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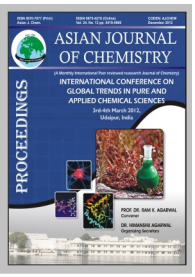

A Convenient Synthesis of Some New Indole Containing Thiazolidinone, Thiohydantoin, Triazine and its Derivatives with Ethoxyphthalimide Moiety†

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Acid catalyzed condensation of thiosemicarbazide with indole-2,3-dione (isatin) yielded indole-2,3-dione-3-thiosemicarbazone (**1**). 3-[(1,3-Thiazolidin-4-one-2-yl)hydrazido]-indole-2-one (**2**) and 3-[(2-thioxoimidazolidin-4-one-3-yl)imino]-indole-2-one (**3**) were obtained by condensation of (**1**) with chloroacetic acid under different conditions. Compounds (**2**) and (**3**) on interaction with various aromatic aldehydes afforded the corresponding 5-substituted benzylidene derivatives (**4a-d**) and (**5a-d**) respectively, which on refluxing with bromoethoxyphthalimide furnished the corresponding 3-[(5-(4-substitutedbenzylidene)-3-*N*-ethoxyphthalimido-1,3-thiazolidin-4-one-2-yl) hydrazido]-1-*N*-ethoxyphthalimido-indole-2-one (**6a-d**) and 3-[(5-(4-substitutedbenzylidene)-2-phthalimidoxyethylsulfanyl-imidazolin-4-one-3-yl)imino]-1-*N*-ethoxyphthalimido-indole-2-one (**7a-d**). Compound (**1**) was also cyclized with KOH in absolute ethanol to yield 2,5-dihydro-3*H*-[1,2,4]triazino[5,6-b]indole-3-thione (**8**). Subsequent treatment with bromoethoxyphthalimide yielded 3-(phthalimidoxyethylsulfanyl)-5-*N*-ethoxyphthalimido-[1,2,4]triazino[5,6-b]indole (**9**). All the synthesized compounds were characterized by their spectral and elemental analysis data.

Key Words: Indole-2,3-dione, Thiazolidinone, Thiohydatoin, Triazine, Bromoethoxy phthalimide, Antimicrobial activity.

INTRODUCTION

Isatin derivatives possess antibacterial[1], antifungal[2], antiviral[3], anti HIV[4], antiprotozoal[5], anticancer[6], muscle relaxant[7], antiallergic activities[8]. The pyrazole unit is one of the core structures in a number of natural products. Spiro compounds are known to possess various biological activities *e.g.* fungicidal[9,10], herbicidal[11], bactericidal[12,13], anticonvulsant[14], anti-inflammatory[15] and antianxiety[16]. These heterocyclic rings attached to alkoxyphthalimide group have been synthesized[17] and tested for antimicrobial[18] and antimalarial[19] activities.

EXPERIMENTAL

Synthesis of indole-2,3-dione-3-thiosemicarbazone (1): An equimolar mixture of isatin and thiosemicarbazide was refluxed in methanol (40 mL) in the presence of catalytic amount (3-4 drops) of glacial acetic acid for 3 h and allowed for cooling. The Schiff base thus obtained was filtered and recrystallized from methanol to give (**1**).

Synthesis of 3-[(1,3-thiazolidin-4-one-2-yl)hydrazido]-indole-2-one (2): A mixture of indole-2,3-dione-3-thiosemicarbazone (1, 0.01 mol), monochloroacetic acid (0.01 mol) and anhydrous sodium acetate (0.02 mol) in ethanol (70 mL) were heated under reflux for 8 h on a water bath with occasional shaking. The solvent was removed and the reaction mixture poured into ice water, filtered and crystallized from ethanol to give (**2**).

Synthesis of 3-[(2-thioxoimidazolidin-4-one-3-yl)imino]-indole-2-one (3): An equimolar mixture of indole-2,3-dione-3-thiosemicarbazone (**1**, 0.01 mol) and monochloroacetic acid (0.01 mol) in pyridine (15 mL) was warmed gently till the exothermic reaction started. After cooling the reaction mixture was treated with ethanol (10 mL) and refluxed for 7 h, poured into ice water, filtered, dried and recrystallized from ethanol, giving yellowish crystals of (**3**).

Synthesis of 3-[(5-(4-chlorobenzylidene)-1,3-thiazolidin-4-one-2-yl)hydrazido]-indole-2-one (4a): To a solution of (**2**, 0.01 mol) in acetic acid were added *p*-chlorobenzaldehyde (0.01 mol) and sodium acetate (0.01 mol). The reaction mixture was refluxed for 5 h, cooled, poured on crushed ice. The separated solid was filtered, washed thoroughly with water, dried and crystallized from a suitable solvent to yield (**4a**). Compounds (**4b-d**) were prepared in a similar manner with a minor change in reflux time.

Synthesis of 3-[(5-(4-chlorobenzylidene)-2-thioxoimidazolidin-4-one-3-yl)imino]-indole-2-one (5a): A mixture of

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(3, 0.01 mol), *p*-chlorobenzaldehyde (0.01 mol) and sodium acetate (0.01 mol) in acetic acid (40 mL) was refluxed for **5** h, cooled and poured onto crushed ice. The yellow solid thus obtained was filtered, washed several times with water and crystallized from acetic acid. Similarly (3.II-Vb-d) were prepared with minor modification in reaction time.

Synthesis of 3-[(5-(4-chlorobenzylidene)-3-*N*-ethoxyphthalimido-1,3-thiazolidin-4-one-2-yl)hydrazido]-1-*N*-ethoxyphthalimido-indole-2-one (6a): Compound (**4a**, 0.01 mol) was dissolved in 15 mL DMF and sodium hydride (0.02 mol) was added portion wise with constant stirring at 0-10 ºC for 5 h. Phthalimidoxyethyl bromide (0.02 mol) was added to above mixture with constant stirring on a magnetic stirrer. Further the reaction mixture was refluxed for 10 h. Excess of solvent was distilled off and the residual reaction mixture was cooled and poured into ice cold water. Solid obtained was recrystallized from ethanol. Compounds (**6b-d**) were also synthesized by similar method using appropriate reactants with required change in reflux time.

Synthesis of 3-[(5-(4-chlorobenzylidene)-2-phthalimidoxyethylsulfanyl-imidazolin-4-one-3-yl)imino]-1-*N*-ethoxyphthalimido-indole-2-one (7a): Compound (**5a**, 0.01 mol) was dissolved in 15 mL DMF and sodium hydride (0.02 mol) was added portion wise with constant stirring at 0-10 ºC for 5 h. Phthalimidoxyethyl bromide (0.02 mol) was added to above mixture with constant stirring on a magnetic stirrer. Further the reaction mixture was refluxed for 10 h. Excess of solvent was distilled off and the residual reaction mixture was cooled and poured into ice cold water. Solid obtained was recrystallized from ethanol. Compounds (3.II-VIIb-d) were prepared in a similar manner with a minor change in reflux time.

Synthesis of 2,5-dihydro-3*H*-[1,2,4]triazino[5,6-b]indole-3-thione (8): Compound (**1**, 0.01 mol) and pottasim hydroxide (0.015 mol) in ethanol was refluxed for 4 h. Excess of solvent was distilled off and the residual reaction mixture was cooled and poured into ice cold water. Solid obtained was recrystallized from ethanol.

Synthesis of 3-(phthalimidoxyethylsulfanyl)-5-*N*-ethoxyphthalimido-[1,2,4]triazino[5,6-b]indole (9): Compound (**8**, 0.01 mol) was dissolved in 15 mL DMF and sodium hydride (0.02 mol) was added portion wise with constant stirring at 0-10 ºC for 5 h. Phthalimidoxyethyl bromide (0.02 mol) was added to above mixture with constant stirring on a magnetic stirrer. Further the reaction mixture was refluxed for 11 h. Excess of solvent was distilled off and the residual reaction mixture was cooled and poured into ice cold water. Solid obtained was recrystallized from ethanol.

RESULTS AND DISCUSSION

Acid catalyzed condensation of thiosemicarbazide with indole-2,3-dione (isatin) was carried out to afford indole-2,3-dione-3-thiosemicarbazone (**1**). Compound (**2**) was prepared by the condensation of (**1**) with chloroacetic acid in ethanol containing anhydrous sodium acetate. The IR spectrum displayed absorption at 1689 cm^{-1} due to imide carbonyl group. The structure was further supported by ^1H NMR spectrum exhibiting signals at δ 9.52 (NH thiazolidinone), 8.61 (NH indole). On the other hand, 3-[(2-thioxoimidazolidin-4-one-3-yl)imino]-indole-2-one (**3**) was obtained by the condensation of (**1**) with chloroacetic acid in presence of pyridine. IR and ^1H NMR data established the structure of (**3**). Its IR spectrum showed absorption bands at 1224 cm^{-1} and 1706 cm^{-1} attributed to C=S and C=O groups, respectively. ^1H NMR spectra showed the signals at 10.32 (NH thiazolidine), 8.97 (NH indole) and 3.93 (-CO-CH_2-). When compounds (**2**) and (**3**) were condensed with aromatic aldehydes in acetic acid and sodium acetate the corresponding arylidene derivatives (**4a-d**) and (**5a-d**) were obtained. The ^1H NMR spectrum of both compounds (**4a**) and (**5a**) were devoid of the singlet at

TABLE-1
IR AND ^1H NMR SPECTRAL DATA OF COMPOUNDS
3.II-I, 3.II-II, 3.II-III, 3.II-IVa -d, 3.II-Va-d and 3.II-VIII

Comp. no	IR (cm^{-1})	^1H NMR (δ)
(2)	3421 (N-H str.), 3103 (C-H str., Ar-H), 2923 (C-H str., CH_2), 1690 (C=O str.),	9.52 (s, 1H, NH of thiazolidinone), 8.60 (s, 1H, NH of indole), 8.57-7.25 (m, 4H, Ar-H), 4.37 (s, 2H, $COCH_2$)
(3)	3321 (N-H str.), 2951 (C-H str., Ar-H), 2830 (C-H str., CH_2), 1706 (C=O str.),	10.32 (s, 1H, NH of thiohydantoin), 8.97 (s, 1H, NH of indole), 8.32-7.33 (m, 4H, Ar-H), 3.93 (s, 2H, $COCH_2$)
(4a)	3342 (N-H str.), 3102 (C-H str., Ar-H), 2976 (C-H str.), 1684 (C=O str.),	9.61 (s, 1H, NH of thiazolidinone), 8.63 (s, 1H, NH of indole), 7.57-7.25 (m, 8H, Ar-H), 6.32 (s, 1H, CH-Ar)
(4b)	3336 (N-H str.), 3107 (C-H str., Ar-H), 2982 (C-H str.), 1692 (C=O str.), 1636 (C=N str.),	9.55 (s, 1H, NH of thiazolidinone), 8.54 (s, 1H, NH of indole), 7.96-6.72 (m, 8H, Ar-H), 6.38 (s, 1H, CH-Ar), 3.82 (s, 3H, OCH_3)
(4c)	3340 (N-H str.), 3104 (C-H str., Ar-H), 2969 (C-H str.), 1682 (C=O str.), 1638 (C=N str.),	9.58 (s, 1H, NH of thiazolidinone), 8.62 (s, 1H, NH of indole), 8.01-6.90 (m, 8H, Ar-H), 6.30 (s, 1H, CH-Ar), 3.27 (s, 6H, $N(CH_3)_2$)
(4d)	3337 (N-H str.), 3109 (C-H str., Ar-H), 2975 (C-H str.), 1685 (C=O str.),	9.60 (s, 1H, NH of thiazolidinone), 8.52 (s, 1H, NH of indole), 7.88-6.72 (m, 9H, Ar-H), 6.34 (s, 1H, CH-Ar)
(5a)	3282 (N-H str.), 3076 (C-H str., Ar-H), 2980 (C-H str.), 1702 (C=O str.),	10.35 (s, 1H, NH of thiohydantoin), 8.79 (s, 1H, NH of indole), 7.06-7.54 (m, 8H, Ar-H), 6.33 (s, 1H, CH-Ar)
(5b)	3275 (N-H str.), 3079 (C-H str., Ar-H), 2985 (C-H str.), 1696 (C=O str.), 1634 (C=N str.),	10.29 (s, 1H, NH of thiohydantoin), 8.75 (s, 1H, NH of indole), 6.85-8.11 (m, 8H, Ar-H), 6.24 (s, 1H, CH-Ar), 3.92 (s, 3H, OCH_3)
(5c)	3266 (N-H str.), 3110 (C-H str., Ar-H), 2974 (C-H str.), 1699 (C=O str.), 1636 (C=N str.),	10.31 (s, 1H, NH of thiohydantoin), 8.68 (s, 1H, NH of indole), 7.20-7.79 (m, 8H, Ar-H), 6.28 (s, 1H, CH-Ar), 3.28 (s, 6H, $N(CH_3)_2$)
(5d)	3281 (N-H str.), 3108 (C-H str., Ar-H), 2972 (C-H str.),	10.18 (s, 1H, NH of thiohydantoin), 8.72 (s, 1H, NH of indole), 6.81-8.05 (m, 9H, Ar-H), 6.22 (s, 1H, CH-Ar)
(8)	3270 (N-H str.), 3109 (C-H str., Ar-H), 1639 (C=N str.), 1226 (C=S str.), 1178 (N-N str.)	9.30 (s, 1H, NH of triazine), 8.12 (s, 1H, NH of indole), 7.21-7.55 (m, 4H, Ar-H)

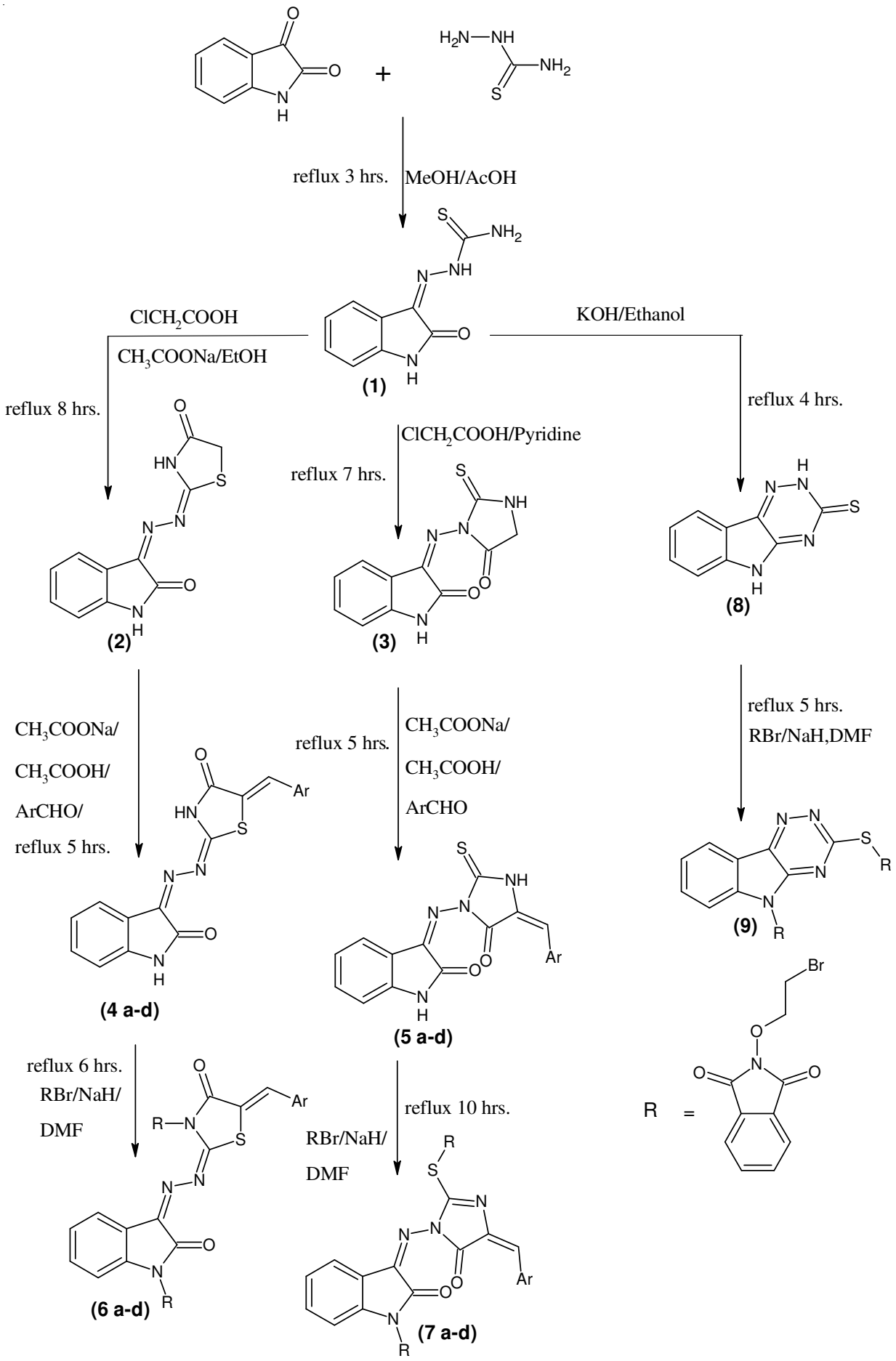
reflux 3 hrs.
MeOH/AcOH
ClCH2COOH
CH3COONa/EtOH
KOH/Ethanol
reflux 8 hrs.
reflux 4 hrs.
ClCH2COOH/Pyridine
reflux 7 hrs.
(1)
(2)
(3)
(8)
CH3COONa/
CH3COOH/
ArCHO/
reflux 5 hrs.
reflux 5 hrs.
CH3COONa/
CH3COOH/
ArCHO
reflux 5 hrs.
RBr/NaH,DMF
(4 a-d)
(5 a-d)
(9)
reflux 6 hrs.
RBr/NaH/
DMF
reflux 10 hrs.
RBr/NaH/
DMF
(6 a-d)
(7 a-d)
R =

TABLE-2
II-III-IR, ^{1}H NMR AND MASS SPECTRAL DATA OF COMPOUND 3.II-Va-l

Comp. No.	IR (cm^{-1})	^{1}H NMR (δ)	Mass (m/z)
(5a)	3322 (N-H str.), 3071 (C-H str., Ar-H), 2981 (C-H str.), 1719 (C=O str.,CONCO str.),	7.27-6.96 (m, 16H, Ar-H), 6.30 (s, 1H, CH-Ar), 4.13 (t, 2H, OCH_2), 3.84 (t, 2H, NCH_2)	761 $[M]^{+\cdot}$, 763 $[M+2]^{+\cdot}$, 709 $[M- C_4H_4]^{+\cdot}$, 649 $[M- C_6H_4Cl]^{+\cdot}$, 629 $[M- C_8H_4O_2]^{+\cdot}$, 570 $[M- C_{10}H_8NO_3]^{+\cdot}$,
(5b)	3328 (N-H str.), 3069 (C-H str., Ar-H), 2972 (C-H str.), 1714 (C=O str., CONCO str.), 1684 (C=O str.),	7.98-6.95 (m, 16H, Ar-H), 6.29 (s, 1H, CH-Ar), 4.09 (t, 2H, OCH_2), 3.62 (s, 3H, OCH_3), 3.80 (t, 2H, NCH_2)	757 $[M]^{+\cdot}$, 705 $[M- C_4H_4]^{+\cdot}$, 649 $[M- C_7H_7O]^{+\cdot}$, 625 $[M- C_8H_4O_2]^{+\cdot}$, 566 $[M- C_{10}H_8NO_3]^{+\cdot}$, 524 $[M- C_{11}H_8N_2O_4]^{+\cdot}$
(5c)	3330 (N-H str.), 3085 (C-H str., Ar-H), 2987 (C-H str.), 1715 (C=O str., CONCO str.), 1680 (C=O str.),	7.63-6.90 (m, 16H, Ar-H), 6.27 (s, 1H, CH-Ar), 4.18 (t, 2H, OCH_2), 3.88 (t, 2H, NCH_2), 3.24 (s, 6H, $N(CH_3)_2$)	770 $[M]^{+\cdot}$, 718 $[M- C_4H_4]^{+\cdot}$, 649 $[M- C_8H_{10}N]^{+\cdot}$, 638 $[M- C_8H_4O_2]^{+\cdot}$, 579 $[M- C_{10}H_8NO_3]^{+\cdot}$, 537 $[M- C_{11}H_8N_2O_4]^{+\cdot}$,
(5d)	3337 (N-H str.), 3082 (C-H str., Ar-H), 2980 (C-H str.), 1717 (C=O str., CONCO str.),	8.12-6.88 (m, 17H, Ar-H), 6.39 (s, 1H, CH-Ar), 4.20 (t, 2H, OCH_2), 3.81 (t, 2H, NCH_2)	727 $[M]^{+\cdot}$, 675 $[M- C_4H_4]^{+\cdot}$, 649 $[M- C_6H_5]^{+\cdot}$, 595 $[M- C_8H_4O_2]^{+\cdot}$, 536 $[M- C_{10}H_8NO_3]^{+\cdot}$,
(7a)	3080 (C-H str., Ar-H), 2931 (C-H str.), 1720 (C=O str., CONCO str.), 1678 (C=O str.), 1633 (C=N str.),	7.11-7.93 (m, 16H, Ar-H), 6.40 (s, 1H, CH-Ar), 3.94 (t, 2H, OCH_2), 3.63 (t, 2H, NCH_2), 2.68 (t, 2H, SCH_2)	761 $[M]^{+\cdot}$, 763 $[M+2]^{+\cdot}$, 709 $[M- C_4H_4]^{+\cdot}$, 615 $[M- C_8H_4O_2N]^{+\cdot}$, 596 $[M- C_9H_5O_4Cl]^{+\cdot}$, 556 $[M- C_{10}H_8N_2O_3]^{+\cdot}$, 426 $[M- C_{18}H_{12}N_3O_4]^{+\cdot}$,
(7b)	3072 (C-H str., Ar-H), 2974 (C-H str.), 1721 (C=O str., CONCO str.), 1683 (C=O str.), 1622 (C=N str.), 1379 (N-O str.),	7.15-7.91 (m, 16H, Ar-H), 6.34 (s, 1H, CH-Ar), 3.90 (t, 2H, OCH_2), 3.74 (s, 3H, OCH_3), 3.64 (t, 2H, NCH_2), 2.73 (t, 2H, SCH_2)	757 $[M]^{+\cdot}$, 705 $[M- C_4H_4]^{+\cdot}$, 611 $[M- C_8H_4NO_2]^{+\cdot}$, 596 $[M- C_{10}H_8O_2]^{+\cdot}$, 552 $[M- C_{10}H_8N_2O_3]^{+\cdot}$, 422 $[M- C_{18}H_{12}N_3O_4]^{+\cdot}$, 364 $[M- C_{19}H_{12}N_4O_4S]^{+\cdot}$,
(7c)	3084 (C-H str., Ar-H), 2983 (C-H str.), 1718 (C=O str., CONCO str.), 1676 (C=O str.), 1625 (C=N str.), 1377 (N-O str.), 684 (C-S-C str.)	7.19-7.95 (m, 16H, Ar-H), 6.38 (s, 1H, CH-Ar), 3.97 (t, 2H, OCH_2), 3.58 (t, 2H, NCH_2), 3.29 (s, 6H, $N(CH_3)_2$), 2.76 (t, 2H, SCH_2)	770 $[M]^{+\cdot}$, 718 $[M- C_4H_4]^{+\cdot}$, 624 $[M- C_8H_4NO_2]^{+\cdot}$, 596 $[M- C_{11}H_{11}NO]^{+\cdot}$, 565 $[M- C_{10}H_8N_2O_3]^{+\cdot}$, 435 $[M- C_{18}H_{12}N_3O_4]^{+\cdot}$, 377 $[M- C_{19}H_{12}N_4O_4S]^{+\cdot}$, 320 $[M- C_{22}H_{19}N_5O_4S]^{+\cdot}$,
(7d)	3089 (C-H str., Ar-H), 2978 (C-H str.), 1716 (C=O str., CONCO str.), 1687 (C=O str.), 1619 (C=N str.),	7.10-7.92 (m, 17H, Ar-H), 6.31 (s, 1H, CH-Ar), 3.99 (t, 2H, OCH_2), 3.60 (t, 2H, NCH_2), 2.70 (t, 2H, SCH_2)	727 $[M]^{+\cdot}$, 675 $[M- C_4H_4]^{+\cdot}$, 596 $[M- C_9H_6O]^{+\cdot}$, 581 $[M- C_8H_4NO_2]^{+\cdot}$, 522 $[M- C_{10}H_8N_2O_3]^{+\cdot}$, 382 $[M- C_{18}H_{12}N_3O_4]^{+\cdot}$,
(9)	3102 (C-H str., Ar-H), 1713 (C=O str., CONCO str.), 1646 (C=N str.),	7.32-7.57 (m, 12H, Ar-H), 4.62 (t, 2H, OCH_2), 3.22 (t, 2H, NCH_2), 2.87 (t, 2H, SCH_2)	580 $[M]^{+\cdot}$, 528 $[M- C_4H_4]^{+\cdot}$, 448 $[M- C_8H_4O_2]^{+\cdot}$, 418 $[M- C_8H_4NO_3]^{+\cdot}$,

δ 4.37 and 3.93 exhibited by their precursors and appearance of the singlet at δ 6.32 and 6.33 for =CH-Ar. This clearly supports that the condensation took place between compounds (2 and 4) and aldehydes. Base catalyzed condensation of (**4a-d**) and (**5a-d**) with phthalimidoxy ethylbromide in DMF yielded compounds (**6a-d**) and compounds (**7a-d**). Absence of singlet at δ 9.52, 8.61 (**4a**) and 10.32, 8.97 (**5a**) for the proton of NH and appearance of two triplets (3.II-VIa) and three triplets (**7a**) corresponding to their phthalimidoxy group in the ^{1}H NMR spectrum confirm the structure of (**6a**) and (**7a**). Formation of compound (**7a**) was further supported by disappearance of the IR band at 1224 cm^{-1} due to the C=S group and appearance of a new band at 699 cm^{-1} due to the C-S bond. In parallel, cyclization of (**1**) with pottasim hydroxide in ethanol compound (**8**). The structure was supported by ^{1}H NMR spectrum exhibiting signals at δ 9.30 (NH triazine), 8.12 (NH indole). When bromoethoxyphthalimide was condensed with (**8**) in NaH and DMF, formation of 3-(phthalimidoxyethyl-sulfanyl)-5-*N*-ethoxyphthalimido-[1,2,4]triazino[5,6-b]indole (**9**) was achieved (Tables 1 and 2).

REFERENCES

1. S.N. Pandeya and D. Sriram, *Acta Pharm. Turc.*, **40**, 33 (1998).
2. S.N. Pandeya, D.N.G. Sriram and E. De Clercq, *Indian J. Pharm. Sci*., **61**, 358 (1999).
3. R.S. Varma and W.L. Nobles, *J. Med. Chem.*, **10**, 972 (1967).
4. S.N. Pandeya, P.S. Yogeeswar, E.D. Clercq and C.W.M. Pannecouque, *Chemotherapy*, **45**, 192 (1999).
5. S.A. Imam and R.S. Varma, *Experientia*, **31**, 1287 (1975).
6. F.D. Popp and H. Pajouhesh, *J. Pharm. Sci.*, **72**, 318 (1983).
7. C. David, T. Marie and G. Roussel, *Chem. Abstr.*, **124**, 343271 (1996).
8. M. Sarangapani and V.M. Reddy, *Indian J. Pharm. Sci.*, **59**, 105 (1997).
9. H.M. Khan, S. Tiwari, K. Begum and Nizamuddin, *Indian J. Chem.*, **37B**, 1075 (1998).
10. M.S.C. Pedras, M. Suchy and P.W.K. Ahiahonu, *Org. Biomol. Chem.*, **4**, 691 (2006).
11. A. Dandia, R. Singh and K. Arya, *Phosphorus Sulfur Silicon Rel. Elem.*, **179**, 551 (2004).
12. J. Kobayashi, M. Tsuda, K. Agemi, H. Shigemori, M. Ishibashi, T. Sasaki and Y. Mikami, *Tetrahedron*, **47**, 6617 (1991).
13. V. Padmavathi, B.J.M. Reddy, A. Baliah, A. Padmaj and D.B. Reddy, *ARKIVOC*, 1 (2005).
14. S. Ali and M. Alam, *Arch. Pharmacol. Res.*, **17**, 131 (1994).
15. M. Rajopadhye and D.F. Popp, *J. Heterocycl. Chem.*, **24**, 1637 (1987).
16. C.C. Rovnyak, V.L. Narayanan and D.U.S. Haugwitz, *Chem. Abstr.*, **88**, 22889 (1978).
17. R.A. Kusanur, M. Ghate and M.V. Kulkarni, *J. Chem Sci.*, **116**, 265 (2004).
18. T. Banu, S. Rajora and G.L. Talesara, *J. Indian Chem. Soc.*, **77**, 300 (2000).
19. C. Sharma, S. Sharma and N. Hussain, *J. Ind. Coun. Chem.*, **26**, 31 (2009).