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Anodic Oxidation of Carboxamides. IV.[1] Anodic Dimerization of 4- and 4′-Substituted Benzanilides in the Presence of a Strong Base

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Anodic oxidation of several 4- and 4′-substituted benzanilides in the presence of a strong base, 1,8-diazabicyclo[5.4.0]undecene-7 (DBU), was investigated by cyclic voltammetry and controlled potential electrolysis at a glassy carbon electrode in acetonitrile. The original voltammetric peak of the anilides shifted to less positive potentials on addition of DBU. The magnitude of the cathodic shift was linearly correlated with the Hammett σ values of the 4′-substituents, and the oxidation process could be explained in terms of a one-electron abstraction from the anilide which exists as a hydrogen-bonded complex with the base. Controlled potential electrolysis of the anilides under basic conditions gave three different types of dimers depending on the nature of the 4′-substituent, X. When X = alkyl group, the 4-(*N*-benzoyl-*N*-phenyl)amino-2,5-cyclohexadienone derivative (**2**) was obtained. Diphenylamine derivatives (**3**) and (**4**) produced through N-2′ and N-4′ dimerization were the main products when X = hetero atom and X = H, respectively. A sandwich-type approach of the radicals formed by the initial one-electron transfer can account for these results.

Keywords——benzanilide; 1,8-diazabicyclo[5.4.0]undecene-7 (DBU); cyclic voltammetry; controlled potential electrolysis; anodic oxidation; anodic dimerization

In our previous studies on anodic oxidation of 4′-methoxybenzanilides[2] and *N*-alkyl-4′-methoxybenzanilides,[3] the oxidation processes of the anilides in acetonitrile and in methanol have been established. In these studies, however, the substrates for macro-scale electrolysis were mainly restricted to the anisidides because of the difficulties in obtaining characterizable products from anilides without a methoxy group on the N-phenyl ring. Hess and co-workers[4] reported on the anodic oxidation of *N*-isopropylacetanilide to give a *para–para* dimerized product, but the yield was low and the mechanism of the reaction was not determined.

In our first study of this series,[2] a cathodic shift (*ca.* 200 mV) of the oxidation potential of carboxanilides was observed in acetonitrile on addition of pyridine, and this was explained in terms of the formation of a hydrogen-bonded complex between the amide hydrogen atom and the added base. This idea has been developed into a general concept which implies a linear relationship between the magnitude of the potential shift and the strength of the added base.[5] Thus, the oxidation potential of the anilides should be lowered further with a base stronger than pyridine, and the possibilities of undesirable follow-up chemical and/or electrochemical reactions can be minimized so that stable products other than intractable tars may be obtained. Another kind of initial electrode process is also expected, as observed in phenol oxidation,[6] where the process involving a two-electron transfer and loss of a proton in acetonitrile is replaced by a one-electron transfer in the presence of a strong base, hydroxide ion.

This paper reports the results of cyclic voltammetry and controlled potential electrolysis of 4- and 4′-substituted benzanilides in acetonitrile in the presence of a strong base, 1,8-diazabicyclo[5.4.0]undecene-7 (DBU).[7]

Results and Discussion

Cyclic Voltammetry

On cyclic voltammetry in acetonitrile containing 0.1 M DBU perchlorate (DBUP) as a supporting electrolyte, 4′-methylbenzanilide (**1a**) showed an irreversible oxidation wave at 1.47 V *vs.* SCE (Fig. 1-A). When DBU was added to the system, an extra wave developed at 0.66 V. The peak current of the extra wave first increased linearly with the amount of DBU and then gradually reached a limiting value (Fig. 2), while the peak potential remained unchanged. On the other hand, the original wave at 1.47 V reduced its peak current concomitantly and completely disappeared when the extra wave attained its limiting value (Fig. 1-B: the wave at 1.10 V is ascribed to the oxidation of DBU itself). These results suggest, as in the case of the 4′-methoxybenzanilide (**1e**)—pyridine system,[2] that a one-to-one hydrogen-bonded complex is formed between **1a** and DBU, and undergoes electron transfer at the extra wave. The position of the hydrogen-bonding in **1a** must be at the amide hydrogen

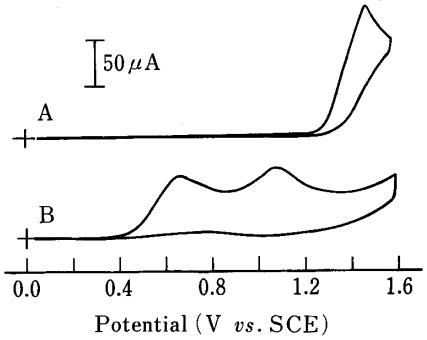


Fig. 1. Cyclic Voltammograms of 4′-Methylbenzanilide (**1a**)

In acetonitrile containing 0.1 M DBUP: (A), without added base; (B), with DBU (2 eq). Concentration of the substrate was *ca.* 5 mM. Glassy carbon anode (area = 0.071 cm^{-1}); voltage sweep rate, 50 mV s^{-1}; at 25 °C.

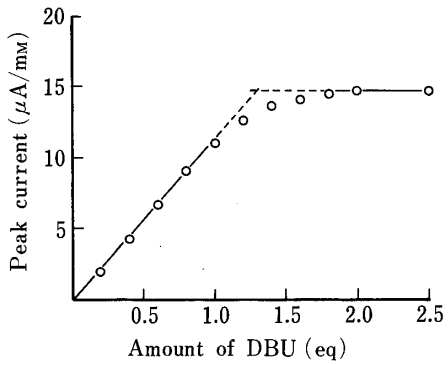
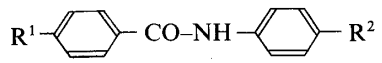


Fig. 2. Plot of Peak Current of the Extra Wave *vs.* Amount of Added Base

TABLE I. Results of Cyclic Voltammetry[a] of Benzanilides **1** in Acetonitrile Containing 0.1 M DBUP



Compound	R^1	R^2	E_p[b]	E'_p[c]	ΔE_p	i_p/C[d]	i'_p/C[d]	i'_p/i_p
1a	H	Me	1.47	0.66	0.81	28.9	15.6	0.54
1b	H	Et	1.51	0.69	0.82	41.0	14.6	0.36
1c	H	iso-Pr	1.41	0.71	0.70	33.4	14.2	0.42
1d	Me	Me	1.48	0.64	0.84	32.0	16.6	0.52
1e	H	OMe	1.24	0.52	0.72	26.8	13.5	0.50
1f	H	Cl	1.67	0.70	0.97	44.9	22.2	0.49
1g	H	NO_2	1.90	0.80	1.10	32.2	23.3	0.72
1h	H	H	1.65	0.88	0.77	39.3	26.4	0.67
1i	Me	H	1.58	0.82	0.76	35.6	23.4	0.66
1j	OMe	H	1.56	0.78	0.78	30.2	24.0	0.79
1k	Cl	H	1.66	0.90	0.76	34.9	19.6	0.56

a) At 25 °C; glassy carbon anode (area = 0.071 cm^2); voltage sweep rate, 50 mV s^{-1}; substrate concentration, *ca.* 5 mM. *b*) Peak potential without added base: V *vs.* SCE. *c*) Peak potential with DBU (>2 eq). *d*) Normalized peak currents without and with DBU, respectively: μA/mM.

atom because *N*-methyl-4′-methylbenzanilide, which showed an irreversible oxidation wave at 1.49 V, did not give an extra wave in the presence of DBU. In Fig. 2, extrapolation of the straight line drawn through the increasing part of the peak current intersects the limiting line near a concentration of DBU equivalent to 1.3 M, rather than 1.0 M, **1a**. However, this is probably due to the effect of water unavoidably contaminating the medium; such water would compete with the anilide for the base. Thus, in the presence of 1% water, the intersection shifted to a DBU concentration equivalent to nearly 1.6 M **1a** (not shown).

Similar voltammetric results were obtained with the other anilides, and pertinent data are summarized in Table I. The i_p'/C values for the extra wave are those obtained with a DBU concentration equivalent to 2.0 M anilide and correspond to the limiting values (*cf.* Fig. 2).

The i_p'/C value for a particular anilide is always smaller than the i_p/C value of the corresponding original wave in the absence of DBU, and the ratio of the two values, i_p'/i_p (Table I, column 9), is distributed between 0.4 and 0.8. Since it has been confirmed that i_p/C values around 30 μA/mM in the cyclic voltammetry of the anilides can be attributed to an electrode process involving overall two-electron transfer,[2,8] these results strongly suggest that the initial electrode process at the extra wave involves a one-electron transfer from the hydrogen-bonded substrate.

The magnitude of the potential shift, $E_p - E_p' = \Delta E_p$ (Table I, column 6), for 4′-substituted benzanilides (**1a**—**h**) can be linearly correlated to the Hammett σ values of the substituents (Fig. 3). The positive slope can also be regarded as an indication of hydrogen-bonding between the amide hydrogen of the anilides and DBU, because the ΔE_p value depends on the degree of partial negative charge developed on the nitrogen atom, which in turn should be a function of the acidity of the amide hydrogen and the strength of the added base. A similar potential shift due to hydrogen-bonding has been observed in the cyclic voltammetry of hydroxamic acid in the presence of various bases, and a linear relationship between the ΔE_p values and the pK_a values of the bases was demonstrated.[5]

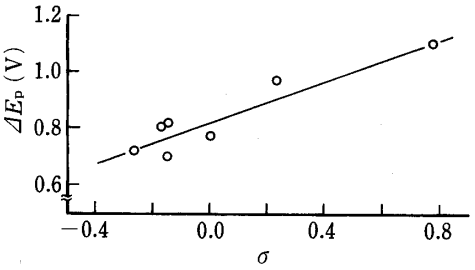


Fig. 3. Plot of ΔE_p *vs.* Hammett σ Value

TABLE II. Results of Controlled Potential Electrolysis of **1**

Compound	E_{app}[a]	F/mol	Product (Yield %)	Recovery of **1** (%)
1a	0.71	1.1	**2a** (61)	2
1b	0.62	1.1	**2b** (37)	2
1c	0.78	1.2	**2c** (19)	—
1d	0.67	1.1	**2d** (48)	—
1e	0.57	1.6	**3e** (11)	23
1f	0.78	1.6	**3f** (15)	16
1g	0.87	2.3	**3g** (3)	23
1h	0.83	1.2	**4h** (33)	25
1i	0.87	1.2	**4i** (36)	23
1j	0.78	1.2	**4j** (39)	26
1k	0.74	1.2	**4k** (22)	15

a) Applied potential: V *vs.* SCE.

Controlled Potential Electrolysis

Table II summarizes typical results on controlled potential electrolysis of **1** in the presence of DBU. The anilides can be classified into three groups according to their behavior in the electrolysis and the type of oxidation products (Chart 1).

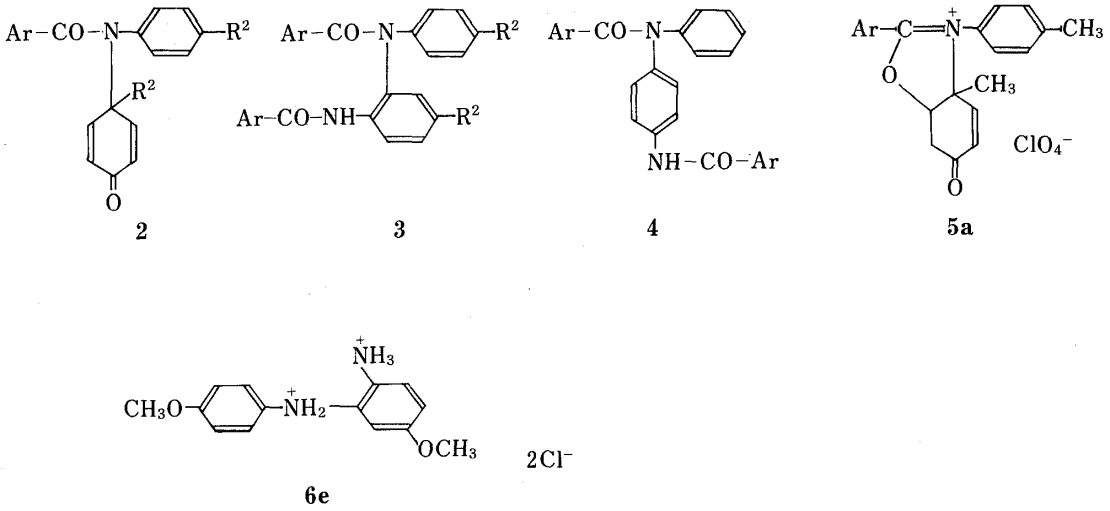


Chart 1

The first group includes the anilides **1a—d**, which consumed 1.1—1.2 F per mol of the substrate until the starting anilides had essentially disappeared in the electrolysis solution. The dimers **2a—d** were isolated from the resulting solution. In the case of **1a**, formation of benzamide was confirmed (45% based on **1a**).[9] Treatment of **2a** with perchloric acid gave the quaternary salt **5a**, which supports the structure assigned to **2a**.

The second group includes **1e—g**. In the electrolysis of these anilides the dimers **3e—g** were obtained in low yields; no other stable product was detected. The position of coupling in **3** is supported by the observation that hydrolysis of **3e** in acid solution afforded the *o*-aminodiphenylamine **6e** (see Experimental). Appreciable amounts of the starting anilides were recovered even when the electolysis was discontinued at various stages with coulometric *n*-values considerably higher than unity (Table II, column 3), and further electrolysis tended to reduce the yields of **3**. The fact that the oxidation potentials of **3** are less positive than those of the corresponding starting anilides may be partly responsible for these results.

Electrolysis of the third group, **1h—k**, gave the dimers **4h—k** as the main products: the structure of **4i** was confirmed by preparing an authentic sample from *p*-aminodiphenylamine and *p*-toluoyl chloride. In these electrolyses, however, in addition to a problem similar to that encountered in the case of the second group, severe electrode filming occurred, so that cleaning of the electrode surface was necessary several times during the experiments to obtain reasonable amounts of the products.

Based on the results of cyclic voltammetry, the initial electrode process in the oxidation of the anilides **1** is considered to be a one-electron transfer from the hydrogen-bonded substrate to form the neutral radical **7**. The proposed pathway, which includes the dimerization of **7**, for the formation of the products **2—4** in the controlled potential electrolysis is shown in Chart 2.

Since the radicals **7** are isoelectronic with phenoxy radicals, the dimerization can be considered to proceed by a mechanism similar to that proposed for the latter radicals, in which the mutual approach of the radicals with sandwich-type geometry has been shown to be energetically favorable.[10] Formation of N-*para*-coupled dimers **4** from **1h—k**, which have no substituent on the N-phenyl ring, suggests that a sandwich-type configuration with the N-

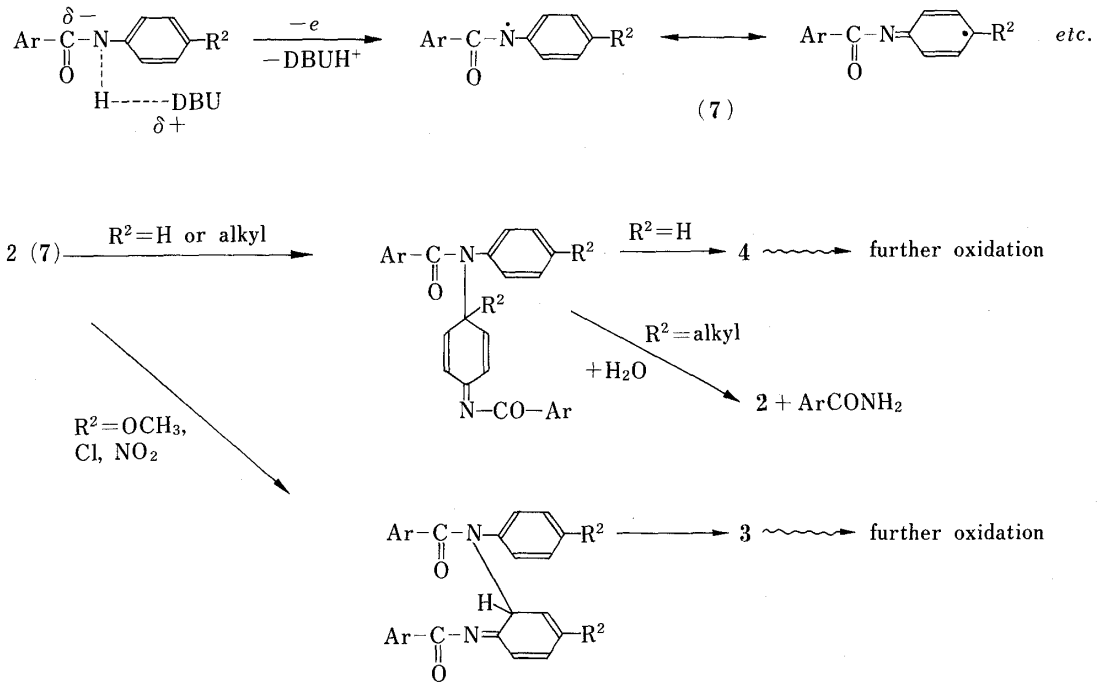


Chart 2

benzoyl groups *trans* to each other is preferred in the dimerization of **7** owing to the electrostatic repulsion between the hetero atoms involved in the two groups. Similar effects of hetero atoms have been reported in the dimerization of phenoxy radicals.[10] The *trans* configuration still seems required for the coupling of **7** derived from **1a—d**. Thus, the dimers **2** were the sole isolated products in the electrolysis of these anilides and the yields decreased with increase in the size of the 4′-alkyl substituents. Bulky alkyl groups will sterically hinder the *trans* configuration and may force the reaction of **7** in the direction of other unstable products.

In the case of the anilides **1e—g** (the second group), however, electrostatic repulsion will also be exerted by the hetero atoms attached to the 4′-position and the *trans* configuration will no longer be the preferred approach. Instead, a sandwich-type approach of the radicals **7e—g** with a staggered configuration[10] will give the N-*ortho* coupled dimers **3**. It is not clear whether the low yield of **3** is simply due to further oxidation of the dimer at the electrolysis potential or not. The possibility of other reactions of the radicals **7e—g** to give unidentified products cannot be ruled out.

Experimental

Materials——The 4- and 4′-substituted benzanilides were prepared from the corresponding benzoyl chlorides and anilines by known methods[2] and recrystallized from methanol or ethanol. All compounds gave analytical results consistent with the expected values. DBUP was prepared by addition of 70% perchloric acid (7 g) to a benzene solution of DBU (7 g, 1 eq in 50 ml) followed by filtration of the precipitate, which was recrystallized from chloroform–ether; mp 78.5—80 °C. *Anal.* Calcd for $C_9H_{17}ClN_2O_4$: C, 42.78; H, 6.78; Cl, 14.03; N, 11.09. Found: C, 42.58; H, 6.91; Cl, 14.18; N, 11.08. DBU was obtained from a commercial source and was purified by vacuum distillation. Acetonitrile was purified as described previously.[11] Preparative thin layer chromatography (PTLC) was carried out on glass plates coated with Merck Kieselgel 60 PF_{254} or Aluminiumoxid 150 PF_{254}.

Apparatus——Cyclic voltammetry and controlled potential electrolysis were carried out essentially as described previously.[11] Infrared (IR), nuclear magnetic resonance (NMR) and mass spectra (MS) were obtained on JASCO A202, Hitachi R-20A or R-22, and JMS-D-300 spectrometers, respectively.

Isolation of Products——A typical example of the procedures as applied to **1a** is described.

4-[*N*-Benzoyl-*N*-(4-methylphenyl)]amino-4-methyl-2,5-cyclohexadienone (**2a**): The anilide **1a** (211 mg) was subjected to electrolysis in acetonitrile (40 ml) containing 0.1 M DBUP and DBU (>2 eq) at 0.71 V at room

temperature until 1.1 F/mol had been consumed. The resulting solution was evaporated under reduced pressure and the residue was subjected to PTLC (silica gel, *n*-hexane : ethyl acetate = 5 : 1). The dienone **2a** (97 mg, 61%) was obtained as the main product; mp 154.5—155.5 °C from ethyl acetate. *Anal.* Calcd for $C_{21}H_{19}NO_2$: C, 79.47; H, 6.03; N, 4.41. Found: C, 79.25; H, 5.99; N, 4.43. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 1660, 1515. NMR ($CDCl_3$) δ: 1.57 (3H, s), 2.23 (3H, s), 6.22 (2H, d, J = 10.2 Hz), 6.85—7.20 (9H, m), 7.21 (2H, d, J = 10.2 Hz). The crude dienone (140 mg) was dissolved in 10 ml of acetonitrile containing 0.3 ml of 70% $HClO_4$. The mixture was stirred for 24 h at room temperature, then the resulting precipitate was filtered off and recrystallized from acetonitrile. This product was identified as 3a,6,7,7a-tetrahydro-7a-methyl-3-(4-methyl)phenyl-6-oxo-2-phenylbenzoxazolium perchlorate (**5a**, 68 mg, 22% from **1a**); mp 191—193 °C. *Anal.* Calcd for $C_{21}H_{20}ClNO_6$: C, 60.36; H, 4.82; Cl, 8.48; N, 3.35. Found: C, 60.31; H, 4.75; Cl, 8.31; N, 3.58. IR ν_{max}^{KBr} cm^{-1}: 1695, 1600, 1575, 1090. NMR (CD_3CN) δ: 1.69 (3H, s), 2.47 (3H, s), 3.28 (2H, d, J = 4 Hz), 5.85 (1H, t, d, J = 4 and 2 Hz), 6.30 (1H, d, J = 11 Hz), 6.72 (1H, dd, J = 11 and 2 Hz), 7.24—7.62 (9H, m).

The following compounds were obtained by essentially the same procedures. The PTLC conditions used for each case are given immediately after the name of the compound. Lack of description of the melting point indicates that the compound was obtained as an oil or an amorphous material, so that the high-resolution mass spectrum (HR-MS) was measured instead of elemental analysis.

4-[*N*-Benzoyl-*N*-(4-ethylphenyl)]amino-4-ethyl-2,5-cyclohexadienone (**2b**): Silica gel PTLC with *n*-hexane : ethyl acetate = 5 : 1. HR-MS: Calcd for $C_{23}H_{23}NO_2$ (M^+) *m/e*: 345.1726, Found: 345.1725. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 1670, 1650, 1515. NMR ($CDCl_3$) δ: 0.95 (3H, t, J = 7.2 Hz), 1.12 (3H, t, J = 7.2 Hz), 2.31 (2H, q, J = 7.2 Hz), 2.52 (2H, q, J = 7.2 Hz), 6.21 (2H, d, J = 10.2 Hz), 6.90—7.35 (9H, m), 7.21 (2H, d, J = 10.2 Hz).

4-[*N*-Benzoyl-*N*-(4-isopropylphenyl)]amino-4-isopropyl-2,5-cyclohexadienone (**2c**): Silica gel with *n*-hexane : ethyl acetate = 1 : 1. HR-MS: Calcd for $C_{25}H_{27}NO_2$ (M^+) *m/e*: 373.2041, Found: 373.2041. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 1675, 1650, 1520. NMR ($CDCl_3$) δ: 1.07 (6H, d, J = 7.2 Hz), 1.09 (6H, d, J = 7.2 Hz), 2.74 (1H, septet, J = 7.2 Hz), 3.79 (1H, septet, J = 7.2 Hz), 6.16 (2H, d, J = 10.2 Hz), 6.82—7.45 (9H, m), 7.29 (2H, d, J = 10.2 Hz).

4-[*N*-(4-Methylbenzoyl)-*N*-(4-methylphenyl)]amino-4-methyl-2,5-cyclohexadienone (**2d**): Silica gel with $CHCl_3$. HR-MS: Calcd for $C_{22}H_{21}NO_2$ (M^+) *m/e*: 331.1572, Found: 331.1575. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 1670, 1655, 1515. NMR ($CDCl_3$) δ: 1.54 (3H, s), 2.20 (3H, s), 2.25 (3H, s), 6.22 (2H, d, J = 10.2 Hz), 6.70—7.18 (8H, m), 7.21 (2H, d, J = 10.2 Hz).

N-Benzoyl-2-(*N*-benzoyl)amino-5,4′-dimethoxydiphenylamine (**3e**): Silica gel with *n*-hexane : $CHCl_3$ = 1 : 1; mp 165.5—166.5 °C from ethyl acetate. *Anal.* Calcd for $C_{28}H_{24}N_2O_4$: C, 74.32; H, 5.35; N, 6.19. Found: C, 74.01; H, 5.28; N, 6.13. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3410, 1665, 1640, 1515. NMR ($CDCl_3$) δ: 1.62 (1H, br s), 3.71 (3H, s), 3.73 (2H, s), 6.52—8.08 (17H, m). MS *m/e*: 452 (M^+). Derivatization to the diphenylamine derivative was also carried out to confirm the structure. When **3e** was refluxed in 10% HCl for 5 h, benzoic acid and a diamine dihydrochloride were produced. The latter (**6e**) was identified as 2-amino-5,4′-dimethoxydiphenylamine dihydrochloride from the similarity of its NMR pattern in the aromatic ring proton region with that of 4-methoxy-2-pyridiniumaniline dihydrochloride[12] rather than that of a 4-methoxy-3-pyridinium derivative.[3] NMR (CD_3OD) δ: 3.84, 3.90 (each 3H, s), 7.10—7.62 (6H, m), 7.76 (1H, d).

N-Benzoyl-2-(*N*-benzoyl)amino-5,4′-dichlorodiphenylamine (**3f**): Silica gel with *n*-hexane : $CHCl_3$ = 1 : 1. HR-MS: Calcd for $C_{26}H_{18}Cl_2N_2O_2$ (M^+) *m/e*: 460.0743, Found: 460.0722. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3440, 1660, 1650, 1515. NMR ($CDCl_3$) δ: 1.65 (1H, br s), 6.90—8.42 (17H, m).

N-Benzoyl-2-(*N*-benzoyl)amino-5,4′-dinitrodiphenylamine (**3g**): Aluminum oxide with *n*-hexane : $CHCl_3$ = 1 : 1. HR-MS: Calcd for $C_{26}H_{18}N_4O_6$ (M^+) *m/e*: 482.1224, Found: 482.1222. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3440, 1680, 1530, 1500. NMR ($CDCl_3$) δ: 1.60 (1H, br s), 7.10—8.23 (17H, m).

N-Benzoylamino-4-benzoylaminodiphenylamine (**4h**): Silica gel with *n*-hexane : ethyl acetate = 4 : 1; mp 207—209 °C from ethyl acetate. *Anal.* Calcd for $C_{26}H_{20}N_2O_2$: C, 79.57; H, 5.14; N, 7.14. Found: C, 79.68; H, 4.97; N, 7.09. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3440, 1660, 1650, 1515. NMR ($CDCl_3$) δ: 1.74 (1H, br s), 6.90—8.30 (19H, m). MS *m/e*: 492 (M^+).

N-(4-Methylbenzoyl)-4-(4-methylbenzoyl)aminodiphenylamine (**4i**): Silica gel with *n*-hexane : ethyl acetate = 4 : 1; mp 194.5—196 °C from ethyl acetate. *Anal.* Calcd for $C_{28}H_{24}N_2O_2$: C, 79.98; H, 5.75; N, 6.66. Found: C, 79.93; H, 5.58; N, 6.62. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3440, 1660, 1640, 1500. NMR ($CDCl_3$) δ: 1.77 (1H, br s), 2.29 (3H, s), 2.38 (3H, s), 6.80—8.25 (17H, m). MS *m/e*: 420 (m^+).

N-(4-Methoxybenzoyl)-4-(4-methoxybenzoyl)aminodiphenylamine (**4j**): Silica gel with *n*-hexane : $CHCl_3$ = 1 : 1, mp 207—208.5 °C from ethyl acetate. *Anal.* Calcd for $C_{28}H_{24}N_2O_4$: C, 74.32; H, 5.35; N, 6.19. Found: C, 74.09; H, 5.29; N, 6.49. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3430, 1665, 1650, 1510. NMR ($CDCl_3$) δ: 1.79 (1H, br s), 3.76 (3H, s), 3.83 (3H, s), 6.57—8.25 (17H, m). MS *m/e*: 452 (M^+).

N-(4-Chlorobenzoyl)-4-(4-chlorobenzoyl)aminodiphenylamine (**4k**): Silica gel with *n*-hexane : ethyl acetate = 4 : 1; mp 213.5—215 °C from ethyl acetate. *Anal.* Calcd for $C_{26}H_{18}Cl_2N_2O_2$: C, 67.69; H, 3.93; Cl, 15.37; N, 6.07. Found: C, 67.83; H, 3.85; Cl, 15.52; N, 6.09. IR $\nu_{max}^{CHCl_3}$ cm^{-1}: 3440, 1660, 1650, 1515. NMR ($CDCl_3$) δ: 1.71 (1H, s), 6.90—8.35 (17H, m). MS *m/e*: 460 (M^+).

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7) If hydroxide ion were to be used, as in the case of phenol oxidation, hydrolysis of the anilides would not be negligible.
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