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A CONVENIENT SYNTHESIS OF UNSYMMETRICAL N,N'-DISUBSTITUTED α,ω-DIAMINOALKANES

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ABSTRACT: A general procedure is described for regiospecific construction of unsymmetrical N-alkyl (or aralkyl)-N'-aryl-α,ω-diaminoalkanes **3** (n=2,3,4) by reduction of N-(ω-arylaminoalkyl)amides **2** with borane. Compounds **2** are readily obtained by condensation of N-(ω-haloalkyl)amides **1** with aromatic amines.

In the course of our research on nitrogen heterocycles, we recently required to prepare several unsymmetrical N,N'-disubstituted α,ω-diaminoalkanes **3** (n= 2,3,4)

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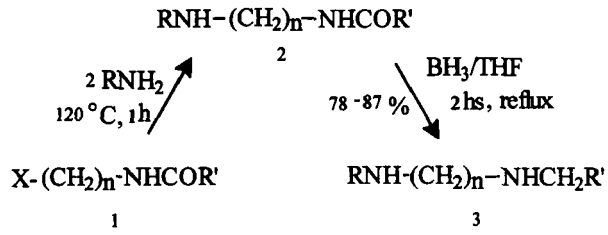
Scheme



Compound (1-3)	n	R	R'	Yield (%) (overall)
a	2	C_6H_5	C_6H_5	75
b	2	p-ClC_6H_4	C_6H_5	67
c	2	β-$C_{10}H_7$	C_6H_5	64
d	2	p-$NO_2C_6H_4$	C_6H_5	55
e	2	p-$CH_3OC_6H_4$	C_6H_5	62
f	3	p-ClC_6H_4	C_6H_5	64
g	3	β-$C_{10}H_7$	C_6H_5	62
h	3	p-$NO_2C_6H_4$	C_6H_5	74
i	3	C_6H_5	p-$NO_2C_6H_4$	63
j	3	p-ClC_6H_4	CH_3	72
k	3	p-ClC_6H_4	C_2H_5	62
l	3	p-ClC_6H_4	i-C_3H_7	67
m	4	p-$CH_3OC_6H_4$	C_6H_5	77
n	4	p-$NO_2C_6H_4$	C_6H_5	70

as precursors of cyclic aminals (imidazolidines, hexahydropyrimidines, and hexahydro-1,3-diazepines). Such compounds are easily prepared by condensation of the suitable α,ω-diaminoalkanes with aldehydes, so the problem devolves to the preparation of unsymmetrical alkylenediamine derivatives. A careful examination

of the literature showed only a few synthetic procedures, and none is general. Besides, compounds **3** are also of interest given their close relationship with biogenic polyamines.[1]

Among accepted procedures to generate the secondary amine functional group, selective alkylation of polyamines is hardly feasible due to polyalkylation reactions which often complicate the isolation of the desired monoalkyl product from the reaction mixture. An alternative method consists in the reaction between an halogen derivative carrying the methylene chain and the suitable amine,[2] though this procedure may present the same limitations as the previous one. To obviate this problem, some methods have been developed to ensure selective polyamine monoalkylation[3] or to generate the required secondary amines from other nitrogenous functions.[2ab,4]

The present paper describes an improved approach to the synthesis of N-alkyl (or aralkyl)-N'-aryl-α,ω-diaminoalkanes **3a-n** (Scheme) in good overall yields, from easily available starting materials in two steps.

Treatment of N-(ω-haloalkyl)amides **1** with two moles of a primary aromatic amine affords N-(ω -arylaminoalkyl)amides **2a-n** which are then readily converted to the corresponding compounds **3** by refluxing with borane in tetrahydrofurane. Instead, reduction of amides **2** with lithium aluminium hydride renders poorer yields. In agreement with Brown,[5] benzaldehyde and benzyl alcohol are by-products. In the case of nitroaryl derivatives **3d,h,n**, the preparation of compounds **2** by the above route requires very drastic conditions and affords low yields . In such cases, direct

acylation of the nitroarylalkylenediamine is the most suitable procedure to obtain the intermediate amide **2**.[6]

The extension of this method to the preparation of unsymmetrical N,N'-dialkyl α,ω–diaminoalkanes is limited by the low yield of the reaction of compounds **1** (n=3,4) with aliphatic amines, due to competitive intramolecular dehydrohalogenation reactions.[6b,7]

Experimental

Melting points were determined with a Büchi capillary apparatus and are uncorrected. ^{1}H NMR spectra were recorded on a Bruker MSL 300 MHz spectrometer, using deuteriochloroform as the solvent. Chemical shifts are reported in ppm (δ) relative to TMS as an internal standard. D_2O was employed to confirm exchangeable protons (ex.). Splitting multiplicities are reported as singlet (s), broad signal (bs), doublet (d), triplet (t), quartet (q) and multiplet (m). Mass spectra (EI) were recorded with a GC-MS Shimadzu QP-1000 spectrometer operating at 20 eV. TLC analyses were carried out on aluminium sheets silica gel 60 F_{254} using chloroform-methanol (9:1) as the solvent. Column chromatographies were performed either on silica gel 60 or on aluminium oxide (90 active, neutral, activity I), with typically 30-50 g of stationary phase *per gram* substance.

N-(ω-Haloalkyl)amides **1** were obtained by acylation of the corresponding ω-bromoalkylamine hydrobromide (n=2,3) in Schotten-Baumann conditions,[6a,b] and by a modified von Braun reaction on N-benzoylpyrrolidine in the case of n= 4.[8]

N–(ω-Arylaminoalkyl)amides 2:

Compounds **2a** (mp 127°C) [6a], **2b** (mp 130°C) [9], **2c** (mp 184°C) [6a], **2d** (mp 162°C) [6a], **2e** (mp 136°C) [6a], **2f** (mp 117°C) [6b], **2g** (mp 139°C) [6b], **2h** (mp 121°C) [6b], **2i** (mp 89°C) [10] and **2n** (mp 145°C) [6c] were prepared following literature procedures.

Compounds **2j-m** were obtained by reaction of the corresponding N-(ω-haloalquil)amides and arylamines.

General Procedure:

A mixture of the corresponding N-(ω-haloalkyl)amide **1** (10 mmol) and arylamine (20 mmol) was heated in an oil bath at 100-120 °C under reflux for 1 h. The crude product was treated three times with boiling water (20 ml) in order to eliminate arylamine hydrohalide.

N[3-(p-Chlorophenylamino)propyl]acetamide **(2j).**

The resulting oil was purified by column chromatography using silica gel 60 (chloroform: ethyl acetate, 8:2 to 1:1) to give **2j** (85%); mp 91-93°C.

MS: m/z= 227 ($M^{+\cdot}$).

^{1}H RMN: δ=1.75 (2H, m, $CH_2CH_2CH_2$), 1.90 (3H, s, $NHCOCH_3$) , 3.10 (2H, t, CH_2NHR), 3.25-3.35 (2H, m, CH_2NHCO), 3.80 (1H, bs, ex., NHR), 6.25 (1H, bs, ex., NHCO), 6.50 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.10 (2H, dd, p-ClC_6H_4, 2 meta H).

Anal. calcd. for $C_{11}H_{15}N_2OCl$: C 58.28, H 6.67, N 12.36, Cl 15.64; found: C 58.36, H 6.80, N 12.48, Cl 15.55.

N[3-(p-Chlorophenylamino)propyl]propanamide **(2k).**

The resulting oil was purified by column chromatography using silica gel 60

(chloroform: ethyl acetate, 8:2 to 1:1) to give **2k** (74%); mp 61-63°C.

MS: m/z= 241 ($M^{+\cdot}$).

^{1}H RMN: δ=1.10 (3H, t, CH_2CH_3), 1.70 (2H, m, CH_2CH_2-CH_2), 2.15 (2H, q, CH_2CH_3), 3.05 (2H, t, CH_2NHR), 3.20-3.35 (2H, m, CH_2NHCO), 4.10 (1H, bs, ex., NHR), 5.60 (1H, bs, ex., NHCO), 6.45 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.05 (2H, dd, p-ClC_6H_4, 2 meta H).

Anal. calcd. for $C_{12}H_{17}N_2OCl$: C 59.87, H 7.12, N 11.64, Cl 14.73; found: C 60.04, H 7.34, N 11.50, Cl 14.85.

N[3-(p-Chlorophenylamino)propyl]-2-methylpropanamide **(2l).**

The resulting oil was purified by column chromatography using silica gel 60 (chloroform: ethyl acetate, 8:2 to 1:1) to give **2k** (82%); mp 49-51°C.

MS: m/z= 255 ($M^{+\cdot}$).

^{1}H RMN: δ=1.10 (6H, d, $CH(CH_3)_2$), 1.75 (2H, m, $CH_2CH_2CH_2$), 2.35 (1H, m, $CH(CH_3)_2$), 3.15 (2H, t, CH_2NHR), 3.30-3.40 (2H, m, CH_2NHCO), 4.20 (1H, bs, ex., NHR), 5.60 (1H, bs, ex., NHCO), 6.55 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.10 (2H, dd, p-ClC_6H_4, 2 meta H).

Anal. calcd. for $C_{13}H_{19}N_2OCl$: C 61.29, H 7.52, N 11.00, Cl 13.92; found: C 61.15, H 7.74, N 10.85, Cl 13.78.

N[4-(p-Methoxyphenylamino)butyl]benzamide **(2m).**

The crude product was recrystallized from cyclohexane to give **2m** (85%); mp 104°C.

MS: m/z= 298 ($M^{+\cdot}$).

^{1}H RMN: δ=1.75 (4H, m, $CH_2CH_2CH_2CH_2$), 2.61 (1H, bs, ex., NHR), 3.10 (2H, t,

CH$_2$NHR), 3.40-3.60 (2H, m, C*H*$_2$NHCO), 3.76 (3H, s, OCH_3), 6.10 (1H, bs, ex., NHCO), 6.55 (2H, d, p-$CH_3OC_6H_4$, 2 ortho H), 6.70 (2H, d, p-$CH_3OC_6H_4$, 2 meta H), 7.26-7.90 (5H, m, C_6H_5).

Anal. calcd. for $C_{18}H_{22}N_2O_2$: C 72.46, H 7.43, N 9.39, found: C 72.35, H 7.62, N 9.23.

N-Alkyl (or aralkyl)-N'-aryl-α,ω-diaminoalkanes (3a-n). General Procedure: Compounds **2** (5 mmol) were treated with BH_3/THF (10 ml satd. soln.)[11] and heated under reflux in a nitrogen atmosphere for 2 h. THF was evaporated *in vacuo* and the residue boiled with concentrated hydrochloric acid (10 ml) for 15 min after which solution was cooled. If the product crystallized, the resulting solid was filtered, washed with ethyl ether, dried and recrystallized (anhydrous ether/ethanol). If not, the acid solution was diluted with water (5 ml) and then made alkaline (pH=12) with sodium hydroxide pellets. The mixture was extracted with chloroform (3 x 20 ml) and the organic layer was washed with water (5 ml), filtered and dried with anhydrous sodium sulphate. The solution was concentrated *in vacuo* and the crude base purified by column chromatography.

N-Benzyl-N'-phenyl-1,2-diaminoethane (**3a**).

Product was isolated as dihydrochloride (87%); mp 188-9°C (lit.[12] mp 188-9°C).

Free base:

MS: m/z= 226 ($M^{+\cdot}$).

^{1}H RMN: δ= 2.40 (1H, bs, ex. N*H*CH_2R'), 2.90 (2H, t, C*H*$_2$NHCH_2R'), 3.40 (2H, t, C*H*$_2$NHR), 3.50 (bs, ex., RN*H*CH_2), 3.90 (2H, s, NHC*H*$_2$R'), 7.00-7.40 (10H, m, aromatics).

N-Benzyl-N'-(p-chlorophenyl)-1,2-diaminoethane (**3b**).

Product was isolated as dihydrochloride (84%); m.p 224-226°C (lit.[4b] monohydrochloride mp 225-7°C).

Anal. calcd. for $C_{15}H_{19}N_2Cl_3$: C 53.99, H 5.74, N 8.40, Cl 31.87; found: C 54.22, H 5.85, N ,8.44, Cl 31.60.

Free base:

MS: m/z= 261 ($M^{+\cdot}$).

^{1}H RMN: δ=2.20 (1H, bs, ex., N*H*CH_2R'), 2.90 (2H, t, C*H*$_2$NHCH_2R'), 3.20 (2H, t, C*H*$_2$NHR), 3.70 (1H, bs, ex., RN*H* CH_2), 3.80 (2H, s, NHC*H*$_2$ R'), 6.40 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.10 (2H, dd, p-ClC_6H_4, 2 meta H), 7.40 (5H, s, C_6H_5).

N-Benzyl-N'-(β-napthyl)-1,2-diaminoethane (**3c**).

Product was isolated as dihydrochloride (81%); mp 236-238°C.

Anal. calcd. for $C_{19}H_{22}N_2Cl_2$: C 65.33, H 6.35, N 8.02, Cl 20.30; found: C 65.51, H 6.55, N 8.07, Cl 20.10.

Free base:

MS: m/z= 276 ($M^{+\cdot}$).

^{1}H RMN: δ=2.50 (1H, bs, ex., N*H*CH_2R'), 3.10 (2H, t, C*H*$_2$ NHCH_2R'), 3.45 (2H, t, C*H*$_2$NHR), 3.80 (1H, bs, ex., RN*H*CH_2), 3.90 (2H, s, NHC*H*$_2$R'), 6.60-7.50 (7H, m, β-naphtyl), 7.30 (5H, s, C_6H_5).

N-Benzyl-N'-(p-nitrophenyl)-1,2-diaminoethane (**3d**).

Product was isolated as dihydrochloride (82%); mp 189-191°C.

Anal. calcd. for $C_{15}H_{19}N_3O_2Cl_2$: C 52.34, H 5.56, N 12.21, Cl 20.60; found: C 52.49, H 5.60, N 12.08, Cl 20.46.

Free base:

MS: m/z= 271 ($M^{+\cdot}$).

^{1}H RMN: δ=2.60 (1H, bs, ex., N*H*CH_2R'), 2.95 (2H, t, C*H*$_2$NHCH_2R'), 3.30 (2H, t, C*H*$_2$NHR), 3.80 (2H, s, NHC*H*$_2$R'), 3.90 (1H, bs, ex., RN*H*CH_2), 6.50 (2H, dd, p-$NO_2C_6H_4$, 2 ortho H), 7.35 (5H, s, C_6H_5), 8.10 (2H, dd, p-$NO_2C_6H_4$, 2 meta H).

N-Benzyl-N'-(p-methoxyphenyl)-1,2-diaminoethane (**3e**)

Product was isolated as dihydrochloride (78%); mp 158-160°C (lit.[4b] mp monohydrochloride 234-6°C).

Anal. calcd. for $C_{16}H_{22}N_2OCl_2$: C 58.36, H 6.73, N 8.51, Cl 21.53; found: C 58.50, H 6.77, N 8.56, Cl 21.30.

Free base:

MS: m/z= 256 ($M^{+\cdot}$).

^{1}H RMN: δ=2.55 (1H, bs, ex., N*H*CH_2R'), 2.90 (2H, t, C*H*$_2$NHCH_2R'), 3.20 (2H, t, C*H*$_2$NHR), 3.60 (1H, bs, ex., RN*H*CH_2), 3.70 (3H, s, CH_3O), 3.80 (2H, s, NHC*H*$_2$ R'), 6.60 (2H, dd, p-$CH_3OC_6H_4$, 2 ortho H), 6.80 (2H, dd, p-$CH_3OC_6H_4$, 2 meta H), 7.40 (5H, s, C_6H_5).

N-Benzyl-N'-(p-chlorophenyl)-1,3-diaminopropane (**3f**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3f** (84%).

MS: m/z= 275 ($M^{+\cdot}$).

^{1}H RMN: δ=1.75 (2H, m, $CH_2CH_2CH_2$), 2.30 (1H, bs, ex., N*H*CH_2R'), 2.55 (2H, t, *CH*$_2$NHCH_2R'), 3.05 (2H, t, RNHC*H*$_2$), 3.60 (2H, s, C*H*$_2$ R'), 3.75 (1H, bs, ex.,

NHR), 6.40 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.05 (2H, dd, p-ClC_6H_4, 2 meta H), 7.30 (5H, s, C_6H_5).

Anal. calcd. for $C_{16}H_{19}N_2Cl$: C 69.93, H 6.97, N 10.19, Cl 12.90; found: C 69.75, H 7.19, N 10.06, Cl 12.70.

N-Benzyl-N'-(β-naphtyl)-1,3-diaminopropane (**3g**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3g** (80%).

MS: m/z= 290 ($M^{+\cdot}$).

^{1}H RMN: δ=1.85 (2H, m, $CH_2CH_2CH_2$), 2.40 (1H, bs, ex., $NHCH_2R'$), 2.60 (2H, t, CH_2NHCH_2R'), 3.20 (2H, t, $RNHCH_2$), 3.60 (2H, s, CH_2R'), 3.70 (1H, bs, ex., NHR), 6.60-6.80 (2H, m, β-naphtyl), 7.10-7.65 (5H, m, β-naphtyl), 7.30 (5H, s, C_6H_5).

Anal. calcd. for $C_{20}H_{22}N_2$: C 82.72, H 7.64, N 9.65; found: C 82.61, H 7.92, N 9.47.

N-Benzyl-N'-(o-nitrophenyl)-1,3-diaminopropane (**3h**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3h** (82%).

MS: m/z= 285 ($M^{+\cdot}$).

^{1}H RMN: δ=1.90 (2H, m, $CH_2CH_2CH_2$), 2.50 (1H, bs, ex., $NHCH_2R'$), 2.80 (2H, t, CH_2NHCH_2R'), 3.30 (2H, t, $RNHCH_2$), 3.70 (2H, s, CH_2R'), 3.85 (1H, bs, ex., NHR), 6.70 (1H, dd, o-$NO_2C_6H_4$,1 ortho H), 6.75-6.80 (1H, m, o-$NO_2C_6H_4$, 1 para H), 7.30 (5H, s, C_6H_5), 7.35-7.45 (1H, m, o-$NO_2C_6H_4$, 1 meta H), 8.10 (1H, dd, o-$NO_2C_6H_4$, 1 meta H).

Anal. calcd. for $C_{16}H_{19}N_3O_2$: C 67.35, H 6.71, N 14.73; found: C 67.17, H 6.92, N 14.60.

N-(p-Nitrobenzyl)-N'-phenyl-1,3-diaminopropane (**3i**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3i** (87%); mp 56-58°C.

MS: m/z= 285 ($M^{+\cdot}$).

^{1}H RMN: δ=1.80 (2H, m, $CH_2CH_2CH_2$), 2.50 (1H, bs, ex., $NHCH_2R'$), 2.75 (2H, t, CH_2NHCH_2R'), 3.20 (2H, t, $RNHCH_2$), 3.90 (2H, s, CH_2R), 3.80 (1H, bs, ex., NHR), 6.50-6.70 (3H, m, C_6H_5, 2 ortho and para H), 7.10-7.20 (2H, m, C_6H_5, 2 meta H), 7.45 (2H, dd, p-$NO_2C_6H_4$, 2 ortho H), 8.15 (2H, dd, p-$NO_2C_6H_4$, , 2 meta H).

Anal. calcd. for $C_{16}H_{19}N_3O_2$: C 67.35, H 6.71, N 14.73; found: C 67.15, H 6.90, N 14.85.

N-(p-Chlorophenyl)-N'-ethyl-1,3-diaminopropane (**3j**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3j** (85%); mp 206-208°C (dihydrochloride).

Anal. calcd. for $C_{11}H_{19}N_2Cl_3$: C 46.25, H 6.70, N 9.81, Cl 37.23; found: C 46.43, H 6.92, N 9.62, Cl 37.04.

Free base:

MS: m/z= 213 ($M^{+\cdot}$).

^{1}H RMN: δ=1.05 (3H, t, CH_3), 1.75 (2H, m, $CH_2CH_2CH_2$), 2.25 (1H, bs, ex., $NHCH_2R'$), 2.60 (2H, q, CH_2CH_3), 2.70 (2H, t, CH_2NHCH_2R'), 3.10 (2H, t,

RNHCH_2), 3.50 (1H, bs, ex., NHR), 6.45 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.05 (2H, dd, p-ClC_6H_4, 2 meta H).

N-(p-Chlorophenyl)-N'-propyl-1,3-diaminopropane (**3k**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3k** (84%); mp 51-53°C.

MS: m/z= 227 (M^+).

^{1}H RMN: δ=0.90 (3H, t, CH_3),1.65 (2H, m, C$H_2$$CH_3$), 1.90 (2H, m, CH_2CH_2 CH_2), 2.50 (1H, bs, ex., N*H*CH_2R'), 2.65 (2H, t, NHCH_2R'), 2.85 (2H, t, CH_2NHCH_2R'), 3.20 (2H, t, RNHCH_2), 3.80 (1H, bs, ex., NHR), 6.55 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.10 (2H, dd, p-ClC_6H_4, 2 meta H).

Anal. calcd. for $C_{12}H_{19}N_2Cl$: C 63.56, H 8.45, N 12.35, Cl 15.64; found: C 63.67, H 8.64, N 12.17, Cl 15.83.

N-(p-Chlorophenyl-N'-(2-methylpropyl)-1,3-diaminopropane (**3l**).

The crude material was purified by column chromatography using aluminium oxide (ethyl acetate:methanol, 9:1 to 0:10) to give **3l** (82%); mp 243-245°C (dihydrochloride).

Anal. calcd. for $C_{13}H_{23}N_2Cl_3$: C 49.77, H 7.39, N 8.93, Cl 33.90; found: C 49.98, H 7.58, N 8.95, Cl 33.88.

Free base:

MS: m/z= 241 (M^+).

^{1}H RMN: δ=0.95 (6H, d, CH_3), 1.75 (1H, m, CH), 1.80 (2H, m, CH_2C$H_2$$CH_2$), 2.45 (3H, bs, $NHCH_2$ R'), 2.75 (2H, t, CH_2NHCH_2R'), 3.15 (2H, t, RNHCH_2),

3.40 (1H, bs, ex., NHR), 6.50 (2H, dd, p-ClC_6H_4, 2 ortho H), 7.10 (2H, dd, p-ClC_6H_4, 2 meta H).

N-Benzyl-N'-(p-methoxyphenyl)-1,4-diaminobutane (**3m**).

The crude material was purified by column chromatography using silica gel 60 (chloroform:methanol, 9:1) to give **3m** (90%).

MS: m/z= 284 (M^+).

^{1}H RMN: δ=2.20-2.25 (4H, m, $CH_2CH_2CH_2CH_2$), 2.60 (1H, bs, ex., N*H*CH_2R'), 2.90 (2H, t, CH_2NHCH_2R'), 3.30 (2H, t, RNHCH_2), 3.60 (1H, bs, ex., NHR), 3.80 (2H, s, NHCH_2R'), 6.65 (2H, dd, p-$CH_3OC_6H_4$, 2 ortho H), 6.85 (2H, dd, p-$CH_3OC_6H_4$, 2 meta H), 7.55 (5H, s, C_6H_5).

Anal. calcd. for $C_{18}H_{24}N_2O$: C 76.02, H 8.51, N 9.85; found: C 76.10, H 8.75, N 9.94.

N-Benzyl-N'-(p-nitrophenyl)-1,4-diaminobutane (**3n**).

The crude material was purified by column chromatography using silica gel 60 (chloroform:methanol, 9:1) to give **3n** (78%).

MS: m/z= 299 (M^+).

^{1}H RMN: δ=2.20-2.15 (4H, m, $CH_2CH_2CH_2CH_2$), 2.65 (1H, bs, ex., N*H*CH_2R'), 2.95 (2H, t, CH_2NHCH_2R'), 3.45 (2H, t, RNHCH_2), 3.80 (2H, s, NHCH_2R'), 3.90 (1H, bs, ex., NHR), 6.55 (2H, dd, p-$NO_2C_6H_4$, 2 ortho H), 7.45 (5H, s, C_6H_5), 8.15 (2H, dd, p-$NO_2C_6H_4$, 2 meta H).

Anal. calcd. for $C_{17}H_{21}N_3O_2$: C 68.20, H 7.07, N 14.04; found: C 68.43, H 7.26, N 14.00.

Reduction of compounds **2** with lithium aluminium hydride afforded moderate yields of **3** (45-60%). Two by-products (benzaldehyde and benzyl alcohol) were detected by GC-MS.

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