Benzotriazole stabilized lithium intermediates as a route to the elaboration of N-substituents in amides

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Abstract

Lithiation of *N*-(benzotriazol-1-ylmethyl)benzamide or *N*-(benzotriazol-1-ylmethyl)-2,2-dimethylbutyramide, readily prepared from the corresponding amides, formaldehyde and benzotriazole, followed by quenching with various electrophiles, such as alkyl halides, ketones or ester, gives the corresponding N-substituted derivatives. Subsequent displacement of the benzotriazole group with Grignard reagents, thiols or alcohols provides access to a wide variety of N-substituted amides in good yields. Treatment of the *N*-(benzotriazol-1-ylalkyl)benzamides with n-BuLi afforded the 1,1-dibenzamidoalkanes.

Key words: Lithium compounds; Benzotriazole compounds; Amide compounds

The amide moiety is an important constituent of many biologically active compounds and the preparation of substituted amides has attracted much attention [1–9]. Alkylation of amides is a common approach for the preparation of N-substituted amides and it has been carried out under neutral, acidic and basic conditions. Treatment of amide anions with alkyl halides has frequently been used for N-alkylation [9], while use of reagents other than alkyl halides is less familiar.

Previous work from our laboratory has demonstrated the versatility of benzotriazole as a synthetic auxiliary in organic synthesis [10]. The benzotriazole anion is a good leaving group and can be used in place of a halogen or other substituents in many transformations. In particular, primary amides [11] or secondary amides [12] are readily converted by treatment with benzotriazole and an aldehyde into intermediates $RCONHCHR^1Bt$ or $RCONR^1CHR^2Bt$. These intermediates react smoothly with Grignard reagents or $LiAlH_4$ to give the corresponding secondary or tertiary amides in which Bt has been replaced by R or H, respectively. These $RCONHCHR^1Bt$ intermediates have also been shown to be effective amidoalkylating reagents; they react readily with active aromatic compounds [13], CH acids [14], thiols [15] and alcohols [16] to give the corresponding amidoalkylated products.

As part of our continuing study dealing with the application of benzotriazole as a synthetic auxiliary, we wish to report the successful conversions of amide–formaldehyde–benzotriazole adducts into important intermediates of type **3** via lithiation of the methylene group followed by quenching with various electrophiles such as alkyl halides, esters, aldehydes and ketones. Compounds of type **3** react readily with Grignard reagents, thiols, alcohols and lithium aluminum hydride ($LiAlH_4$) to provide more complex N-substituted amides **7**. Although these N-substituted amides **7** can be alternatively obtained by direct condensation of the corresponding amides, aldehydes and benzotriazole followed by displacement of the benzotriazole group, our new method provides a convenient approach for the preparation of more complex N-substituted amides especially in the cases where the aldehydes required for the direct condensation are unstable or not readily available.

Lithiation alpha to nitrogen has been observed for various N-substituted amides and N,N-disubstituted amides [17–24]. Generally, such α-lithiation is restricted to amides without α-hydrogen such as benzamides [25, 26] or 2,2-dialkylbutyramides [27, 28]. Treatment of an amide bearing an α hydrogen adjacent to the carbonyl group with lithium agents leads to deprotonation at this position instead of lithiation alpha to nitrogen. Thus, in our experiments, treatment of *N*-(benzotriazol-1-ylmethyl)acetamide with 2 or 3 equiv. of lithium

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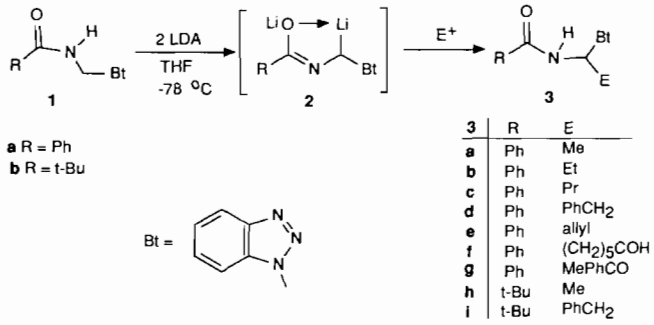


3	R	E
a	Ph	Me
b	Ph	Et
c	Ph	Pr
d	Ph	$PhCH_2$
e	Ph	allyl
f	Ph	$(CH_2)_5COH$
g	Ph	MePhCO
h	t-Bu	Me
i	t-Bu	$PhCH_2$

Scheme 1.

diisopropylamide in THF followed by quenching with methyl iodide resulted only in *N*-(benzotriazol-1-ylmethyl)propylamide rather than any product derived from lithiation at methylene. An electron-withdrawing group adjacent to the methylene group of an amide such as aryl [29], carboxyl [26] or ester [30] facilitates the methylene metalation. Thus, benzotriazole should also serve as a good activating group for such deprotonations.

Treatment of *N*-(benzotriazol-1-ylmethyl)benzamide (**1a**), which was easily prepared by heating benzamide and 1-hydroxymethylbenzotriazole in acetic acid [11], with 2 equiv. of lithium diisopropylamide at −78 °C under nitrogen formed a dianion **2** (see Scheme 1). This intermediate was quenched with methyl iodide to give **3a** in 80% yield. Compounds **3b–g** were similarly obtained from *N*-(benzotriazol-1-ylmethyl)benzamide (**1a**) and the corresponding electrophiles in good to excellent yields. The products were easily purified by recrystallization from suitable solvents (see Table 1). It is noteworthy that compound **3e**, obtained via lithiation followed by trapping with allyl bromide, is not easily accessible by previous methods [11] due to the expected isomerization of a β,γ-unsaturated aldehyde in the reaction. Furthermore, the present method allows easy introduction of a β-hydroxy or β-carbonyl group into the N-substituted amides when dianions **2** are reacted with a ketone or an ester.

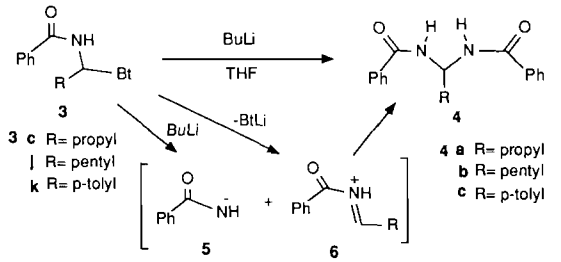
N-(Benzotriazol-1-ylmethyl)trimethylacetamide (**1b**) underwent similar lithiation with LDA, and reaction of the resulting dianion with methyl iodide and benzyl bromide gave **3h** and **3i** in 30% and 22% yields, respectively. These low yields probably reflect the decomposition of the strongly basic dianion in the reaction system.

When the *N*-[(α-benzotriazol-1-yl)alkyl]benzamides **3c, j, k** were treated with 1 equiv. of butyllithium in THF at −78 °C, the symmetrical 1,1-dibenzamidoalkanes **4a–c** were obtained, respectively. This is probably due to base catalyzed scission of **3** to give the amide anion **5**, which reacted with the iminium ion **6** produced in the solution to yield the final product **4** (Scheme 2). Symmetrical amide derivatives of type **5** are easily prepared by reaction of 2 equiv. of amide with an α,α-(dimorpholin-1-yl)toluene [31], with an acetal [32] or

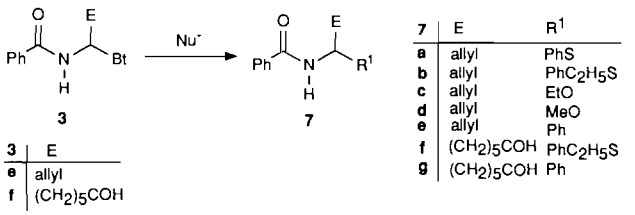
TABLE 1. Preparation of *N*-(benzotriazol-1-ylmethyl)amides and their substituted products

Compound	R	E	R^1	Yield (%)	m.p. (°C)	Purif.	Lit. m.p. or Calc./Found		
							C	H	N
3a	Ph	Me		78	156–158	MeOH	67.64/67.88	5.30/5.29	21.05/21.30
3b	Ph	Et		81	143–144	EtOH	68.55/68.72	5.75/5.79	19.99/20.16
3c	Ph	Pr		80	157–159	MeOH		156–159 [11]	
3d	Ph	$PhCH_2$		76	180–181	MeOH	73.65/73.48	5.30/5.29	16.37/16.05
3e	Ph	allyl		73	144–145	MeOH/ hexane	69.85/69.73	5.52/5.48	19.16/19.12
3f	Ph	$(CH_2)_5OH$		74	203–205	hexane	68.55/68.50	6.33/6.35	15.99/16.09
3g	Ph	MePhCO		82	183–185	MeOH	63.38/63.45	7.37/7.45	22.76/23.00
3h	t-Bu	Me		30	145–146	MeOH	62.03/62.09	6.95/6.99	24.13/24.43
3i	t-Bu	$PhCH_2$		33	228–229	acetone		[a]	
7a	Ph	allyl	PhS	88	76–78	EtOH/ H_2O		[b]	
7b	Ph	allyl	PhC_2H_5S	93	99–100	EtOH/ H_2O	73.27/73.39	6.80/6.90	4.50/4.46
7c	Ph	allyl	EtO	76	63–65	hexane	71.21/71.08	7.81/7.81	6.39/6.31
7d	Ph	allyl	MeO	89	50–51	[c]	70.22/70.24	7.37/7.35	6.82/6.98
7e	Ph	allyl	Ph	89	118–120	MeOH/ H_2O	81.24/81.12	6.82/6.84	5.57/5.47
7f	Ph	$(CH_2)_5OH$	PhC_2H_5S	94	142–143	EtOH	71.51/71.32	7.36/7.42	3.79/3.73
7g	Ph	$(CH_2)_5OH$	Ph	80	207–208	EtOH		[d]	

[a] $C_{19}H_{22}N_4O$ Calc. $MW = 322.1793$; Found: $M+1 = 323.1829$. [b] $C_{17}H_{17}NOS$ Calc. $MW = 283.1031$; Found: $M+1 = 284.1110$. [c] Purified by column chromatography on silica gel, eluent: $CHCl_3$. [d] $C_{20}H_{23}NO_2$ Calc. $MW = 309.1729$; Found: $M+1 = 310.1710$.



Scheme 2.



Scheme 3.

by direct refluxing with an aldehyde in the presence of 2-naphthalenesulfonic acid [33–35].

Displacements of the benzotriazole group in the amide derivatives of type **3a–d, h, i** by diverse nucleophiles including Grignard reagents [36], hydrides ($NaBH_4$ or $LiAlH_4$) [11], thiols [15], alcohols [16], active methylene compounds [14], and electron-rich aromatic and heterocyclic compounds [13] have been previously carried out in our group. However, no such displacements of the benzotriazole moiety in amide derivatives containing additional functionality as in **3e, f** have previously been reported. We have now carried out a number of such transformations to demonstrate the generality of the present method.

Reaction of *N*-[(benzotriazol-1-yl)allylmethyl]benzamide (**3e**) with sodium thiophenolate at room temperature for 12 h gave product **7a** in 87% yield (Scheme 3). Similarly, compounds **7b** and **7f** were obtained in 93% and 94% yields, respectively. The by-product, benzotriazole, was readily removed during the workup, and the products were purified by recrystallization. Oxygen nucleophiles behaved similarly: thus, sodium alkoxides reacted with **3e** to afford the *N*-(1-alkoxy-3-butenyl)benzamide **7c, d** in good yields. Heating **3e** or **3f** with phenylmagnesium bromide in THF gave the corresponding **7e** and **7g** in very good yields.

The structures of the lithiated intermediates **3**, the displacement products **7** and the 1,1-dibenzamido derivatives **5** were confirmed by their NMR spectral data and elemental analyses. The methine signals of compounds **3** in both ^1H and ^{13}C spectra were shifted downfield compared to the signal of methylene (Tables 2 and 3). The data for known compounds are in agreement with those reported in the literature.

Conclusions

The presently reported method provides an alternative approach for the preparation of amides with complex N-substituents. Our method is versatile in the sense that (α-hydroxyalkyl), carbonyl and γ,δ-unsaturated groups can be easily introduced to give compounds which are not readily accessible by previous methods. Furthermore, treatment of the *N*-(benzotriazol-1-ylalkyl)benzamides with a lithium agent could afford the symmetrical 1,1-dibenzamido substituted derivatives. The mild reaction conditions, simple workup procedure and high overall yields make this method an attractive procedure for the preparation of N-substituted benzamides and 2,2-dialkylbutyramides.

Experimental

Melting points were determined on a Kofler hot stage apparatus without correction. ^1H NMR and ^{13}C NMR spectra were recorded on a Varian VXR 300 MHz spectrometer using TMS as an internal reference for ^1H spectra and solvent $CDCl_3$ or d^6-DMSO for ^{13}C spectra. Elemental analyses were performed on a Carlo Erba-1106 instrument and high resolution mass spectra were measured on an AEL MS-30 mass spectrometer. Column chromatography was carried out on MCB silica gel (230–400 mesh).

N-(Benzotriazol-1-ylmethyl)benzamides **1a** and **3j, k** were prepared according to the previously described methods [11]. Compound **1b** was not previously reported and was prepared according to the literature procedure [11].

N-(Benzotriazol-1-ylmethyl)trimethylacetamide (1b)

This was obtained as white needles (from MeOH). Yield 80%. m.p. 172–174 °C. *Anal.* Found: C, 62.09; H, 6.99; N, 24.43. Calc. for $C_{12}H_{16}N_4O$: C, 62.03; H, 6.95; N, 24.13%. ^1H NMR: δ 1.19 (s, 9H), 6.12 (d, 2H, J=6.5 Hz), 7.35 (t, 1H, J=7.8 Hz), 7.47 (t, 1H, J=6.8 Hz), 7.96 (dd overlapped, 2H, J=7.1, 7.3 Hz), 8.51 (t, 1H, J=6.5 Hz). ^{13}C NMR: δ 26.7, 38.2, 51.1, 110.5, 118.5, 123.5, 126.9, 131.8, 145.2, 178.9.

Lithiation and alkylation of N-(benzotriazol-1-ylmethyl)-amides (3e, 3f). General procedure

A solution of **1a** or **1b** (10 mmol) under a nitrogen atmosphere was cooled to −78 °C in a dry ice-acetone bath. Lithium diisopropylamide (10 ml, 2.0 M) was added slowly. The solution was stirred at this temperature for 2 h, and then the appropriate electrophile (10 mol) was added. The mixture was stirred for a further 4 h at −78 °C, then cold water (25 ml) was added. The resultant mixture was extracted with diethyl

TABLE 2. ^{1}H NMR spectral data of compounds **3a–i** and **7a–g**

Compound	E	R^1	CH_2 or CH	Others
3a	2.13 (d, 3H, J=6.8)		[a]	7.1–7.5 (m, 6H), 7.8–8.0 (m, 4H), 8.78 (d, 1H, J=8.8)
3b	0.99 (t, 3H, J=7.6), 2.4–2.7 (m, 2H)		6.88 (m, 1H)	7.3–7.5 (m, 5H), 7.82 (d, 2H, J=7.1), 7.92 (d, 1H, J=8.1), 7.99 (d, 1H, J=7.8), 8.12 (d, 1H, J=9.1)
3c	0.91 (t, 3H, J=7.4), 1.3–1.5 (m, 2H), 2.4–2.6 (m, 2H)		6.97 (m, 1H)	7.3–7.5 (m, 5H), 7.85 (d, 2H, J=7.1), 7.95 (d, 1H, J=8.5), 7.98 (d, 1H, J=8.3), 8.44 (d, 1H, J=8.7)
3d	3.83 (dd, 2H, J_1=7.0, J_2=3.9)[b]		[a]	7.0–7.5 (m, 3H), 7.7–7.8 (m, 3H), 7.95 (d, 1H, J=8.2), 8.38 (d, 1H, J=9.2)
3e	3.2–3.4 (m, 2H), 5.04 (d, 1H, J=10.3), 5.13 (d, 1H, J=15.8), 5.7–5.8 (m, 1H)		7.02 (m, 1H)	7.3–7.5 (m, 5H), 7.83 (d, 2H, J=9.4), 7.93 (d, 1H, J=9.5), 7.96 (d, 1H, J=8.4), 8.45 (d, 1H, J=9.0)
3f	1.0–2.0 (m, 10H), 5.13 (s, 1H)		6.86 (d, 1H, J=8.4)	7.4–7.6 (m, 5H), 7.91 (d, 2H, J=6.8), 8.00 (d, 1H, J=8.4), 8.10 (d, 1H, J=8.4), 9.12 (d, 1H, J=8.5)
3g	2.30 (s, 3H)[b]		7.12 (d, 1H, J=8.4)	7.2–7.4 (m, 6H), 7.8–7.9 (m, 4H), 8.03 (d, 1H, J=8.4), 8.12 (d, 1H, J=8.4), 8.20 (d, 1H, J=7.5), 8.43 (d, 1H, J=7.4)
3h	2.02 (d, 3H, J=6.8)		6.88 (m, 1H)	1.19 (s, 9H), 7.3–7.5 (m, 2H), 7.61 (d, 1H, J=9.0), 7.85 (d, 1H, J=8.3), 8.0 (d, 1H, J=7.2)
3i	3.72 (d, 2H, J=7.6)		6.99 (m, 1H)	1.09 (s, 9H), 7.1–7.2 (m, 5H), 7.25–7.45 (m, 3H), 7.65 (d, 1H, J=7.9), 8.00 (d, 1H, J=8.1)
7a	1.7–1.8 (m, 2H), 4.13 (d, 1H, J=10.4), 4.23 (d, 1H, J=15.8), 4.9–5.0 (m, 1H)	[a]	4.78 (m, 1H)	6.3–6.5 (m, 3H), 6.5–6.7 (m, 5H), 6.9–7.0 (m, 2H), 8.09 (d, 1H, J=9.3)
7b	5.06 (d, 1H, J=10.3), 5.16 (d, 1H, J=15.6), 5.8–5.9 (m, 1H)[b]	2.5–3.0 (m, 6H)[b]	5.45 (m, 1H)	7.1–7.3 (m, 5H), 7.4–7.6 (m, 3H), 7.93 (d, 2H, J=7.1), 8.88 (d, 1H, J=9.5)
7c	2.4–2.6 (m, 2H), 5.1–5.2 (m, 2H), 5.8–6.0 (m, 1H)	1.20 (t, 3H, J=7.0), 3.5–3.7 (m, 2H)	5.55 (m, 1H)	6.50 (d, 1H, J=8.6), 7.4–7.6 (m, 3H) 7.78 (dd, 2H, J=7.0, 1.5)
7d	2.4–2.5 (m, 2H), 5.1–5.2 (m, 2H), 5.8–6.0 (m, 1H)	3.39 (s, 3H)	5.45 (m, 1H)	6.81 (d, 1H, J=8.1), 7.4–7.5 (m, 3H) 7.81 (d, 2H, J=7.0)
7e	2.65 (t, 2H, J=7.0), 5.0–5.1 (m, 2H), 5.7–5.8 (m, 1H)	[a]	5.27 (m, 1H)	6.78 (d, 1H, J=7.6), 7.2–7.5 (m, 8H) 7.76 (d, 2H, J=7.0)
7f	1.2–1.8 (m, 9H), 2.6–3.3 (m, 5H), 4.41 (s, 1H)	[a]	5.44 (d, 1H, J=10.0)	7.1–7.3 (m, 5H), 7.4–7.6 (m, 3H) 7.90 (d, 2H, J=6.7), 8.15 (br s, 1H)
7g	1.1–1.6 (m, 10H), 1.90 (s, 1H)	[a]	5.09 (d, 1H, J=5.7)	7.2–7.5 (m, 9H), 7.79 (d, 2H, J=7.3)

[a]Signals are overlapped and are reported in other groups. [b]Other signals are overlapped and are reported in other groups.

ether (3×100 ml), washed with water and dried over magnesium sulfate. Removal of the solvent under reduced pressure yielded the crude product which was purified by recrystallization from the appropriate solvent (see Table 1).

*Preparation of N-(α-alkylthioalkyl)benzamides (**7a, b, f**). General procedure*

A solution of the appropriate thiol (6.2 mmol) and sodium (6.0 mmol) in absolute ethanol (16 ml) was added dropwise to a solution of *N*-[1-(benzotriazol-1-yl)-3-butenyl]benzamide (**3e**) (6.0 mmol) in absolute ethanol (24 ml) over 5 min with stirring at room temperature. The mixture was stirred for 24 h at this temperature. Then, approximately half of the EtOH was evaporated *in vacuo*, and ice-water (60 ml) was added. After stirring for 24 h a white solid was formed, which was purified by recrystallization. Compounds **7a, b, f** have been prepared and the preparative data are given in Table 1.

*Preparation of N-(α-alkoxyalkyl)benzamides (**7c, d**). General procedure*

N-[1-(Benzotriazol-1-yl)-3-butenyl]benzamide (**3e**) (2.92 g, 10 mmol) was added in one portion to a solution of sodium salt (12 mmol) in the corresponding alcohol

TABLE 3. ^{13}C NMR spectral data of compounds **3a–i** and **7a–g**

Compound	E	R^1	CH_2 or CH	Others
3a	20.0		59.2	110.6, 119.0, 124.1, 127.3, 127.4, 128.2, 131.8, 132.4, 132.7, 145.3, 167.1
3b	10.0, 27.5		64.0	110.5, 119.3, 124.2, 127.3, 127.7, 128.4, 132.0, 132.8, 133.1, 145.4, 167.3
3c	13.3, 18.7, 35.9		62.5	110.5, 119.3, 124.2, 127.4, 127.7, 128.3, 132.0, 132.7, 133.0, 145.3, 167.4
3d	40.4[b]		63.8	110.3, 119.2, 124.2, 127.1, 127.4, 127.7, 128.4, 128.6, 129.1, 132.1, 132.7, 133.1, 135.0, 145.2, 167.3
3e	38.1[b]		62.1	110.5, 119.2, 119.9, 124.2, 127.4, 127.7, 128.3, 131.4, 132.0, 132.7, 133.0, 145.3, 167.3
3f	20.8, 21.0, 24.9, 33.4, 33.5, 72.8		71.8	112.6, 118.8, 123.4, 126.7, 127.5, 128.2, 131.8, 132.5, 133.0, 145.1, 166.9
3g	21.8, 187.9[b]		62.8	110.4, 120.1, 124.6, 127.4, 128.6, 128.7, 129.0, 129.8, 130.3, 132.2, 132.5, 132.6, 145.9, 146.1, 167.2
3h	20.0		58.7	27.0, 38.5, 110.4, 119.0, 124.0, 127.2, 132.2, 145.3, 178.2
3i	40.4[b]		63.1	27.0, 38.6, 110.0, 119.2, 124.0, 127.1, 127.5, 128.5, 129.0, 132.8, 135.0, 145.2, 178.3
7a	39.2[b]	[a]	56.6	117.9, 127.3, 128.2, 128.9, 131.4, 132.0, 133.5, 133.9, 134.0, 134.2, 165.8
7b	39.1[b]	31.3, 36.0[b]	53.2	117.4, 126.0, 127.4, 128.2, 128.3, 131.3, 133.9, 134.5, 140.5, 166.0
7c	40.0[b]	15.1, 63.9	78.8	118.8, 126.9, 128.6, 131.8, 132.5, 133.9, 167.2
7d	39.8[b]	55.9	80.6	118.8, 127.1, 128.6, 131.8, 132.5, 133.9, 167.5
7e	40.5[b]	[a]	52.8	118.3, 126.5, 127.0, 127.3, 128.5, 128.6, 131.4, 134.1, 134.5, 141.7, 166.8
7f	21.5, 21.6, 25.3, 31.2, 35.1, 72.8	34.7, 36.1[b]	63.7	126.0, 127.4, 128.2, 128.4, 131.5, 133.9, 140.7, 166.2
7g	21.5, 21.7, 25.2, 30.5, 35.1, 72.9	[a]	60.6	126.8, 127.2, 127.9, 128.2, 128.3, 131.4, 134.1, 138.8, 167.5

[a]Signals are overlapped and are reported in other groups. [b]Other signals are overlapped and are reported in other groups.

at room temperature. The solution was stirred at this temperature for 12 h and then poured into ice-water (100 ml). The resultant mixture was extracted with diethyl ether (3×100 ml), washed with Na_2CO_3 solution (2 N) and water and dried over magnesium sulfate. Removal of the solvent gave a residue, which was chromatographed to give the product. Compounds **7c, d** were prepared and their preparative data are also presented in Table 1.

Preparation of N-alkylbenzamides (7e, g). General procedure

To a solution of **3e** or **3f** (2 mmol) in THF (20 ml) under nitrogen was added phenylmagnesium bromide solution (10 ml, 1 mmol/ml). The Et_2O was distilled off and the solution was refluxed for the appropriate time (for **7e**, 1h; **7g**, 4 h) and stirred for 6 h at room temperature. The reaction mixture was then poured into a solution of ice-water (20 ml) and saturated NH_4Cl (15 ml) then extracted with Et_2O (3×60 ml). The combined extracts were washed with NaOH solution (2×15 ml, 1 N) and water (2×20 ml). Evaporation of the solvent gave the crude product which was purified by recrystallization (see Table 1).

Preparation of 1,1-dibenzamido derivatives (4a–c). General procedure

To a solution of *N*-[α(benzatriazol-1-ylalkyl)]-benzamide (3 mmol) in THF (80 ml) was added n-BuLi (3 mmol) at −78 °C. The solution was kept at −78 °C for 4 h and room temperature overnight. The solution was poured into water (40 ml), extracted with ether (3×60 ml) and dried over Na_2SO_4. Evaporation of the solvent gave a crude product, which was purified by recrystallization from MeOH.

1,1-Dibenzamidobutane (4a). This was obtained as needles from MeOH. Yield 85%; m.p. 218–220 °C. *Anal.* Found: C, 72.95; H, 6.88; N, 9.32. Calc. for $C_{18}H_{20}N_2O_2$: C, 72.94; H, 6.81; N, 9.46%. ^1H NMR: δ 0.96 (t, 3H, J=7.3 Hz), 1.4–1.6 (m, 2H), 1.9–2.1 (m, 2H), 5.7–5.9 (m, 1H), 7.3–7.6 (m, 6H), 7.82 (d, 4H, J=7.2 Hz), 8.15 (d, 2H, J=7.1 Hz). ^{13}C NMR: δ 13.2, 18.6, 36.1, 57.8, 126.8, 127.9, 131.0, 133.9, 166.5.

1,1-Dibenzamidohexane (4b). Obtained as white needles (MeOH). Yield 82%; m.p. 207–209 °C. *Anal.* Found: C, 74.00; H, 7.50; N, 8.58. Calc. for $C_{20}H_{24}N_2O_2$: C, 74.03; H, 7.46; N, 8.64%. ^{1}H NMR: δ 0.86 (t, 3H, J=6.1 Hz), 1.2–1.5 (m, 6H), 2.01 (q, 2H, J=7.9 Hz), 5.8–5.9 (m, 1H), 7.39 (t, 4H, J=6.4 Hz), 7.45 (t, 2H, J=6.1 Hz), 7.85 (d, 4H, J=8.2 Hz), 8.25 (d, 2H, J=8.0 Hz). ^{13}C NMR: δ 13.4, 21.9, 24.8, 30.7, 34.0, 57.6, 126.7, 127.7, 130.8, 133.8, 166.2.

Dibenzamido(4-methylphenol)methane (4c). Obtained as white needles (MeOH). Yield 80%; m.p. 243–245 °C. *Anal.* Found: C, 76.43; H, 5.78; N, 8.14. Calc. for $C_{22}H_{20}N_2O_2$: C, 76.71; H. 5.86; N, 8.14%. ^{1}H NMR: δ 2.32 (s, 3H), 7.01 (t, 1H, J=7.4 Hz), 7.15 (d, 2H, J=7.8 Hz), 7.8–8.1 (m, 8H), 7.91 (d, 4H, J=6.6 Hz), 8.57 (d, 2H, J=8.2 Hz). ^{13}C NMR: δ 20.6, 58.9, 125.6, 127.0, 128.0, 128.7, 131.2, 133.7, 136.7, 137.0, 166.3.

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