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## SHORT COMMUNICATIONS

# Oxidative Addition of *N*-Aminophthalimide to Cinnamic Aldehyde Phthaloylhydrazone

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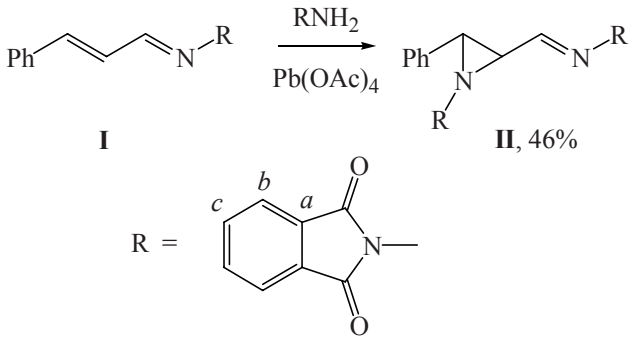
The oxidative addition of a series of *N*-aminoheterocycles, first of all, of N aminophthalimide and various 3-aminoquinazolinones, to olefins proceeds at the π-bond and serves as a general method of synthesis of *N*-aminoaziridine derivatives [1–3]. Under these conditions in the azo group the unshared electron pair is attacked on one of the nitrogen atoms leading to the formation of polynitrogen 1,3-dipoles, N-aminoazimines [4]. A question arises, what path would take this reaction with the C=N group? No published data exist on such reactions with imines, and our attempts to carry out the oxidative addition of N-aminophthalimide to compounds conteining isolated, conjugated, or cumulated C=N bonds have been unsuccessful up till now [5]. Moreover, it turned out that in compounds containing a fragment C=C–C=N in this reaction did not enter not only the C=N bond, but also the commonly active conjugated C=C bond [6]. In particular, the investigation of oxidative aziridination with phthalimide of conjugated alkenylpyrazolines and alkenylpyrazoles [6] showed that the taken alkenylpyrazolines did not react, although the similarly substituted alkenylpyrazoles gave the corresponding aziridines in good yields. Evidently this result originated from the inclusion of the C=N bond into an aromatic heterocycle where this bond considerably lost its individual character.

Yet by an example of hydrazone **I** we were fortunate to carry out the first successful aminoaziridination of a compound containing a fragment C=C–C=N whose C=N bond was not included in an aromatic system.

The composition of obtained aziridine **II** was confirmed by elemental analysis and by the presence in its mass spectrum of the molecular ion peak, and its




structure was proved by $^1$H and $^{13}$C NMR spectra where the signals of all fragments of the molecule were present in the corresponding regions and with the expected multiplicity. In keeping with the $^1$H NMR spectrum due to the typical for the N-aminoaziridine derivatives slow inversion of the endocyclic nitrogen [7] this compound existed in DMSO in the form of two invertomers in a ratio ~8:1. Inasmuch as in these aziridines the phthalimide substituent commonly deshielded the syn-located protons, the main invertomer contained the phthalimide and phenyl groups on the opposite sides of the plane of the three-membered ring, in agreement with the steric reasons.

Still on attempt to perform the oxidative addition of N-aminophthalimide to analogous compounds containing a C=C–C=N fragment, cinnamic aldehyde phenylimine (R = Ph) and to its dimethylhydrazone (R = $Me_2N$), we did not detect in the $^1$H NMR spectra of the reaction mixtures the characteristic signals of the aziridine protons.

**2-{[(1*E*,2*E*)-3-Phenylprop-2-en-1-ylidene]-amino}-1*H*-isoindole-1,3(2*H*)-dione (I)**. To a suspension of 1.62 g (10 mmol) of *N*-aminophthalimide [8] in

30 ml of ethanol was added 1.45 g (11 mmol) of cinnamic aldehyde, and the mixture was boiled at reflux for 2 h. The precipitate settled on cooling was filtered off and dried in air. Yield 2.10 g (76%). Light-yellow crystalline powder, mp 197–198°C (mp 199–200°C [8]). $^1$H NMR spectrum ($CDCl_3$), δ, ppm: 7.1–7.2 m (2H, CH=CH), 7.35–7.45 m (3H, $H^m + H^n$), 7.50–7.57 m (2H, $H^O$), 7.77–7.82 m (2H, Ht), 7.90–7.96 m (2H, Ht), 9.29 d.d (1H, CH=N, *J* 5.5 and 2.7 Hz).

**2-(2-{(*E*)-[(1,3-Dioxo-1,3-dihydro-2*H*-isoindol-2-yl)imino]methyl-3-phenyl-aziridin-1-yl)-1*H*-isoindole-1,3(2*H*)-dione (II)**. To a solution of 2.76 g (10 mmol) of hydrazone **I** in 50 ml of dichloromethane cooled to 0°C where was dispersed 5.0 g of potassium carbonate was added at stirring by small portions within 15 min 1.62 g (10 mmol) of *N*-aminophthalimide and 4.43 g (10 mmol) of lead tetraacetate. On completion of addition the mixture was stirred for 10 min more, then it was filtered, the precipitate of inorganic salts was washed with 20 ml of dichloromethane. The solution was evaporated in a vacuum to a volume of ~30 ml, cooled to–5°C, the separated precipitate was filtered off. Yield 2.00 g (46%). Colorless powder, mp 178–179°C. According to $^1$H NMR spectrum the product existed in the form of two invertomers in a ratio ~8:1. $^1$H NMR spectrum (DMSO-$d_6$), δ, ppm (*J*, Hz), main invertomer: 3.76 d.d (1H, CHN, *J* 7.3 and 5.5), 4.50 d (1H, PhCHN, *J* 5.5), 7.33–7.45 m (3H, $H^m + H^p$), 7.51 d (2H, $H^o$, *J* 7.3), 7.79–7.83 m (8H, Ht), 8.68 d (CH=N, *J* 7.3); minor invertomer: 4.14 d (1H, PhCHN, *J* 5.5), 4.70 d.d (1H, CHN, *J* 7.3 and *J* 5.5), 7.67–7.95 m (8H, Ht), 8.74 d (CH=N, *J* 7.3). $^{13}$C NMR spectrum of the main invertomer (DMSODMCO-$d_6$), δ, ppm: 48.53 (CHN), 49.26 (CHN), 122.95 and 123.50 ($2C_{Ht}$, $C^b$), 127.32 and 128.48 (Ph, $C^o$ and $C^m$), 128.30 (Ph, $C^p$), 129.61 and 130.06 ($2C_{Ht}$, $C^a$), 134.51 and 135.00 ($2C_{Ht}$, $C^c$), 135.18 (Ph, $C^i$), 157.29 (C=N), 163.99 and 164.65 (2CO). Mass spectrum, *m/z* ($I_{rel}$, %): 436 (1.3) $[M]^+$, 290 (32) $[M - R]^+$, 289 (79) $[M - RH]^+$, 236 (58), 187 (16), 185 (16), 147 (98) $[RH]^+$, 132 (32), 130 (28), 105 (26), 104 (99) $[C_6H_4CO]^+$, 103 (44), 90 (18), 77 (38), 76 (100), 75 (28), 74 (25), 63 (12), 51 (21), 50 (87), 39 (17), 38 (18), 37 (17). Found, %: C 68.71; H 3.73; N 12.80. $C_{25}H_{16}N_4O_4$. Calculated, %: C 68.81; H 3.67; N 12.84. *M* 436.43.

$^1$H (300 MHz) and $^{13}$C (75.4 MHz) NMR spectra were registered on a spectrometer Bruker DPX-300. Mass spectrum was measured on an MKh-1321 instrument (energy of ionizing electrons 70 eV). Elemental analysis was performed on an automatic CHN-analyzer Hewlett Packard 185B.

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