Open-Chain Reissert Compounds: One-Pot Synthesis and Utility in Synthesis of Unsymmetrical Imides, α-Acylamino Carboxamides, Imidazolinones, and Hydantoins

Jean-Pierre Leblanc and Harry W. Gibson*

Department of Chemistry, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061-0212

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Acyclic Reissert compounds **2** and bis(Reissert compound)s **3–5** can be conveniently prepared in a biphasic system without isolation of the intermediate α-amino nitriles. Treatment of **2** with sodium hydride affords substituted unsymmetrical imides such as **8**. Oxidative hydrolysis of **2** by hydrogen peroxide converts the nitrile groups to primary amides, giving acyl derivatives of α-amino carboxamides **9**, whereas substituted analogs **7** undergo cyclization to imidazolinones **11**. Hydrogen peroxide treatment of α-cyano carbamate Reissert compounds, substituted (**13**) or not (**12**), affords substituted hydantoins **14**.

One-Pot Synthesis

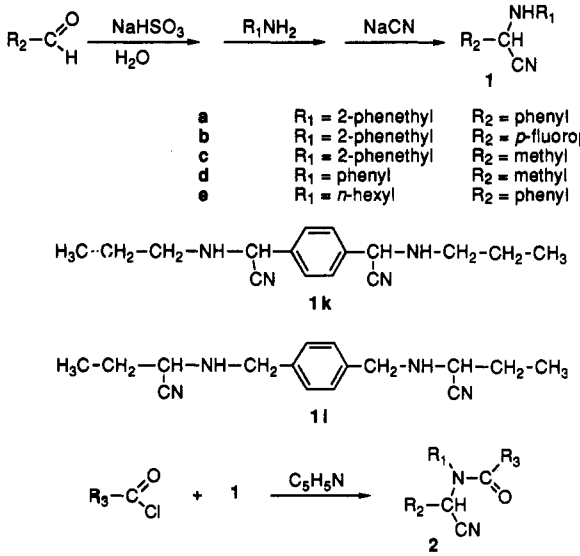
Several reviews have addressed the use of acyclic or open-chain Reissert compounds (α-acylamino nitriles) **2** in organic synthesis.[1,2] Their preparation, as opposed to that of heterocycle-based Reissert compounds,[2] involves two stages. In the first, an α-amino nitrile (**1**), more often in an aqueous-based system, is prepared. Acylation in pyridine usually follows, yielding the acyclic Reissert compound (**2**, Scheme 1).[1]

In the case of oily α-amino nitriles, McEwen et al.[1] reported conversion to the *N*-benzoyl derivative without purification. However, the aminonitrile had to be isolated before undergoing the acylation step in pyridine. Similarly, Elliott converted crude *N*-phenylglycinonitrile to the corresponding *N*-benzoyl Reissert compound.[3]

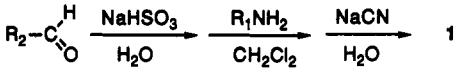
During the course of synthesis of α-amino nitriles from the bisulfite salts of aldehydes, amines, and sodium cyanide in water,[4] we found that *N*-phenethyl-α-cyanobenzylamine (**1a**) was obtained in a purer form (i.e., containing no *N*-phenethylbenzylidene) in 76% purified yield when prepared in a two-phase system (Scheme 2) as opposed to a wholly aqueous solvent (Scheme 1) which gave a 72% purified yield. A similar result was obtained for *N*-*n*-hexyl-α-cyanobenzylamine (**1e**).

The feasability of obtaining α-amino nitriles using a biphasic system prompted us to carry the reaction one step further by acylating the α-amino nitrile directly in the organic phase—where it remains preponderantly. Hence, various acyclic Reissert compounds **2a–e** and bis(Reissert compound)s **3–5** (Table 1) were prepared using this shortened procedure (Scheme 3) (several new acyclic Reissert compounds, **2f–h,j**, have only been prepared using the classical two-step procedure).

Scheme 1



Scheme 2



The purified yields of Reissert compounds by the one-pot method were comparable to those obtained when the two-pot sequence outlined in Scheme 1 was used (purified yield being taken as product of crude yield for the amino nitrile times the purified yield for the Reissert compound). The reaction is, however, limited to aromatic acid halides. With the more water-sensitive *p*-fluorobenzoyl chloride, the yield was diminished (**2d**). As for the two-pot method, the main impurity turned out to be the amide resulting from condensation of the acid chloride with the amine. The presence of such amides (or diamides) accounts for the overall low yield obtained in some cases (**2c,d**). The use of a hydrogen chloride acceptor such as pyridine is not necessary and even appeared detrimental if added too soon after the acid chloride.

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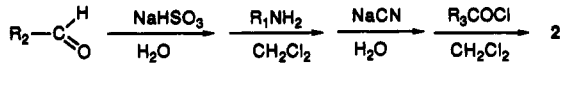
Table 1. Comparison of Yields of Acyclic Reissert and Bis(Reissert) Compounds by Two-Pot and One-Pot Methods

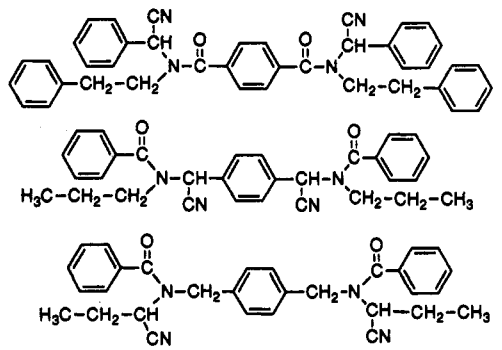
	two-pot synthesis					one-pot synthesis
	crude yield α-amino nitrile or bis(α-amino nitrile) (%)	mp (corr) (°C)	purified yield of Reissert compound (%)	mp (corr) (°C)	overall yield of purified Reissert compound (%)	overall yield of purified Reissert compound (%)
2a	91 (**1a**)	72.8–73.8 lit.[6] 82–7	86	124–125 lit.[7] 124	78	74
2b	94 (**1b**)	75.6–78.0	83	113.5–114.0	78	66
2c	88 (**1c**)	oil	53	72.3–74.0	47	38
2d	100 (**1d**)	91.8–93.5 lit.[8] 90	67	oil	67	33
2e	83 (**1e**)	oil	62	69.3–70.1 lit.[1] 65–66	51	56
3	91 (**1a**)	72.8–73.8 lit.[6] 82–7	40	194.5	36	44
4	66 (**1k**)	85–86.5	31	214.5–215.5	21	37
5	91 (**1l**)	92.1–95.4 (ref 9)	80	205.5–206	78	79

Scheme 3



a	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl
b	R_1 = 2-phenethyl	R_2 = *p*-fluorophenyl	R_3 = phenyl
c	R_1 = 2-phenethyl	R_2 = methyl	R_3 = phenyl
d	R_1 = phenyl	R_2 = methyl	R_3 = *p*-fluorophenyl
e	R_1 = *n*-hexyl	R_2 = phenyl	R_3 = phenyl
f	R_1 = phenyl	R_2 = *p*-methoxyphenyl	R_3 = *p*-fluorophenyl
g	R_1 = *p*-fluorophenyl	R_2 = *p*-fluorophenyl	R_3 = methyl
h	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = methyl
j	R_1 = 2-phenethyl	R_2 = *p*-fluorophenyl	R_3 = *p*-fluorophenyl

(f,g,h, and I prepared using the two-pot procedure)

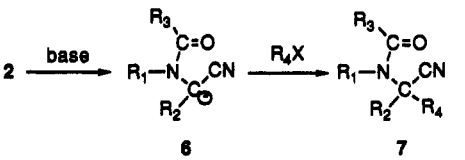


There are a number of chemical reactions involving a two-phase liquid–liquid medium.[5] The approach presented here includes transfer of the amino nitrile from the aqueous to the organic phase, followed by acylation in the latter and irreversible transfer from the aqueous medium. Compared to the classical two-pot procedure, the one-pot procedure avoids extraction and purification steps, reducing the amount of vessels, time, solvent, and labor involved.

Sodium Hydride Treatment

There have been extensive studies on the hydrolysis of heterocycle-based Reissert compounds.[1,2] Probably the best known studies regard the acid-catalyzed hydrolysis for the preparation of aldehydes. Their open-chain or acyclic analogs **2** have drawn less interest since acid hydrolysis yields only carboxylic acids.[3] Open-chain Reissert compounds **2** can readily be alkylated to **7** by the action of alkyl halides on their conjugate bases **6** (Scheme 4, Table 2). The reaction can be conveniently performed

Scheme 4



a	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl	R_4 = methyl
f	R_1 = phenyl	R_2 = *p*-methoxyphenyl	R_3 = *p*-fluorophenyl	R_4 = methyl
g	R_1 = *p*-fluorophenyl	R_2 = *p*-fluorophenyl	R_3 = methyl	R_4 = methyl
h	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = methyl	R_4 = methyl
i	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl	R_4 = ethyl
j	R_1 = 2-phenethyl	R_2 = *p*-fluorophenyl	R_3 = *p*-fluorophenyl	R_4 = methyl

Table 2. Synthesis and Properties of (Alkylated) Acyclic Reissert Compounds and Their Hydrogen Peroxide Hydrolysis Products

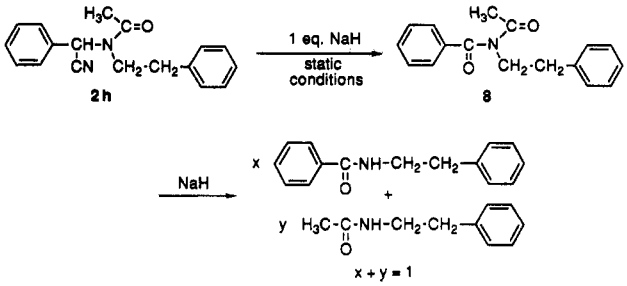
	yield (%, pur.)[a]	mp (°C)	hydrolysis procedure[b]	yield of hydrolysis product (%, pur.)	mp (°C)
2a	86	124–125	A	70	174.9–175.9
2c	38	72.3–74.0	A	81	170.5–172.5
2d	76	oil	A	95	138.5–140
2h	87	80.2	B	100	189.4–190.8
2j	82	107.8–108.5	A	61	173–174.5
2k	68	88.4–89.4	B	71	176.5–177.3
2l	78	124.5–126.0			
7a	82	157–158	A	85	157.5–158.5
7h	83[c]	96.6–102.4	A	92	133.2–134.6
7i	67	174.1–176.3	C	52	186.5–187.0
7j	68	139.7–140.4	A	85	157.5–159.5
7k	57	139.0–140.0	C	80[c]	158.5–159.5
7l	90	156.5–158.5	C	45	140–141

[a] For compounds **2** from amino nitrile by acylation with R_3COCl; for compounds **7** by alkylation of **2** with R_4X as per Scheme 4. [b] Cf. Experimental Section. [c] Crude yield.

at room temperature in dimethylformamide (DMF) using sodium hydride as the base.[10] However when we attempted the alkylation of **2h** with a relatively unreactive α-iodo-ω-methoxy oligo(ethylene glycol) in a stoichiometric ratio, new signals arising from degeneration of the Reissert compound were seen by ^1H NMR spectroscopy. The behavior of **2h** in the absence of any electrophile was therefore investigated. Unexpectedly the anion was found to remain stable in *stirred solutions* for days under an inert atmosphere and no significant amounts of decom-

(10) Popp, F. D.; Wefer, J. M. *J. Chem. Soc., Chem. Commun.* **1966**, 207.

Scheme 5



position products were detected. However, when sodium hydride was added to a *static solution* of **2h** in DMF or THF, the decomposition rate increased greatly. The unsymmetrical imide **8** resulted in 82% yield from the unstirred sodium hydride treatment of **2h** (Scheme 5).

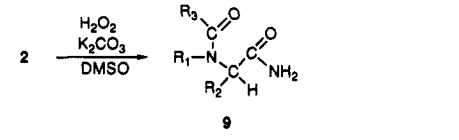
No Stevens-type rearrangement can be expected in this case[11] and rather than being cleaved to an aldehyde (action of potassium hydride on a substituted compound gives a ketone[1]), the post-sodium hydride treatment hydrolysis manifested the acyl equivalence of α-cyano compounds.[12] The reaction was conducted under an inert atmosphere in DMF or THF with about stoichiometric quantities of sodium hydride. It is plausible that oxidation of the anion upon exposure to the atmosphere could have occurred during workup, as air oxidation of closely related α-dialkylamino nitriles leads to the amide equivalents.[13] When a larger amount of sodium hydride was used, complete cleavage of the amide **8** occurred to yield *N*-(2-phenethyl)-benzamide and *N*-(2-phenethyl)acetamide (Scheme 5). Alkaline cleavage into these amides is indiscriminate since various ratios of the two products were obtained in subsequent experiments.

Oxidative Hydrolysis with Hydrogen Peroxide

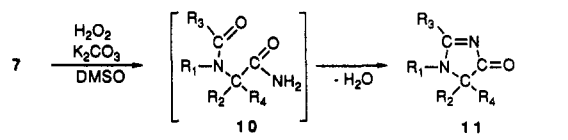
Oxidation of heterocycle-based Reissert compounds by hydrogen peroxide in basic DMSO converts the nitrile group into a primary amide.[14,15] We extended this reaction to acyclic analogs **2** and their substituted counterparts **7**. The selective oxidative hydrolysis of **2** also resulted in the formation of primary amide groups. Compounds **9** were generated from **2** in excellent yields (Scheme 6, Table 2). On the other hand, oxidation of the alkylated analogs **7** proceeded directly to the entirely substituted 2-imidazolin-4-ones **11**, possibly via the amide intermediates **10** (Scheme 6, Table 2). The reaction proceeded with less ease than for the unsubstituted analogs **2**, requiring harsher conditions (Table 2). Difficulty in purification further arose when a large amount of hydrogen peroxide was used because dimethyl sulfone was also produced.[15]

This type of cyclization was observed upon heating an alkylated acyclic Reissert compound **7** with sodium hydroxide at 100 °C.[16] A similar cyclization occurred when acyclic Reissert compounds **2** were hydrogenated over

Scheme 6

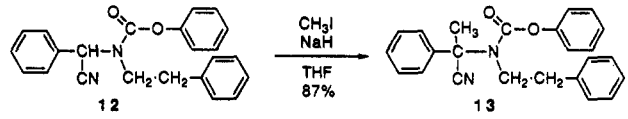


a	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl
c	R_1 = 2-phenethyl	R_2 = methyl	R_3 = phenyl
d	R_1 = phenyl	R_2 = methyl	R_3 = *p*-fluorophenyl
f	R_1 = phenyl	R_2 = *p*-methoxyphenyl	R_3 = *p*-fluorophenyl
h	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = methyl
j	R_1 = 2-phenethyl	R_2 = *p*-fluorophenyl	R_3 = *p*-fluorophenyl



a	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl	R_4 = methyl
f	R_1 = phenyl	R_2 = *p*-methoxyphenyl	R_3 = *p*-fluorophenyl	R_4 = methyl
g	R_1 = *p*-fluorophenyl	R_2 = *p*-fluorophenyl	R_3 = methyl	R_4 = methyl
h	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = methyl	R_4 = methyl
i	R_1 = 2-phenethyl	R_2 = phenyl	R_3 = phenyl	R_4 = ethyl
j	R_1 = 2-phenethyl	R_2 = *p*-fluorophenyl	R_3 = *p*-fluorophenyl	R_4 = methyl

Scheme 7



Raney nickel, though the carbonyl function resulting from the hydrolysis of the nitrile group was also reduced.[17] Imidazolethiols, structurally related to **11**, were prepared from substituted and unsubstituted acyclic Reissert compounds via thionoamide intermediates.[18] Although cyclizations are common with conventional Reissert compounds[2,19] and known for their acyclic analogs,[1,20] those have involved the Reissert salts and not the substituted analogs. Thus oxidative hydrolysis by hydrogen peroxide provides a novel method for cyclization of open-chain Reissert analogs. The alkylation of *N*-unsubstituted analogs **11**, R_1 = H, has also been reported to yield entirely substituted 2-imidazolin-4-ones **11**, R_1 = alkyl.[21]

Though less studied, Reissert compounds from heterocycles and chloroformates have been found to behave similarly regarding alkylation.[22] We prepared the open-chain α-cyano carbamate **12**, which, as with α-cyano amides **2**, was easily alkylated to **13** (Scheme 7). However, oxidative hydrolysis of neither **12** nor **13** with hydrogen peroxide occurs similarly to that of the α-cyano amide analogs **2** and **7**. A large excess of hydrogen peroxide had to be used (for a lower yield than for **9** or **11**) to generate the substituted hydantoins **14**, with loss of the phenol

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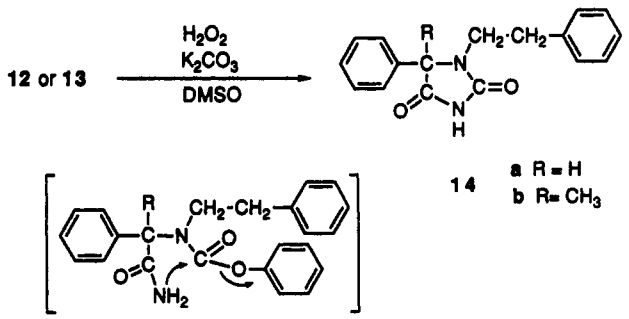
Scheme 8



portion of the Reissert compound (Scheme 8). Hydantoins, as imidazolinones 11, are five-membered rings containing nitrogen atoms at the 1 and 3 positions, reminiscent of the α-cyanoacyl amino unit of open-chain Reissert compounds.

Conclusions

We have reported a new one-pot synthesis of acyclic Reissert compounds 2, involving sequential addition of a primary amine, dichloromethane, sodium cyanide, and an acid chloride in dichloromethane to an aqueous solution of an aldehyde and sodium metabisulfite (Scheme 3). These open-chain Reissert compounds are shown to be readily alkylated via their anions (Scheme 4). In the absence of electrophiles in static solutions they produce imides 8 (Scheme 5). The Reissert compounds 2 upon treatment with basic hydrogen peroxide yield α-(acylamino) carboxamides 9, while under similar conditions alkylated Reissert compounds 7 produce imidazolinones 11. Carbamate-type Reissert compounds 12, or their alkylated derivatives, yield hydantoins 14 with basic hydrogen peroxide. These reactions constitute new synthetic routes to imides 8, diamides 9, imidazolinones 11, and hydantoins 14.

Experimental Section

Chemicals were used as received. Terephthaloyl chloride was recrystallized from hexanes. Recrystallizations were carried out by saturation at reflux and allowing cooling to room temperature. Chromatographic elutions were performed using 25–30 g of silica gel per gram of compound. Melting points are corrected. ^{1}H NMR spectra were recorded on a Bruker 270 MHz instrument using TMS as the reference and $CDCl_3$ as the solvent. Coupling constants are given in hertz. FTIR spectra were obtained in KBr. Elemental analyses were performed by Atlantic Microlab, Inc., Norcross, GA.

Preparation of α-Amino Nitriles in a Two-Phase System. The synthesis of ***N*-(2-phenethyl)-α-cyanobenzylamine (1a)** is given as a general procedure.

A mixture of 21.85 g (0.206 mol) of benzaldehyde and 21.58 g (0.110 mol) of sodium metabisulfite was stirred in 250 mL of water for 2 h. Phenethylamine (26.0 mL, 0.207 mol) was then added in 100 mL of dichloromethane and the mixture was stirred for 2 h. The addition of sodium cyanide (10.2 g, 0.208 mol) followed and the mixture was stirred overnight. The organic phase was washed with water, dried over sodium sulfate, and evaporated to give a light yellow oil, which crystallized into a pale yellow solid (44.50 g, yield 91%). The product was recrystallized from ethyl acetate/hexanes (37 g, 76%): IR 3700–3090, 3023, 2942, 2921, 2840, 2233 cm^{-1}; ^{1}H NMR δ 7.5–7.2 (m, 10H), 4.80 (s, 1H), 3.2–2.95 (m, 2H), 2.95–2.75 (m, 2H), 1.5 (s, 1H). Anal. Calcd for $C_{16}H_{16}N_2$: C, 81.32; H, 6.83. Found: C, 81.04; H, 6.82.

***N*-(2-Phenethyl)-α-cyano-*p*-fluorobenzylamine (1b)**: recrystallization from ethyl acetate/hexanes; IR 3318, 3063, 3027, 2932, 2902, 2832, 2227, 1237 cm^{-1}; ^{1}H NMR δ 7.48 (q, J = 4, 2H), 7.38–7.2 (m, 5H), 7.10 (t, J = 10, 2H), 4.79 (d, J = 9, 1H), 3.15–2.95 (m, 2H), 2.9–2.8 (m, 2H), 1.51 (q, J = 11, 1H). Anal. Calcd for $C_{16}H_{15}FN_2$: C, 75.56; H, 5.95; N, 11.02. Found: C, 75.61; H, 5.99; N, 10.99.

***N*,*N*'-Di-*n*-propyl-α,α'-dicyano-*p*-xylylenediamine (1k)**: recrystallization from ethyl acetate/hexanes: IR 3300, 3040, 2960, 2930, 2230 cm^{-1}; ^{1}H NMR δ 7.58 (s, 4H), 4.81 (s, 2H), 2.54 (m, J = 9, 4H), 1.53 (m, 6H), 0.98 (t, J = 9, 6H). Anal. Calcd for $C_{16}H_{22}N_4$: C, 71.07; H, 8.20. Found: C, 71.13; H, 8.20.

Preparation of Acyclic Reissert Compounds in a One-Pot Process. The preparation of ***N*-(2-phenethyl)-*N*-(α-cyanobenzyl)benzamide (2a)** is given as a general procedure.

A mixture of benzaldehyde (5.80 g, 54.7 mmol) and sodium metabisulfite (5.38 g, 28.3 mmol) in 120 mL of water was stirred for 2 h and 6.90 mL (54.9 mmol) of phenethylamine was then added, followed by 100 mL of CH_2Cl_2. The mixture was stirred for 2 h. Then NaCN (2.95 g, 60.1 mmol) was added and the mixture was stirred overnight. Benzoyl chloride, 7.50 mL (63.5 mmol), in 10 mL of CH_2Cl_2 was added and the stirring was continued for 3 h. The aqueous phase was discarded and the organic phase was washed with a saturated solution of sodium bicarbonate and then water, dried over Na_2SO_4, and evaporated to give a very light yellow solid. Recrystallization from absolute ethanol afforded 13.70 g (74%): IR 3060, 3024, 2927, 2243, 1647 cm^{-1}; ^{1}H NMR δ 7.65–7.4 (m, 10H), 7.2–7.1 (m, 4H), 6.9–6.6 (m, 2H), 3.65–3.48 + 3.42–3.25 (m, 2H), 2.95–2.75 + 2.55–2.3 (2H).

***N*-(2-Phenethyl)-*N*-(α-cyano-*p*-fluorobenzyl)-benzamide (2b)**: recrystallization from absolute ethanol and absolute ethanol/hexanes; IR 3063, 3027, 1651 cm^{-1}; ^{1}H NMR δ 7.48 (m, 8H), 7.17 (m, 5H), 6.78 (bs, 2H), 3.58 (m) + 3.33 (m) (2H), 2.83 + 2.48 (bs, 2H). Anal. Calcd for $C_{23}H_{19}FN_2O$: C, 77.08; H, 5.34; N, 7.82. Found: C, 77.09; H, 5.39; N, 7.83.

***N*-(α-Cyanoethyl)-*N*-(2-phenethyl)benzamide (2c)** was chromatographed on a silica gel column (CH_2Cl_2/hexanes 50:50 then CH_2Cl_2 alone); a colorless oil was obtained which crystallized slowly upon standing: IR 3086, 3066, 3030, 2953, 2244, 1643 cm^{-1}; ^{1}H NMR δ 7.48 (t, J = 5, 3H), 7.39 (q, J = 2, 2H), 7.25 (d, J = 6, 3H), 7.03 (bs, 2H), 5.5–5.1 (bs, 1H), 3.64 (t, J = 8, 2H), 3.1–2.8 (m, 2H), 1.57 (d, J = 8, 3H). Anal. Calcd for $C_{18}H_{18}N_2O$: C, 77.67; H, 6.52; N, 10.07. Found: C, 77.70; H, 6.58; N, 10.12.

***N*-Phenyl-*N*-(α-cyanoethyl)-*p*-fluorobenzamide (2d)** was filtered through a neutral alumina column and then eluted through a neutral alumina column (CH_2Cl_2/hexanes 45:55). Further purification was achieved through a silica gel column (CH_2Cl_2): IR (neat) 3070, 2995, 2949, 2246, 1657 cm^{-1}; ^{1}H NMR δ 7.34 (m, 5H), 7.20 (m, 2H), 6.85 (t, J = 9, 2H), 5.95 (q, J = 7, 1H), 1.52 (d, J = 7, 3H). Anal. Calcd for $C_{16}H_{13}FN_2O$: C, 71.63; H, 4.88; N, 10.44. Found: C, 71.44; H, 4.93; N, 10.34.

***N*-Phenyl-*N*-(α-cyano-*p*-methoxybenzyl)-*p*-fluorobenzamide (2f)**. Recrystallization from methanol or ethyl acetate/hexanes gave white needles: IR 3077, 3015, 2976, 2240, 1650 cm^{-1}; ^{1}H NMR δ 7.34 (m, 2H), 7.28 (m, 3H), 7.18 (m, 3H), 6.84 (m, 6H), 3.80 (s, 3H). Anal. Calcd for $C_{22}H_{17}FN_2O_2$: C, 73.32; H, 4.75; N, 7.77. Found: C, 73.38; H, 4.79; N, 7.75.

***N*-(*p*-Fluorophenyl)-*N*-(α-cyano-*p*-fluorobenzyl)acetamide (2g)**. Recrystallization from methanol (activated carbon), methanol, and ethyl acetate/hexanes afforded shiny, white crystals: IR 3065, 3049, 2917, 2244, 1663 cm^{-1}; ^{1}H NMR δ 7.26 (t, J = 5, 4H), 7.03 (t, J = 9, 4H), 7.1–6.65 (bs, 1H), 1.86 (s, 3H). Anal. Calcd for $C_{16}H_{12}F_2N_2O$: C, 67.13; H, 4.23; N, 9.79. Found: C, 67.16; H, 4.27; N, 9.74.

***N*-(2-Phenethyl)-*N*-(α-cyanobenzyl)acetamide (2h)**. Acylation in CH_2Cl_2 in the presence of triethylamine and recrystallization from ethyl acetate/hexanes afforded large, orthorhombic crystals: IR 3084, 3060, 3032, 2937, 2243, 1658 cm^{-1}; ^{1}H NMR δ 7.55–7.4 (m, 5H), 7.3–7.17 (m, 3H), 7.11 (s, 1H), 7.1–7.0 (d, J = 9, 2H), 3.6 + 3.33 (m, 2H), 2.94–2.77 + 2.6–2.45 (m, 2H), 2.3 + 2.15 (s, 3H, CH_3, E/Z). Anal. Calcd for $C_{18}H_{18}N_2O$: C, 77.67; H, 6.52; N, 10.07. Found: C, 77.71; H, 6.56; N, 10.08.

***N*-(2-Phenethyl)-*N*-(α-cyano-*p*-fluorobenzyl)-*p*-fluorobenzamide (2j)**. Recrystallization from hexanes/very little ethyl acetate afforded white crystals (82%): IR 3070, 3060, 2935, 2225, 1630, 1225 cm^{-1}; ^{1}H NMR δ 7.54 (m, 2H), 7.44 (q, J = 8, 2H), 7.18 (m, 9H), 6.81 (bs, 1H), 3.59 + 3.36 (m, 2H), 2.81 + 2.50 (bs, 2H).

Anal. Calcd for $C_{23}H_{18}F_2N_2O$: C, 73.39; H, 4.82; N, 7.44. Found: C, 73.45; H, 4.84; N, 7.43.

***N,N'*-(α-Cyanobenzyl)-*N,N'*-bis(2-phenethyl)terephthalamide (3)** was recrystallized from ethyl acetate/hexanes: IR 3018, 2918, 1639 cm^{-1}; ^1H NMR (RD = 1s) δ 7.7–7.35 (bs, 15H), 7.20–7.05 (s, 7.5H), 7.0–6.6 (bs, 3.5H), 3.7–3.5 (bs) + 3.42–3.30 (m, 4H), 3.0–2.65 + 2.65–2.35 (bs) (4H). Anal. Calcd for $C_{40}H_{34}N_4O_2$: C, 79.71; H, 5.69; N, 9.30. Found: C, 79.72; H, 5.69; N, 9.31.

α,α'-Dicyano-*N,N'*-dibenzoyl-*N,N'*-di-*n*-propyl-*p*-xylylenediamine (4) was recrystallized from toluene and dried under vacuum at 110 °C for 1 week: IR 3042, 2976, 2956, 2242, 1642 cm^{-1}; ^1H NMR δ 7.64 (s) + 7.48 (s) + 6.85 (bs) (16H), 3.39 (m) + 3.10 (m) (4H), 1.55 (bs) + 1.31 (bs) (4H), 0.65 (bs, 6H). Anal. Calcd for $C_{30}H_{30}N_4O_2$: C, 75.29; H, 6.32; N, 11.71. Found: C, 75.35; H, 6.36; N, 11.68.

***N,N'*-Dibenzoyl-*N,N'*-bis(α-cyanopropyl)-*p*-xylylenediamine (5)** was recrystallized from DMSO. An analytical sample was obtained by elution through a silica gel column (ethyl acetate/hexanes 50:50) for complete removal of DMSO: IR 3058, 2985, 2940, 2236, 1630 cm^{-1}; ^1H NMR δ 7.44 (s, 10H), 7.27 (s, 4H), 4.93 (bs, 2H), 4.71 (ABq, Δν = 0.21 δ, J = 16, 4H, CH_2Ar), 1.82 (m, 4H), 0.97 (t, J = 7, 6H). Anal. Calcd for $C_{30}H_{30}N_4O_2$: C, 75.29; H, 6.32; N, 11.71. Found: C, 75.04; H, 6.36; N, 11.64.

***N*-(2-Phenethyl)-*N*-(α-cyano-α-methylbenzyl)benzamide (7a).** The DMF reaction mixture[10] was partitioned between water and CH_2Cl_2. The organic phase was further washed with water, dried over sodium sulfate, and evaporated. The residue was recrystallized from absolute ethanol or absolute ethanol/very little ethyl acetate to afford white needles (82%): IR 3060, 3024, 2232, 1649 cm^{-1}; ^1H NMR δ 7.66 (d, J = 10, 2H), 7.55–7.3 (m, 8H), 7.12 (m, 3H), 6.75 (m, 2H), 3.9–3.5 (m, 2H), 3.0–2.75 (m, 2H), 2.19 (s, 3H). Anal. Calcd for $C_{24}H_{22}N_2O$: C, 81.32; H, 6.26; N, 7.91. Found: C, 81.41; H, 6.30; N, 7.95.

***N*-Phenyl-*N*-(α-cyano-α-methyl-*p*-methoxybenzyl)-*p*-fluorobenzamide (7f)** was chromatographed on a silica gel column (hexanes/ethyl acetate 80:20) and recrystallized from ethyl acetate/hexanes: IR 3064, 2997, 2235, 1654 cm^{-1}; ^1H NMR δ 7.64 (d, J = 9, 2H), 7.40 (m, 2H), 7.30 (m, 5H), 6.93 (t, J = 9, 2H), 6.81 (t, J = 9, 2H), 3.80 (s, 3H), 1.68 (s, 3H). Anal. Calcd for $C_{23}H_{19}FN_2O_2$: C, 73.78; H, 5.11; N, 7.48. Found: C, 73.87; H, 5.15; N, 7.43.

***N*-(*p*-Fluorophenyl)-*N*-(α-cyano-α-methyl-*p*-fluorobenzyl)acetamide (7g)** was recrystallized from ethyl acetate/hexanes: IR 3119, 3080, 3001, 2233, 1679 cm^{-1}; ^1H NMR δ 7.55 (q, J = 5, 2H), 7.36 (m, 2H), 7.21 (m, 2H), 7.08 (t, J = 9, 2H), 1.80 (s, 3H), 1.57 (s, 3H). Anal. Calcd for $C_{17}H_{14}F_2N_2O$: C, 67.99; H, 4.70; N, 9.33. Found: C, 68.03; H, 4.74; N, 9.31.

***N*-(2-Phenethyl)-*N*-(α-cyano-α-methylbenzyl)acetamide (7h)** was recrystallized from ethyl acetate/hexanes to give white needles: IR 3062, 3029, 2997, 2951, 2236, 1662 cm^{-1}; ^1H NMR δ 7.5–7.2 (m, 8H), 7.12 (d, J = 7, 2H), 3.35–3.10 (m, 2H), 3.05–2.9 (m, 2H), 2.12 (s, 3H), 2.10 (s, 3H). Anal. Calcd for $C_{19}H_{20}N_2O$: C, 78.05; H, 6.90; N, 9.58. Found: C, 77.95; H, 6.92; N, 9.62.

***N*-(2-Phenethyl)-*N*-(α-cyano-α-ethylbenzyl)benzamide (7i)** was recrystallized from ethyl acetate: IR 3060, 3019, 2992, 2941, 2231, 1645 cm^{-1}; ^1H NMR δ 7.64 (d, J = 9, 2H), 7.45–7.34 (m, 8H), 7.14 (t, J = 4, 3H), 6.74 (m, 2H), 3.71 (m, 2H), 3.0–2.8 (m, 2H), 2.26 (hex, J = 7, 2H), 1.00 (t, J = 7, 3H). Anal. Calcd for $C_{25}H_{24}N_2O$: C, 81.49; H, 6.57; N, 7.60. Found: C, 81.43; H, 6.55; N, 7.53.

***N*-(2-Phenethyl)-*N*-(α-cyano-α-methyl-*p*-fluorobenzyl)-*p*-fluorobenzamide (7j)** was recrystallized from ethyl acetate/hexanes: IR 3064, 2997, 2235, 1654 cm^{-1}; ^1H NMR δ 7.60 (q, J = 5, 2H), 7.31 (q, J = 5, 2H), 7.18 (t, J = 4, 2H), 7.05 (m, 5H), 6.86 (q, J = 4, 2H), 3.79 (m, 2H), 2.86 (m, 2H), 2.16 (s, 3H). Anal. Calcd for $C_{24}H_{20}F_2N_2O$: C, 73.83; H, 5.16; N, 7.17. Found: C, 73.86; H, 5.21; N, 7.14.

***N*-Benzoyl-*N*-(2-phenethyl)acetamide (8).** To an unstirred solution of 0.56 g (2 mmol) of *N*-(2-phenethyl)-*N*-(α-cyanobenzyl)-acetamide (2h) in 5 mL of DMF was added 0.10 g (2.5 mmol) of a 60% dispersion of NaH in mineral oil. The mixture turned orange and after 20 h, CH_2Cl_2 (10 mL) was added. The resulting mixture was washed twice with 10 mL of water and dried over sodium sulfate. Chromatography through a silica gel column (CH_2Cl_2, then CH_2Cl_2/ethyl acetate 9:1) afforded 0.45 g (82%) of a very light yellow oil: IR 3063, 3028, 2955, 1690, 1660 cm^{-1}; ^1H NMR δ 7.54 (m) + 7.46 (t, J = 7) + 7.22 (m) + 7.11 (d, J = 8) (10H), 4.00 (t, J = 8, 2H), 2.91 (t, J = 8, 2H), 2.15 (s, 3H). Anal. Calcd for $C_{17}H_{17}NO_2$: C, 76.38; H, 6.41; N, 5.24. Found: C, 76.24; H, 6.45; N, 5.23.

Hydrogen Peroxide Hydrolysis of Reissert Compounds 2 and 7. The following variations of the method described by Katritzky[15] were used.

Procedure A. The open-chain Reissert compound (1.5 g) was dissolved in 15 mL of DMSO. A stoichiometric amount of potassium carbonate was added, followed by the addition under slow stirring of 1.7 times the stoichiometric equivalent of a 30% solution of H_2O_2. If no exothermicity was noted the mixture was warmed to about 50 °C. After cooling, the mixture was taken up in CH_2Cl_2 (150 mL). The organic phase was washed twice with 100 mL of water, dried over sodium sulfate, and evaporated to give the crude product.

Procedure B. The open-chain Reissert compound (1.5 g) was dissolved in 15 mL of DMSO. A stoichiometric amount of potassium carbonate was added, followed by the addition under slow stirring of 1 mL of a 30% solution of H_2O_2. The mixture was heated until reflux started, cooled, and worked up as above.

Procedure C. Procedure B was repeated, allowing the mixture to cool before adding a second portion of hydrogen peroxide.

***N*-(α-(Aminocarbonyl)benzyl)-*N*-(2-phenethyl)benzamide (9a)** was recrystallized from ethyl acetate/hexanes: IR 3460, 3293, 3233, 3218, 3182, 3164, 3060, 3027, 2961, 2925, 1701, 1681 cm^{-1}; ^1H NMR δ 7.7–7.35 (bs) + 7.10 (s) (15H), 6.7–6.45 (bs, 1H), 6.2–5.9 + 5.7–5.5 (bs, 2H), 3.6–3.35 (bs, 2H), 2.7–2.4 (bs) + 2.2–2.05 (m) (2H). Anal. Calcd for $C_{23}H_{22}N_2O_2$: C, 77.07; H, 6.19; N, 7.82. Found: C, 76.96; H, 6.20; N, 7.83.

***N*-(α-(Aminocarbonyl)ethyl)-*N*-(2-phenethyl)benzamide (9c)** was recrystallized from ethyl acetate/hexanes and ethyl acetate: IR 3417, 3196, 3058, 2987, 2943, 1693 cm^{-1}; ^1H NMR δ 7.42 (m, 3H), 7.36 (m, 2H), 7.18 (s, 3H), 6.84 (bs, 2H), 5.45 (bs, 2H), 5.09 (bs, 1H), 3.51 (bs, 2H), 2.87 + 2.71 (bs, 2H), 1.53 (bs, 3H). Anal. Calcd for $C_{18}H_{20}N_2O_2$: C, 72.95; H, 6.80; N, 9.45. Found: C, 72.95; H, 6.81; N, 9.49.

***N*-(α-(Aminocarbonyl)ethyl)-*N*-phenyl-*p*-fluorobenzamide (9d)** was chromatographed through a silica gel column (ethyl acetate): IR 3406, 3325, 3218, 3077, 2992, 1685, 1639 cm^{-1}; ^1H NMR δ 7.25 (m, 5H), 7.07 (m, 2H), 6.83 (t, J = 9, 2H), 6.62 (bs) + 5.5 (bs) (2H), 5.42 (q, J = 7, 1H), 1.27 (d, J = 7, 3H). Anal. Calcd for $C_{16}H_{15}FN_2O_2$: C, 67.12; H, 5.28; N, 9.78. Found: C, 67.00; H, 5.32; N, 9.71.

***N*-(α-(Aminocarbonyl)-*p*-methoxybenzyl)-*N*-phenyl-*p*-fluorobenzamide (9f)** was recrystallized from ethyl acetate/hexanes: IR 3450, 3351, 3067, 3017, 2959, 1713, 1627 cm^{-1}; ^1H NMR δ 7.31 (m, 2H), 7.18 (d, J = 9, 2H), 7.06 (m, 3H), 6.98 (bs, 2H), 6.77 (q, J = 7, 4H), 6.15 (s, 1H), 5.83 (bs) + 5.66 (bs) (2H), 3.76 (s, 3H). Anal. Calcd for $C_{22}H_{19}FN_2O_3$: C, 69.83; H, 5.06; N, 7.40. Found: C, 69.96; H, 5.10; N, 7.38.

***N*-(α-(Aminocarbonyl)benzyl)-*N*-(2-phenethyl)acetamide (9h)** was recrystallized from ethyl acetate/hexanes: IR 3375, 3211, 3023, 2951, 1683, 1635 cm^{-1}; ^1H NMR δ 7.50 (m, 2H), 7.44 (d, J = 8, 3H), 7.19 (m, 3H), 6.89 (d, J = 10, 2H), 6.02 (s, 1H), 5.83 + 5.50 (bs, 2H), 3.50 (t, J = 9, 2H), 2.58 + 2.72 (m) + 2.20 (s) (5H). Anal. Calcd for $C_{18}H_{20}N_2O_2$: C, 72.95; H, 6.80; N, 9.45. Found: C, 72.79; H, 6.76; N, 9.42.

***N*-(α-(Aminocarbonyl)-*p*-fluorobenzyl)-*N*-(2-phenethyl)-*p*-fluorobenzamide (9j)** was recrystallized from toluene and ethyl acetate/hexanes; IR 3401, 3336, 3072, 3027, 1688 cm^{-1}; ^1H NMR δ 7.51 (bs) + 7.41 (m) (5H), 7.10 (m, 8H), 6.67 (bs, 1H), 6.2–5.7 (bs, 2H), 3.46 (m, 2H), 2.61 (bs) + 2.18 (m) (2H). Anal. Calcd for $C_{23}H_{20}F_2N_2O_2$: C, 70.04; H, 5.11; N, 7.10. Found: C, 69.94; H, 5.15; N, 7.02.

1-(2-Phenethyl)-2,5-diphenyl-5-methyl-2-imidazolin-4-one (11a) was recrystallized from ethyl acetate/hexanes to give white dendritic crystals: IR 3060, 3029, 3004, 2939, 2870, 1706, 1483 cm^{-1}; ^1H NMR δ 7.81 (d, J = 8, 2H), 7.60 (m, 3H), 7.38 (m, 5H), 7.19 (m, 3H), 6.80 (m, 2H), 3.8–3.67 + 3.6–3.47 (m, 2H), 2.69–2.58 + 2.50–2.39 (m, 2H), 1.92 (s, 3H). Anal. Calcd for $C_{24}H_{22}N_2O$: C, 81.32; H, 6.26; N, 7.91. Found: C, 81.31; H, 6.31; N, 7.88.

2-(*p*-Fluorophenyl)-5-(*p*-methoxyphenyl)-1-phenyl-5-methyl-2-imidazolin-4-one (11f) was recrystallized from ethyl acetate/hexanes. The solid was dried under vacuum at 110 °C to allow sublimation of contaminating dimethyl sulfone: ^{1}H NMR δ 7.73 (m, 2H), 7.24 (m, 5H), 7.01 (t, J = 8, 2H), 6.90 (d, J = 9, 2H), 6.76 (d, J = 8, 2H), 3.82 (s, 3H), 1.72 (s, 3H). Anal. Calcd for $C_{23}H_{19}FN_2O_2$: C, 73.78; H, 5.11; N, 7.48. Found: C, 73.52; H, 5.19; N, 7.37.

1-(*p*-Fluorophenyl)-5-(*p*-fluorophenyl)-2,5-dimethyl-2-imidazolin-4-one (11g) was chromatographed through silica gel (CH_2Cl_2, then ethyl acetate gradually to isocratic ethyl acetate). Further purification was achieved by recrystallization from ethyl acetate/hexanes; IR 3121, 3070, 2974, 1715, 1489, 1222 cm^{-1}; ^{1}H NMR δ 7.15 (m, 2H), 7.04 (m, 4H), 6.79 (m, 2H), 2.24 (s, 3H), 1.70 (s, 3H). Anal. Calcd for $C_{17}H_{14}F_2N_2O$: C, 67.99; H, 4.70; N, 9.33. Found: C, 67.86; H, 4.72; N, 9.29.

1-(2-Phenethyl)-5-phenyl-2,5-dimethyl-2-imidazolin-4-one (11h) was recrystallized from ethyl acetate/hexanes to give white needles: IR 3055, 3020, 2945, 1696 cm^{-1}; ^{1}H NMR δ 7.45–7.20 (m, 8H), 7.05 (d, J = 7, 2H), 3.7–3.35 (2m, 2H), 2.65 (m, 2H), 2.28 (s, 3H), 1.8 (s, 3H); ^{13}C NMR ($CDCl_3$, 270 MHz) showed signals at 192 and 180 ppm (CO and C=N). Anal. Calcd for $C_{19}H_{20}N_2O$: C, 78.05; H, 6.90; N, 9.58. Found: C, 77.98; H, 6.86; N, 9.62.

1-(2-Phenethyl)-2,5-diphenyl-5-ethyl-2-imidazolin-4-one (11i). Chromatography through silica gel column (ethyl acetate) and then recrystallization from ethanol/water gave short, white, fluffy needles: IR 3058, 3029, 2976, 2959, 2930, 1721, 1482 cm^{-1}; ^{1}H NMR δ 7.83 (d, J = 8, 2H), 7.60 (m, 3H), 7.40 (m, 5H), 7.16 (t, J = 3, 3H), 6.74 (m, 2H), 3.63 (hex, J = 6) + 3.46 (hex, J = 6, 2H), 2.56 (m, 2H), 2.34 (m, 2H), 1.01 (s, 3H). Anal. Calcd for $C_{25}H_{24}N_2O$: C, 81.49; H, 6.57; N, 7.60. Found: C, 81.36; H, 6.64; N, 7.55.

2,5-Bis(*p*-fluorophenyl)-1-(2-phenethyl)-5-methyl-2-imidazolin-4-one (11j) was recrystallized from toluene and then ethyl acetate/hexanes: IR 1705 (CO), 1642 cm^{-1} (C=N); ^{1}H NMR δ 7.81 (m, 2H), 7.35–7.30 (m, 3H), 7.25–7.20 (m, 4H), 7.11 (t, J = 9, 2H), 6.84 (m, 2H), 3.79–3.67 + 3.60–3.48 (m, 2H), 2.71–2.60 + 2.54–2.43 (m, 2H), 1.90 (s, 3H). Anal. Calcd for $C_{24}H_{20}F_2N_2O$: C, 73.83; H, 5.16; N, 7.17. Found: C, 73.56; H, 5.20; N, 7.15.

Carbamate Reissert Compounds and Their Hydrolysis. *N*-(α-Cyanobenzyl)-*N*-(2-phenethyl)phenylcarbamate (12) was obtained from the amino nitrile **1a** using the procedure employed for the preparation of the Reissert compounds **2** with phenyl chloroformate instead of the acid chloride. Recrystallization from toluene/hexanes and hexanes afforded white crystals (67%), mp 81–82 °C: IR 3010, 2934, 1717 cm^{-1}; ^{1}H NMR δ 7.57 (m, 2H), 7.48 (m, 3H), 7.42 (t, J = 8, 2H), 7.26 (m, 4H), 7.19 (t, J = 6, 2H), 7.09 (d, J = 6, 2H), 6.64 (bs, 1H), 3.54 (bs, 2H), 3.02 (m) + 2.68 (m) (2H). Anal. Calcd for $C_{23}H_{20}N_2O_2$: C, 77.51; H, 5.66; N, 7.86. Found: C, 77.56; H, 5.67; N, 7.88.

***N*-(α-Cyano-α-methylbenzyl)-*N*-(2-phenethyl)phenylcarbamate (13).** The alkylation was performed using the procedure employed for the preparation of compounds **7** with THF as the solvent. Recrystallizations from 95% ethanol and ethyl acetate/hexanes afforded white, fluffy needles (87%), mp 127.1–128.4 °C: IR 3061, 3028, 2999, 2235, 1716 cm^{-1}; ^{1}H NMR δ 7.52 (m, 2H), 7.42 (m) + 7.26 (m) (11H), 6.85 (d, J = 9, 2H), 3.96 (qn, J = 8) + 3.77 (qn, J = 8) (2H), 3.11 (t, J = 8, 2H), 2.09 (s, 3H). Anal. Calcd for $C_{24}H_{22}N_2O_2$: C, 77.81; H, 5.99; N, 7.56. Found: C, 77.93; H, 6.02; N, 7.56.

4-Phenyl-3-(2-phenethyl)hydantoin (14a). Hydrolysis of **12** was performed using procedure C described for the hydrolysis of compounds **2** and **7**. Recrystallization from ethyl acetate/hexanes afforded colorless crystals (58% yield), mp 176.5–177.5 °C; IR 3156, 3050, 2952, 1767, 1708 cm^{-1}; ^{1}H NMR δ 7.41 (bs, 1H), 7.39 (t, J = 4, 3H), 7.27 (m, 3H), 7.13 (d, J = 7, 4H), 4.54 (s, 1H), 4.00 (qn, J = 7, 1H), 3.01 (m, 1H), 2.88 (m, 2H). Anal. Calcd for $C_{17}H_{16}N_2O_2$: C, 72.84; H, 5.75; N, 9.99. Found: C, 72.76; H, 5.78; N, 10.02.

4-Phenyl-3-(2-phenethyl)-4-methylhydantoin (14b). Hydrolysis of **13** was also performed using procedure C. Recrystallization from 95% ethanol allowed removal of unreacted starting material. The filtrate was evaporated and recrsytallized from ethyl acetate/hexanes to afford dimethyl sulfone. The filtrate was evaporated to give crystals, mp 158.5–159.5 °C (11%): IR 3178, 3057, 1774, 1697 cm^{-1}; ^{1}H NMR δ 7.55 (bs, 1H), 7.38 (t, J = 7, 3H), 7.28 (m, 5H), 7.12 (d, J = 7, 2H), 3.60 (m, 1H), 3.07 (m, 1H), 2.90 (m, 2H), 1.72 (s, 3H). Anal. Calcd for $C_{18}H_{18}N_2O_2$: C, 73.45; H, 6.16; N, 9.52. Found: C, 73.22; H, 6.21; N, 9.47.

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