Photoinduced Reactions of Phenyl-Substituted Acetonitriles in the Presence of Amine

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Irradiation of phenylacetonitrile and its derivatives (1) in the presence of triethylamine gave α-benzylated triethylamine (2), bibenzyl (3), and toluene (4) derivatives. The formation of these products was explained in terms of a benzylic radical intermediate formed by electron transfer between the substrate and triethylamine, followed by elimination of a cyanide anion from the radical anion of 1.

Key words photoreaction; phenylacetonitrile; electron transfer; aliphatic amine

Photochemical reactions of aromatic compounds with a good leaving group, such as halogen, hydroxyl, alkoxyl or acyloxyl, at a benzylic position have been reported.[1-12] The major reaction paths are homolysis and/or heterolysis depending on the reaction conditions.[11,12] For instance, benzyl acetate gives benzyl methyl ether as a benzyl cation-derived product, as well as toluene, bibenzyl, and 2-phenylethanol as radical products, on irradiation in methanol.[8] However, the presence of an electron acceptor[5,12,13] or a donor[14,15] alters the reaction path. For instance, Ohashi *et al.*[14] irradiated benzylic acetates in the presence of triethylamine to obtain products derived only from a benzylic radical. The electron-donating nature of aliphatic amines is well established. We report here photochemical reactions of phenylacetonitrile derivatives possessing a cyano group at the benzylic position in the presence of an amine.

When phenylacetonitrile (**1a**) was irradiated with a low-pressure mercury lamp (150 W) in an acetonitrile–triethylamine (9 : 1 v/v) mixture in a quartz tube under a nitrogen atmosphere, photochemical reaction proceeded smoothly within a few minutes. After repeated chromatographies of the tarry product mixture, an α-benzylated triethylamine (**2a**), bibenzyl (**3a**), and toluene (**4a**) were obtained in 18%, 16%, and 16% yields, respectively (Table 1, run 1) Irradiation of *p*-methoxyphenylacetonitrile (**1b**) or diphenylacetonitrile (**1c**) under similar conditions gave corresponding products (**2**—**4**) in varying yields (runs 2,3). Triphenylacetonitrile (**1d**) also produced **2d** and triphenylmethane (**4d**), but no dimeric compound[16] was found in the mixture (run 4). In the absence of triethylamine, starting materials were recovered except for **1d**, which gave a trace of biphenyl[17] on prolonged irradiation. Under the irradiation conditions **3** and **4** were photostable, but **2** were converted to **4** (see below).

p-Cyanophenylacetonitrile (**1e**) behaved quite differently: on irradiation for 15 min, **1e** gave 2-(*p*-cyanophenyl)butyronitrile (**5**) and a diastereomeric pair of **6** in low yields (run 5). Compound **6** did not produce **5** under similar conditions.

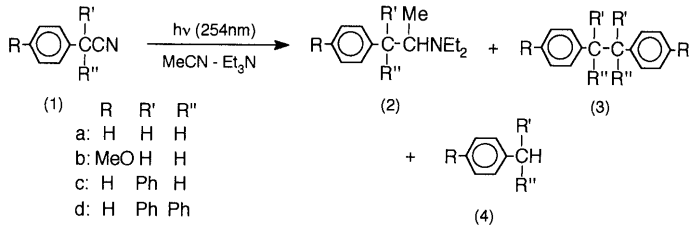

Chart 1

Table 1. Products and Yields of the Photoreactions of Phenylacetonitrile Derivatives (**1**)

Run	Substrate				Solvt.	Amine	Time (min)	Convn. (%)	Products and yields (%)[a]			
		R	R′	R″					**2**	**3**	**4**	Others
1	**1a**	H	H	H	MeCN	Et_3N	3	44	18	16	16	
2	**1b**	MeO	H	H	MeCN	Et_3N	3	14	38	15	28	
3	**1c**	H	Ph	H	MeCN	Et_3N	3	45	26	7	18	
4	**1d**	H	Ph	Ph	MeCN	Et_3N	3	53	10	0	67	
5	**1e**	NC	H	H	MeCN	Et_3N	15	57	0	0	0	**5** (11), **6** (8)
6	**1c**	H	Ph	H	MeCN	DABCO	3	35	0	30	0	
7	**1c**	H	Ph	H	c-C_6H_{12}	Et_3N	3	35	10	3	21	
8	**1c**	H	Ph	H	MeOH	Et_3N	3	55	8	3	21	**10** (16)
9	**1c**	H	Ph	H	MeOH	DABCO	3	18	0	12	0	**10** (34)

a) Based on the substrate consumed.

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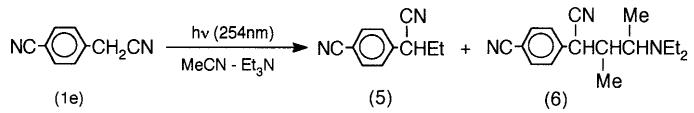

Chart 2

Table 2. Fluorescence Quenching of Phenylacetonitrile Derivatives with Amine in Acetonitrile

Substrate	Amine	$k_q\tau$/l mol^{-1}	τ[a]/ns	k_q/10^{10} l mol^{-1} s^{-1}	r[b]
1a	Et_3N	152	10.7	1.4	0.988
1b	Et_3N	53.8	6.1	0.88	0.990
1c	Et_3N	285	9.3	3.1	0.995
1d	Et_3N	69.3	—	—	0.997
1e	Et_3N	143	9.4	1.5	0.994
1c	DABCO	433	9.3	4.7	0.980

a) Determined in the present study. *b*) Relative coefficient of the Stern–Volmer plot.

Table 3. Free Energy Changes for Electron Transfer Reactions of Phenylacetonitrile Derivatives (**1**) with Amine in Acetonitrile

Substrate	Amine	E_{ox}[a]/V *vs.* SCE	E_{red}[b]/V *vs.* SCE	ΔG/kJ mol^{-1}
1a	Et_3N	0.96	[c]	—
1b	Et_3N	0.96	−2.20	−118
1c	Et_3N	0.96	−1.82	−179
1e	Et_3N	0.96	−2.34	−113
1c	DABCO	0.68	−1.82	−207

a) Ref. 25. *b*) Determined in the present study. *c*) Not determined due to decomposition of the supporting electrolyte.

Fluorescence of phenylacetonitriles (**1**) was quenched by an amine. Stern–Volmer plots of Φ_0/Φ against [amine] were linear ($r \geqq 0.98$) and the quenching rate constants (k_q) obtained from the slope of the plot were almost of the order of a diffusion-controlled rate of 10^{10} l mol^{-1} s^{-1} (Table 2), implying the interaction of an excited singlet state with an amine. Reduction potentials of **1** were measured in acetonitrile and the free energy change (ΔG) for a one-electron transfer[18] from the excited singlet state of **1** was calculated (Table 3). All the compounds showed a large negative ΔG value. These physicochemical data strongly suggest an electron transfer as the first step and the subsequent formation of a radical anion of **1**. Formation of benzylic radical-derived products (**3**, **4**) can be explained in terms of elimination of cyanide anion from the radical anion formed. Formation of **2** is also explicable by recombination of a benzylic radical and 1-(*N*,*N*-diethylamino)ethyl radical (**8**) derived from a triethylamine radical cation (**7**) by deprotonation. Similar recombination of radicals derived from a radical anion–radical cation pair has been reported.[19] Although we have not obtained direct evidence, *e.g.*, by physical methods such as CIDEP or CIDNP (chemically induced dynamic electron/nuclear polarizations), for the recombination of a radical pair, the above mechanism is consistent with the structures and the distribution of the products.

To clarify further the course and the mechanism of these reactions, the reactions of **1c** were carried out under various conditions. When **1c** was irradiated in CD_3CN–triethylamine mixture, no deuterium was incorporated into **4c**. This suggests that **4c** is not a direct hydrogen

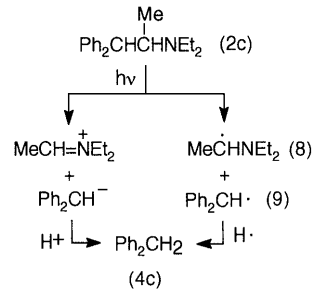

Chart 3

abstraction product of a diphenylmethyl radical (**9**), although the possibility that the radical abstracted the α-hydrogen of the triethylamine radical cation in their solvent cage can not be excluded. Alternatively, the radical (**9**) may recombine with the 1-(*N*,*N*-diethylamino)ethyl radical (**8**) to give the α-benzylated product **2c**. When **2c** was irradiated, **4c** was produced as a major product, but no **3c** was obtained even in the absence of triethylamine. Furthermore, when 1,4-diazabicyclo[2.2.2]octane (DABCO) was used as the amine in the irradiation, **1c** gave the dimer **3c** as a sole product (run 6). Thus, hydrogen abstraction by the radical (**9**) from the solvent or other hydrogen source does not occur, implying intermediacy of **2c** in the formation of **4c**. The C–C bond cleavage of the Ar–C–C–N system by irradiation without an electron acceptor is interesting.[20] On irradiation in MeOD for 10 min, **2c** gave **4c**-d_0, -d_1, and -d_2 in a ratio of 40 : 52 : 8.[21] This means that both heterolysis and homolysis occurred in the fragmentation of **2c** (Chart 3), probably *via* an intramolecular electron transfer between the β-amino group and aromatic moieties.[22] Relatively low yields of the dimer (**3**) compared with those of the radical recombination products (**2**, **4**) may be due to in-cage recombination of the radicals and a low concentration of escaped radicals.

In cyclohexane–triethylamine, the reaction was almost the same as in acetonitrile (run 7). However, in methanol–triethylamine (run 8), **1c** gave diphenylmethyl methyl ether (**10**) in 16% yield along with the other products. This can be explained in terms of oxidation of the radical (**9**) by the amine radical cation (**7**), followed by nucleophilic attack of methanol.[23] Oxidation of **9** by **7** is exothermic as calculated by means of the equation,[23]

$$\Delta G_{ox} = E^{ox}_{1/2}(Ph_2CH\cdot) - E^{red}_{1/2}(Et_3N^{+\cdot}) = E^{ox}_{1/2}(Ph_2CH\cdot) - E^{ox}_{1/2}(Et_3N)$$

Substituting +0.35[24] and +0.96[25] V *vs.* SCE for $E^{ox}_{1/2}(Ph_2CH\cdot)$ and $E^{ox}_{1/2}(Et_3N)$, we obtained −0.61 V for this process. This mechanism is supported by the finding that when the reaction was done in methanol–DABCO, the same product (**10**) was obtained in 34% yield (run 9). Possible pathways for the above reactions are summarized in Chart 4.

The mechanism of the unusual reaction of **1e** is unclear.

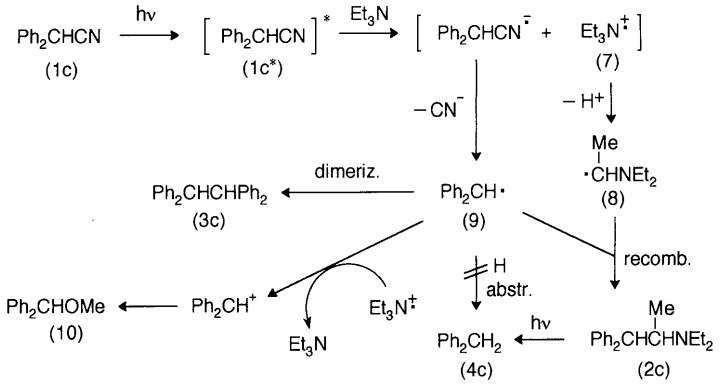

Chart 4

However, the large negative ΔG values for the electron transfer for both *p*-methoxy- (**1b**) and *p*-cyano- (**1e**) phenylacetonitriles (Table 3) suggests that the substituent does not affect the electron transfer step. We are inclined to ascribe the difference to the elimination step of cyanide anion from the benzylic radical anion. An electron-withdrawing *p*-cyano group does not facilitate the elimination of cyanide anion, leading to low conversion in the reaction of **1e** (14%).

In conclusion we have found that phenylacetonitriles are photocleaved at the benzylic position to afford the benzylic radical under photoinduced electron transfer conditions.

Experimental

Irradiations were carried out in a quartz tube with a 125 W low-pressure mercury lamp (Sen Lamp UBL-125) under a nitrogen atmosphere. The structures of volatile products were determined by GC-MS (Shimadzu QP-1000) and the product yields were determined by GC (Shimadzu GC-14B) using an appropriate internal standard. Capillary columns (CBP-1-m25-050, CBP-1-s25-050) were used, respectively. ^{1}H- and ^{13}C-NMR spectra were measured with a Varian Unity plus 500 (500 and 125 MHz, respectively) or a JEOL GX-270 (270 and 67.5 MHz, respectively) spectrometer in $CDCl_3$. HPLC (Nihon Bunseki Kogyo, LC-908) was run on GPC-type columns (1H, 2H) with toluene as an eluent. Chromatography was carried out over alumina (Wako, activated) or aminoalkylated silica (Fuji Davison, DM1020).

Irradiation of Phenylacetonitrile (1a) **1a** (100 mg) was irradiated in acetonitrile–triethylamine (9 : 1, 10 ml) for 3 min. Chromatography of the brown tar product mixture over alumina and then aminoalkylated silica gave 1,2-diphenylethane (**3a**) and *N*,*N*-diethyl-1-methyl-2-phenylethylamine (**2a**) as an oil. **2a**: ^{1}H-NMR ($CDCl_3$) δ: 0.92 (3H, d, J=7.0 Hz), 1.07 (6H, t, J=7.0 Hz), 2.38 (1H, dd, J=13.0, 9.5 Hz), 2.55 (4H, dq, J=14.0, 7.0 Hz), 2.94 (1H, m), 3.03 (1H, m), 7.15—7.31 (5H, m). ^{13}C-NMR ($CDCl_3$) δ: 14.29 (q), 14.72 (q), 39.59 (t), 43.51 (t), 57.00 (d), 125.65 (d), 128.14 (d), 129.23 (d), 141,05 (s). MS (70 eV) *m*/*z* (%): 190 (M^+-1, 0.1), 100 (100), 91 (10), 72 (12). Formation of toluene (**4a**) was determined by GC-MS.

Irradiation of *p*-Methoxyphenylacetonitrile (1b) **1b** (105 mg) was irradiated in acetonitrile–triethylamine (9 : 1, 10 ml) for 3 min. Chromatography of the mixture over alumina gave 1,2-bis(*p*-methoxyphenyl)ethane (**3b**) as colorless crystals, mp 126 °C (EtOH, lit[26] 130 °C). Further chromatography over aminoalkylated silica gave *N*,*N*-diethyl-1-methyl-2-(*p*-methoxyphenyl)ethylamine (**2b**) as a yellow oil. ^{1}H-NMR ($CDCl_3$) δ: 0.90 (3H, d, J=6.6 Hz), 1.07 (6H, t, J=7.0 Hz), 2.37 (1H, dd, J=13.0, 9.5 Hz), 2.56 (4H, m), 2.84 (1H, m), 2.97 (1H, m), 3.78 (3H, s), 6.81 (2H, d, J=8.4 Hz), 7.07 (2H, d, J=8.4 Hz). ^{13}C-NMR ($CDCl_3$) δ: 14.29 (q), 14.54 (q), 38.74 (t), 43.51 (t), 55.17 (q), 57.10 (d), 113.57 (d), 130.05 (d), 133.08 (s), 157.68 (s). MS (70 eV) *m*/*z* (%): 121 (4), 100 (100). Formation of *p*-methoxytoluene (**4b**) was determined by GC-MS.

Irradiation of Diphenylacetonitrile (1c) **1c** (101 mg) was irradiated for 3 min. Chromatography of the mixture over alumina gave **1c** and diphenylmethane (**4c**) as a colorless oil. Further chromatography over aminoalkylated silica gave 1,1,2,2-tetraphenylethane (**3c**), mp 210—212 °C (lit[27] 208—210 °C), and *N*,*N*-diethyl-1-methyl-2,2-diphenylethylamine (**2c**) as an oil. ^{1}H-NMR (270 MHz, $CDCl_3$) δ: 0.82 (6H, t, J=7.0 Hz), 0.85 (3H, d, J=7.0 Hz), 2.27 (2H, dq, J=13.0, 7.0 Hz), 2.55 (2H, dq, J=13.0, 7.0 Hz), 3.60 (1H, dq, J=11.0, 7.0 Hz), 3.85 (1H, d, J=11.0 Hz), 7.10—7.32 (10H, m). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 11.29 (q), 14.22 (q), 43.20 (t), 57.48 (d), 57.77 (d), 125.46 (d), 126.06 (d), 127.64 (d), 128.28 (d), 128.43 (d), 128.47 (d), 144.37 (s). MS (70 eV) *m*/*z*(%): 266 (M^+-1, 0.1), 167 (1), 100 (100), 72 (11).

Irradiation of **1c** in methanol–triethylamine (9 : 1) was conducted in a similar manner and diphenylmethyl methyl ether (**10**) was obtained as an oil identical with a standard sample,[28] in addition to **2c**, **3c**, and **4c**.

Irradiation of **1c** in acetonitrile or methanol (4 ml)–DABCO (449 mg) was conducted in a similar manner and the product yields were determined as above.

Irradiation of Triphenylacetonitrile (1d) **1d**[29] (103 mg) was irradiated for 3 min. Unreacted **1d** was removed by chromatography over alumina and further chromatography over aminoalkylated silica gave triphenylmethane (**4d**) as colorless needles, mp 94.5—94.8 °C, and *N*,*N*-diethyl-1-methyl-2,2,2-triphenylethylamine (**2d**) as an oil. ^{1}H-NMR (300 MHz, $CDCl_3$) δ: 0.81 (6H, t, J=6.9 Hz), 1.11 (3H, d, J=6.9 Hz), 1.20 (2H, dq, J=13.8, 6.9 Hz), 2.41 (2H, dq, J=13.8, 6.9 Hz), 4.55 (1H, q, J=6.9 Hz), 7.10—7.46 (15H, m). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 12.81 (q), 12.81 (q), 44.64 (t), 61.97 (d), 63.37 (s), 125.38 (d), 127.06 (d), 130.54 (d), 146.40 (s). MS (70eV) *m*/*z* (%): 328 (M^+-15, 0.2), 270 (2), 165 (41), 100 (100), 72 (45).

Irradiation of *p*-Cyanophenylacetonitrile (1e) **1e**[30] (99 mg) was irradiated in acetonitrile–triethylamine (9 : 1, 10 ml) for 15 min. Chromatography of the mixture over alumina and aminoalkylated silica followed by HPLC gave 2-(*p*-cyanophenyl)butyronitrile (**5**) as a yellow oil. ^{1}H-NMR (270 MHz, $CDCl_3$) δ: 1.10 (3H, t, J=7.3 Hz), 1.97 (2H, dq, J=7.3 Hz), 3.83 (1H, t, J=7.3 Hz), 7.47 (2H, d, J=8.4 Hz), 7.70 (2H, d, J=8.4 Hz). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 11.26 (q), 28.90 (t), 38.82 (d), 112.19 (s), 118.06 (s), 19.41 (s), 128.11 (d), 132.77 (d), 140.83 (s). MS (70 eV) *m*/*z* (%): 170 (M^+, 20), 142 (100), 115 (23) and the diastereomeric 2-(*p*-cyanophenyl)-4-(diethylamino)-3-methylvaleronitriles (**6**). Isomer A: ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 0.87 (3H, d, J=7.0 Hz), 0.98 (3H, d, J=6.5 Hz), 1.14 (6H, t, J=7.0 Hz), 1.78 (1H, m), 2.37 (2H, dq, J=13.5, 7.0 Hz), 2.59 (2H, dq, J=13.5, 7.0 Hz), 2.73 (1H, dq, J=10.5, 6.5 Hz), 5.20 (1H, d, J=3.0 Hz), 7.42 (2H, d, J=8.0 Hz), 7.68 (2H, d, J=8.0 Hz). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 10.51 (q), 12.45 (q), 15.19 (q), 40.38 (d), 43.70 (t), 44.17 (d), 57.89 (d), 111.96 (s), 118.75 (s), 118.78 (s), 128.88 (d), 133.01 (d), 142.53 (s). MS (70 eV) *m*/*z* (%): 269 (M^+, 0.5), 254 (0.3), 141 (5), 100 (100), 72 (12). Isomer B: ^{1}H-NMR (300 MHz, $CDCl_3$) δ: 0.97 (3H, d, J=6.6 Hz), 1.01 (6H, t, J=6.9 Hz), 1.12 (3H, d, J=6.6 Hz), 1.84 (1H,m), 2.37 (2H, dq, J=13.2, 6.9 Hz), 2.51 (2H, dq, J=13.2, 6.9 Hz), 2.74 (1H, dq, J=8.7, 6.6 Hz), 4.18 (1H, d, J=3.9 Hz), 7.47 (2H, d, J=8.4 Hz), 7.69 (2H, d, J=8.4 Hz). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 10.94 (q), 12.73 (q), 14.09 (q), 40.46 (d), 43.29 (d), 43.47 (t), 57.67 (d), 111.84 (s), 118.02 (s), 118.18 (s), 128.46 (d), 132.59 (d), 141.12 (s). MS (70 eV) *m*/*z* (%): 269(M^+, 0.1), 254 (0.4), 141 (4), 100 (100), 72 (7).

Fluorescence Experiments Fluorescence quenching experiments with the phenylacetonitriles (**1a—d**) were carried out in acetonitrile, methanol, or cyclohexane solutions using a Hitachi F-4000 fluorometer. Concentrations of the substrates were $1—3\times10^{-3}$ mol/l, due to the weak

fluorescence. Spectra were obtained at five different amine concentrations (10^{-2}—10^{-3} mol/l) for each substrate and the results are summarized in Table 2. Fluorescence lifetimes were measured by a single photon counting method using a Horiba NAES-550 spectrometer.

Reduction Potential Measurements Reduction potentials of **1a**—**c**, **e** were obtained in acetonitrile with 0.1 mol/l tetraethylammonium perchlorate as a supporting electrolyte using a potentio-galvanostat (Nikko Keisoku NPGS-301). The results are summarized in Table 3.

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