

Papers

Synthesis and antimicrobial activity of some new 1-substituted-3-pyrrolyl aminocarbonyl/oxadiazolyl/triazolyl/5-methoxy-2-methylindoles and benz[g]indoles

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1-Substituted-5-methoxy-3-carbethoxy-2-methylindoles **2a-c** on reaction with hydrazine hydrate in ethyl alcohol give back the starting materials **2a-c**. Then ethylene glycol is used as solvent instead of ethyl alcohol in the above reaction and obtained conveniently the desired 1-substituted-5-methoxy-3-hydrazinocarbonyl-2-methylindoles **3a-c** and they are further reacted separately with acetonyl acetone; carbon disulphide and alcoholic potassium hydroxide; followed by hydrazine hydrate; allyl isothiocyanate and phenyl isothiocyanate to obtain the desired 1-substituted-5-methoxy-3-(2,5-dimethylpyrrol-1-yl)amino carbonyl-2-ethyl-indoles **4a-c**, 1-substituted-5-methoxy-3-(5-mercapto-1,3,4-oxadiazol-2-yl) 2-methylindoles **5a-c**, 1-substituted-5-methoxy-3-(4-amino-5-mercapto-1,2,4-triazol-3-yl)-2-methylindoles **6a-c**, 1-substituted-5-methoxy-3-(*N*-allylthiosemicarbazinocarbonyl)-2-methylindoles **7a-c** and 1-substituted-5-methoxy-3-(*N*-phenylthiosemicarbazinocarbonyl)-2-methyl indoles **8a-c** respectively. The thiosemicarbazides **7a-c** and **8a-c** are heated with 4% NaOH to secure 1-substituted-3-(4-allyl-5-mercapto-1, 2,4-triazol-3-yl)-5-methoxy-2-methylindoles **9a-c** and 1-substituted-3-(4-phenyl-5-mercapto-1,2,4-triazol-3-yl)-5-methoxy-2-methylindoles **10a-c** respectively. Similar set of reactions are also carried out on 1-furfuryl-3-carbethoxy-5-methoxy-2-methylbenz[g]indole **12**. Structures of all these newly synthesised compounds are confirmed by their spectral and analytical data and the compounds are also screened for their antibacterial and antifungal activities.

Key words: Carbethoxy methylindoles, hydrazine hydrate, ethyl alcohol, ethylene glycol, ethylindoles, thiosemicarbazides, antibacterial activity, antifungal activities

IPC: Int.Cl.⁷ C 07 D

The diverse pharmacological properties¹⁻⁴ like anti-microbial, anticancer, antihypertension, anti-inflammatory associated with indole derivatives make them an important class of heterocyclic compounds. Pyrroles⁵⁻⁷, oxadiazoles⁸⁻¹⁰ and triazoles¹¹⁻¹³ also have made significant contribution as therapeutic agents. In the light of these biological activities and also continuation of our earlier work¹⁴⁻¹⁸, it was envisaged to prepare biologically potent new indole derivatives carrying pyrroles, oxadiazoles and triazoles at position-3 of 1-substituted-5-methoxy-2-methylindoles **2a-c** and also their corresponding benz[g]indole derivatives **12-17**.

When 1-furfuryl-5-methoxy-3-carbethoxy-2-methylindole **2a** was refluxed with hydrazine hydrate in ethylene glycol furnished the much desired 1-furfuryl-5-methoxy-3-hydrazino-carbonyl-2-methylindole **3a**. Similarly, other 1-substituted-5-methoxy-3-

hydrazinocarbonylindoles **3b,c** were also prepared. However, 1-substituted-5-hydroxy-3-carbethoxy-2-methylindoles wherein the C₅-OH is free, did not react with hydrazine hydrate (99%) even in boiling ethylene glycol to yield the expected 1-substituted – 5 – hydroxy-3-hydrazinocarbonylindoles under identical reaction conditions. These hydrazinocarbonylindoles **3a-c** were further reacted separately with acetonyl acetone; carbon disulfide and alcoholic potassium hydroxide; carbon disulphide, alcoholic KOH, followed by hydrazine hydrate; allyl isothiocyanate and phenyl isothiocyanate to yield the desired 1-substituted-5-methoxy-3 - (2, 5-dimethylpyrrol -1-yl)-aminocarbonyl-2-methylindoles **4a-c**, 1-substituted-5-methoxy-3-(5-mercapto-1, 3, 4 – oxadiazol – 2 - yl)-2-methylindoles **5a-c**, 1-substituted-5-methoxy-3-(4-amino-5-mercapto-1, 2, 4-triazol-3-yl)-2-methylindoles **6a-c**, 1-substituted-5-methoxy-3-(*N*-allylthio-semicarba-

nocarbonyl)-2-methyl indoles **7a-c** and 1-substituted-5-methoxy-3-(*N*-phenylthiosemicarbazino carbonyl)-2-methylindoles **8a-c**, respectively. The thiosemicarbazides **7a-c** and **8a-c** were heated with 4% NaOH to secure 1-substituted-5-methoxy-3-(4-allyl-5-mercapto-1,2,4-triazol-3-yl)-2-methylindoles **9a-c** and 1-substituted-5-methoxy-3-(4-phenyl-5-mercapto-1,2,4-triazol-3-yl)-2-methylindoles **10a-c**, respectively. Similar set of reactions were carried out on 1-furfuryl-3-carbethoxy-5-methoxy-2-methylbenz[*g*]indole **12** (Schemes I and II). The structures of all these compounds were confirmed on the basis of their spectral and analytical data (Table I).

Antimicrobial activity

All the newly synthesised compounds were screened for their antimicrobial activity *in vitro* at doses of 100 µg in 0.1 mL of DMF against the bacteria *Escherichia coli* and *Bacillus cirroflagellosus* using norfloxacin as standard and for their antifungal activity *in vitro* against the fungi *Aspergillus niger* and *Penicillium* using griseofulvin as standard. DMF was used as culture medium and the method employed was cup-plate method¹⁹. The zone of inhibition was measured in mm and was compared with standard drugs. Compounds **4**, **5**, **10a**, **13** and **16** displayed higher activity towards *E. coli* while none of the compound was highly active against *B. cirroflagellosus*. Compounds **3-5c**, **6**, **9a**, **10b** and **14** were moderately active towards *E. coli* and compounds **3c**, **4**, **5a**, **5c**, **6a**, **10a**, **10b**, and **15** showed moderate activity towards *B. cirroflagellosus*. Compounds **7a** and **12** were inactive towards *E. coli* and compounds **8b** and **11** were inactive against *B. cirroflagellosus*. Rest of the compounds were weakly active towards both the bacteria. Compounds **5a** and **7a** were highly active towards *Penicillium*, while compounds **7a** and **14** were highly active towards *A. niger*. The compounds **4b**, **6**, **9a**, **10c**, **12**, **14** and **16** were moderately active towards *Penicillium* and compounds **4a**, **4c**, **5**, **6**, **9**, **10a**, **10c**, **11**, **15** and **16** were moderately active towards *A. niger*. Compound **8b** was inactive against *Penicillium*, while the compounds **7c** and **8c** were inactive towards *A. niger*. Rest of the compounds showed weak antifungal activity.

Experimental Section

Melting points were determined in open capillary tubes and are uncorrected. IR spectra were recorded

on a Perkin-Elmer 881 spectrometer; ¹H NMR spectra in CDCl₃ or DMSO-*d*₆ on a Bruker's 300 MHz NMR spectrometer and mass spectra on a Autospec EI mass spectrometer. Elemental analysis was carried out on a Heraeus CHN repid analyser.

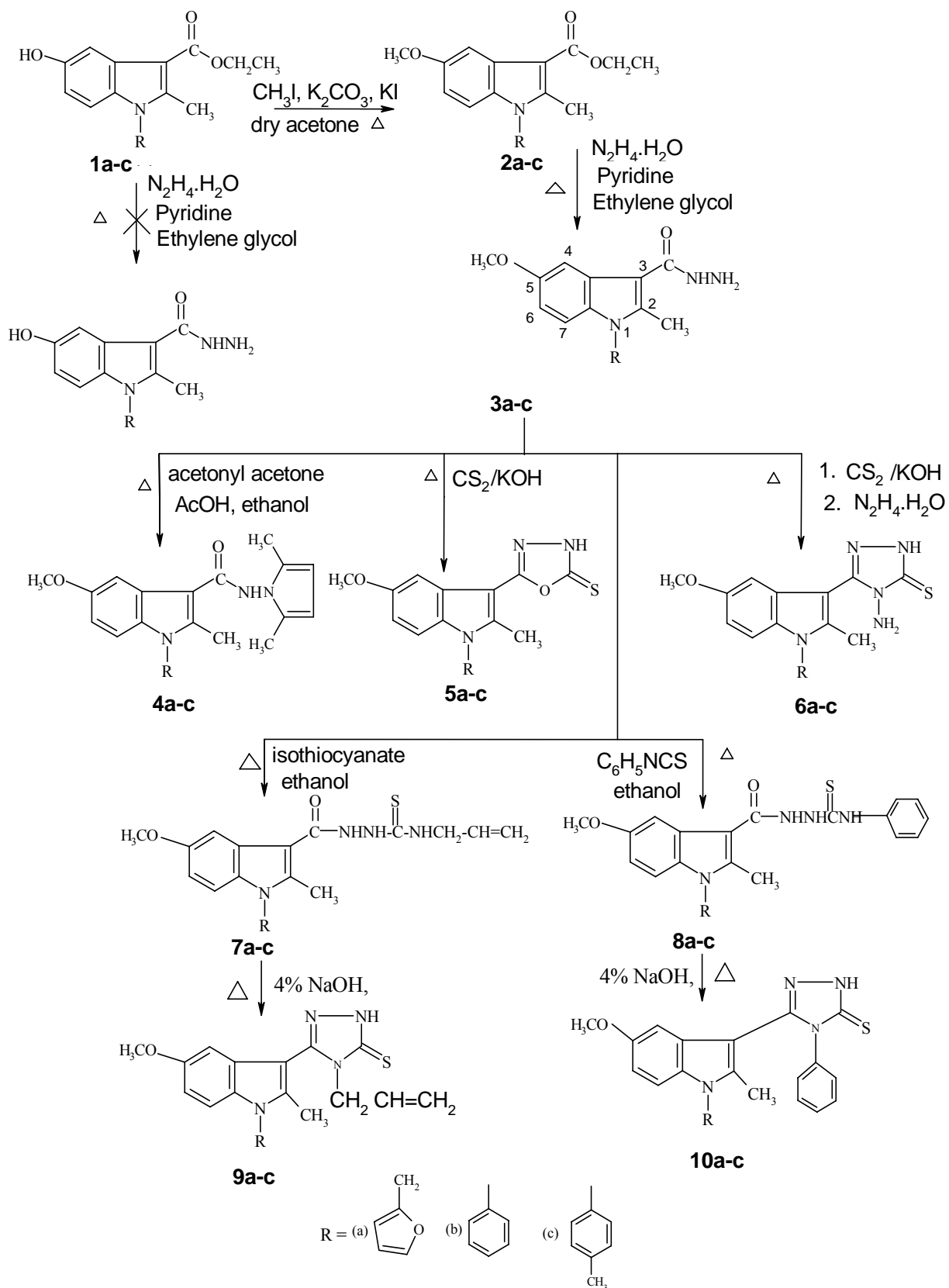
1-Substituted-3-hydrazinocarbonyl-5-methoxy-2-methylindoles 3a-c. To a solution of **2a-c** (0.02 mole) in ethylene glycol (50 mL) was added excess of hydrazine hydrate (99%, 4 mL, 0.08 mole) and 0.5 mL of pyridine. The mixture was heated in an oil-bath at 155-60°C for about 20 hr. It was then cooled and poured on crushed ice (100 g) and acidified with dil. hydrochloric acid. The separated solid was filtered, washed with cold water, dried and recrystallised from suitable solvents (Table I).

Compound **3a**. IR (KBr, cm⁻¹): 1598 (C₃-amide C=O) 1617 (NH bending) and 3285, 3325 (NH/NH₂); ¹H NMR (CDCl₃/TMS): δ 2.76 (s, 3H, C₂-CH₃), 3.85 (s, 3H, C₅-OCH₃), 5.20 (s, 2H, 1-CH₂), 6.14 (d, *J*=3Hz, 1H, C₃-H of furan), 6.27 (m, 1H, C₄-H of furan), 6.85 (dd, *J*=8.5 and 2.5 Hz, 1H, C₆-H), 7.26 (s, 1H, NH, vanished on D₂O exchange), 7.21-7.32 (m, 3H, C₄-H and C₇-H of indole and C₅-H of furan); MS (*m/z*): 299 (M⁺, 11), 300 (M+1, 5), 284(8), 268(55), 241(6), 187(4), 172(6), 79(100).

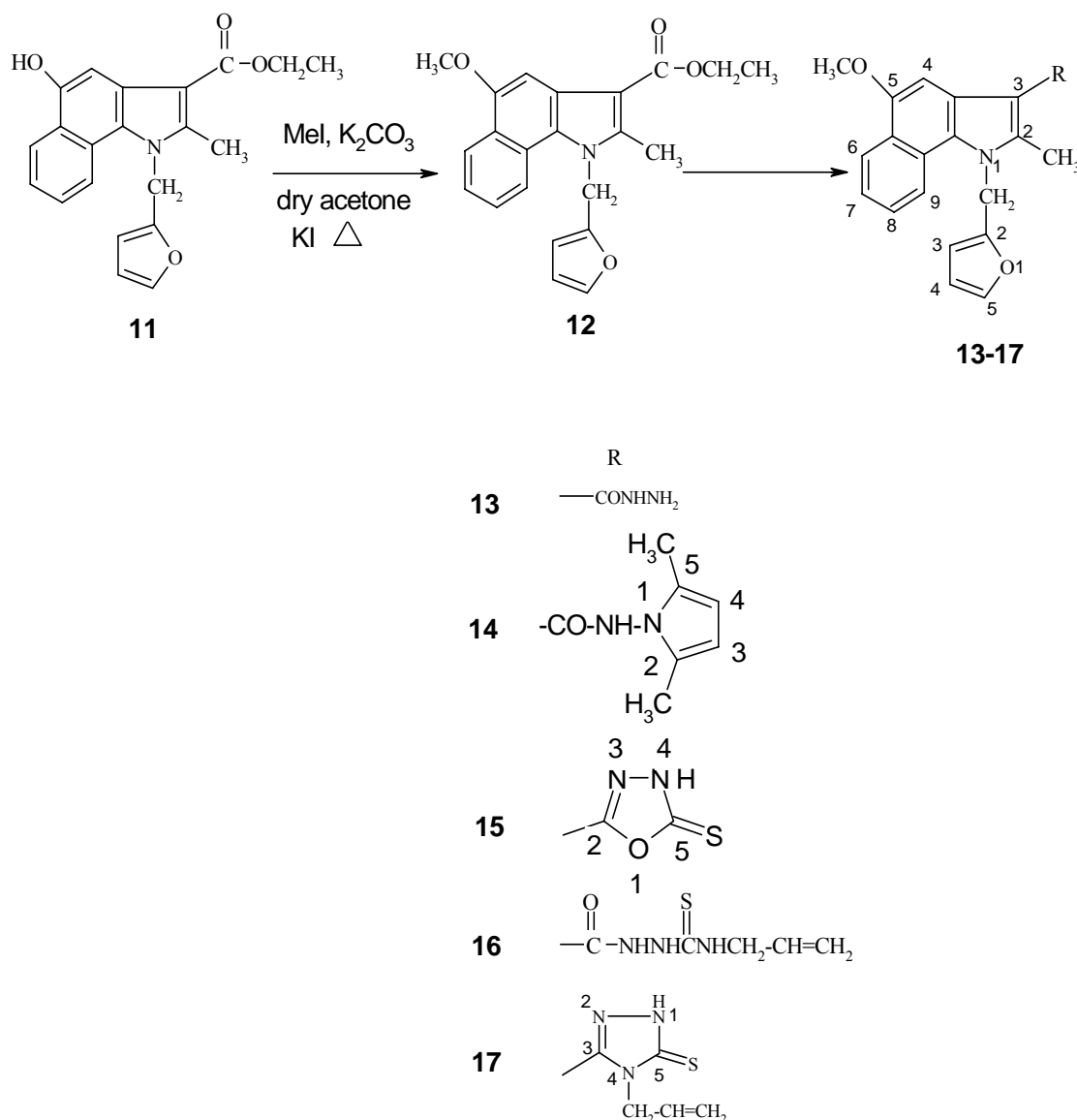
1-Substituted-5-methoxy-3-(2,5-dimethylpyrrol-1-yl)-aminocarbonylmethoxy-2-methylindoles 4a-c. To a suspension of **3a-c** (0.001 mole) in ethanol (10 mL) was added acetonyl acetone (0.002 mole) and glacial acetic acid (1 mL) and the reaction mixture was heated on a boiling water-bath for 4 hr. The reaction mixture was concentrated to half of its original volume and poured into crushed ice (50 g). The separated solid was filtered, washed with water, dried and recrystallised from suitable solvents (Table I).

Compound **4a**: IR (KBr, cm⁻¹): 1645 (C₃-amide C=O) and 3235 (NH); ¹H NMR (CDCl₃/TMS): δ 2.25 (s, 6H, pyrrole CH₃), 2.82 (s, 3H, C₂-CH₃), 3.86 (s, 3H, C₅-OCH₃), 5.26 (s, 2H, 1-CH₂), 5.86 (s, 2H, 2H of pyrrole), 6.21(d, *J*=3 Hz, 1H, C₃-H of furan), 6.30 (m, 1H, C₄-H of furan), 6.92 (dd, *J*=8.5 Hz and 2.5 Hz, 1H, C₆-H), 7.31-7.40 (m, 3H, C₄-H and C₇-H of indole and C₅-H of furan), 7.91(s, 1H, NH disappeared on D₂O exchange); MS (*m/z*): 377(M⁺, 24), 378(M+1, 8), 284(4), 268(100), 187(6), 172(6), 109(3), 81(92), 53(8).

1-Substituted-5-methoxy-3-(5-mercapto-1, 3, 4-oxadiazol-2-yl)methoxy-2-methylindole 5a-c. A mixture of **3a-c** (0.001 mole) in ethanol (20 mL), potassium hydroxide (0.003 mole) dissolved in water (3 mL) and carbon disulphide (0.0045 mole) was



Scheme I



Scheme II

heated under reflux until the evolution of H_2S ceased (about 25 hr). The reaction mixture was cooled to room temperature and poured into ice cold water (100 mL). It was then neutralised with dilute hydrochloric acid. The precipitated solid was filtered, washed with water and dried product was recrystallised from suitable solvents (**Table I**).

Compound **5a**: IR (KBr, cm^{-1}): 1147 ($\text{C}=\text{S}$), 1617 (NH bending) and 3125 (NH); ^1H NMR ($\text{DMSO}-d_6/\text{TMS}$): δ 2.82 (s, 3H, $\text{C}_2\text{-CH}_3$), 3.86 (s, 3H, $\text{C}_5\text{-OCH}_3$), 5.30 (s, 2H, 1- CH_2), 6.26 (d, $J=3$ Hz, 1H, $\text{C}_3\text{-H}$ of furan), 6.30 (m, 1H, $\text{C}_4\text{-H}$ of furan), 6.89 (dd, $J=8.7$ Hz and 2.5 Hz, 1H, $\text{C}_6\text{-H}$), 7.34 (d, $J=8.5$ Hz, 1H, $\text{C}_7\text{-H}$), 7.39 (d, $J=3$ Hz, 1H, $\text{C}_5\text{-H}$ of furan), 14.00 (s, 1H, NH, vanished on D_2O exchange); ^{13}C NMR ($\text{DMSO}-$

d_6/TMS): δ 12.66 ($\text{C}_2\text{-CH}_3$), 40.49 (1- CH_2), 56.27 ($\text{C}_5\text{-OCH}_3$), 96.66 (C_3), 102.73 (C_6), 109.51 (C_3 of furan), 111.43 (C_4 of furan), 112.57 (C_4), 112.65 (C_7), 125.67 ($\text{C}[\text{b}]$), 131.87 ($\text{C}[\text{a}]$), 141.82 (C_5 of furan), 143.99 (C_2), 150.68 (C_2 of furan), 156.12 (C_5), 160.37 (C_2 of oxadiazole), 176.37 (C_5 of oxadiazole); MS (M/z): 341 (M^+ , 38), 342 ($\text{M}+1$, 10), 266 (4), 200 (3), 184 (4), 173 (3), 81 (100), 53 (12).

1-Substituted-3-(4-amino-5-mercapto-1-2,4-triazol-3-yl)-5-methoxy-2-methyl indoles 6a-c. To the ice-cold mixture of **3a-c** (0.001 mole) and KOH (0.0045 mole) in dry ethanol (25 mL), carbon disulphide (0.3 mL, 0.004 mole) was added with magnetic stirring. The reaction mixture was stirred further at room temperature for 30 hr. It was then diluted with dry

Table I — Physical data of new synthesized compounds **3** to **10a-c**

Compd	Substituent (R) (Mol. Formula)	m.p °C	Yield (%)	Nature (Solvent)
3a	Furfuryl (C ₁₆ H ₁₇ N ₃ O ₃)	190-91	76	Colourless tiny needles (ethanol)
3b	Phenyl (C ₁₇ H ₁₇ N ₃ O ₂)	178-79	67	Colourless needles (ethanol)
3c	<i>p</i> -tolyl (C ₁₈ H ₁₉ N ₃ O ₂)	184-85	69	Colourless flowers (ethanol)
4a	Furfuryl (C ₂₂ H ₂₃ N ₃ O ₃)	165-66	84	Pinkish granules (ethanol)
4b	Phenyl (C ₂₃ H ₂₃ N ₃ O ₂)	181-82	81	Brown needles (ethanol)
4c	<i>p</i> -tolyl (C ₂₄ H ₂₅ N ₃ O ₂)	168-69	76	Brown granules (ethanol)
5a	Furfuryl (C ₁₇ H ₁₅ N ₃ O ₃ S)	225-26	80	Colourless flakes (ethanol)
5b	Phenyl (C ₁₈ H ₁₅ N ₃ O ₂ S)	210-11	82	Colourless flakes (ethanol)
5c	<i>p</i> -tolyl (C ₁₉ H ₁₇ N ₃ O ₂ S)	204-05	77	Colourless flakes (ethanol)
6a	Furfuryl (C ₁₇ H ₁₇ N ₂ O ₂ S)	186-87	68	Colourless granules (ethanol)
6b	Phenyl (C ₁₈ H ₁₇ N ₃ OS)	198-99	69	Colourless granules (ethanol)
6c	<i>p</i> -tolyl (C ₁₉ H ₁₉ N ₅ OS)	200-01	79	Colourless granules (ethanol)
7a	Furfuryl (C ₂₀ H ₂₇ N ₄ O ₃ S)	162-63	76	Colourless flakes (ethanol)
7b	Phenyl (C ₂₁ H ₂₂ N ₄ O ₂ S)	171-72	71	Colourless flakes (ethanol)
7c	<i>p</i> -tolyl (C ₂₂ H ₂₄ N ₄ O ₂ S)	155-56	77	Colourless flakes (ethanol)
9a	Furfuryl (C ₂₀ H ₂₀ N ₄ O ₂ S)	145-46	72	Colourless granules (ethanol)
9b	Phenyl (C ₂₁ H ₂₀ N ₄ OS)	182-83	69	Colourless granules (ethanol)
9c	<i>p</i> -tolyl (C ₂₂ H ₂₂ N ₄ OS)	168-69	65	Colourless granules (ethanol)
8a	Furfuryl (C ₂₃ H ₂₂ N ₂ O ₃ S)	185-86	76	Colourless flakes (ethanol)
8b	Phenyl (C ₂₄ H ₂₂ N ₄ O ₂ S)	178-79	78	Colourless flakes (ethanol)
8c	<i>p</i> -tolyl (C ₂₅ H ₂₄ N ₄ O ₂ S)	192-93	70	Colourless flakes (ethanol)
10a	Furfuryl (C ₂₃ H ₂₀ N ₄ O ₂ S)	195-96	72	Colourless granules (ethanol)
10b	Phenyl (C ₂₄ H ₂₀ N ₄ OS)	202-03	69	Colourless granules (ethanol)
10c	<i>p</i> -tolyl (C ₂₅ H ₂₂ N ₄ OS)	207-08	73	Colourless granules (ethanol)

Elemental analyses for C, H, and N were in good agreement with the calculated values.

ether (50 mL) and the separated solid was filtered, washed with dry ether and dried to obtain the potassium salt in quantitative yield. It was magnetically stirred with hydrazine hydrate (1 mL) and water (0.5 mL) on boiling water-bath till the evolution of H_2S ceased (2 hr). The reaction mixture was poured into ice-cold water (20 mL) and acidified with acetic acid. The separated solid was filtered, washed with water, dried and recrystallised from suitable solvents (**Table I**). Compound **6a**: IR (KBr, cm^{-1}): 1623 (NH bending), 3263, 3295 (NH/NH₂); ^1H NMR (CDCl_3 +DMSO- d_6 /TMS): δ 2.57 (s, 3H, C_2 -CH₃), 3.82 (s, 3H, C_5 -OCH₃), 5.25 (s, 2H, 1-CH₂), 6.20 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.28 (m, 1H, C_4 -H of furan), 6.90 (dd, $J=8.5$ and 2.5 Hz, 1H, C_6 -H), 6.99 (d, $J=3$ Hz, 1H, C_5 -H of furan), 7.35 (d, $J=8.5$ Hz, 1H, C_7 -H), 7.52 (d, $J=2.5$ Hz, 1H, C_4 -H), 10.5 (s, 1H, NH, disappeared on D_2O exchange); MS (m/z): 355 (M^+ , 40), 356 ($\text{M}+1$, 9), 341 (11), 340 (5), 267 (3), 81 (100), 53 (10).

1-Substituted-5-methoxy-3-(*N*-allyl / phenyl-thiosemicarbazinocarbonyl) methoxy-2-methyl-indoles 7,8a-c. To a mixture of **3a-c** (0.002 mole) in ethanol (50 mL) was added allyl isothiocyanate (0.003 mole)/phenyl isothiocyanate (0.003 mole). The mixture was heated under reflux for 6 hr, part of the solvent was evaporated. The solid that separated on cooling to room temperature was filtered, washed with ethanol and recrystallised from suitable solvents (**Table I**).

Compound **7a**: IR (KBr, cm^{-1}): 1146 (C=S), 1634 (C_3 -amide C=O) and 3186, 3322 (NH); ^1H NMR (DMSO- d_6 /TMS): δ 2.80 (s, 3H, C_2 -CH₃), 3.88 (s, 3H, C_5 -OCH₃), 4.24-4.28 (m, 2H, allyl methylene protons), 5.13-5.29 (m, 4H, 1-CH₂ and alkene protons of allyl group), 5.85-5.96 (m, 1H, methine proton of allyl group), 6.21 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.29 (m, 1H, C_4 -H of furan), 6.86 (dd, $J=8.5$ and 2.5 Hz, 1H, C_6 -H), 7.32-7.35 (m, 2H, C_5 -H of furan and C_7 -H), 7.45 (s, 1H, NH vanished on D_2O exchange), 7.57 (d, $J=2.5$ Hz, 1H, C_4 -H), 9.50 (s, 1H, NH, disappeared on D_2O exchange), 9.50 (s, 1H, NH, disappeared on D_2O exchange).

Compound **8a**: IR (KBr, cm^{-1}): 1228 (C=S), 1607 (C_3 -amide C=O) and 3171, 3312 (NH); ^1H NMR (CDCl_3 +DMSO- d_6 /TMS): δ 2.79 (s, 3H, C_2 -CH₃), 3.90 (s, 3H, C_5 -OCH₃), 5.22 (s, 2H, 1-CH₂), 6.15 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.26 (m, 1H, C_4 -H of furan), 6.90 (dd, $J=8.5$ and 2.5 Hz, 1H, C_6 -H), 7.17-7.48 (m, 7H, 5H of phenyl and C_5 -H of furan and C_7 -H of indole), 7.69 (d, $J=2.5$ Hz, 1H, C_4 -H), 8.88 (s,

1H, NH, disappeared on D_2O exchange), 9.16 (s, 1H, NH, vanished on D_2O exchange), 10.20 (s, 1H, NH disappeared on D_2O exchange).

1-Substituted-5-methoxy-(4-allyl/phenyl-5-mercapto-1,2,4-triazol-3-yl)-2-methyl indoles. 9, 10a-c. The suspension of thiosemicarbazides **7, 8a,c** (0.008 mole)/ **8a-c** (0.008 mole) in sodium hydroxide (4%, 10 mL) was heated on a water-bath for about 4 hr. The reaction mixture after cooling to room temperature was poured into crushed ice (20 g) and acidified carefully with dilute acetic acid. The precipitate thus obtained was filtered, washed with water, dried and recrystallised from suitable solvents (**Table I**).

Compound **9a**: IR (KBr, cm^{-1}): 1170 (C=S) and 3229 (NH); ^1H NMR (CDCl_3 +DMSO- d_6 /TMS): δ 2.42 (s, 3H, C_2 -CH₃), 3.79 (s, 3H, C_5 -OCH₃), 4.59 (d, $J=7$ Hz, 2H, allyl methylene protons), 4.73-5.04 (m, 2H, alkene protons of allyl group), 5.24 (s, 2H, 1-CH₂), 5.67-5.80 (m, 1H, methine proton of allyl group), 6.15 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.28 (m, 1H, C_4 -H of furan), 6.73 (d, $J=8.5$ and 2.5 Hz, 1H, C_6 -H), 7.26-7.35 (m, 2H, C_4 -H and C_7 -H of indole), 11.7 (br, s, 1H, NH disappeared on D_2O exchange); MS (m/z): 380 (M^+ , 6), 381 ($\text{M}+1$, 2), 341 (4), 313 (6), 285 (7), 241 (10), 81 (100), 53 (11).

Compound **10a**: IR (KBr, cm^{-1}): 1159 (C=S) and 3142 (NH); ^1H NMR (CDCl_3 +DMSO- d_6 /TMS): δ 2.20 (s, 3H, C_2 -CH₃), 3.70 (s, 3H, C_5 -OCH₃), 5.11 (s, 2H, 1-CH₂), 5.96 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.24 (m, 1H, C_4 -H of furan), 6.67 (d, $J=3$ Hz, 1H, C_5 -H of furan), 6.77 (dd, $J=8.5$ and 2.5 Hz, 1H, C_6 -H), 7.16-7.32 (m, 7H, 5H of triazole phenyl and C_4 -H and C_7 -H of indole ring), 11.3 (s, 1H, NH disappeared on D_2O exchange); MS (m/z): 416 (M^+ , 48), 417 ($\text{M}+1$, 12), 384 (6), 335 (4), 307 (8), 276 (84), 262 (4), 81 (100), 53 (5).

1-Furfuryl-3-carbethoxy-5-methoxy-2-methyl-benz[g]indole 12. To a solution of **11** (10 g, 0.028 mole) in dry acetone (100 mL) was added methyl iodide (3.58 mL, 0.066 mole), anhydrous potassium carbonate (6 g) and potassium iodide (0.1 g). The mixture was heated at reflux for 50 hr. It was filtered hot and the solvent was removed under reduced pressure. The residue was recrystallised from ethanol as colourless needles, m.p. 102-03°C, yield 82%. IR (KBr, cm^{-1}): 1694 (C_3 -ester C=O); ^1H NMR (CDCl_3 /TMS): δ 1.50 (t, $J=7.1$ Hz, 3H, C_3 -ester CH₃), 2.86 (s, 3H, C_2 -CH₃), 4.08 (s, 3H, C_5 -OCH₃), 4.46 (q, $J=7.1$ Hz, 2H, C_3 -ester CH₂), 5.71 (s, 2H, 1-CH₂), 6.04 (d, $J=3$ Hz, 1H, C_3 -H of furan), 6.29 (m, 1H, C_4 -H of

furan), 7.41-7.51 (m, 3H, C₇-H & C₈-H of indole and C₅-H of furan), 7.81 (s, 1H, C₄-H), 8.16 (d, *J*=8.7 Hz, 1H, C₆-H), 8.42 (d, *J*=8.7 Hz 1H, C₉-H). Anal. Found: C, 72.62; H, 5.71; N, 3.97. Calcd for C₂₂H₂₁NO₄: C, 72.70; H, 5.82; N, 3.85%.

1-Furfuryl-3-hydrazinocarbonyl-5-methoxy-2-methylbenz[g]indole 13. This compound was prepared from **12** (6 g, 0.016 mole) as per the procedure depicted for compound **3a** and recrystallised from ethanol-dioxane as pinkish granules, m.p. 184-85°C, yield 68%. IR (KBr, cm⁻¹): 1600 (C₃-amide C=O), 1619 (NH bending) and 3296, 3311 (NH/NH₂); ¹H NMR (CDCl₃/TMS); δ 2.76 (s, 3H, C₂-CH₃), 4.07 (s, 3H, C₅-OCH₃), 5.67 (s, 2H, 1-CH₂), 6.05 (d, *J*=3 Hz, 1H, C₃-H of furan), 6.29 (m, 1H, C₄-H of furan), 7.07 (s, 1H, NH, vanished on D₂O exchange), 7.29-7.53 (m, 4H, C₄-H, C₇-H, C₈-H of indole and C₅-H of furan), 8.18 (d, *J*=8.7 Hz, 1H, C₆-H), 8.41 (d, *J*=8.7 Hz, 1H, C₉-H). Anal. Found: C, 68.86; H, 5.32; N, 12.12. Calcd. for C₂₀H₁₉N₃O₃: C, 68.74; H, 5.48; N, 12.02%.

1 – Furfuryl - 5-methoxy-3-(2,5-dimethylpyrrol-1-yl)aminocarbonyl-2-methyl benz[g]indole 14. The compound **14** was prepared from **13** (0.5 g, 0.001 mole) as per the procedure given for compound **4a** and recrystallised from ethanol as pinkish needles, m.p. 238-39°C, yield 92%. IR (KBr, cm⁻¹): 1649 (C₃-amide C=O) and 3180 (NH); ¹H NMR (CDCl₃/TMS): δ 2.29 (s, 6H, 2CH₃ of pyrrole), 2.84 (s, 3H, C₂-CH₃), 4.06 (s, 3H, C₅-OCH₃), 5.74 (s, 2H, 1-CH₂), 5.91 (s, 2H, 2H of pyrrole), 6.14 (d, *J*=3 Hz, 1H, C₃-H of furan), 6.32 (m, 1H, C₄-H of furan), 7.28 (s, 2H, CHCl₃ and NH, the signal due to NH vanished on D₂O exchange), 7.43-7.57 (m, 3H, C₅-H of furan and C₇-H and C₈-H of indole ring), 8.04 (s, 1H, C₄-H), 8.23 (d, *J*=8.7 Hz, 1H, C₆-H), 8.45 (d, *J*=8.7 Hz, 1H, C₉-H); ¹³C NMR (CDCl₃/TMS): 12.00 (Pyrrole CH₃), 12.03 (C₂-CH₃), 44.27 (1-CH₂), 56.12 (C₅-OCH₃), 96.48 (C₃), 104.58 (C₃ and C₄ of pyrrole), 108.13 (C₉), 108.42 (C₃ of furan), 111.12 (C₄ of furan), 120.73 (C₆), 122.67 (C₇), 123.14 (C₄), 123.94 and 123.99 (two junction carbons of naphthalene), 124.56 (C₈), 125.22 (C[b]), 127.17 (C[a]), 128.48 (C₂ and C₅ of pyrrole), 140.39 (C₅ of furan), 143.16 (C₂) 150.01 (C₂ of furan), 152.13 (C₅), 165.85 (C₃ amide carbonyl carbon); MS (m/z): 427 (M⁺, 28.6), 345 (0.8), 333 (3.2), 318 (14.3), 317 (66.7), 236 (30.3), 221 (17.5), 81 (100). Anal. Found: C, 73.18; H, 5.11; N, 9.72. Calcd for C₂₆H₂₅N₃O₃: C, 73.04; H, 5.89; N, 9.82%.

1-Furfuryl-5-methoxy-3- (5-mercapto-1,3,4-oxadiazol-2-yl-2-methylbenz[g] indole 15. The compound **15** was prepared from **13** (0.5 g, 0.001 mole) as per the procedure depicted for compound **5a** and recrystallised from ethanol-dioxane as colourless flakes, m.p. 243-44°C yield (79%). IR (KBr, cm⁻¹): 1114 (C=S), 1617 (NH bending) and 3142 (NH); ¹H NMR (DMSO-*d*₆ /TMS); δ 2.81 (s, 3H, C₂-CH₃), 3.98 (s, 3H, C₅-OCH₃), 5.86 (s, 2H, 1-CH₂), 6.38 (m, 2H, C₃-H and C₄-H of furan), 7.45-7.63 (m, 4H, C₄-H, C₇-H, C₈-H of indole and C₅-H of furan), 8.28 (d, *J*=8.7 Hz, 1H, C₆-H), 8.45 (d, *J*=8.7 Hz, 1H, C₉-H), 14.5 (br, s, 1H, NH disappeared on D₂O exchange); ¹³C NMR (DMSO-*d*₆ /TMS): 12.68 (C₂-CH₃), 43.91 (1-CH₂), 56.37 (C₅-OCH₃), 97.51 (C₃), 98.64 (C₉), 108.93 (C₆), 111.56 (C₄), 121.79 (C₇), 122.39 (C₃ of furan), 122.95 (C₄ of furan), 123.62 (C[b]), 124.23 and 124.44 (two junction carbons of naphthalene), 125.17 (C₈), 127.54 (C[a]), 140.29 (C₅ of furan), 144.08 (C₂), 150.55 (C₂ of furan), 151.60 (C₅), 160.22 (C₂ of oxadiazole), 176.67 (C₅ of oxadiazole). Anal. Found: C, 64.32; H, 4.52; N, 10.81. Calcd for C₂₁H₁₇N₃O₃S: C, 64.43; H, 4.37; N, 10.73%.

1-Furfuryl-3-(N-allylthiosemicarbazinocarbonyl)-5-methoxy-2-methylbenz[g]indole 16. This compound was prepared from **13** (0.6 g, 0.0017 mole) according to the procedure given for compound **7a** and recrystallised from ethanol-dioxane as colourless granules, m.p. 212-13°C, yield (86%). IR (KBr, cm⁻¹): 1143 (C=S), 1616, 1630 (NH bending/C₃-amide C=O) and 3186, 3317 (NH); ¹H NMR (DMSO-*d*₆ /TMS): δ 2.75 (s, 3H, C₂-CH₃) 3.99 (s, 3H, C₅-OCH₃), 4.16 (d, *J*=7 Hz, 2H, allyl methylene protons), 5.06-5.24 (m, 2H, two alkene protons of allyl group), 5.84-5.94 (m, 3H, 1CH₂ and methine protons of allyl group), 6.33 (d, *J*=3 Hz, 1H, C₃-H of furan), 6.39 (m, 1H, C₄-H of furan), 7.40-7.59 (m, 4H, C₄-H, C₇-H, C₈-H indole and C₅-H of furan), 8.08 (br s, 1H, NH disappeared on D₂O exchange), 8.25 (d, *J*=8.7 Hz, 1H, C₆-H), 8.45 (d, *J*=8.7 Hz, 1H, C₉-H), 9.40 (br s, 1H, NH vanished on D₂O exchange). Anal. Found: C, 64.11; H, 5.22; N, 12.39. Calcd for C₂₄H₂₄N₄O₃S: C, 64.26; H, 5.39; N, 12.49%.

1-Furfuryl-5-methoxy – 3- (4-allyl-5-mercapto-1, 2, 4-triazol-3-yl)-2-methylbenz[g]indole 17. The compound **17** was prepared from **16** (0.4 g, 0.0008 mole) as per the procedure given for compound **9a** and recrystallised from ethanol as colourless granules, m.p. 260-61°C, yield 79%. IR (KBr, cm⁻¹): 1114 (C=S), 1623 (NH bending) and 3103 (NH); ¹H NMR

(CDCl₃/TMS): δ 2.46 (s, 3H, C₂-CH₃), 3.99 (s, 3H, C₅-OCH₃), 4.63 (d, $J=7$ Hz, 2H, allyl methylene protons), 4.90 (m, 2H, alkene protons of allyl group), 5.72-5.83 (m, 3H, 1-CH₂ and methine proton of allyl group), 6.07 (d, $J=3$ Hz, 1H, C₃-H of furan), 6.30 (m, 1H, C₄-H of furan), 6.68 (s, 1H, C₄-H of indole), 7.42-7.57 (m, 3H, C₇-H, C₈-H of indole and C₅-H of furan), 8.23 (d, $J=8.7$ Hz, 1H, C₆-H), 8.42 (d, $J=8.7$ Hz, 1H, C₉-H), 11.80 (s, 1H, NH disappeared on D₂O exchange); ¹³C NMR (CDCl₃/TMS): δ 11.84 (C₂-CH₃), 44.47 (1-CH₂), 47.18 (N-CH₂ of triazole), 56.36 (C₅ OCH₃), 95.69 (C₃), 100.45 (C₉), 108.25 (C₃ of furan), 111.09 (C₄ of furan), 111.01 (C₆), 120.01 (C₄), 123.29 (C₇), 123.78 (C₈), 123.83 and 124.06 (two junction carbons of naphthalene), 124.71 and 125.19 (alkene protons of allyl group), 137.81 (C₅ of furan), 143.08 (C₂), 148.77 (C₂ of furan), 150.24 (C₅), 127.07 (C[b]), 130.94 (C[a]), 151.84 (C₂ of triazole), 168.03 (C₅ of triazole). Anal. Found: C, 66.82; H, 5.24; N, 13.17. Calcd for C₂₄H₂₂N₄O₂S: C, 66.95; H, 5.14; N, 13.01%.

Antimicrobial testing

Base layer was obtained by pouring about 15 mL of base layer medium (prepared by dissolving peptone (0.6%), yeast extract (0.3%), beef extract (0.13%) and agar (2.5%) in distilled water adjusted to pH 5.2 and sterilized) into each sterilized petridish and were allowed to attain room temperature. This solid layer after attaining room temperature is called base layer. Overnight grown sub-cultures of bacteria were mixed with seed layer medium and immediately poured into petridishes containing base layer and allowed to attain room temperature. The cups were made by scooping out nutrient agar with a sterile cork borer. To these cups, solutions of test compounds (0.1 mL) were added using sterile pipettes and these plates were subsequently incubated at 37°C for 48 hr. the zones of inhibitions if any were measured in mm for the particular test compound with organism.

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