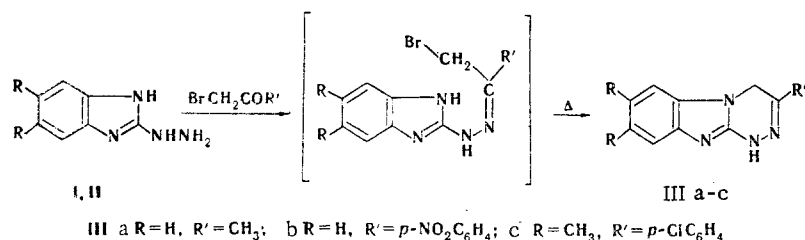


NEW SYNTHESIS OF 1,4-DIHYDRO-1,2,4-TRIAZINO [3,4-*a*]  
BENZIMIDAZOLES

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A 1,2,4-triazine ring is formed (evidently through a step involving the formation of hydrazones) when 2-hydrazinobenzimidazoles (I, II) are refluxed with bromoacetone, p-nitrophenacyl bromide, or p-chlorophenacyl bromide in ethanol or dimethylformamide (DMF) in the presence of triethylamine. This procedure was used to obtain 1,4-dihydro-3-methyl-1,2,4-triazino [3,4-*a*] benzimidazole (IIIa) [with mp 294-295° (from aqueous DMF) in 43% yield], the 3-(p-nitrophenyl)-substituted derivative (IIIb) [with mp 298-299° (dec., from aqueous DMF) in 51% yield], and the 3-(p-chlorophenyl)-7,8-dimethyl-substituted derivative (IIIc) [with mp 309-310° (dec., from aqueous DMF) in 45% yield].



The individuality of IIIa-c was confirmed by thin-layer chromatography on silica gel (Silufol UV-254) in a chloroform-acetic acid (5:0.1) system. The structure was confirmed by the results of elementary analysis, the IR and PMR spectra, and alternative synthesis from the corresponding 1-acylmethyl-2-chlorobenzimidazole and hydrazine hydrate.

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