fluorobutadiene, 381-61-3; 2-cyanobutadiene, 5167-62-4; butadiene, 106-99-0.

References and Notes

(1) (a) K. Ziegler and H. Wilms, *Justus Liebigs Ann. Chem.*, **567**, 1 (1950); (b) W. von E. Doering, M. Franck-Neumann, D. Hasselmann, and R. L. Kaye, *J. Am. Chem. Soc.*, **94**, 3833 (1972); (c) J. A. Berson and P. B. Dervan, *ibid.*, **95**, 267 (1973).
(2) (a) O. Wallach, *Justus Liebigs Ann. Chem.*, **227**, 277 (1895); (b) J. L. Binder, K. C. Eberly, and G. E. P. Smith, Jr., *J. Polym. Sci.*, **38**, 229, (1959); (c) C. Walling and J. Peisach, *J. Am. Chem. Soc.*, **80**, 5819 (1958).
(3) (a) N. C. Billingham, J. R. Ebdon, R. S. Lehrle, J. L. Markham, and J. C. Robb, *Trans. Faraday Soc.*, **65**, 481 (1969); (b) C. A. Stewart, Jr., *J. Am. Chem. Soc.*, **93**, 4815 (1971); (c) C. A. Stewart, Jr., *ibid.*, **94**, 635 (1972).
(4) (a) H. R. Snyder and G. I. Poos, *J. Am. Chem. Soc.*, **71**, 1395 (1949); (b) J. L. Charlish and W. H. Davies, *J. Chem. Soc.*, 1385 (1950).
(5) C. S. Marvel and N. O. Brace, *J. Am. Chem. Soc.*, **70**, 1775 (1948); **71**, 37 (1949).
(6) A. A. Petrov and A. V. Tumanova, *J. Gen. Chem. USSR (Engl. Transl.)*, **26**, 3687 (1956).
(7) (a) F. J. Weigert and J. D. Roberts, *J. Am. Chem. Soc.*, **93**, 2361 (1971); (b) J. B. Grutzner, M. Jautelat, J. B. Dence, R. A. Smith, and J. D. Roberts, *ibid.*, **92**, 7107 (1970).
(8) C. W. Wilson III and E. R. Santee, Jr., *J. Polym. Sci., Part C*, **8**, 97 (1967).
(9) J. Schaefer, *Macromolecules*, **4**, 105 (1971).
(10) Def. Publ. U.S. Pat. Off. T, 11516 (Nov 1973).
(11) O. Eisenstein, J. M. Lefour, N. T. Anh, and R. F. Hudson, *Tetrahedron*, **33**, 523 (1977).

Acylanthranils. 5. Reaction of Acetylanthranil with β-Substituted Amines that Associate by Intramolecular Hydrogen Bonding[1]

L. A. Errede* and J. J. McBrady

Central Research Laboratories, 3M Company, Saint Paul, Minnesota 55101

Received January 28, 1977

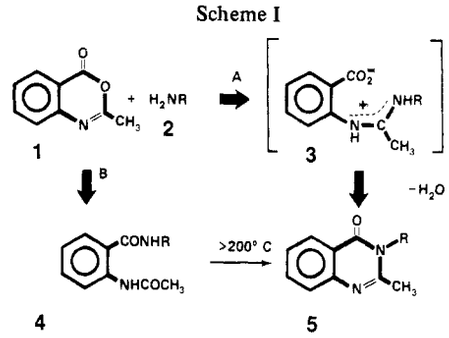
The reaction of acetylanthranil (1) with anthranilic acid (**2a**) in toluene follows pathway B to give *o*-(*o*-acetamidobenzamido)benzoic acid (**4a**), but in a polar solvent it follows pathway A to give *N*-(2-carboxyphenyl)-2-methylquinazol-4-one (**5a**), as shown in Scheme I. Similarly, reaction of 1 with ethanolamine (**2b**) neat follows pathway B to give the corresponding *o*-acetamidobenzamide **4b**, but in pyridine it follows pathway A to give the corresponding quinazolone **5b**. This change in selectivity with change in solvent is attributed to steric hindrance that is manifested by the amine when it is held in a cyclic configuration by intramolecular hydrogen bonding, but which is precluded when the reaction is made to occur in a polar solvent.

It was reported by us[2] that acetylanthranil (1) reacts with anilines to give the corresponding acetamidine intermediate **3** via pathway A as shown in Scheme I, but that anthranilic acid is an exception, which gives the corresponding *o*-acetamidobenzamide **4a** via pathway B. Since it was shown subsequently[3,4] that 1 reacts with aliphatic amines via pathway A preferentially unless steric hindrance on the part of the amine causes the reaction to occur via the slower alternate pathway B, it might be reasonable to attribute the exception with anthranilic acid to classical bulk interference by the ortho substituent were it not for the observation that other ortho-substituted anilines, such as 2,4,6-trimethylaniline, 2,6-diethylaniline, and *o*-anisidine, follow pathway A exclusively to give the corresponding quinazolone **5** in very good yields.

These results indicate that an ortho substituent per se does not interfere with the approach of the amino group to the 2 position of acetylanthranil. Neither can this exception be attributed to simple decreased reactivity of the aniline, owing to the electronegative withdrawing effect of a carboxylic acid group, because it was observed[2] that other similarly substituted anilines, such as *p*-aminobenzoic acid and *m*-trifluoromethylaniline, also follow pathway A exclusively, despite the retarding effect of the electronegative substituent, and like anthranilic acid, required several hours for reaction completion at reflux in toluene.

We postulated, therefore, that this observed exception in selectivity in a nonpolar solvent was caused by the direct association of the *o*-carboxylic acid group with the nucleophilic amine group to form a relatively rigid six-membered ring by intramolecular hydrogen bonding. In this form the anthranilic acid molecule resembles geometrically, but not electronically, heterocyclic secondary amines, which have been shown to exhibit steric hindrance in this reaction.[4] It is reasonable to expect, therefore, that so long as the geometric integrity of this quasi-heterocyclic amine structure remains intact, it should encounter about the same magnitude of steric hindrance in its approach to the electrophilic center at the 2 position of acetylanthranil as that encountered by a true cyclic secondary amine such as pyrrolidine and piperidine, which follow pathway B.

Scheme I



Others have shown[5] that *o*-aminobenzoic acids do indeed exhibit intramolecular hydrogen bonding, especially when dissolved in nonpolar solvents, and that the magnitude of force supporting the cyclic configuration is 7–14 kcal.[6] It is assumed that this force is sufficient to ensure the integrity of the quasi-cyclic amine structure in its reaction with acetylanthranil in a nonpolar solvent.

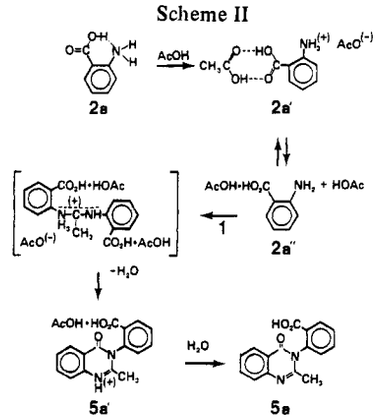
If this hypothesis is correct, then it also follows that pathway A should be favored, when reaction of anthranilic acid with 1 is made to occur in a polar solvent such as acetic acid, which would preclude the formation of the quasi-heterocyclic amine structure, **2a**, by salt formation and strong intermolecular association with the solvent to give an open structure, **2a'**, as shown in Scheme II. Equilibrium dissociation of the salt form **2'** makes available for reaction with 1 the free base form, **2a''**, in low concentration, but not fettered by the *o*-carboxylic acid group, which is still associated with the solvent by strong intermolecular hydrogen bonding. In the form of **2a''**, an-

Table I. Reaction Products of Acetylanthranil with Difunctional Amines

Registry no.	Amine	Reaction conditions: Solv	Temp, °C	Time, h	% Acetylanthranil units isolated as products: 3	5	4	Selectivity ratio, (3 + 5)/4
118-92-3	**2a,** Anthranilic acid	*a*	Reflux	24		5	92	1:18
		b	Reflux	3		95		>50:1
		c	Room temp	3	40	55		>50:1
141-43-5	**2b,** Ethanolamine	*d*	Room temp	6			95	<1:25
		c	Room temp	4		92		>50:1
		b	Room temp	24		*f*		*f*
156-87-6	**2c,** 3-Aminopropanol	*d*	Room temp	3		81	3	27:1
		c	Room temp	4		75	4	19:1
		e	80	0.5		63	28	2:1

[a] Toluene. [b] Acetic acid. [c] Pyridine. [d] Neat. [e] Molar equivalents used at 80 °C. [f] 98% of the acetylanthranil units were isolated as **5a.** Characterization data for products **3, 5,** and **4** are collected in Table II.

Scheme II



thranilic acid is little different from ordinary ortho-substituted anilines, which are known[2] to react readily with **1** to give the corresponding quinazolone **5** via pathway A.

Accordingly, the reaction of anthranilic acid with an equivalent of acetylanthranil was repeated in acetic acid at reflux temperature, and the product distribution was established as described previously.[2,4] The product *N*-(2-carboxyphenyl)-2-methylquinazol-4-one (**5a**) was isolated in 95% yield, as expected on the basis of the reaction sequence outlined in Scheme II.

It might be argued that this change in selectivity with anthranilic acid from A/B = 1:18 in toluene to >50:1 in acetic acid, as indicated in Table I, was caused primarily by the reaction of the nucleophile **2a''** with the protonated form of acetylanthranil, which would be expected to render the 2 position much more electrophilic than the 4 position and therefore responsible in large part for the sharp change in selectivity. To determine whether or not this electronic contribution was indeed the dominant factor, the experiment was repeated with pyridine as the solvent. In this case, the quasi-heterocyclic amine configuration is precluded by salt formation with the *o*-carboxylic acid group and weak hydrogen bonding of the amino group with solvent. Equilibrium dissociation of the amino carboxylic acid–solvent complex would provide an ample concentration of free amine as described previously to enable reaction to occur selectively via pathway A at an appreciable rate even at room temperature. Again as expected, the product distribution indicated that the reaction favored overwhelmingly pathway A (i.e., A/B >50:1). About 55% of the acetylanthranil was recovered as **5a** and about 40% was recovered as *N,N'*-di(2-carboxyphenyl)acetamidine (**3a**), which was converted in turn to **5a** by cyclodehydration in aqueous solution at room temperature.

One could question whether or not this change in reaction selectivity from A/B = ca. 1:25 in a nonpolar solvent to ca. 50:1 in a polar solvent might also obtain for those monofunctional amines, which we reported earlier[4] favor pathway B instead of A owing to steric hindrance. Therefore we reexamined the reaction of acetylanthranil with *tert*-butylamine using pyridine at reflux instead of benzene as the reaction solvent. Again the reaction selectivity A/B was found to be <1:25 as reported earlier.[4]

These results support the hypothesis that the anomalous selectivity noted in the reaction of **1** with anthranilic acid in a nonpolar solvent is caused by intramolecular hydrogen bonding, which is precluded in a polar solvent. To determine whether or not this result is unique to anthranilic acid or is perhaps general for any bifunctional primary amine that can form a quasiheterocyclic ring by intramolecular hydrogen bonding, it was decided to determine the corresponding selectivity with a β-substituted amino alcohol. Accordingly, acetylanthranil was allowed to react at room temperature with excess ethanolamine used as solvent. Reaction under these conditions required several hours for completion which usually is the case with amines that encounter steric hindrance to reaction with acetylanthranil at the 2 position. The only product isolated was identified as 2-acetamido-*N*-(β-hydroxyethyl)benzamide (**4b**), and it accounted for 95% of the acetylanthranil. This result indicates that reaction of **1** with ethanolamine neat follows pathway B almost exclusively (i.e., A/B <1:25) and infers steric hindrance. This result is also in sharp contrast to those obtained under the same reaction conditions with *n*-propylamine and allylamine, which have comparable geometry and bulk to that of ethanolamine. Reaction with these monofunctional amines was complete within 5 min and, in both instances, it followed pathway A exclusively[4] (i.e., A/B >50:1).

If this change in selectivity is indeed due to the formation of a quasi-heterocyclic structure as postulated above, then by analogy with the rationale applied to the case with anthranilic acid, reaction of **2b** with **1** should follow pathway A when made to occur in a polar solvent. Accordingly, acetylanthranil was allowed to react at room temperature with an equivalent of ethanolamine dissolved in pyridine. Under these conditions, the pyridinium salt of *N*-(2-carboxyphenyl)-*N'*-(β-hydroxyethyl)acetamidine (**3b**) was produced as expected, and it was isolated as an oil that was readily soluble in water. Cyclodehydration occurred slowly in aqueous solution at room temperature, and the daughter product separated over a period of seven days to give *N*-(β-hydroxyethyl-2-methylquinazol-4-one (**5b**) in the form of long fine needles easily collected by filtration. The amount of **5b** isolated in this way accounted for 92% of the acetylanthranil made to react with ethanolamine in pyridine. Since none of the more easily isolable benzamide **4b** was obtained, it was concluded that reaction of **1** with **2b** in pyridine follows pathway A exclusively

Table II. Characterization Data for Reaction Products 3, 4, and 5 of the Difunctional Amines Listed in Table I

Registry no.	Product	Mp, °C	Key IR absd. bands, μ	NMR data, τ values[a]
	Amidine salt			
1954-87-6	**3a**	166–168	2.9 → 4.3 5.9, 6.0, 6.3	1.4–2.9 cpx, 8 H (Ar'c) 7.85 s, 3 H (CH_3)
	Quinazolones			
4005-06-5	**5a**	254–256	3.3 → 4.2 5.8, 5.9, 6.1	1.7–2.6 cpx, 8 H (Ar'c) 7.86 s, 3 H (CH_3)
10376-59-7	**5b**	156–157	3.1 m, br 5.9, 6.3	1.8–2.6 cpx, 4 H (Ar'c) 5.85 t, 2 H ($N-CH_2$) 6.29 t, 2 H (CH_2O) 7.3 s, 3 H (CH_3) 5.1 br, 1 H (OH)
63703-20-0	**5c**	78–79	3.2 m, br 6.0, 6.3	1.8–2.6 cpx, 4 H (Ar'c) 5.82 t, 2 H ($N-CH_2$) 8.10 cpx, 2 H ($C-CH_2-C$) 6.40 q, 2 H (CH_2O) 7.31 s, 3 H (CH_3) 5.26 t, 1 H (OH)
	o-Acetamidobenzamides			
58426-37-2	**4a**	215–216	3.2 → 4.3 5.9, 6.1, 6.3 6.5	
63703-31-1	**4b**	158–159	3.0 6.0, 6.1, 6.3 6.6	
63703-32-2	**4c**	195–196	3.1 6.0, 6.1, 6.3 6.6	

[a] Legend for NMR data; cpx, complex; s, singlet; t, triplet; q, quadruplet; br, broad.

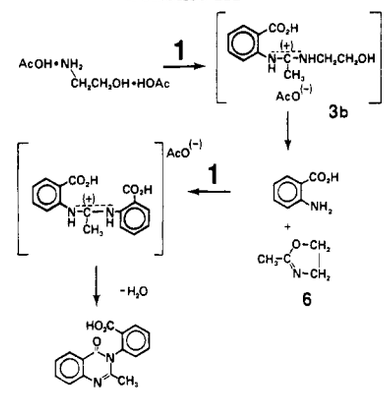
(i.e., A/B >50:1) inferring that reaction under these conditions occurs without steric hindrance, which is in sharp contrast to the selectivity exhibited in the reaction with ethanolamine neat (i.e., A/B <1:25).

When acetylanthranil was made to react with ethanolamine in acetic acid, however, neither **4b** nor **5b** was produced. Virtually all of the acetylanthranil was isolated instead as **5a**, the quinazolone product isolated previously when **1** was made to react with anthranilic acid. The unexpected formation of this product can be rationalized by assuming that the expected intermediate, *N*-(2-carboxyphenyl)-*N'*-(2-hydroxyethyl)-acetamidine (**3b**) undergoes decomposition in acetic acid to give 2-methyl-4,5-dihydrooxazole (**6**) and anthranilic acid, which reacts in turn with residual **1** to give ultimately **5a** as outlined in Scheme III. Although mass spectrometric data indicated that **6** was indeed present in the acetic acid distillate obtained as a result of the separation procedure, it cannot be offered as supporting evidence for the pathway outlined in Scheme III because **6** could have formed by direct reaction of ethanolamine and acetic acid. More work is obviously needed to establish unambiguously the reaction pathway for the unexpected formation of **5a** when acetylanthranil is made to react with ethanolamine in acetic acid.

Despite this uncertainty, the sharp change in selectivity realized in the reaction of **1** with **2b** neat and in pyridine supports the point of view that ethanolamine can assume a cyclic configuration in its neat liquid state, owing to *intra*molecular hydrogen bonding, whereas it has a more open configuration when dissolved in a polar solvent owing to *inter*molecular hydrogen bonding.

One would expect that the ratio of *inter*- to *intra*-molecular hydrogen bonding of amino alcohols in the liquid state should increase with increase in the number of methylene units that separate the polar end groups, and that this ratio should be reflected in the product selectivity ratio with acetylanthranil. Accordingly, acetylanthranil was dissolved in 3-aminopropanol at room temperature and the product mixture was separated as described in the reaction with ethanolamine neat.

Scheme III



The major product isolated (81%) was *N*-(3-hydroxypropyl)-2-methylquinazol-4-one (**5c**), and the minor product isolated (3%) was 2-acetamido-*N*-(3-hydroxypropyl)benzamide (**4c**). This reaction selectivity with **2c** neat, which favors pathway A (i.e., A/B = 27:1), is in sharp contrast to that observed with ethanolamine **2b** neat (i.e., A/B <1:25). This change in selectivity implies that intramolecular hydrogen bonding is much less important in **2c** neat relative to that in **2b** neat. Similar results were obtained using pyridine as solvent (A/B = 19:1) as noted in Table I. When equivalents of acetylanthranil and 3-aminopropanol were made to react by fusion at 80 °C, however, the product distribution corresponded to a selectivity ratio A/B = 2:1, which implies a marked increase in intramolecular hydrogen bonding at the higher temperature.

Experimental Section

The general procedure for reaction of acetylanthranil (**1**) with amines (**2**) in a suitable solvent and the separation and identification of the product to ensure a good materials balance is described in ref 2. The percent acetylanthranil units isolated as product **3**, **4**, and **5** and the corresponding reaction selectivity, A/B = (**3** and/or **5**)/**4**, calculated therefrom are collected in Table I. The characterization data for the products are collected in Table II. Slight modifications

of the general procedure to accomodate the bifunctional amines of this study are given below.

A. Reaction of Acetylanthranil 1 with Anthranilic Acid 2a. 1. In Toluene to Give 4a and 5a. Equivalent amounts of 1 and **2a** were allowed to react in toluene at reflux temperature overnight and the products were separated and identified by IR and NMR (Table II) as described in ref 2. The materials balance showed that 92% of 1 was recovered as *o*-(*o*-acetamidobenzamido)benzoic acid (**4a**) (mp 215–216 °C) and 5% as *N*-(2-carboxyphenyl)-2-methylquinazol-4-one (**5a**) (mp 255–257 °C).

2. In Acetic Acid to Give 5a. A solution of 1 (3.2 g) and **2a** (2.8 g) in acetic acid (30 mL) was allowed to react at reflux for 3 h. The solvent was removed by evaporation in a rotary film evaporator at 60 °C (8 mmHg). The residue was dissolved in dilute aqueous base. The clear solution was neutralized to pH 5 and allowed to remain at room temperature for about 1 week, during which time the product separated as a white powder (5.3 g; mp 254–256 °C). The product was identified as **5a** by its molecular weight, 280, which was determined by mass spectrometry, and by its melting point and IR spectrum, which were identical with those of an authentic sample.

3. In Pyridine to Give *N,N'*-Di(*o*-carboxyphenyl)acetamidine (3a). A solution of 1 (5 g) and **2a** (4.1 g) in pyridine (50 mL) was allowed to remain at room temperature overnight. The solvent was removed in a rotary film evaporator at 50 °C (8 mmHg). The residue was leached with 10% aqueous HCl. The acid insoluble residue (4.8 g; mp 252–254 °C) was identified as **5a** by mixture melting point with an authentic sample and by its IR spectrum. The acidic mother liquor was neutralized to pH 7 with aqueous NaOH to give a light brown precipitate (4.3 g; mp 192–193 °C). The IR and NMR spectra were consistent with the structure of *N,N'*-di(*o*-carboxyphenyl)acetamidine (**3a**) (Table II). A sample of **3a** (1 g) was dissolved in dilute aqueous base. It was allowed to remain at room temperature overnight. The solution was then made acidic with aqueous HCl. The acid insoluble precipitate (0.8 g; mp 252–254 °C) was identified as **5a** by comparison with an authentic sample.[2]

B. Reaction of 1 with Ethanolamine (2b). 1. Neat to Give 4b. Acetylanthranil (10 g) was added slowly to ethanolamine (20 g) kept below 20 °C in a water bath. The solution was allowed to warm to room temperature, and the product began to precipitate within 20 min. The mixture became a semisolid mass overnight. The white crystalline product (13.3 g) was collected by filtration and then recrystallized from ethyl acetate to give *o*-acetamido-*N*-(2-hydroxyethylbenzamide) (**4b**) in the form of long delicate needles (10.1 g; mp 158–159 °C). The product was identified by its IR spectrum (Table II) and its elemental analyses.

Anal. Calcd for $C_{11}H_{14}N_2O_3$: C, 59.45; H, 6.35; N, 12.61. Found: C, 59.5; H, 6.4; N, 12.6.

2. In Pyridine to Give *N*-(2-Hydroxyethyl)-2-methylquinazol-4-one (5b). A solution of 1 (5.0 g) and **2b** (20 g) in pyridine (30 mL) was allowed to react at room temperature overnight. The solution was evaporated to dryness in a rotary film evaporator. The viscous residue (7.5 g) was crystallized from a minimum amount of hot water to give **5b** in the form of white crystals (5.7 g; mp 156–157 °C) that resembled in physical appearance **4b** (mp 155–156 °C). The mixture melting point of **5b** and **4b,** however, was markedly depressed (128–142 °C). The assigned structure for **5b** was verified by its IR and NMR spectra (Table II).

3. In Acetic Acid to Give 5a. A solution of 1 (5.0 g) and **2b** (1.8 g) in acetic acid (20 mL) was allowed to react overnight at room temperature. The solution was evaporated to dryness in a rotary film evaporator. The viscous residue (7.1 g) was dissolved in hot water and allowed to remain at room temperature overnight, during which time the product separated as dense white crystals (4.2 g; mp 248–251 °C). The product was identified as *N*-(2-carboxyphenyl)-2-methylquinazol-4-one (**5a**) by its NMR spectrum (Table II) and by comparison of its IR spectrum with that of an authentic sample.

C. Reaction of 1 with 3-Aminopropanol (2c). 1. As a Fused Melt to Give 4c and 5c. A mixture of 1 (5 g) and **2c** (2.3 g) was warmed to about 80 °C to give a clear amber solution that solidified within 20 min to a glass-like solid. The mass was leached with hot water leaving a powdery residue (1.9 g) which was recrystallized from methanol–water to give *o*-acetamido-*N*-(3-hydroxypropyl)benzamide (**4c**) in the form of white crystals (mp 195–196 °C). The assigned structure was verified by its IR spectrum (Table II). The aqueous extract was allowed to remain at room temperature for 2 weeks, during which time *N*-(3-hydroxypropyl)-2-methylquinazol-4-one (**5c**) separated as a white powder (4.3 g; mp 78–79 °C). The assigned structure was verified by its IR and NMR spectra (Table II).

2. In Pyridine to Give 5c and Some 4c. A solution of 1 (5 g) and **2c** (2.3 g) in pyridine (10 mL) was allowed to react at room temperature overnight. The solvent was removed by evaporation under vacuum at 60 °C in a rotary film evaporator. The oily residue was leached with hot water leaving a powdery residue (0.3 g; mp 185–188 °C), which was identified as **4c** by its melting point and IR spectrum (Table II). The aqueous extract was allowed to remain at room temperature for about 1 month, during which time **5c** separated in the form of white crystals (5.1 g; mp 78–79 °C).

3. Neat to Give 5c and Some 4c. A solution of 1 (5 g) and **2c** (8 g) was allowed to react at room temperature overnight. The solution was diluted with water to about 50 mL. A small amount (0.2 g) of a white precipitate formed and was removed by filtration. The product was identified as **4c.** The mother liquor was stored at room temperature for about 1 month. After 5 days, a precipitate (mp 78–79 °C) began to separate from the clear solution and accumulate to 3.7 g after 12 days. An additional 1.8 g accumulated over the next 7 days. This product was identified as **5c** by its melting point and IR spectrum (Table II). Presumably, **5c** was formed by cyclodehydration of the acetamidine (**3c**) in aqueous solution at room temperature. The results obtained in experiments C-1, -2, and -3 show that under these conditions, conversion from **3c** to **5c** is only half complete after about 12 days.

Registry No.—**1,** 525-76-8; **3c,** 64056-87-7.

References and Notes

(1) Presented in part before the 10th Great Lakes Regional Meeting of the American Chemical Society, Northwestern University, Evanston, Ill., June 1976, Abstract No. 276.

(2) Part 2: L. A. Errede, J. J. McBrady, and H. T. Oien, *J. Org. Chem.*, **41,** 1765 (1976).

(3) Part 3: L. A. Errede, H. T. Oien, and D. R. Yarian, *J. Org. Chem.*, **42,** 12 (1977).

(4) (a) Part 4: L. A. Errede, J. J. McBrady, and H. T. Oien, *J. Org. Chem.*, **42,** 656 (1977). (b) The product distributions listed in Table I of this reference for NH_3 (**2d**) should read 0% for **4d,** 0% for **3d,** and 65% for **5d;** that for CH_3NH_2 (**2e**) should read 0% for **4e,** 100% for **3e,** and 0% for **5e.** Also the selectivity ratios k_A/k_B calculated for the amines **2f, 2i, 2k, 2p,** and **2q** should all read <1:25 rather than >1:25.

(5) (a) G. Chuchani in "The Chemistry of the Amino Group", S. Patai, Ed., Interscience, New York, N.Y., 1968, pp 205–275; (b) H. C. Brown, D. H. McDaniel, and O. Haflinger in "Determination of Organic Structures by Physical Methods", E. A. Braude and F. C. Nachod, Ed., Academic Press, New York, N.Y., 1955, pp 567–662; (c) B. Legate and G. E. Dunn, *Can. J. Chem.*, **43,** 1158 (1965); (d) H. C. Saraswat and U. D. Tripathi, *Bull. Chem. Soc. Jpn.*, **38,** 1555 (1965).

(6) P. Schuster in "The Hydrogen Bond/I-Theory", P. Schuster, G. Zunsten, and C. Sandorfy, Ed., North-Holland Publishing Co., Amsterdam, 1976, p 117.