

Synthesis of 2-Aryl-3*H*-naphtho[1,2-*d*]imidazoles Containing an Adamantane Fragment

T. A. Frolenko^a, E. S. Semichenko^a, A. A. Kondrasenko^b, N. A. Gavrilova^a, and G. A. Suboch^a

^a Siberian State Technological University, pr. Mira 82, Krasnoyarsk, 660049 Russia
e-mail: organic@sibgtu.ru

^b Institute of Chemistry and Chemical Technology, Siberian Branch, Russian Academy of Sciences,
Akademgorodok 50/24, Krasnoyarsk, 660036 Russia

Received August 22, 2012

Abstract—*N*²-[1-(1-Adamantyl)alkyl]naphthalene-1,2-diamines reacted with benzoyl chlorides in chloroform in the presence of triethylamine to give *N*-{2-[1-(1-adamantyl)alkylamino]naphthalen-1-yl}benzamides which underwent intramolecular cyclization to 2-aryl-3*H*-naphtho[1,2-*d*]imidazoles on heating in toluene in the presence of *p*-toluenesulfonic acid. 3-[(1-Adamantyl)methyl]-2-(3-nitrophenyl)-3*H*-naphtho[1,2-*d*]imidazole was synthesized from *N*²-[1-(1-adamantyl)methyl]naphthalene-1,2-diamine and 3-nitrobenzaldehyde.

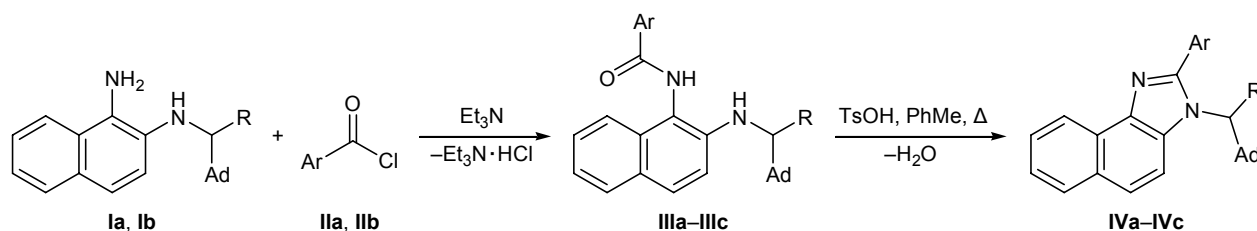
DOI: 10.1134/S1070428013120075

Some 3*H*-naphtho[1,2-*d*]imidazoles were reported as nonsteroidal antiinflammatory agents [1]; they also exhibited antihypertensive activity [2]. It is known that introduction of an aromatic substituent into the 2-position of 3*H*-naphtho[1,2-*d*]imidazoles may enhance biological activity [2] and that the presence of a lipophilic adamantyl fragment in their molecules facilitates penetration through biological membranes [3]. We previously synthesized *N*²-[1-(1-adamantyl)alkyl]naphthalene-1,2-diamines **Ia** and **Ib** by reduction of *N*-[1-(1-adamantyl)alkyl]-1-nitronaphthalen-2-amines, and compounds **Ia** and **Ib** were then used to obtain 2-alkyl-3*H*-naphtho[1,2-*d*]imidazoles possessing an adamantane fragment [4]. While continuing our studies on heterocyclizations of naphthalene-1,2-diamines **Ia** and **Ib**, in the present work we examined their reactions with benzoyl chlorides **IIa** and **IIb** (Scheme 1).

By heating diamines **Ia** and **Ib** with benzoyl chlorides **IIa** and **IIb** and triethylamine in chloroform we obtained the corresponding benzamides **IIIa–IIIc**. In the ¹³C NMR spectra of **IIIb** and **IIIc** (CDCl₃), the carbonyl carbon signal appeared at δ_C 165–167 ppm. Benzamides **IIIa–IIIc** were subjected to intramolecular cyclization to previously unknown 2-aryl-3*H*-naphtho[1,2-*d*]imidazoles **IVa–IVc** by heating in boiling toluene in the presence of *p*-toluenesulfonic acid. The progress of the cyclization was monitored by TLC. After removal of the solvent, the products were isolated by column chromatography.

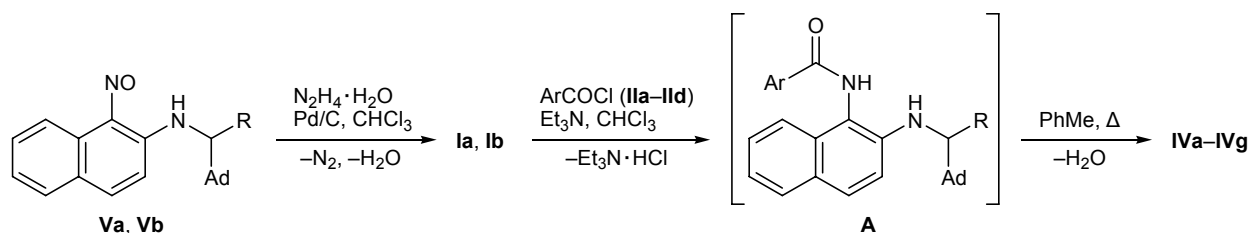
Diamines **Ia** and **Ib** readily undergo oxidation; therefore, it is advisable to perform their benzylation *in situ* immediately after the reduction of *N*-[1-(1-adamantyl)alkyl]-1-nitronaphthalen-2-amines **Va** and **Vb** with hydrazine hydrate over Pd/C in chloroform (Scheme 2). Following this procedure, we have synthe-

Scheme 1.



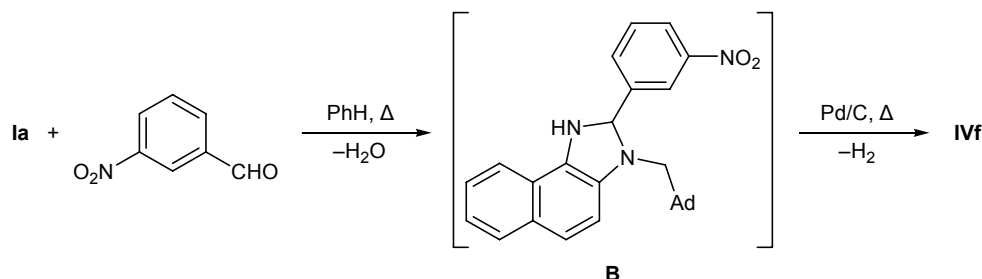
I, R = H (**a**), Me (**b**); **II**, Ar = Ph (**a**), 4-ClC₆H₄ (**b**); **III**, **IV**, R = H, Ar = Ph (**a**); R = Me, Ar = Ph (**b**), 4-ClC₆H₄ (**c**).

Scheme 2.



V, R = H (a), Me (b); IV, R = H (a, d-f), Me (b, c, g); Ar = Ph (a, b), 4-ClC₆H₄ (c, d), 4-O₂NC₆H₄ (e), 3-O₂NC₆H₄ (f, g).

Scheme 3.



sized 2-aryl-3H-naphtho[1,2-d]imidazoles **IVa–IVg**. 3-Alkyl-2-aryl-3H-naphtho[1,2-d]imidazoles were successfully obtained previously via condensation of aromatic aldehydes with naphthalene-1,2-diamines and subsequent dehydrogenation [1]. We examined the reaction of compound **Ia** with 3-nitrobenzaldehyde (Scheme 3) and thus isolated a compound which was identical to 3H-naphtho[1,2-d]imidazole **IVf** synthesized from diamine **Ia** and 3-nitrobenzoyl chloride.

The mass spectra of compounds **IVa**, **IVb**, and **IVf** contained the molecular ion peaks with m/z values corresponding to the assumed structure. In addition, a ion peak with m/z 135 due to adamantyl cation was observed; its further fragmentation gave ions with m/z 93 and 79, which are typical of 1-alkyl-substituted adamantanes regardless of the alkyl group nature [5]. The elemental compositions of **IVc–IVe** and **IVg** were consistent with the calculated values.

Compounds **IVa** and **IVd–IVf** displayed in the ¹H and ¹³C NMR spectra signals from the AdCH₂ methylene group at δ 4.19–4.25 ppm (s) and δ_c 56.1–56.5 ppm. The corresponding fragment in the spectra of **IVc** and **IVg** was represented by a doublet at δ 1.9 ppm (CH₃, δ_c 13 ppm) and a quartet at δ 4.4 ppm (CH, δ_c 62 ppm). Signals from the adamantyl substituent were also present. Signals from the benzene and naphthalene fragments were assigned using ¹H–¹H COSY, HMBC, and HSQC homo- and heteronuclear correlation techniques (see figure). The 4'-H and 6'-H

signals in the spectra of **IVf** and **IVg** were assigned on the basis of calculations of chemical shifts according to the additivity scheme with the following increments for the nitro group: Z_{ortho} = 0.93, Z_{para} = –0.39 [6].

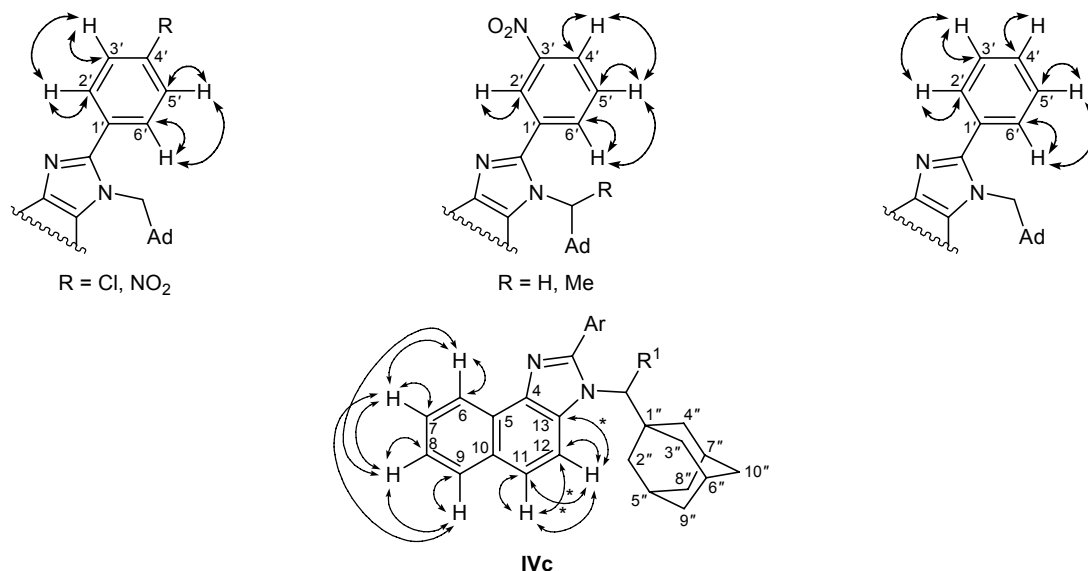
EXPERIMENTAL

The ¹H and ¹³C NMR spectra were recorded at the Krasnoyarsk Regional Joint Center (Siberian Branch, Russian Academy of Sciences) on a Bruker Avance III spectrometer (600.13 and 125.86 MHz, respectively) at 223 K using the solvent signals as reference (for atom numbering in the description of the NMR spectra, see figure). The mass spectra were obtained on a Finnigan MAT-8200 mass spectrometer. GC/MS analyses were performed on an Agilent Technologies 6890N instrument (HP-5MS quartz capillary column, 30 m × 0.25 mm, film thickness 0.33 μ m; electron impact, 70 eV); the components were identified in the total ion current mode.

The products were isolated by column chromatography on Silicagel L 100/400 (Chemapol). Analytical thin-layer chromatography was performed on Sorbfil PTSKh-AF-V plates with a UV indicator; eluent heptane–ethyl acetate (9:1).

Naphthalene-1,2-diamines **Ia** and **Ib** were synthesized according to the procedure described in [7].

N-{2-[1-(1-Adamantyl)alkylamino]naphthalen-1-yl}benzamides IIIa–IIIc (general procedure).



^1H - ^1H COSY, ^1H - ^{13}C HSQC, and ^1H - ^{13}C HMBC correlations in compounds **IVa** and **IVc-IVg**. Weak ^1H - ^{13}C HMBC correlations in **IVc** are marked with an asterisk.

A mixture of 1.6 mmol of naphthalene-1,2-diamine **Ia** or **Ib**, 3 mmol of benzoyl chloride **IIa** or **IIb**, and 0.22 mL (1.5 mmol) of triethylamine in 30 mL of chloroform was heated for 2 h under reflux (TLC). The solution was evaporated under reduced pressure, and the residue was purified by chromatography in a 2 × 50-cm column charged with 50 g of silica gel using hexane-ethyl acetate (9:1) as eluent.

N-{2-[(1-Adamantyl)methylamino]naphthalen-1-yl}benzamide (IIIa). Yield 0.43 g (65%), light yellow crystals soluble in acetone, ethanol, and chloroform; mp 169°C. Found, %: C 82.15; H 6.94; N 6.43. C₂₈H₃₀N₂O. Calculated, %: C 81.91; H 7.37; N 6.82.

N-{2-[1-(1-Adamantyl)ethylamino]naphthalen-1-yl}benzamide (IIIb). Yield 0.37 g (57%), light yellow crystals soluble in acetone, ethanol, and chloroform; mp 168°C. ^1H NMR spectrum, δ , ppm: 1.16 d (3H, CH₃, J = 6.77 Hz), 1.51–1.73 m (12H, CH₂, Ad), 2.02 m (3H, CH, Ad), 3.31 m (1H, CH), 7.21–7.24 m (2H, 3-H, 7-H), 7.42 m (1H, 8-H), 7.56–7.61 m (3H, 3'-H, 5'-H, 9'-H), 7.64–7.66 m (1H, 4'-H), 7.74–7.76 m (2H, 4-H, 6-H), 8.07 m (2H, 2'-H, 6'-H). ^{13}C NMR spectrum (CDCl₃), δ_{C} , ppm: 15.2 (CH₃), 28.5 (3C, CH, Ad), 37.2 (3C, CH₂, Ad), 36.8 (C, Ad), 38.8 (3C, CH₂, Ad), 57.8 (CH), 112.1 (C¹), 115.1 (C³), 120.0 (C⁹), 121.8 (C⁷), 127.3 (C¹⁰), 127.3 (C^{2'}, C^{6'}, C⁸), 128.0 (C⁶, C⁴), 129.0 (C^{3'}, C^{5'}), 131.8 (C⁵), 132.0 (C^{4'}), 134.5 (C^{1'}), 143.6 (C²), 166.6 (C=O). Found, %: C 82.27; H 7.14; N 6.33. C₂₉H₃₂N₂O. Calculated, %: C 82.04; H 7.60; N 6.60.

N-{2-[1-(1-Adamantyl)ethylamino]naphthalen-1-yl}-4-chlorobenzamide (IIIc). Yield 0.46 g (66%), light yellow crystals soluble in acetone, ethanol, and chloroform; mp 170°C. ^1H NMR spectrum, δ , ppm: 1.16 d (3H, CH₃, J = 6.35 Hz), 1.51–1.73 m (12H, CH₂, Ad), 2.00 m (3H, CH, Ad), 3.31 m (1H, CH), 7.21–7.24 m (2H, 3-H, 7-H), 7.41–7.44 m (1H, 8-H), 7.55 m (2H, 3'-H, 5'-H), 7.61–7.64 m (1H, 9-H), 7.74–7.76 m (2H, 4-H, 6-H), 8.00 m (2H, 2'-H, 6'-H). ^{13}C NMR spectrum, δ_{C} , ppm: 15.1 (CH₃), 28.4 (3C, CH, Ad), 37.2 (3C, CH₂, Ad), 36.8 (C, Ad), 38.8 (3C, CH₂, Ad), 57.7 (CH), 112.1 (C¹), 115.1 (C³), 119.8 (C⁹), 121.9 (C⁷), 127.0 (C¹⁰), 127.2 (C⁸), 128.7 (C^{2'}, C^{6'}), 128.4 (C⁴), 128.7 (C⁶), 129.3 (C^{3'}, C^{5'}), 131.7 (C⁵), 138.4 (C^{4'}), 132.6 (C^{1'}), 143.0 (C²), 165.6 (C=O). Found, %: C 75.48; H 6.16; Cl 7.61 N 6.83. C₂₉H₃₁ClN₂O. Calculated, %: C 75.88; H 6.81; Cl 7.72; N 6.10.

2-Aryl-3H-naphtho[1,2-d]imidazoles IVa-IVf (general procedure). *a.* A mixture of 0.6 mmol of benzamide **IIIa-IIIc** and 0.05 g of *p*-toluenesulfonic acid in 30 mL of toluene was heated for 48 h under reflux. The mixture was cooled, washed with a 5% solution of sodium carbonate and water, and evaporated, and the residue was purified by column chromatography (2 × 50 cm) on 50 g of silica gel using hexane-ethyl acetate (4:1) as eluent.

b. To a mixture of 1.6 mmol of compound **Va** or **Vb** and 0.5 g of 0.5% Pd/C in 30 mL of chloroform we added under vigorous stirring 0.3 mL (7.2 mmol) of

95% hydrazine hydrate, and the mixture was stirred for 1 h. The catalyst was filtered off, 3 mmol of benzoyl chloride **Ila–IId** and 0.22 mL (1.5 mmol) of triethylamine were added in succession to the filtrate, and the mixture was heated for 2 h under reflux. The mixture was then evaporated under reduced pressure on a rotary evaporator, 30 mL of toluene was added to the residue, and the mixture was heated for 48 h under reflux, filtered, and evaporated. An analytical sample of the product was obtained by chromatographic purification in a 2×50-cm column charged with 50 g of silica gel using hexane–ethyl acetate (4:1) as eluent.

3-[(1-Adamantyl)methyl]-2-phenyl-3H-naphtho[1,2-*d*]imidazole (IVa). Yield 0.15 g (64%) (*a*), 0.13 g (21%) (*b*); light yellow crystals soluble in acetone, ethanol, and chloroform; mp 230°C. ¹H NMR spectrum, δ, ppm: 1.30–1.75 m (12H, CH₂, Ad), 1.84 m (3H, CH, Ad), 4.22 s (2H, CH₂), 7.52 m (1H, 4'-H), 7.53 m (1H, 8-H), 7.55 m (2H, 2'-H, 6'-H), 7.62 d (1H, 12-H), 7.66 m (1H, 7-H), 7.73 d (1H, 11-H), 7.75 m (2H, 3'-H, 5'-H), 7.97 m (1H, 9-H), 8.77 m (1H, 6-H). ¹³C NMR spectrum, δ_C, ppm: 28.1 (C^{5''}, C^{7''}), 36.4 (C^{8''}, C^{9''}, C^{10''}), 36.9 (C^{1''}), 41.1 (C^{2''}, C^{3''}, C^{4''}), 56.1 (CH₂), 112.3 (C¹²), 122.0 (C⁶), 123.2 (C¹¹), 124.5 (C⁸), 126.4 (C⁷), 127.0 (C⁵), 128.3 (C⁹), 128.7 (C^{2'}, C^{6'}), 129.1 (C^{4'}), 130.2 (C¹⁰), 130.1 (C^{3'}, C^{5'}), 132.3 (C^{1'}), 133.1 (C⁴, C¹³), 152.5 (C²). Mass spectrum, *m/z* (*I*_{rel}, %): 392 (9) [*M*]⁺, 135 (20), 93 (15), 79 (100), 55 (38). Found, %: C 85.90; H 7.22; N 7.21 C₂₈H₂₈N₂. Calculated, %: C 85.67; H 7.19; N 7.14. *M* 392.54.

3-[1-(1-Adamantyl)ethyl]-2-phenyl-3H-naphtho[1,2-*d*]imidazole (IVb). Yield 0.14 g (57%) (*a*), 0.4 g (35%) (*b*); mp 168°C. Mass spectrum, *m/z* (*I*_{rel}, %): 406 (7) [*M*]⁺, 327 (5), 178 (10), 135 (100), 113 (12), 93 (25), 79 (18), 67 (5), 55 (5), 44 (12), 32 (11). Found, %: C 85.70; H 7.47; N 7.05. C₂₉H₃₀N₂. Calculated, %: C 85.67; H 7.44; N 6.89. *M* 406.57.

3-[1-(1-Adamantyl)ethyl]-2-(4-chlorophenyl)-3H-naphtho[1,2-*d*]imidazole (IVc). Yield 0.13 g (51%) (*a*), 0.13 g (31%) (*b*); light yellow crystals soluble in acetone, ethanol, and chloroform; mp 171°C. ¹H NMR spectrum, δ, ppm: 1.30–1.71 m (12H, CH₂, Ad), 1.85 m (3H, CH, Ad), 4.19 s (2H, CH₂), 7.54 m (1H, 8-H), 7.55 m (2H, 2'-H, 6'-H), 7.61 m (1H, 12-H), 7.67 m (1H, 7-H), 7.71 m (2H, 3'-H, 5'-H), 7.98 m (1H, 9-H), 8.74 m (1H, 6-H). ¹³C NMR spectrum, δ_C, ppm: 13.7 (CH₃), 28.2 (C^{5'}, C^{7''}), 36.5 (C^{8''}, C^{9''}, C^{10''}), 39.4 (C^{1''}), 39.9 (C^{2''}, C^{3''}, C^{4''}), 62.7 (CH), 115.0 (C¹²), 122.0 (C⁶), 122.8 (C¹¹), 124.7 (C⁸), 126.4 (C⁷), 127.1 (C¹⁰), 128.0 (C⁹), 129.0

(C^{2'}, C^{6'}), 129.8 (C¹³), 131.3 (C^{1'}), 131.6 (C^{3'}, C^{5'}), 135.3 (C^{4'}), 152.3 (C²). Found, %: C 79.27; H 6.59; Cl 8.00; N 6.38. C₂₉H₂₉ClN₂. Calculated, %: C 78.98; H 6.63; Cl 8.04; N 6.35.

3-[(1-Adamantyl)methyl]-2-(4-chlorophenyl)-3H-naphtho[1,2-*d*]imidazole (IVd). Yield 0.27 g (40%) (*b*), mp 230°C. ¹H NMR spectrum, δ, ppm: 1.30–1.71 m (12H, CH₂, Ad), 1.85 m (3H, CH, Ad), 4.19 s (2H, CH₂), 7.54 m (1H, 8-H), 7.55 m (2H, 2'-H, 6'-H), 7.61 m (1H, 12-H), 7.67 m (1H, 7-H), 7.71 m (2H, 3'-H, 5'-H), 7.98 m (1H, 9-H), 8.74 m (1H, 6-H). ¹³C NMR spectrum, δ_C, ppm: 28.1 (C^{5''}, C^{6''}, C^{7''}), 36.4 (C^{8''}, C^{9''}, C^{10''}), 36.9 (C^{1''}), 41.2 (C^{2''}, C^{3''}, C^{4''}), 56.2 (CH₂), 112.2 (C¹²), 121.9 (C⁶), 123.5 (C¹¹), 124.7 (C⁸), 126.6 (C⁷), 127.0 (C⁵), 128.3 (C⁹), 129.1 (C^{2'}, C^{6'}), 130.2 (C¹⁰), 130.8 (C^{1'}), 131.2 (C^{3'}, C^{5'}), 133.1 (C¹³), 135.3 (C^{4'}, C⁴), 151.2 (C²). Found, %: C 79.17; H 6.54; Cl 8.16; N 6.23. C₂₈H₂₇ClN₂. Calculated, %: C 78.76; H 6.37; Cl 8.30; N 6.56.

3-[(1-Adamantyl)methyl]-2-(4-nitrophenyl)-3H-naphtho[1,2-*d*]imidazole (IVe). Yield 0.3 g (43%) (*b*), yellow crystals soluble in acetone, ethanol, and chloroform; mp 220°C. ¹H NMR spectrum, δ, ppm: 1.28–1.60 m (12H, CH₂, Ad), 1.84 m (3H, CH, Ad), 4.23 s (2H, CH₂), 7.56 m (1H, 8-H), 7.62 m (1H, 12-H, *J* = 8.9 Hz), 7.69 m (1H, 7-H), 7.77 m (1H, 11-H), 7.97 m (2H, 2'-H, 6'-H), 7.98 m (1H, 9-H), 8.42 m (2H, 3'-H, 5'-H), 8.74 m (1H, 6-H). ¹³C NMR spectrum, δ_C, ppm: 28.0 (C^{5''}, C^{6''}, C^{7''}), 36.3 (C^{8''}, C^{9''}, C^{10''}), 37.1 (C^{1''}), 41.2 (C^{2''}, C^{3''}, C^{4''}), 56.5 (CH₂), 112.1 (C¹²), 121.8 (C⁶), 124.0 (C^{3'}, C^{5'}), 124.3 (C¹¹), 125.0 (C⁸), 126.9 (C⁷), 127.0 (C⁵), 128.4 (C⁹), 130.4 (C¹⁰), 130.7 (C^{2'}, C^{6'}), 133.6 (C^{1'}), 138.4 (C¹³), 138.7 (C⁴), 147.9 (C^{4'}), 149.7 (C²). Found, %: C 77.24; H 5.89; N 9.13. C₂₈H₂₇N₃O₂. Calculated, %: C 76.86; H 6.22; N 9.60.

3-[(1-Adamantyl)methyl]-2-(3-nitrophenyl)-3H-naphtho[1,2-*d*]imidazole (IVf). Yield 0.55 g (80%) (*b*), 0.21 g (31%) (according to [1]); light yellow crystals soluble in acetone, ethanol, and chloroform, and DMSO; mp 158°C. ¹H NMR spectrum, δ, ppm: in CDCl₃: 1.30–1.61 m (12H, CH₂, Ad), 1.86 m (3H, CH, Ad), 4.25 s (2H, CH₂), 7.56–7.58 m (1H, 8-H), 7.63 m (1H, 12-H), 7.69 m (1H, 7-H), 7.77–7.79 m (2H, 5-H, 11-H), 7.97 m (1H, 9-H), 8.18–8.19 m (1H, 6'-H), 8.40 m (1H, 4'-H), 8.66 s (1H, 2'-H), 8.75 br.s (1H, 6-H); in DMSO-*d*₆: 1.21 m (6H, CH₂, Ad), 1.32–1.49 m (6H, CH₂, Ad), 1.71 (3H, CH, Ad), 4.35 s (2H, CH₂), 7.50–7.54 m (1H, 8-H), 7.6–7.66 m (1H, 7-H), 7.78–7.81 m (1H, 12-H), 7.85–7.90 m (1H, 5'-H), 7.93–7.96 m (1H, 11-H), 8.01–8.04 m (1H, 9-H), 8.32–

8.35 m (1H, 6'-H), 8.37–8.40 m (1H, 4'-H), 8.50–8.53 m (1H, 6-H), 8.64 s (1H, 2'-H). ^{13}C NMR spectrum, δ_{C} , ppm: 28.0 ($\text{C}^{5''}$, $\text{C}^{6''}$, $\text{C}^{7''}$), 36.3 ($\text{C}^{8''}$, $\text{C}^{9''}$, $\text{C}^{10''}$), 37.1 ($\text{C}^{1''}$), 41.2 ($\text{C}^{2''}$, $\text{C}^{3''}$, $\text{C}^{4''}$), 56.5 (CH_2), 112.1 (C^{12}), 121.9 (C^6), 123.9 ($\text{C}^{4'}$), 124.2 (C^{11}), 124.6 ($\text{C}^{2'}$), 125.0 (C^8), 126.9 (C^7), 127.0 (C^{10}), 128.4 (C^9), 130.0 ($\text{C}^{5'}$), 130.4 ($\text{C}^{1'}$), 133.3 (C^{13}), 135.9 ($\text{C}^{6'}$), 148.3 (C^4), 149.5 ($\text{C}^{3'}$, C^2). Mass spectrum, m/z (I_{rel} , %): 437 (70) [M] $^+$, 407 (100), 281 (15), 259 (15), 207 (30), 156 (27), 135 (30), 114 (12), 93 (20), 79 (18), 67 (12), 55 (5), 44 (13). Found, %: C 77.05; H 6.40; N 9.74. $\text{C}_{28}\text{H}_{27}\text{N}_3\text{O}_2$. Calculated, %: C 76.86; H 6.22; N 9.60. M 437.54.

3-[1-(1-Adamantyl)ethyl]-2-(3-nitrophenyl)-3H-naphtho[1,2-*d*]imidazole (IVg). Yield 0.5 g (70%) (*b*), light yellow crystals soluble in acetone, ethanol, and chloroform; mp 148°C. ^1H NMR spectrum, ppm: 1.44–1.60 m (12H, CH_2 , Ad), 1.91 m (3H, CH, Ad), 1.91–1.95 d (3H, CH_3 , $J = 7.40$ Hz), 4.41 q (1H, CH), 7.56 m (1H, 8-H), 7.68 m (1H, 7-H), 7.73 m (1H, 11-H), 7.78 m (1H, 5'-H), 7.83 (1H, 12-H), 7.97 m (1H, 9-H), 8.06 m (1H, 6'-H), 8.41 m (1H, 4'-H), 8.56 m (1H, 2'-H), 8.75 (1H, 6-H). ^{13}C NMR spectrum, δ_{C} , ppm: 13.7 (CH_3), 28.1 ($\text{C}^{5''}$, $\text{C}^{6''}$, $\text{C}^{7''}$), 36.4 ($\text{C}^{8''}$, $\text{C}^{9''}$, $\text{C}^{10''}$), 39.5 ($\text{C}^{1''}$), 39.9 ($\text{C}^{2''}$, $\text{C}^{3''}$, $\text{C}^{4''}$), 63.3 (CH), 114.9 (C^{12}), 122.1 (C^6), 123.8 ($\text{C}^{4'}$), 124.2 (C^{11}), 125.2 ($\text{C}^{2'}$),

125.2 (C^8), 126.9 (C^7), 128.1 (C^9), 130.0 (C^4 , $\text{C}^{5'}$, $\text{C}^{1'}$), 136.3 ($\text{C}^{6'}$). Found, %: C 77.47; H 5.96; N 8.83. $\text{C}_{29}\text{H}_{29}\text{N}_3\text{O}_2$. Calculated, %: C 77.13; H 6.47; N 9.31.

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