Reactions of 3-Benzylindole-2-carbohydrazides: Synthesis of New 10-Benzyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles and 3-Benzyl-2-(1,3,4-oxadiazol-2-yl)indoles

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3-Benzylindole-2-carbohydrazides (**4**) on reaction with triethylorthoformate in a polar solvent like DMF yielded only 10-benzyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles (**5**) while (**4**) on reaction with triethylorthoacetate in DMF yielded both 10-benzyl-4-methyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles (**5**) and 3-benzyl-2-(5-methyl-1,3,4-oxadiazol-2-yl)indoles (**6**) instead of only the triazinoindoles as expected. The oxadiazolylindoles (**6**) were also synthesized by refluxing (**4**) with excess of orthoesters. The structures of the compounds formed were characterized by their analytical and spectral data.

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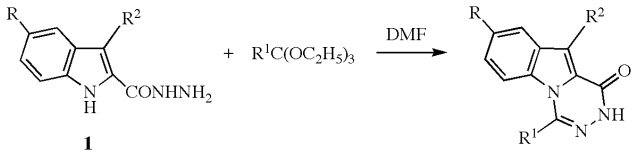
The reactions of both aromatic and heteroaromatic acid hydrazides with orthoesters have been of both synthetic and mechanistic interest. The reactions of indole-2-carbohydrazides with orthoesters have been widely studied. Ainsworth [1,2] reported that a neat reaction leading to the formation of exclusively oxadiazoles occurs on boiling indole-2-carbohydrazides with orthoesters. However, refluxing of indole-2-carbohydrazide with orthoester in presence of a polar solvent, like DMF, results in the formation of 1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles [3]. Robba and co-workers [4,5,6] have reported the synthesis of 1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles as a base induced ring expansion rearrangement of oxadiazolylindoles. 3-Substituted indole-2-carbohydrazides (**1**) with appropriate orthoester in boiling DMF yielded triazines (**2**) as the only products of the reaction; with a view to study the role of stereoelectronic factors in the reaction of (**1**) with orthoesters, different substituents at C-3 and a variety of groups of different electronic nature at C-5 position in the indole nucleus were incorporated [7] (Scheme 1).

Scheme 1



1 **2**

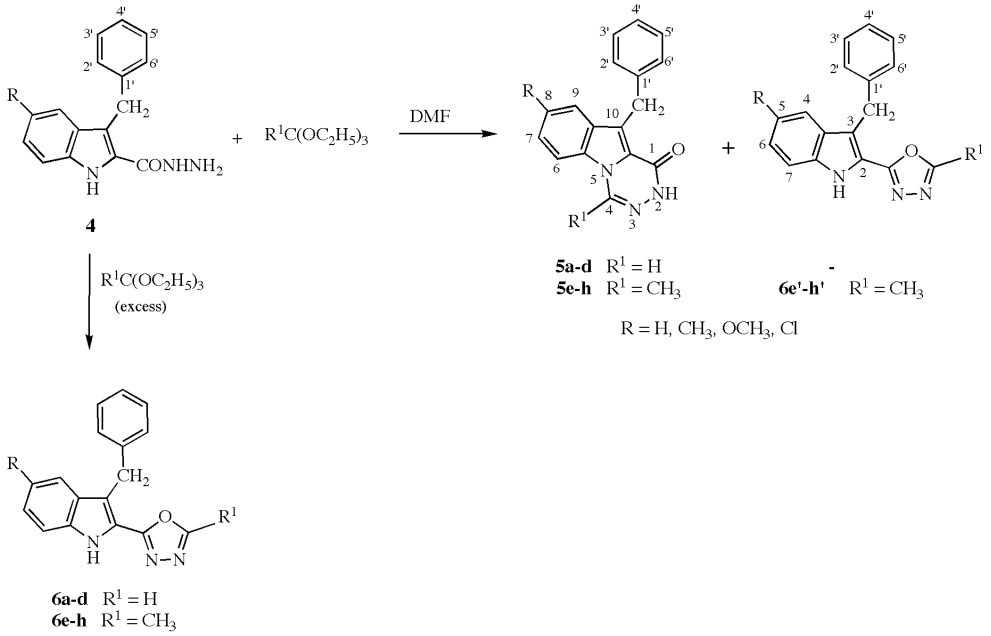
R = H, CH_3, OCH_3, OC_2H_5, Cl ; R^2 = CH_3, C_6H_5; R^1 = H, CH_3

In continuation of our interest in the synthesis of triazino[4,5-*a*]indoles, herein we report the results of the reactions of various 5-substituted 3-benzylindole-2-carbohydrazides (**4**) with orthoesters. Reaction of ethyl 3-benzylindole-2-carboxylates (**3**) [8] with hydrazine hydrate gave 3-benzylindole-2-carbohydrazides (**4**) in good yields. Refluxing compounds (**4**) with triethylorthoformate in DMF gave exclusively 10-benzyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles (**5a-d**), while with excess of triethylorthoformate only, the corresponding oxadiazolylindoles (**6a-d**) were obtained as anticipated (Scheme 2). However, hydrazides (**4**) when refluxed with triethylorthoacetate in DMF yielded corresponding triazinoindoles (**5e-h**) along with compounds which were characterized, by their analytical and spectral data, to be corresponding 3-benzyl-2-(5-methyl-1,3,4-oxadiazol-2-yl)indoles (**6e'-h'**). The structures of compounds (**6e'-h'**) were further confirmed by their independent syntheses, by refluxing appropriate hydrazides with excess of triethylorthoacetate only (Scheme 2). The m.p., infrared (KBr) and NMR spectra of (**6e'-h'**) were identical to those of (**6e-h**) respectively; no depression in their mixed melting points was observed.

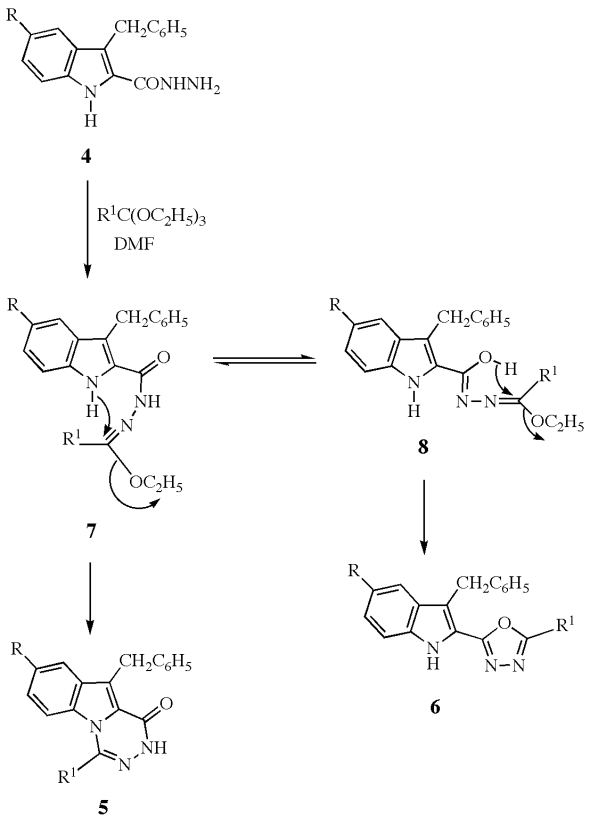
It appears, therefore, that the reactions of 5-substituted-3-benzylindole-2-carbohydrazides with orthoesters provide an interesting example of competitive cyclisation to five and six-membered ring products. A pathway consistent with these results may be given as follows. Although the hydrazides normally yield the oxotriazines (**5**) directly on reaction with orthoesters, in a polar solvent like dimethylformamide, it appears that the stepwise mechanism for the cyclisation involves initial condensation with the more nucleophilic hydrazide nitrogen with subsequent ring closure by the indole nitrogen. Initial condensation would be expected to occur at the more nucleophilic site, the terminal hydrazide nitrogen, producing the non-isolable intermediate (**7**). Intermediates of this type have been postulated and in few cases isolated during the reactions of heterocyclic carbohydrazides with orthoesters [9,10].

Loss of a molecule of ethanol, directly from the intermediate (**7**), would yield 3-benzyl-1,2-dihydro-1-oxo-1,2,4-triazinoindoles (**5**), while its iminol tautomer (**8**) would give 3-benzyl-2-(1,3,4-oxadiazol-2-yl)indoles (**6**) (Scheme 3). Direct evidence for the possible intermediate (**7**) was sought but not obtained in the present study. The failure to detect the intermediate suggests the rapid nucle-

Scheme 2



Scheme 3



ophilic involvement of either the indole nitrogen or iminol oxygen with consequent formation of products.

Formation of both the triazinoindoles and oxadiazolylindoles together in the same reaction suggests that both the compounds are sufficiently stable under the reaction conditions. Further, the formation of both the products in the case of benzyl substituent and not with methyl or phenyl substituent at 3-position of indole [7] suggests that steric factors also and not the electronic factors alone control the course of reaction. It may, therefore, be concluded that both the steric implications of the orthoester substituent and substituent at 3-position of indole-2-carbohydrazides appear to influence the mode of cyclisation and the type of products obtained from these cyclisation reactions.

EXPERIMENTAL

All melting points are uncorrected. Infrared spectra were recorded on a Nicolet Impact-410 FT-IR spectrophotometer using KBr pellets; the frequencies are expressed in cm^{-1}. The ^1H NMR spectra were recorded on Bruker Avance –200 MHz FT-NMR, and Bruker AMX-400 400 MHz instruments with tetramethylsilane as the internal reference; the chemical shifts are reported in ppm and coupling constants (J) are given in Hertz (Hz). Elemental analyses were performed using Heraus CHN rapid analyzer.

3-Benzylindole-2-carbohydrazides (**4a-d**).

General Procedure.

A solution of appropriate ethyl 3-benzylindole-2-carboxylate (**3a-d**) (0.05 mole) in pyridine (25 ml) and 30 g of hydrazine hydrate (99%) was heated under reflux for 2 hours and cooled. The crystalline solid that separated was collected by filtration and recrystallised from dioxane to give the corresponding 3-benzylindole-2-carbohydrazides. The physical data and spectral

Table 1

Physical and Analytical Data of 3-Benzylindole-2-carbohydrazides

Compound	R	Yield (%)	Mp (°C)	Molecular Formula	Analyses % Calcd./Found		
					C	H	N
4a	H	78	217-218	$C_{16}H_{15}N_3O$	72.43/72.51	5.70/5.79	15.84/15.92
4b	CH_3	75	205-206	$C_{17}H_{17}N_3O$	73.10/73.19	6.13/6.21	15.04/15.27
4c	OCH_3	74	208-209	$C_{17}H_{17}N_3O_2$	69.14/69.25	5.80/5.92	14.23/14.31
4d	Cl	72	208-209	$C_{16}H_{14}ClN_3O$	64.11/64.22	4.71/4.82	14.02/14.14

Table 2

Physical and Analytical Data of 10-Benzyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles

Compound	R	R^1	Yield (%)	Mp (°C)	Molecular Formula	Analyses % Calcd./Found		
						C	H	N
5a	H	H	62	265-266	$C_{17}H_{13}N_3O$	74.17/74.25	4.76/4.82	15.26/15.34
5b	CH_3	H	68	272-273	$C_{18}H_{15}N_3O$	74.72/74.81	5.23/5.32	14.52/14.64
5c	OCH_3	H	58	235-236	$C_{18}H_{15}N_3O_2$	70.81/70.92	4.95/5.07	13.76/13.82
5d	Cl	H	53	268-269	$C_{17}H_{12}ClN_3O$	65.92/65.99	3.90/4.12	13.57/13.68
5e	H	CH_3	42	275-276	$C_{18}H_{15}N_3O$	74.72/74.91	5.23/5.00	14.52/14.61
5f	CH_3	CH_3	50	290-291	$C_{19}H_{17}N_3O$	75.23/75.29	5.65/5.72	13.85/13.99
5g	OCH_3	CH_3	60	260-261	$C_{19}H_{17}N_3O_2$	71.46/71.52	5.37/5.49	13.16/13.21
5h	Cl	CH_3	50	299-300	$C_{18}H_{14}ClN_3O$	66.77/66.82	4.36/4.49	12.98/13.09

Table 3

Physical and Analytical Data of 3-Benzyl-2-(1,3,4-oxadiazol-2-yl)indoles

Compound	R	R^1	Yield (%)	Mp (°C)	Molecular Formula	Analyses % Calcd./Found		
						C	H	N
6a	H	H	55	225-226	$C_{17}H_{13}N_3O$	74.17/74.05	4.76/4.86	15.26/15.34
6b	CH_3	H	58	235-236	$C_{18}H_{15}N_3O$	74.72/74.61	5.23/5.32	14.52/14.61
6c	OCH_3	H	58	230-231	$C_{18}H_{15}N_3O_2$	70.81/70.70	4.95/5.05	13.76/13.84
6d	Cl	H	58	248-249	$C_{17}H_{12}ClN_3O$	65.92/66.10	3.90/3.98	13.57/13.68
6e'	H	CH_3	33	217-218	$C_{18}H_{15}N_3O$	74.72/74.82	5.23/5.34	14.52/14.67
6e	H	CH_3	64	218-219	$C_{18}H_{15}N_3O$	74.72/74.68	5.23/5.20	14.52/14.49
6f'	CH_3	CH_3	37	235-236	$C_{19}H_{17}N_3O$	75.23/75.33	5.65/5.72	13.85/13.92
6f	CH_3	CH_3	55	237-238	$C_{19}H_{17}N_3O$	75.23/75.19	5.65/5.60	13.85/13.80
6g'	OCH_3	CH_3	28	234-235	$C_{19}H_{17}N_3O_2$	71.46/71.52	5.37/5.48	13.16/13.24
6g	OCH_3	CH_3	56	234-235	$C_{19}H_{17}N_3O_2$	71.46/71.40	5.37/5.30	13.16/13.08
6h'	Cl	CH_3	32	267-268	$C_{18}H_{14}ClN_3O$	66.77/66.83	4.36/4.42	12.98/13.08
6h	Cl	CH_3	56	267-268	$C_{18}H_{14}ClN_3O$	66.77/66.70	4.36/4.31	12.98/12.90

analyses of these substituted 3-benzylindole-2-carbohydrazides are recorded in Table 1 and Table 4.

General Procedure for the Reaction of 3-Benzylindole-2-carbohydrazides (**4**) with Triethylorthoformate.

A mixture of appropriate 3-benzylindole-2-carbohydrazide (**4a-d**) (0.011 mole) and triethylorthoformate (2.07 g, 0.014 mole) in dimethylformamide (20 ml) was refluxed for 12 hours while protecting it from moisture. The reaction mixture was poured onto crushed ice. The white solid that separated was collected by filtration and crystallized from dioxane ethanol mixture to obtain the various 8-substituted 10-benzyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-*a*]indoles (**5a-d**). The physical data and spectral analyses of these triazinoindoles are recorded in Table 2 and Table 4.

3-Benzyl-2-(1,3,4-oxadiazol-2-yl)indoles (**6a-d**).

General Procedure.

A mixture of the appropriate 3-benzylindole-2-carbohydrazide (**4a-d**) (0.01 mole) and triethylorthoformate (50 ml) was heated under reflux for 12 hours. Excess of the reagent was removed under reduced pressure. The residue was treated with pet. ether and crystallized from ethanol to obtain the 5-substituted 3-benzyl(1,3,4-oxadiazol-2-yl)indoles. The physical data and spectral analyses of these oxadiazolylindoles are recorded in Table 3 and Table 4.

General Procedure for the Reaction of 3-Benzylindole-2-carbohydrazides (**4**) with Triethylorthoacetate.

A mixture of the appropriate 3-benzylindole-2-carbohydrazide (**4a-d**) (0.011 mole) and triethylorthoacetate (2.27 g, 0.014 mole) in dimethylformamide (20 ml) was refluxed for 12 hours while protecting it from moisture. The reaction mixture was left

Table 4

IR and ^{1}H-NMR Spectral Analyses for Compounds **4a**-**d**, **5a**-**h**, **6a**-**d** and **6e'**-**h'**

Compound	IR (KBr) (ν/cm^{-1})	^{1}H NMR (Solvent/ δ values in ppm)
4a	1616 (C=O), 3350-3260 (br) $NHNH_2$	(DMSO-d_6): 4.43 (s, 2H, CH_2Ph), 4.53 (s, 2H, -NH_2), 6.96-7.00 (m, 1H, 5-H), 7.07-7.11 (m, 1H, 6-H), 7.16-7.21 (m, 3H, phenyl protons), 7.27-7.29 (m, 2H, phenyl protons), 7.37 (d, J = 8.2 Hz, 1H, 4-H), 7.53 (d, J = 7.98Hz, 1H, 7-H), 9.28 (s, 1H, indole NH), 11.15 (s, 1H, -CONH).
4b	1616 (C=O), 3320-3270 (br) $NHNH_2$	(DMSO-d_6): 2.32 (s, 3H, CH_3), 4.41 (s, 2H, CH_2Ph), 4.53 (s, 2H, -NH_2), 7.01 (d, J = 8.36 Hz, 1H, 6-H), 7.10 (t, J = 7.16 Hz, 1H, 4'-H), 7.20 (t, J = 7.16 Hz, 2H, 3' and 5'-H), 7.28 (d, J = 8.39 Hz, 3H, 2', 6' and 7-H), 7.31 (s, 1H, 4-H), 9.25 (s, 1H, indole NH), 11.04 (s, 1H, -CONH).
4c	1635 (C=O), 3438-3259 (br) $NHNH_2$	(DMSO-d_6): 3.69 (s, 3H, OCH_3), 4.39 (s, 2H, CH_2Ph), 4.48 (s, 2H, -NH_2), 6.83 (dd, J = 8.61 Hz, 2.35 Hz, 1H, 6-H), 6.95 (d, J = 2.35 Hz, 1H, 4-H), 7.09 (t, J = 7.04 Hz, 1H, 4'-H), 7.19 (t, J = 7.04 Hz, 2H, 3'-H and 5'-H), 7.26-7.29 (m, 3H, 2'-H, 6'-H and 7-H), 9.18 (s, 1H, indole NH), 10.98 (s, 1H, -CONH).
4d	1612 (C=O), 3350-3256 (br) $NHNH_2$	(DMSO-d_6): 4.38 (s, 2H, CH_2Ph), 4.54 (s, 2H, -NH_2), 7.11 (t, J = 7.04 Hz, 1H, 4'-H), 7.16 (dd, J = 1.95 Hz, 8.61Hz, 1H, 6-H), 7.20 (t, J = 7.43 Hz, 2H, 3'-H and 5'-H), 7.26 (d, J = 7.42 Hz, 2H, 2'-H and 6'-H), 7.39 (d, J = 8.61 Hz, 1H, 7-H), 7.52 (d, J = 1.57 Hz, 4-H), 9.35 (s, 1H, indole NH), 11.37 (s, 1H, -CONH).
5a	1659 (C=O), 3184 (NH)	(DMSO-d_6): 4.59 (s, 2H, CH_2Ph), 7.14 (t, J = 7.32 Hz, 1H, 4'-H), 7.24 (t, J = 7.33 Hz, 2H, 3'- and 5'-H), 7.34-7.39 (m, 3H, 8-, 2'- and 6'-H), 7.51 (ddd, J = 8.1 Hz and J = 0.9 Hz, 1H, 7-H), 7.82 (d, J = 8.08 Hz, 1H, 9-H), 8.18 (d, J = 8.42 Hz, 1H, 6-H), 9.05 (s, 1H, -CH=N-), 11.80 (s, 1H, -NH).
5b	1652 (C=O), 3173 (NH)	(DMSO-d_6): 2.08 (s, 3H, CH_3), 4.56 (s, 2H, CH_2Ph), 7.12 (t, J = 7.3 Hz, 1H, 4'-H), 7.23 (t, J = 7.3 Hz, 2H, 3'- and 5'-H), 7.31-7.36 (m, 3H, 7-, 2'- and 6'-H), 7.58 (s, 1H, 9-H), 8.05 (d, J = 8.5 Hz, 1H, 6-H), 8.95 (s, 1H, -CH=N-), 11.74 (s, 1H, -NH).
5c	1658 (C=O), 3170 (NH)	(DMSO-d_6): 3.78 (s, 3H, OCH_3), 4.57 (s, 2H, CH_2Ph), 7.11-7.39 (m, 7H, 7-H, 9-H, and phenyl protons), 8.08 (d, J = 9.05 Hz, 1H, 6-H), 8.97 (s, 1H, -CH=N-), 11.75 (s, 1H, -NH).
5d	1659 (C=O), 3194 (NH)	(DMSO-d_6): 4.58 (s, 2H, CH_2Ph), 7.14 (t, J = 6.9 Hz, 1H, 4'-H), 7.24 (t, J = 7.1 Hz, 2H, 3'- and 5'-H), 7.37 (d, J = 7.8 Hz, 2H, 2'- and 6'- H), 7.53 (dd, J = 8.9 Hz, J = 1.3Hz, 1H, 7-H), 7.87 (d, J = 1.5 Hz, 1H, 9-H), 8.21 (d, J = 8.90 Hz, 1H, 6-H), 9.04 (s, 1H, -CH=N-), 11.88 (s, 1H, -NH).
5e	1659 (C=O), 3172 (NH)	(DMSO-d_6): 2.84 (s, 3H, CH_3), 4.67 (s, 2H, CH_2Ph), 7.13 (t, J = 7.3 Hz, 1H, 4'-H), 7.23 (t, J = 7.3 Hz, 2H, 3'-H and 5'-H), 7.36 (d, J = 7.1 Hz, 2H, 2'-H and 6'-H), 7.38 (t, J = 7.8 Hz, 1H, 8-H), 7.47 (ddd, J = 8.5 Hz, 7.1 Hz and 1.2 Hz, 1H, 7-H), 7.85 (d, J = 7.91 Hz, 1H, 9-H), 8.11 (d, J = 8.66 Hz, 1H, 6-H), 11.70 (s, 1H, NH).
5f	1647 (C=O), 3178 (NH)	(DMSO-d_6): 2.41 (s, 3H, CH_3Ph), 2.78 (s, 3H, CH_3), 4.61 (s, 2H, CH_2Ph), 7.07-7.35 (m, 6H, 7-H and phenyl protons), 7.59 (s, 1H, 9-H), 7.96 (d, J = 8.82 Hz, 1H, 6-H), 11.63 (s, 1H, NH).
5g	1652 (C=O), 3166 (NH)	(DMSO-d_6): 2.79 (s, 3H, CH_3), 3.35 (s, 3H, OCH_3), 4.63 (s, 2H, CH_2Ph), 7.09-7.38 (m, 7H, 9-H and Phenyl protons), 7.99 (d, J = 9.31 Hz, 1H, 6-H), 11.67 (s, 1H, NH).
5h	1648 (C=O), 3180 (NH)	(DMSO-d_6): 2.82 (s, 3H, CH_3), 4.64 (s, 2H, CH_2Ph), 7.13 (t, J = 7.2 Hz, 1H, 4'-H), 7.23 (t, J = 7.4 Hz, 2H, 3'-H and 5'-H), 7.35 (d, J = 7.5 Hz, 2H, 2'-H and 6'-H), 7.48 (dd, J = 9.0 Hz and J = 1.3 Hz, 1H, 7-H), 7.89 (d, J = 1.3 Hz, 1H, 9-H), 8.12 (d, J = 9.17 Hz, 1H, 6-H), 11.80(s, 1H, NH).
6a	3231 (NH), 1623 (C=N)	(DMSO-d_6): 4.52 (s, 2H, CH_2Ph), 7.06 (t, J = 7.16 Hz, 1H, 5-H), 7.14 (t, J = 7.07 Hz, 1H 6-H), 7.21-7.43 (m, 5H, phenyl protons), 7.48 (d, J = 8.2 Hz, 1H, 4-H), 7.62 (d, J = 8.0 Hz, 1H, 7-H), 9.41(s, 1H, 5-H of oxadiazole), 12.05 (s, 1H, -NH).
6b	3245 (NH), 1606 (C=N)	($CDCl_3$): 2.41 (s, 3H, CH_3), 4.51(s, 2H, CH_2Ph), 7.14-7.39 (m, 8H, Phenyl protons), 8.43(s, 1H, 5-H of oxadiazole), 9.22 (brs, 1H, indole NH).
6c	3229 (NH), 1609 (C=N)	($CDCl_3$): 3.78(s, 3H, OCH_3), 4.51 (s, 2H, -CH_2Ph), 6.93 (d, J = 2.35 Hz, 1H, 4-H), 7.00 (dd, J = 9 Hz, J = 2.35 Hz, 1H, 6-H), 7.17-7.26 (m, 5H, phenyl protons), 7.36 (d, J = 9 Hz, 1H, 7-H), 8.43 (s, 1H, 5-H of oxadiazole), 8.96 (brs, 1H, -NH).
6d	3201 (NH), 1606 (C=N)	($CDCl_3$): 4.50 (s, 2H, -CH_2Ph), 7.18-7.29 (m, 6H, 6-H and phenyl protons), 7.44 (d, J = 8.61 Hz, 1H, 7-H), 7.54 (s, 1H, 4-H), 8.48 (s, 1H, 5-H of oxadiazole), 9.41 (brs, 1H, -NH).
6e'	3221 (NH), 1625 (C=N)	($CDCl_3$): 2.61(s, 3H, CH_3), 4.51(s, 2H, CH_2Ph), 7.09-7.33 (m, 7H, phenyl protons), 7.46 (d, J = 8.22 Hz, 1H, 4-H), 7.56 (d, J = 7.83 Hz, 1H, 7-H), 9.19 (brs, 1H, -NH).
6f'	3222 (NH), 1616 (C=N)	($CDCl_3$): 2.41(s, 3H, CH_3Ph), 2.59 (s, 3H, CH_3), 4.48 (s, 2H, -CH_2Ph), 7.12-7.34 (m, 8H, phenyl protons), 8.97 (brs, H, -NH).
6g'	3216 (NH), 1616 (C=N)	($CDCl_3$): 2.60 (s, 3H, CH_3), 3.77 (s, 3H, OCH_3), 4.48 (s, 2H, -CH_2Ph), 6.91(d, J = 2.35 Hz, 1H, 4-H), 6.97 (dd, J = 8.61 Hz, J = 2.35 Hz, 1H, 6-H), 7.14-7.26 (m, 5H, phenyl protons), 7.38 (d, J = 9Hz, 1H, 7-H), 9.19 (brs, 1H, -NH).
6h'	3172 (NH), 1625 (C=N)	($CDCl_3$): 2.63 (s, 3H, CH_3), 4.46 (s, 2H, CH_2Ph), 7.24-7.28 (m, 6H, 6-H and phenyl protons), 7.45 (d, J = 8.8 Hz, 1H, 7- H), 7.52 (d, J = 2 Hz, 1H, 4- H), 9.60 (brs, 1H, NH).

overnight and then poured onto crushed ice. The white solid that separated was collected by filtration and crystallized from dioxane to obtain the 8-substituted 10-benzyl-4-methyl-1,2-dihydro-1-oxo-1,2,4-triazino[4,5-a]indoles (**5e-h**).

The mother liquors, from which triazinoindoles (**5e-h**) were isolated, on concentration yielded solid products which were recrystallised from ethanol to get various 5-substituted 3-benzyl-2-(5-methyl-1,3,4-oxadiazol-2-yl)indoles (**6e'-h'**). The physical data and spectral analyses of these triazinoindoles and oxadiazolylindoles are recorded in Table 2, Table 3 and Table 4.

3-Benzyl-2-(5-methyl-1,3,4-oxadiazol-2-yl)indoles (**6e-h**).

General Procedure.

Appropriate 3-benzylindole-2-carbohydrazide (**4a-d**) (0.01 mole) and triethylorthoacetate (50 ml) were heated under reflux for 12 hours. Excess of the reagent was removed under reduced pressure. The residue was treated with pet. ether and crystallized from ethanol to get 5-substituted 3-benzyl-2-(5-

methyl-1,3,4-oxadiazol-2-yl)indoles. The physical data and spectral analyses of these oxadiazolylindoles are recorded in Table 3 and Table 4.

REFERENCES AND NOTES

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