Intramolecular Diels-Alder Cyclization of *N*-Furfurylpropargylamines: A Novel General Synthesis of Isoindoles [1]

Alan R. Katritzky*, Maria H. Paluchowska [2a], and John K. Gallos [2b]

Department of Chemistry, University of Florida,
Gainesville, Florida 32611
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Benzotriazole mediated synthesis from furfurylamines, aldehydes and lithium acetylides readily provides high yields of *N*-furfurylpropargylamines. These undergo intramolecular Diels-Alder cyclization to give isoindoles, which are characterized as their Diels-Alder reaction products with dimethyl acetylenedicarboxylate.

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Isoindoles have recently attracted considerable theoretical and synthetic interest [3], but difficulties in their preparation together with their instability, has restricted their study and synthetic use. The existing most generally applicable methods [3,4] for the synthesis of isoindoles, include the dehydrogenation of isoindolines, eliminations (from isoindolinium salts, from isoindoline *N*-oxides or from 2-substituted isoindolines) retrocycloadditions, synthesis from phthalimidines, and condensations of *o*-disubstituted benzenes and of pyrroles.

Intramolecular Diels-Alder reactions have been widely utilized for the synthesis of polycyclic heterocycles and natural products [5]. This kind of methodology has been developed for the synthesis of substituted isoindolines from *N*-allyl-*N*-furfurylamines and related compounds [6]. No such intramolecular Diels-Alder reactions of corresponding *N*-furfuryl-*N*-propargylamines have been recorded in the literature, evidently because of the difficulties in the synthesis of such amines and the lower reactivity of the triple bond. Although, there are examples [7] of intramolecular Diels-Alder cyclizations of *N*-fufuryl-*N*-propargylammonium salts to tetrahydroepoxyisoindolinium salts, these have not been aromatized to isoindoles.

Results and Discussion.

We now report a new general method for the synthesis of isoindoles utilizing intramolecular Diels-Alder reactions of *N*-furfuryl-*N*-propargylamines catalyzed by strong bases such as potassium *t*-butoxide. Recently, we described the one-pot high yield synthesis [8] of tertiary propargylamines under mild conditions. We have now synthesized *N*-furfuryl-*N*-propargylamines **6** analogously from acetylene lithium salts **5** and the benzotriazole adducts **4**. Adducts **4** are produced almost quantitatively by the Mannich reactions of benzotriazole (**1**), *N*-substituted furfurylamines **3** and formaldehyde (**2**, R' = H) (Scheme 1). Some of the benzotriazole adducts **4** were isolated and characterized (Table 1) but the synthesis of propargyl-

Scheme 1

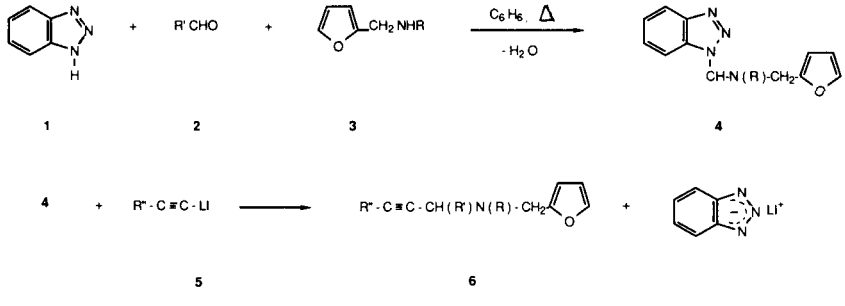


Table 1

Synthesis and Characterization of Benzotriazole Adducts **4**.

Compound	R	R'	Yield (%)	mp (°C)	Molecular Formula	Found (%) C	H	N	Required (%) C	H	N
4a	Ph	H	64	90-92	$C_{18}H_{16}N_4O$	70.87	5.28	18.12	71.03	5.30	18.41
4b	*p*-Tolyl	H	90	70-72	$C_{19}H_{18}N_4O$	71.67	5.73	17.46	71.68	5.70	17.60
4c	CH_3	H	90	43-45	$C_{13}H_{14}N_4O$	64.60	5.85	23.20	64.44	5.82	23.13

amines **6** (Table 4) is advantageously carried out without isolation of **4**.

The spectral characterization of adducts **4** is recorded in Tables 2 and 3. The benzotriazole adducts **4** show in the ^{1}H nmr spectra doubled singlets for the methylene groups (*i.e.* Bt-CH_2-N= and =N-CH_2-Fu) of the 1- and 2-benzotriazole isomers; the intensities indicate that the former dominates. Among the complex aromatic multiplets, signals within 8.00-7.82 ppm from protons at the C-4 position in 1-benzotriazole isomer and at the C-4 and C-7 positions in 2-benzotriazole isomer could be distinguished. Both methyl singlets for the 1- and 2-isomer of **4b** (tolyl moiety) and

Table 2

^{1}H NMR Assignments of Benzotriazole Adducts **4**, δ (deuteriochlorofrorm), J (Hz)

Signals	Compound 4a	4b	4c
Bt-CH2-N=	6.14 (s)	6.08 (s)	5.48 (s)
1*H*-isomer =N-CH2-Fu	4.50 (s)	4.46 (s)	3.76 (s)
H at C-4 in Bt	8.00 (d, J = 7.5)	8.08-7.97 (m)	8.07 (d, J = 8.2)
Bt-CH2-N=	6.09 (s)	6.13 (s)	5.60 (s)
2*H*-isomer =N-CH2-Fu	4.83 (s)	4.81 (s)	3.85 (s)
H at C-4 and C-7 in Bt	7.90-7.82 (m)	7.92-7.82 (m)	7.95-7.84 (m)
Furan at C-3 and C-4	6.31 (s) 6.26 (s)	6.33-6.24 (m)	6.37-6.28 (m)
Other aromatic	7.50-7.00 (m, 8H) 6.90 (t, J = 7.1 , 1H)	7.40-7.22 (m, 3H) 7.22-6.94 (m, 5H)	7.60-7.29 (m, 4H)
Others	--------	2.26 (s) 2.22 (s) (3H)	2.43 (s) 2.39 (s) (3H)

Table 3

^{13}C NMR Assignments of Benzotriazole Adducts **4** δ (deuteriochloroform)

Compound	Benzotriazole 3a	4	5	6	7	7a	Bt-CH_2-N	N-CH_2-Fu	Furan 2	3	4	5	Others
4a	145.8	119.7	123.7	127.2	108.8	132.6	64.4	46.6	150.6	108.7	110.2	142.2	147.2, 116.5, 129.3, 120.7
1*H*-isomer **4b**	145.0	119.7	123.8	127.3	110.0	132.8	65.0	46.9	150.8	108.8	110.3	142.3	146.0, 130.6, 129.9, 117.3, 20.4
4c	145.7	119.7	123.8	127.4	109.8	133.7	67.4	50.8	151.0	109.2	110.2	142.4	39.9
4a	144.2	118.2	126.3	—	—	—	70.4	47.5	151.3	108.0	110.2	142.2	146.9, 129.1, 119.6, 114.4
2*H*-isomer **4b**	144.3	118.3	126.3	—	—	—	70.9	47.7	151.6	108.1	110.3	142.3	144.7, 129.7, 129.1, 114.9, 20.3
4c	144.1	118.2	126.2	—	—	—	75.4	50.7	151.2	109.1	110.0	142.4	39.3

Table 4

Synthesis and Characterization of Propargylamines **6**

Compound	R	R′	R″	Yield (%)	mp (°C)	Molecular Formula	Found (%) C	H	N	Required (%) C	H	N
6a	Ph	H	Ph	63	43-45	$C_{20}H_{17}NO$	83.62	6.00	4.92	83.62	5.96	4.88
6a$_1$	Ph	H	*n*-C_6H_{13}	38	oil	$C_{20}H_{25}NO$	81.38	8.57	4.71	81.31	8.53	4.74
6a$_2$	Ph	$CH(CH_3)_2$	Ph	63	120-122 [a]	$C_{29}H_{26}N_4O_8$	62.39	4.70	9.94	62.36	4.69	10.03
6b	*p*-Tolyl	H	Ph	65	38-40	$C_{21}H_{19}NO$	84.10	6.39	4.60	83.69	6.36	4.65
6b$_1$	*p*-Tolyl	H	*n*-C_6H_{13}	60	62-64 [a]	$C_{27}H_{30}N_4O_8$	59.89	5.54	10.43	60.21	5.62	10.40
6b$_2$	*p*-Tolyl	$CH(CH_3)_2$	Ph	49	119-120 [a]	$C_{30}H_{28}N_4O_8$	62.50	4.92	9.61	62.93	4.93	9.78
6c	CH_3	H	Ph	89	125-127 [a]	$C_{21}H_{16}N_4O_8$	55.44	3.93	12.39	55.50	3.99	12.33

[a] Mp and analytical data refer to the corresponding picrate; all these propargylamines were oils.

Table 5

^{1}H NMR Assignments of Propargylamines **6**, δ (deuteriochloroform), J (Hz)

Compound	2-Furyl S	m	J	H	Fur-CH_2-N= (s, 2H)	=N-CH-C≡C or =N-CH_2-C≡C-	Aromatics	Others
6a	7.4-7.3	m	-	3*	4.5	4.2 (s, 2H)	7.4-7.3 (m, 3H*)	—
	6.3	dd	1.7, 3.2	1			7.3-7.2 (m, 2H)	—
	6.2	d	3.2	1			7.0 (d, J = 8.3, 2H)	—
6a$_1$	7.3	d	1.8	1	4.5	4.0 (t, J = 1.9, 2H)	7.3-7.2 (m, 2H)	2.2-2.1 (m, 2H)
	6.3	dd	1.8, 3.2	1			7.0 (d, J = 8.7, 2H)	1.5-1.2 (m, 8H)
	6.2	d	3.2	1			6.8 (t, J = 7.6, 1H)	0.9 (t, J = 6.6, 3H)
6a$_2$	7.4-7.1	m	—	8*	4.5	4.2 (d, J = 9.2, 1H)	7.4-7.1 (m, 8H*)	2.3-2.1 (m, 1H)
	6.3-6.2	m	—	1			7.0 (d, J = 8.9, 2H)	1.1 (d, J = 6.5, 3H)
	6.2-6.1	m	—	1			6.8 (t, J = 7.4, 1H)	1.0 (d, J = 6.6, 3H)
6b	7.4-7.3	m	—	3*	4.5	4.2 (s, 2H)	7.4-7.3 (m, 3H*)	2.2 (s, 3H)
	6.3-6.2	m	—	2			7.3-7.2 (m, 3H)	—
							7.1 (d, J = 8.8, 2H)	—
							6.9 (d, J = 8.7, 2H)	—
6b$_1$	7.3	d	1.7	1	4.4	3.9 (t, J = 2.1, 2H)	7.0 (d, J = 8.7, 2H)	2.2 (s, 3H)
	6.3	dd	3.1, 1.7,	1			6.9 (d, J = 8.6, 2H)	2.2-2.1 (m, 2H)
	6.2	d	3.1	1				1.5-1.2 (m, 8H)
								0.9 (t, J = 6.6, 3H)
6b$_2$	7.5-7.2	m	—	6*	4.5	4.1 (d, J = 9.5, 1H)	7.5-7.2 (m, 6H*)	2.2 (s, 3H)
	6.3-6.2	m	—	1			7.0 (d, J = 8.2, 2H)	2.2-2.1 (m, 1H)
	6.2-6.1	m	—	1			6.9 (d, J = 8.6, 2H)	1.1 (d, J = 6.6, 3H)
								1.0 (d, J = 6.5, 3H)
6c	7.4	d	1.9	1	3.7	3.5 (s, 2H)	7.5-7.4 (m, 2H)	2.4 (s, 3H)
	6.3-6.2	m	—	2			7.3-7.2 (m, 3H)	

*1Hα in furan ring, the rest in benzene or *p*-toluene ring.

Table 6

^{13}C NMR Assignments of Propargylamines **6**, δ (deuteriochloroform)

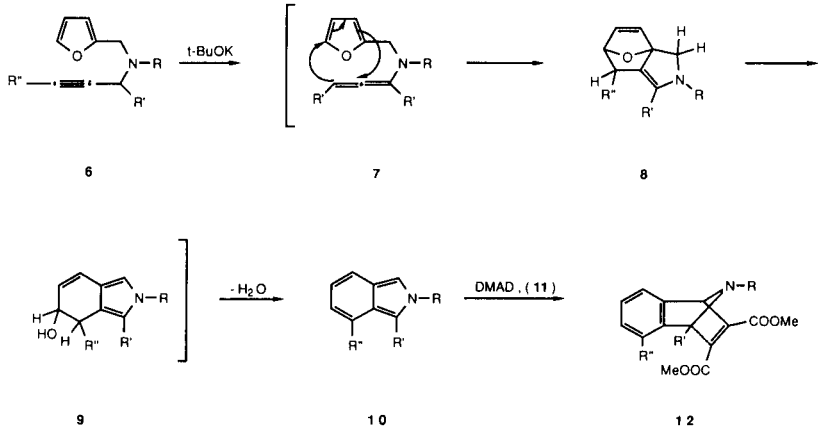
Compound	R″	-C≡C-	CHR′	R	CH_2	2-Furyl
6a	131.7, 128.2, 128.1, 122.9	85.1, 84.3	40.6	148.5, 129.1, 118.6, 114.8	48.2	152.0, 142.0, 110.2, 107.9
6a	31.3, 28.7, 28.4, 22.5, 18.7, 14.0	84.7, 75.4	40.1	148.6, 129.0, 118.3, 114.7	48.0	152.2, 141.9, 110.1, 107.6
6a	131.6, 128.2, 128.0, 123.2	87.5, 85.8	46.4, 32.2, 20.0, 19.9	149.2, 128.9, 118.9, 116.4	60.4	153.3, 141.3, 110.3, 107.4
6b	131.7, 128.1, 128.0, 123.0	85.2, 84.4	40.9	146.5, 129.6, 128.2, 115.5, 20.3	48.4	152.1, 142.0, 110.2, 107.9
6b$_1$	31.3, 28.7, 28.5, 22.5, 18.7, 14.0	84.8, 75.5	40.5	146.6, 129.5, 127.9, 115.4, 20.3	48.3	152.4, 141.9, 110.1, 107.7
6b$_2$	131.5, 128.0, 127.8, 123.0	87.6, 85.7	46.6, 32.0, 19.9, 19.8	147.0, 129.3, 128.1, 117.2, 20.3	60.8	153.4, 141.1, 110.1, 107.3
6c	131.7, 128.3, 128.1, 123.2	85.7, 84.0	45.8	41.6	52.1	151.9, 142.3, 110.1, 108.8

of **4c** (*N*-methyl group) are present.

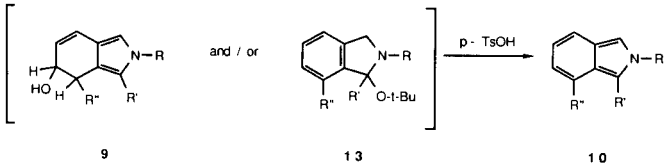
The propargylamines **6** were characterized by their ^{1}H and ^{13}C nmr spectra (Tables 5 and 6). For amines **6** in the ^{1}H nmr, methylene singlets of the 2-furfuryl moiety appear at 4.83-4.76 ppm. The propargyl methylene of **6a**, **6b** and **6c** gives singlets, while the same group in the rest of amines **6** gives doublets or triplets when R′ or R″ are the alkyl substituents. In the ^{13}C nmr spectra of the propargylamines **6** the acetylenic carbon peaks appear at 84.0-87.6 ppm, except that when R″ is a *n*-hexyl group, the peak of the C-1 carbon is shifted about 10 ppm upfield.

Intramolecular cyclization of *N*-furfuryl-*N*-propargylamine **6a** required refluxing in *p*-xylene with potassium *t*-butoxide for a few minutes, and gave the corresponding

Scheme 2



Scheme 3



isoindole **10a** in ~20% overall yield. Without added catalyst the propargyl amine **6a** was recovered quantitatively after 24 hours reflux. The potassium *t*-butoxide isomerized the propargyl groups to allenes, which are more reactive [9] in the intramolecular Diels-Alder cyclization (Scheme 2). However, attempts at the cyclization of amines **6a**$_1$, **6b**$_1$, **6c** were unsuccessful even after 6 days heating with a ten fold excess of potassium *t*-butoxide.

Because of their instability, the isoindoles were trapped and characterized by intermolecular Diels-Alder reactions with dimethyl acetylenedicarboxylate (**11**) to give cycloadducts **12**. After refluxing the propargylamines **6** in *t*-butanolic potassium *t*-butoxide for 2-26 hours, *p*-toluenesulfonic acid was added to neutralize the strongly basic solution and avoid hydrolysis of the subsequently added dimethyl acetylenedicarboxylate. It is also possible that a hydroxydihydroisoindole species **9** and/or butoxy derivative **13** still exists under the basic conditions, *p*-toluenesulfonic acid should convert these species into the desired isoindoles **10** (Scheme 3), as happens in the case of isobenzofurans [9]. A similar method was used [9] for the intramolecular cyclization of furfuryl propargyl ethers.

Table 7 reports the prepared isoindole derivatives **12**. The structure of adducts **12** was confirmed by their ^{1}H and ^{13}C nmr spectra. In the ^{1}H nmr spectra the characteristic signals for isoindoline aliphatic protons appear at 5.9-5.2 ppm as singlets for **12a**$_2$ and **12b**$_2$. These signals are finely split doublets for **12a** and **12b** (J = 1-2 Hz) (Table 8). Chemical shifts for the isoindole carbons C-1 and C-3 in **12a** and **12b** are similar (72.3, 71.1 and 72.5, 71.3 ppm, respectively) (Table 9). In ^{13}C spectra of **12a**$_2$ and **12b**$_2$ the peaks at 90.8 and 90.7 ppm were identified by ATP test as the quarternary carbons (C-3 in isoindole system). The remaining signals in ^{1}H as well as in ^{13}C spectra are in good agreement with the structure of isoindole adducts **12** (Tables 8 and 9).

EXPERIMENTAL

All melting points are uncorrected and were taken either in open glass capillary tubes with a Thomas-Hoover melting point apparatus or in a Kofler-stage microscope. The ^{13}C nmr spectra were obtained at 50 MHz on a Varian XL-200 NMR spectrometer or at 75 MHz on a Varian VRX-300 NMR spectrometer, referred to deuteriochloroform (δ = 77.0). The ^{1}H nmr spectra were obtained at 200 MHz or 300 MHz on the same NMR spectrometers,

Table 7

Synthesis and Characterization of Isoindole Adducts **12**

Product	R	R′	R″	Yield (%)	mp (°C)	Molecular Formula	Found (%) C	H	N	Required (%) C	H	N
12a	Ph	H	Ph	32	161-163	$C_{26}H_{21}NO_4$	75.92	5.26	3.31	75.90	5.14	3.40
12a$_2$	Ph	$CH(CH_3)_2$	Ph	30	175-176	$C_{29}H_{27}NO_4$	76.56	6.09	3.00	76.80	6.00	3.09
12b	*p*-Tolyl	H	Ph	60	138-139	$C_{27}H_{23}NO_4$	76.36	5.44	3.23	76.22	5.45	3.29
12b$_2$	*p*-Tolyl	$CH(CH_3)_2$	Ph	12	150-151	$C_{30}H_{29}NO_4$	76.98	6.31	2.97	77.06	6.25	3.00

Table 8

^{1}H NMR Assignments of Isoindole Adducts **12**, δ (deuteriochloroform) J (Hz)

Compound	Isoindoline aliphatic protons	Aromatic protons	CH_3O	Others
12a	5.9 (d, J = 1.9, 1H), 5.8 (d, J = 1.9, 1H)	7.6-7.4 (m, 6H), 7.2-7.1 (m, 4H)	3.8 (s, 3H), 3.7 (s, 3H)	— —
12a$_2$	5.2 (s, 1H)	7.7-7.5 (b, 2H), 7.5-7.2 (m, 4H), 7.2-6.9 (m, 7H)	3.9 (s, 3H), 3.8 (s, 3H)	2.8-2.6 (m, 1H), 0.7 (d, J = 7.0, 3H), 0.4 (d, J = 7.0, 3H)
12b	5.8 (d, J = 0.9, 1H) 5.7 (d, J = 1.9, 1H)	7.6-7.4 (m, 6H), 7.1 (d, J = 4.6, 2H), 7.0 (d, J = 8.1, 2H), 6.7 (d, J = 8.4, 2H)	3.8 (s, 3H), 3.7 (s, 3H)	2.2 (s, 3H)
12b$_2$	5.2 (s, 1H)	7.7-7.5 (b, 2H) 7.4-7.2 (m, 4H), 7.1-7.0 (m, 1H), 7.0-6.9 (m, 5H)	3.9 (s, 3H), 3.8 (s, 3H)	2.8-2.6 (m, 1H), 2.2 (s, 3H), 0.7 (d, J = 7.0, 3H), 0.4 (d, J = 7.0, 3H)

Table 9

^{13}C NMR Assignments of Isoindole Adducts **12**, δ (deuteriochloroform)

Compound	Isoindoline aliphatic carbons	Aromatic quaternary carbons	Other aromatic carbons	C=C	C=O	CH_3O	Others
12a	72.3, 71.1	146.2, 145.3, 143.4, 139.3, 137.3	129.1, 128.5, 128.5, 127.5, 126.5, 126.2 121.8, 121.2, 117.9	149.9, 149.7	163.6, 163.6	52.3, 52.2	—
12a$_2$	90.8, 72.9	155.7, 146.6, 145.7, 141.0, 138.3	129.7, 129.3, 128.3, 127.6, 127.2, 126.2, 125.4, 120.6	150.5, 149.7	166.6, 163.0	52.2, 52.2	26.3, 18.7, 18.7
12b	72.5, 71.3	146.3, 143.4, 142.9 139.4, 137.3, 131.1	129.6, 128.5, 128.5, 127.4, 126.4, 126.2, 121.2, 117.9	149.8, 149.6	163.7, 163.7	52.3, 52.1	20.5
12b$_2$	90.7, 73.1	155.8, 145.8, 144.0, 141.0, 138.3, 136.0	129.7, 129.2, 129.0, 127.6, 127.2, 127.1, 125.3, 120.6	150.6, 149.9	166.6, 163.1	52.2, 52.2	26.3, 28.7, 18.8

with tetramethylsilane as an internal standard. Microanalyses were obtained on a Carlo Erba 1106 elemental analyzer.

Commercially available reagent grade solvents and reagents were used without further purification except tetrahydrofuran, which was distilled from benzophenone-sodium. *N*-Phenyl-, *N*-*p*-tolyl- and *N*-methylfurfurylamines were obtained from the appropriate *N*-substituted furfurylideneimines by reduction with sodium borohydride [10].

General Procedure for the Preparation of Benzotriazole Adducts **4**.

Equimolar amounts (10 mmoles) of the amine, benzotriazole and the aldehyde were refluxed in 100 ml of benzene in a Dean-Stark apparatus until the theoretical amount of water was removed. The solution was washed with saturated sodium carbonate and dried over magnesium sulfate. Solvent was evaporated and the oily residue was dissolved in ethyl ether and cooled to give adducts **4** (Table 1).

General Procedure for the Preparation of Propargylamines **6**.

To a cold (−70°) solution of 10 mmoles of the aryl- or alkylacetylene in freshly distilled tetrahydrofuran (20 ml), a solution (2.5*M*) of *n*-butyllithium (4.4 ml, 11 mmoles) in hexane was added under an argon atmosphere. Stirring at 20° for 2 hours was followed by the addition of 10 mmoles of the appropriate benzotriazole adduct **4a**, **4b**, **4c** in tetrahydrofuran (20 ml) or benzene; crude material from the previous step was used for the preparation of **6a$_2$** and **6b$_2$**. The reaction mixture was refluxed for 5 hours, and after cooling to 20°, quenched with water and extracted with ethyl ether. The organic layer was washed with saturated sodium carbonate and dried (magnesium sulfate). Solvent was removed under reduced pressure and the oily residue was purified by column chromatography (silica gel; petroleum ether: chloroform/5:1/ as an eluent) to give the propargylamine (Table 4).

General Procedure for the Preparation of Isoindole Adducts **1**.

The propargylamine (2 mmoles) and an equimolar amount (for the preparation of **12a**), or a two fold excess (for preparation of **12b**), or a 10 fold excess (for the preparation of **12a$_2$** and **12b$_2$**) of potassium *t*-butoxide in 25-50 ml of *t*-butyl alcohol were refluxed for 2-26 hours with stirring, under argon. Then an equimolar (to the potassium *t*-butoxide) amount of *p*-toluenesulphonic acid in *t*-butyl alcohol was added and the reaction mixture was stirred for 1 hour at 20°. Dimethyl acetylenedicarboxylate (3 mmoles) was added and the reaction mixture was stirred overnight at 20°. The reaction mixture was poured into water and extracted with ethyl ether. The ethereal phase was washed with brine and dried (magnesium sulfate). Evaporation of the solvent and purification of the residue by column chromatography (silica gel, methylene chloride as an eluent) gave an oily fraction which crystallized on scratching with ethanol or ethanol/petroleum (see Table 7).

REFERENCES AND NOTES

[1] Part **21** of the series, "The Chemistry of *N*-Substituted Benzotriazoles". For Part **20** see, A. R. Katritzky and J. Vanden Eynde, *J. Chem. Soc., Perkin Trans. 1*, in press.

[2a] Permanent address: Department of Organic Chemistry, Academy of Medicine, Dzierzynskiego 14b str., 30048 Cracow, Poland. [b] Permanent address: Department of Chemistry, Aristotelian University of Thessaloniki, Thessaloniki 540 06, Greece.

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