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SYNTHESIS OF ARYLSUBSTITUTED IMIDAZO[1,2-*b*]PYRIDAZINO[6,5-*e*]-1,2,4-TRIAZINE AND PYRIDAZINO[6,5-*a*]-1,2,4-TRIAZINO[2,3-*a*]BENZIMIDAZOLE

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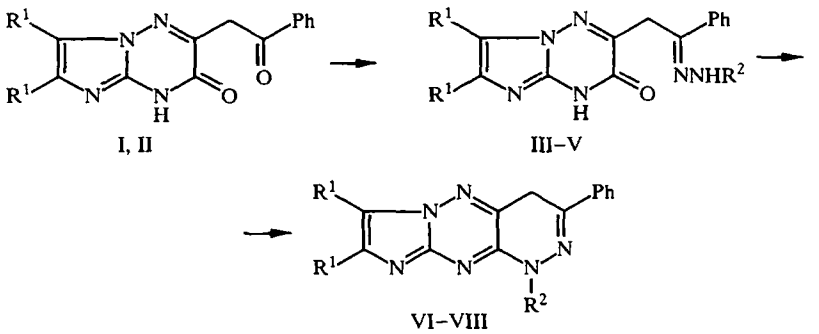
We earlier demonstrated that the reaction of 2-benzoylmethylsubstituted 6,7-diphenylimidazo[1,2-*b*]-1,2,4-triazin-4H-3-one (I) or 1,2,4-triazino[2,3-*a*]benzimidazol-4H-3-one (II) with primary amines produces alkyl(aryl)-iminophenethyl derivatives of imidazotriazine or triazinobenzimidazole that can cyclize to the corresponding 1,2,6,7-tetraphenylpyrrolo[2,3-*e*]imidazo[1,2-*b*]-1,2,4-triazine [1] and its benzo analogs [2].

The behavior of arylhetarylketones I and II in the reaction with phenyl- or *p*-tolylhydrazines was studied in order to expand their synthetic application.

Compound I reacts in boiling propan-2-ol (4 h) with phenylhydrazine (1:1 molar ratio) to form the phenylhydrazone of 2-benzoylmethyl-6,7-diphenylimidazo[1,2-*b*]-1,2,4-triazin-4H-3-one (III). Yield 65%; mp 246-247°C (from propan-2-ol). R_f 0.67. IR spectrum (KBr): $\nu_{C=N}$ 1600, $\nu_{C=O}$ 1620, ν_{NH} 3110, 3220 cm^{-1}. UV spectrum (DMF), λ_{max} (log ε): 345 (4.24).

Heating of mixture of ketone II (2 mmol) and phenylhydrazine (70 mmol) at 135-140°C for 4 h with subsequent 5-fold dilution with ether gives the phenylhydrazone of 2-benzoylmethyl-1,2,4-triazino[2,3-*a*]-benzimidazol-4H-3-one (IV). Yield 57%; mp 259-260°C (from DMF). R_f 0.55. IR spectrum (KBr): $\nu_{C=N}$ 1620, $\nu_{C=O}$ 1640, ν_{NH} 3055, 3180 cm^{-1}. UV spectrum (dioxane), λ_{max} (log ε): 325 (4.29), 410 (3.75).



$^1R = C_6H_5$, $^1R + {}^1R$ = condensed benzene core, ^2R = Ph, 4-$CH_3C_6H_4$.

Refluxing of equimolar amounts of compound II and *p*-tolylhydrazine hydrochloride in hexan-1-ol containing sodium methoxide (2 h) produces 4-methylphenylhydrazone of 2-benzoylmethyl-1,2,4-triazino[2,3-*a*]-benzimidazol-4H-3-one (V). Yield 52%; mp 262-263°C (from DMF). R_f 0.68. IR spectrum (KBr): $\nu_{C=N}$ 1615, $\nu_{C=O}$ 1640, ν_{NH} 3050, 3170 cm^{-1}.

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Cyclization of hydrazones III-V in boiling $POCl_3$ (3 h) produced the novel compounds 1,3,7,8-tetraphenylimidazo[1,2-*b*]pyridazino[6,5-*e*]-1,2,4-triazine (VI) and 1,3-diarylpyridazino[6,5-*e*]-1,2,4-triazino[2,3-*a*]-benzimidazoles (VII and VIII).

1,3,7,8-Tetraphenylimidazo[1,2-*b*]pyridazino[6,5-*e*]-1,2,4-triazine (VI). Yield 59%; mp 238-239°C (from aqueous DMF). R_f 0.71. PMR spectrum (CF_3COOH): 3.82 (2H, s, CH_2), 7.12 ppm (20H, m, H_{arom}).

1,3-Diphenylpyridazino[6,5-*e*]-1,2,4-triazino[2,3-*a*]benzimidazole (VII); mp 335-336°C (from aqueous DMF). R_f 0.62. Mass spectrum, *m/z* (I_{rel}, %): 377 (26), 376 (100), M^+; 375 (53), $[M-H]^+$; 348 (3), $[M-N_2]^+$; 299 (4), $[M-Ph]^+$; 273 (22), $[M-PhCN]^+$; 272 (23), $[(M-H)-PhCN]^+$; 103 (3), $[PhCN]^+$; 102 (6), $[PhCCH]^+$; 77 (17), $[Ph]^+$. HRMS (M/ΔM 10000, PFK): M^+ 376.1416, $C_{23}H_{16}N_6$; $[M+H]^+$ 375.1361, $C_{23}H_{15}N_6$.

1-(4-Methyphenyl)-3-phenylpyridazino[6,5-*e*]-1,2,4-triazino[2,3-*a*]benzimidazole (VIII). Yield 89%; mp 313-315°C (from aqueous DMF). R_f 0.76. PMR spectrum (CF_3COOH): 1.87 (3H, s, CH_3), 4.53 (2H, s, CH_2), 7.25 ppm (13H, m, H_{arom}).

Elemental analyses of III-VIII agree with the calculated values.

REFERENCES

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