 2H, $–N–CH_2$), 6.04–7.65 (*m*, 8H, *J* = 6.3, 6.5 Hz, ArH), 8.0 (s, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 40.0 ($–CH_2$), 164.4 (amide carbon), 181.1 (–C=S), 17.9, 14.4 ($–CH_2CH_3$ at benzene)142.20, 136.217, 135.02, 134.12, 130.24, 129.24, 127.16, 126.12, 125.80, 119.8, 110.02 (aromatic carbons). MS (*m*/*z*): 367 [M^+].

*2-[(2-Methyl-1*H*-benzimidazol-1-yl)acetyl]*-N-*(2-propylphenyl)hydrazine-carbothioamide (**5d**)*. IR (KBr, cm^{-1}): 1654 (–C=O amide), 1222 (–C=S). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 0.91 (*t*, 3H, *J* = 4.5 Hz, $–CH_2CH_2CH_3$), 1.62 (*m*, 2H, *J* = 6.5, 1.2 Hz, $–CH_2CH_2CH_3$), 2.51 (*s*, 3H, $–CH_3$ at C-2 of Bz), 2.60 (*t*, 2H, *J* = 6.5, 1.5 Hz, $–CH_2CH_2CH_3$), 4.62 (*s*, 2H, $–N–CH_2$), 6.04–7.65 (*m*, 8H, *J* = = 8.1 and 4.5 Hz, ArH), 8.0 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / / ppm): 14.4 ($–CH_3$ at C-2 of benzimidazole), 40.0 ($–CH_2$), 164.4 (amide carbon), 181.1 (–C=S), 17.9, 14.4, 10.6 ($–CH_2CH_2CH_3$ at benzene) 142.20, 136.217, 135.02, 134.12, 130.24, 129.24, 127.16, 126.12, 125.80, 119.8, 110.02 (aromatic carbons). MS (*m*/*z*): 381 [M^+].

N-*(2-Hydroxyphenyl)-2-[(2-methyl-1*H*-benzimidazol-1-yl)acetyl]hydrazine-carbothioamide (**5e**)*. IR (KBr, cm^{-1}): 1654 (–C=O amide), 1225 (–C=S) 990 (Ar–OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.35 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.67 (*s*, 2H, $–N–CH_2$), 7.42–7.85 (*m*, 8H, *J* = 7.8 and 4.5 Hz, Ar–H), 7.47 (*s*, 1H, –CONH), 12.03 (*s*,1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.64 ($–CH_3$ at C-2 of benzimidazole), 50.25 ($–CH_2$), 171.54 (amide carbon), 178.18 (–C=S), 143.12, 137.45, 119.23, 108.41, 123.24, 123.44, 148.15, 13.25, 126.14, 128.47, 125.91 (aromatic carbons). MS (*m*/*z*): 355 [M^+], 340, 329, 327, 313, 311, 304.

N-*(4-Hydroxyphenyl)-2-[(2-methyl-1*H-*benzimidazol-1-yl)acetyl]hydrazine-carbothioamide (**5f**)*. IR (KBr, cm^{-1}): 1649 (–C=O amide), 1220 (–C=S) 1022 (Ar–OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.31 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.62 (*s*, 2H, $–N–CH_2$), 7.40–7.81 (*m*, 8H, *J* = 7.8 and 4.5 Hz, Ar–H), 7.44 (*s*, 1H, –CONH), 9.48 (*s*, 1H, Ar–OH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.60 ($–CH_3$ at C-2 of benzimidazole), 50.14 ($–CH_2$), 171.54 (amide carbon), 178.18 (–C=S), 143.12, 137.45, 119.23, 108.41, 123.24, 123.44, 148.27, 13.32, 126.29, 128.54, 125.98 (aromatic carbons). MS (*m*/*z*): 355 [M^+], 340, 329, 327, 313, 311, 304.

N-*(2-Methoxyphenyl)-2-[(2-methyl-1*H-*benzimidazol-1-yl)acetyl]hydrazine-carbothioamide (**5g**)*:IR (KBr, cm^{-1}): 1653 (–C=O amide), 1232 (–C=S) 2829 ($O–CH_3$). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.35 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.69 (*s*, 2H, $–N–CH_2$), 7.49 (*s*, 1H, –CONH), 7.54–8.35 (*m*, 8H, *J* = 7.7 and 5.0 Hz, Ar–H), 9.48 (*s*, 3H, $O-CH_3$). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.88 ($–CH_3$ at C-2 of benzimidazole), 50.18 ($–CH_2$), 171.45 (amide carbon), 178.14 (–C=S), 55.53 ($O–CH_3$), 143.21, 137.49, 119.28, 108.47, 123.26, 123.25, 148.45, 127.87, 126.08, 123.54, 111.44, 123.27, 151.67 (aromatic carbons). MS (*m*/*z*): 369 [M^+], 354, 343, 338, 326.

N-*(4-Methoxyphenyl)-2-[(2-methyl-1*H-*benzimidazol-1-yl)acetyl]hydrazine-carbothioamide (**5h**)*. IR (KBr, cm^{-1}): 1653 (–C=O amide), 1232 (–C=S) 2835 ($O-CH_3$). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.31 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.65 (*s*, 2H, $–N–CH_2$), 3.80 (*s*, 3H, $O–CH_3$), 7.30–8.25 (*m*, 8H, *J* = 7.7 and 5.1 Hz, Ar–H), 7.44 (s, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.82 ($–CH_3$ at C-2 of benzimidazole), 50.13 ($–CH_2$), 171.41 (amide carbon), 178.28 (–C=S), 55.50 ($O–CH_3$), 143.15, 137.32, 119.21, 108.44, 123.21, 123.24, 148.43, 132.58, 158.38, 113.84, 125.49 (aromatic carbons). MS (*m*/*z*): 369 [M^+], 354, 343, 338, 326.

N-*(2-Chlorophenyl)-2-[(2-methyl-1*H-*benzimidazol-1-yl)acetyl]hydrazinecar-bothioamide (**5i**)*. IR (KBr, cm^{-1}): 1665 (–C=O amide), 1245 (–C=S) 775 (C–Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.37 (*s*, 3H, $–CH_3$ at C-2 of benzimida-zole), 3.69 (*s*, 2H, $–N–CH_2$), 7.48 (*s*, 1H, –CONH), 7.11–8.28 (*m*, 8H, Ar–H) 2.37 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.69 (*s*, 2H, $–N–CH_2$), 7.48 (*s*, 1H, –CONH), 7.11–8.28 (*m*, 8H, *J* = 8.5 and 5.9 Hz, Ar–H). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / / ppm): 12.96 ($–CH_3$ at C-2 of benzimidazole), 50.24 ($–CH_2$), 171.55 (amide carbon), 178.36 (–C=S), 143.11, 137.25, 119.24, 108.40, 123.20, 123.21, 148.47, 137.14, 130.19, 128.93, 124.48 (aromatic carbons). MS (*m*/*z*): 373 [M^+], 358, 347, 345, 331.

N-*(4-Chlorophenyl)-2-[(2-methyl-1*H-*benzimidazol-1-yl)acetyl]hydrazinecar-bothioamide (**5j**)*. IR (KBr, cm^{-1}): 1660 (–C=O amide), 1240 (–C=S) 782 (C–Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.34 (*s*, 3H, $–CH_3$ at C-2 of Bz), 3.62 (*s*, 2H, $–N–CH_2$), 7.04–8.12 (*m*, 8H, *J* = 8.5 and 5.8 Hz, Ar–H), 7.42 (*s*, 1H,

–CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.96 (–CH_3 at C-2 of benzimidazole), 50.24 (–CH_2), 171.55 (amide carbon), 178.36 (–C=S), 143.02, 137.27, 119.20, 108.47, 123.19, 123.32, 148.48, 137.25, 130.14, 128.18, 124.58 (aromatic carbons). MS (*m*/*z*): 373 [M^+], 358, 347, 345, 331.

N-*(4-Aminophenyl)-2-[(2-methyl-1*H*-benzimidazol-1-yl)acetyl]hydrazinecarbothioamide (**5k**)*. IR (KBr, cm^{-1}): 1658 (–C=O amide), 1232 (–C=S), 3340 (–C–NH_2). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.45 (*s*, 3H, –CH_3 at C-2 of benzimidazole), 3.54 (*s*, 2H, –N–CH_2), 8.31 (*s*, 1H, –CONH), 6.42 (*s*, 2H, –C–NH_2) 7.40–8.59 (*m*, 8H, Ar–H) 2.45 (*s*, 3H, –CH_3 at C-2 of Bz), 3.54 (*s*, 2H, –N–CH_2), 6.42 (*s*, 2H, –C–NH_2), 7.40–8.59 (*m*, 8H, *J* = 6.2, 4.8 Hz, Ar–H), 8.31 (*s*, 1H, –CONH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 12.90 (–CH_3 at C-2 of benzimidazole), 50.11 (–CH_2), 171.53 (amide carbon), 178.41 (–C=S), 143.45, 137.40, 119.27, 108.60, 123.21, 123.44, 132.56, 145.96, 113.77, 125.09 (aromatic carbons). MS (*m*/*z*): 354 [M^+], 339, 328, 326, 312, 310, 278.

*5-[(2-Methyl-1*H*-benzimidazol-1-yl)methyl]-4-phenyl-4*H*-1,2,4-triazole-3--thiol (**6a**)*. IR (KBr, cm^{-1}): 2550 (SH).^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.56 (s, 3H, -CH_3 at C-2 of Bz), 6.00 (*s*, 2H, –N–CH_2), 6.61–7.57 (*m*, 9H, *J* = 7.5 and 6.6 Hz, Ar–H), 12.90 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / / ppm): 13.12 (–CH_3 at C-2 of benzimidazole), 41.73 (–CH_2), 141.84, 133.22, 119.55, 106.84, 122.30, 126.56, 147.72, 161.16, 172.51, 146.11, 127.08, 129.35, 132.60, 127.08, 129.35 (aromatic carbons). MS (*m*/*z*): 321 [M^+], 306, 295, 245, 270.

*5-[(2-Methyl-1H-benzimidazol-1-yl)methyl]-4-(2-methylphenyl)-4H-1,2,4--triazole-3-thiol (**6b**)*. IR (KBr, cm^{-1}): 2538 (SH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / / ppm): 2.25 (*s*, 3H, –CH_3), 2.51 (*s*, 3H, –CH_3 at C-2 of Bz), 4.99 (*s*, 2H, –N–CH_2), 7.22–7.68 (*m*, 8H, *J* = 7.2, 6.6 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.7 (–CH_3 at C-2 of benzimidazole), 49.1 (–CH_2), 149.2, 142.2, 164.2, 128.3, 119.8, 110.0, 131.9, 129.5, 128.7, 123.0, 125.9, (aromatic carbons). MS (*m*/*z*): 321 [M^+], 335.

*4-2-Ethylphenyl-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4-triazole-3-thiol (**6c**)*. IR (KBr, cm^{-1}): 2536 (SH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / / ppm): 1.25 (*t*, 3H, *J* = 6.2 Hz, –CH_2CH_3), 2.51 (*s*, 3H, –CH_3 at C-2 of Bz), 2.60 (*q*, 2H, *J* = 6.5 and 4.2 Hz, –CH_2CH_3),4.99 (*s*, 2H, –N–CH_2), 7.22–7.62 (*m*, 8H, *J* = 7.2 and 4.9 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.7 (–CH_3 at C-2 of benzimidazole), 49.1 (–CH_2), 29.1 and 14.6 (–CH_2CH_3) 168.9, 149.3, 145.7, 142.2, 134.2, 128.3, 119.8, 110.0, 131.9, 129.5, 128.7, 123.0, 125.9, (aromatic carbons). MS (*m*/*z*): 321 [M^+], 349.

*5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4-(2-propylphenyl)-4H-1,2,4-triazole-3-thiol (**6d**)*. IR (KBr, cm^{-1}): 2538 (SH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / / ppm): 0.90 (*t*, 3H, *J* = 4.5 Hz, –$CH_2CH_2CH_3$), 1.65 (*m*, 2H, *J* = 6.5, 1.8 Hz, –$CH_2CH_2CH_3$), 2.51 (*s*, 3H, –CH_3 at C-2 of Bz), 2.62 (*t*, 2H, *J* = 6.5 Hz,

–$CH_2CH_2CH_3$), 4.99 (*s*, 2H, –N–CH_2), 7.22–7.68 (*m*, 8H, *J* = 7.7, 4.9 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 14.7 (–CH_3 at C-2 of benzimidazole), 49.1 (–CH_2), 32.8, 24.1 and 13.7 (–$CH_2CH_2CH_3$) 168.9, 149.3, 145.7, 142.2, 134.2, 128.3, 119.8, 110.0, 131.9, 129.5, 128.7, 123.0, 125.9 (aromatic carbons). MS (*m*/*z*): 321 [M^+], 363.

*2-{3-[(2-Methyl-1*H*-benzimidazol-1-yl)methyl]-5-sulfanyl-4*H*-1,2,4-triazol--4-yl}phenol (**6e**)*. IR (KBr, cm^{-1}): 2548 (SH), 997 (Ar-OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.58 (*s*, 3H, –CH_3 at C-2 of Bz), 6.02 (*s*, 2H, –N–CH_2), 6.60––7.69 (*m*, 8H, *J* = 7.9 and 5.1 Hz, Ar–H), 10.01 (*s*, 1H, Ar–OH), 11.15 (*s*, 1H, –C-SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.13 (–CH_3 at C-2 of benzimidazole), 41.58 (–CH_2), 141.84, 133.22, 119.55, 106.84, 122.30, 126.56, 147.72, 160.05, 171.40, 125.16, 148.63, 116.65, 126.71, 117.31, 120.03 (aromatic carbons). MS (*m*/*z*): 337 [M^+], 322, 311, 295, 261, 286.

*4-{3-[(2-Methyl-1*H*-benzimidazol-1-yl)methyl]-5-sulfanyl-4*H*-1,2,4-triazol-4-yl}phenol (**6f**)*. IR (KBr, cm^{-1}): 2545 (SH), 982 (Ar–OH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.60 (*s*, 3H, –CH_3 at C-2 of Bz), 6.08 (*s*, 2H, –N–CH_2), 6.78––7.89 (*m*, 8H, *J* = 7.9 and 8.5 Hz, Ar–H), 9.40 (*s*, 1H, Ar–OH), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.11 (–CH_3 at C-2 of benzimidazole), 41.52 (–CH_2), 141.84, 133.22, 119.55, 106.84, 122.30, 126.56, 147.72, 160.05, 171.62, 137.13, 124.26, 115.34, 158.34, (aromatic carbons). MS (*m*/*z*): 337 [M^+], 322, 311, 295, 261, 286.

*4-(2-Methoxyphenyl)-5-[(2-methyl-1*H*-benzimidazol-1-yl)methyl]-4*H*-1,2,4--triazole-3-thiol (**6g**)*. IR (KBr, cm^{-1}): 2548 (SH), 2817 (OCH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.52 (*s*, 3H, –CH_3 at C-2 of Bz), 3.88 (*s*, 3H, –OCH_3), 6.06 (*s*, 2H, –N–CH_2), 6.59–7.70 (*m*, 8H, *J* = 9.0 and 6.1 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.10 (–CH_3 at C-2 of benzimidazole), 41.66 (–CH_2), 55.87 (OCH_3), 159.24, 170.59, 125.85, 138.40, 112.50, 129.55, 117.27, 120.86 (aromatic carbons). MS (*m*/*z*): 351 [M^+], 336, 325, 320, 300.

*4-(4-Methoxyphenyl)-5-[(2-methyl-1*H*-benzimidazol-1-yl)methyl]-4*H*-1,2,4--triazole-3-thiol (**6h**)*. IR (KBr, cm^{-1}): 2549 (SH), 2810 (OCH_3). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.50 (*s*, 3H, –CH_3 at C-2 of Bz), 3.82 (*s*, 3H, –OCH_3), 6.08 (*s*, 2H, –N–CH_2), 6.66–7.65 (*m*, 8H, *J* = 8.9 and 6.2 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.15 (–CH_3 at C-2 of benzimidazole), 41.71 (–CH_2), 55.66 (OCH_3), 161.15, 172.50, 167.96, 124.22, 114.18, 163.03, 124.22, 114.18 (aromatic carbons). MS (*m*/*z*): 351 [M^+], 336, 325, 320, 308, 375, 300.

*4-(2-chlorophenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4--triazole-3-thiol (**6i**)*. IR (KBr, cm^{-1}): 2550 (SH), 750 (C-Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.62 (*s*, 3H, –CH_3 at C-2 of Bz), 6.07 (*s*, 2H, –N–CH_2), 6.56–7.72 (*m*, 8H, *J* = 8.9 and 7.2 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR

(300 MHz, $CDCl_3$, δ / ppm): 13.18 ($–CH_3$ at C-2 of benzimidazole), 41.75 ($–CH_2$), 154.14, 172.12, 167.87, 124.11, 114.24, 163.54, 124.78, 114.77 (aromatic carbons). MS (*m*/*z*): 355 [M^+], 340, 329, 319, 279, 304.

*4-(4-chlorophenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4--triazole-3-thiol (**6j**)*. IR (KBr, cm^{-1}): 2552 (SH), 742 (C-Cl). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / ppm): 2.61 (*s*, 3H, $–CH_3$ at C-2 of Bz), 6.07 (*s*, 2H, $–N–CH_2$), 6.50–7.70 (*m*, 8H, *J* = 8.9 and 7.2 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.12 ($–CH_3$ at C-2 of benzimidazole), 41.72 ($–CH_2$), 160.71, 172.06, 142.58, 120.24, 126.97, 135.38, 120.24, 126.97 (aromatic carbons). MS (*m*/*z*): 355 [M^+], 340, 329, 319, 279, 304.

*4-(4-aminophenyl)-5-[(2-methyl-1H-benzimidazol-1-yl)methyl]-4H-1,2,4--triazole-3-thiol (**6k**)*. IR (KBr, cm^{-1}): 2560 (SH). ^{1}H-NMR (300 MHz, $CDCl_3$, δ / / ppm): 2.61 (*s*, 3H, $–CH_3$ at C-2 of Bz), 6.07 (*s*, 2H, $–N–CH_2$), 6.50–7.70 (*m*, 8H, *J* = 8.9 and 7.2 Hz, Ar–H), 11.15 (*s*, 1H, –C–SH). ^{13}C-NMR (300 MHz, $CDCl_3$, δ / ppm): 13.11 ($–CH_3$ at C-2 of benzimidazole), 41.71 ($–CH_2$), 141.84, 133.22, 119.55, 106.84, 122.30, 126.56, 147.72, 162.66, 174.01, 138.00, 105.48, 148.27, 105.48, 116.19 (aromatic carbons). MS (*m*/*z*): 336 [M^+], 321, 310, 260, 285, 309.