The Thermal Decomposition of *N,O*-Diacyl-*N*-*t*-butylhydroxylamines. II.[1] Thermal Rearrangement of *O*-Acyl-*N*-[2-(methylthio)-benzoyl]-*N*-*t*-butylhydroxylamines

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Several *O*-acyl-*N*-[2-(methylthio)benzoyl]-*N*-*t*-butylhydroxylamines (**1**) were prepared and their thermal decompositions were studied. The thermal decomposition of **1** at 200 °C in *o*-dichlorobenzene gave *N*-*t*-butyl-2-(acyloxymethylthio)benzamide (**4**), the carboxylic acid derived from the acyl part of **1**, and 2-*t*-butyl-1,2-benzothiazol-3(2*H*)-one as the main products, together with small amounts of 4*H*-3,1-benzoxathiin-4-one, *N*-*t*-butyl-2-(methylthio)benzamide, methyl ester of the carboxylic acid, and *N*-*t*-butylamide. The benzamide (**4**) was found to be an initial product of the thermolysis and the subsequent decomposition gave the carboxylic acid and other products. Pummerer type reaction, *via* acylaminosulfonium ion as the intermediate, was suggested for the thermal decomposition of **1** since similar products were also obtained by the Pummerer reaction of *N*-*t*-butyl-2-(methylsulfinyl)benzamide with acylating reagents.

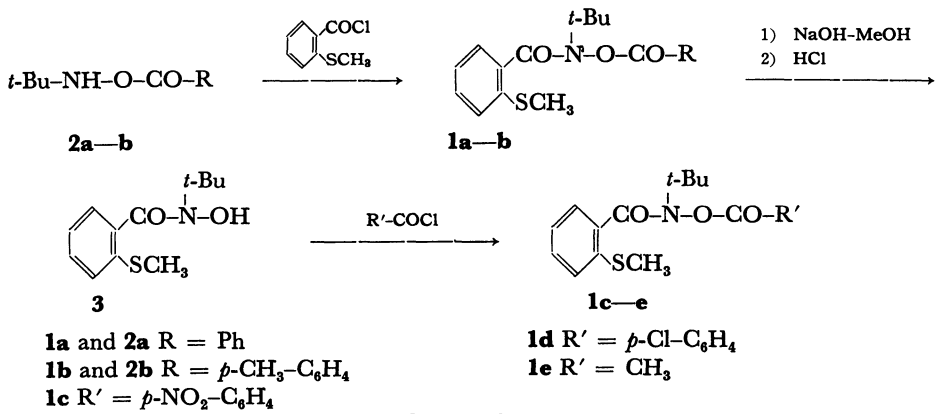
N,O-Diacylhydroxylamines have been known to react with nucleophiles both at the carboxyl carbon atom and at the nitrogen atom depending on the nature of the nucleophiles. In general, nucleophiles such as amines,[2] sulfide,[3] alkoxide,[1,3] azide,[3] and cyanide ions[3] attack the carboxyl carbon atom of the *N,O*-diacylhydroxylamines. On the other hand, Ohta *et al.* have shown the substitution reaction at the nitrogen atom by the reaction of *O*-acetyl-*N*-benzoyl-*N*-(4-methoxyphenyl)hydroxylamine with phenols, pyrrole, and indoles.[4] In the previous paper, we have reported that *N,O*-diacyl-*N*-*t*-butylhydroxylamines were decomposed thermally to afford amides and carboxylic acids in good yields. A mechanism involving nucleophilic substitution at the nitrogen atom has been suggested for the thermolysis of the hydroxylamines.[1]

Meanwhile, formations of acylaminosulfonium salts by the nucleophilic substitution at the amide-nitrogen atom have been known for the reaction of *N*-haloamides with dialkyl sulfides.[5] In this connection, thermolysis of a *N,O*-diacylhydroxylamine bearing a nucleophilic group at an appropriate position would take place by a different way, that is, an intramolecular substitution. Thus, we have prepared several *N,O*-diacyl-*N*-*t*-butylhydroxylamines (**1**) and studied their thermal decompositions. An intramolecular substitution reaction at the nitrogen atom by the methylthio-sulfur atom has been found as was expected.

Results and Discussion

Preparation of O-*Acyl*-N-[*2-(methylthio)benzoyl*]-N-t-*butylhydroxylamines (1).* Several title hydroxylamines (**1**) were prepared by the acylation of *O*-acyl-*N*-*t*-butylhydroxylamines (**2**) or *N*-[2-(methylthio)-benzoyl]-*N*-*t*-butylhydroxylamine (**3**) with corresponding acyl chlorides in the presence of pyridine (Scheme 1). Syntheses of **1a** and **1b** were performed by the reactions of 2-(methylthio)benzoyl chloride with **2a** and **2b**, respectively, in good yields. By the reaction of **2c** and **2d**, **1c** and **1d**, however, were obtained, respectively, in only poor yields. Alternatively, **1a** was hydrolized giving **3** which was acylated by appropriate acyl chlorides giving **1c—e** in satisfactory yields. Yields, physical properties, and analyses of **1** are summarized in Table 1.

Thermal Decomposition of N,O-*Diacyl*-N-t-*butylhydroxylamine (1).* On heating at 200 °C for 25 h in *o*-dichlorobenzene, 78% of **1a** was decomposed to give *N*-*t*-butyl-2-(benzoyloxymethylthio)benzamide (**4a**), benzoic acid (**5a**), 2-*t*-butyl-1,2-benzothiazol-3(2*H*)-one (**6**), 4*H*-3,1-benzoxathiin-4-one (**7**), *N*-*t*-butyl-2-(methylthio)benzamide (**8**), methyl benzoate (**9a**), and *N*-*t*-butylbenzamide (**10a**). The yields of the products

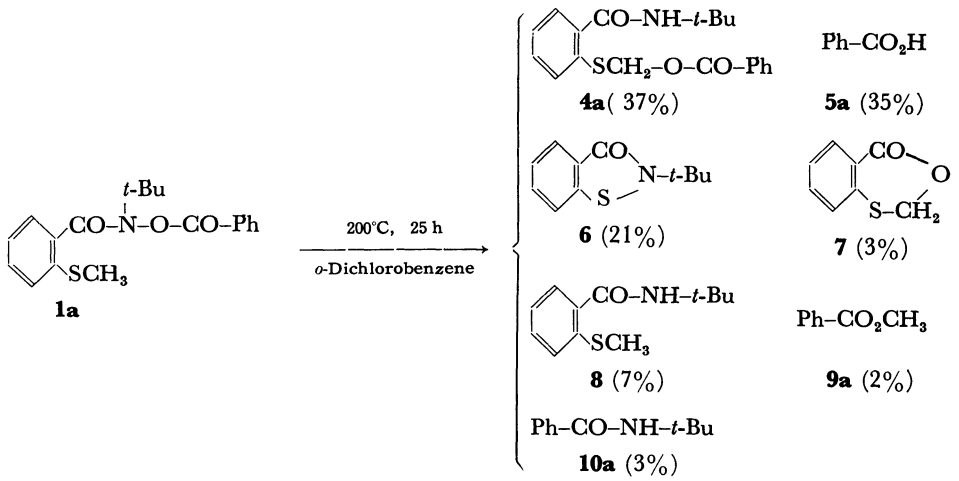


Scheme 1.

Table 1. Yields and physical properties of *O*-acyl-*N*-[2-(methylthio)-benzoyl]-*N*-*t*-butylhydroxylamines (**1a—e**)

Compd	Starting compd	Yield %	Mp(Bp) °C	IR (KBr) $\tilde{\nu}$(C=O)/cm^{-1}	Found (Calcd) (%) C	H	N
1a	**2a**	83	116.5—117.5	1760, 1640	66.37 (66.45)	5.98 (6.16)	4.02 (4.08)
1b	**2b**	69	70.5— 71.5	1750, 1650	67.27 (67.20)	6.43 (6.49)	3.75 (3.92)
1c	**3**	82	119.0—120.0	1760, 1670	58.58 (58.75)	5.13 (5.19)	7.30 (7.21)
1d	**3**	80	97.0— 98.0	1760, 1650	60.33 (60.39)	5.30 (5.34)	3.57 (3.71)
1e	**3**	86	(156—157/3 mmHg)††	1790, 1660	59.95 (59.76)	6.89 (6.81)	5.01 (4.98)

††Through this paper 1 mmHg=133.322 Pa.



Scheme 2.

were estimated by means of GLC (Scheme 2).

The structures of the products were ascertained on the basis of the spectral and chemical evidences. The NMR spectrum of **4a** in carbon tetrachloride showed singlets at δ 1.38 (9H) and δ 5.57 (2H), broad singlet at δ 6.10 (1H), and multiplet at around δ 7.1—8.1 (9H). The characteristic amide and ester bands of **4a** were observed at 3290 (N–H), 1720 (ester C=O), 1630 (amide C=O), and 1540 cm^{-1} (N–H) in its IR spectrum. In addition, the structure of **4a** was confirmed by comparision these physical properties with those of an authentic sample prepared by the reaction of *N*-*t*-butyl-2-(methylsulfinyl)benzamide (**11**) with benzoic anhydride.

The NMR spectrum of **6** in carbon tetrachloride showed a singlet at δ 1.68 (9H) and aromatic proton signals at δ 7.3—8.0 (4H). The IR spectrum of **6** consisted with amide carbonyl absorption at 1650 cm^{-1}. The elemental analysis also did not conflict with the structure. The compound **6** was synthesized by a different route. Namely, the reaction of *N*-*t*-butyl-2-(methylsulfinyl)benzamide (**11**) with acetyl or thionyl chlorides gave **6** in good yields. The structure of **7** was confirmed by comparing physical properties with those of the authentic sample prepared by the method of Numata and Oae.[6]

Table 2. Thermal decomposition of *O*-acyl-*N*-[2-(methylthio)benzoyl]-*N*-*t*-butylhydroxylamines at 200 °C in *o*-dichlorobenzene (0.1 mol/dm^3)

Compd	Time h	Conversion %	Products yield/%[a] 4	5[b]	6	7	8	9	10
1a	25	78	37	35	21	3	7	2	3
1a	50	94	11	60	36	7	11	1	9
1b	25	67	30	14	30	2	4	1	2
1b	50	85	22	51	38	3	6	2	5
1c	7	100	50[c]	23	9	7	2	1	11
1d	25	96	44	43	26	7	7	2	4
1e	25	56	7	[d]	20	2	1	[d]	[d]
1e	50	70	6	[d]	28	5	7	[d]	[d]

a) Based on the starting **1**. Estimated by GLC. b) The amounts of the acid were estimated after methylation with diazomethane. c) Isolated yield. d) Not determined.

The rearranged products (**4b—e**) were obtained similarly together with **5**, **6**, **7**, **8**, **9**, and **10** by the thermal decompositions of **1b—e**. These results are summarized in Table 2. Physical properties and analyses of **4** are summarized in Table 3.

Inspection of the results given in Table 2 reveals that

Table 3. Physical properties of *N*-*t*-butyl-2-(acyloxymethylthio)benzamides

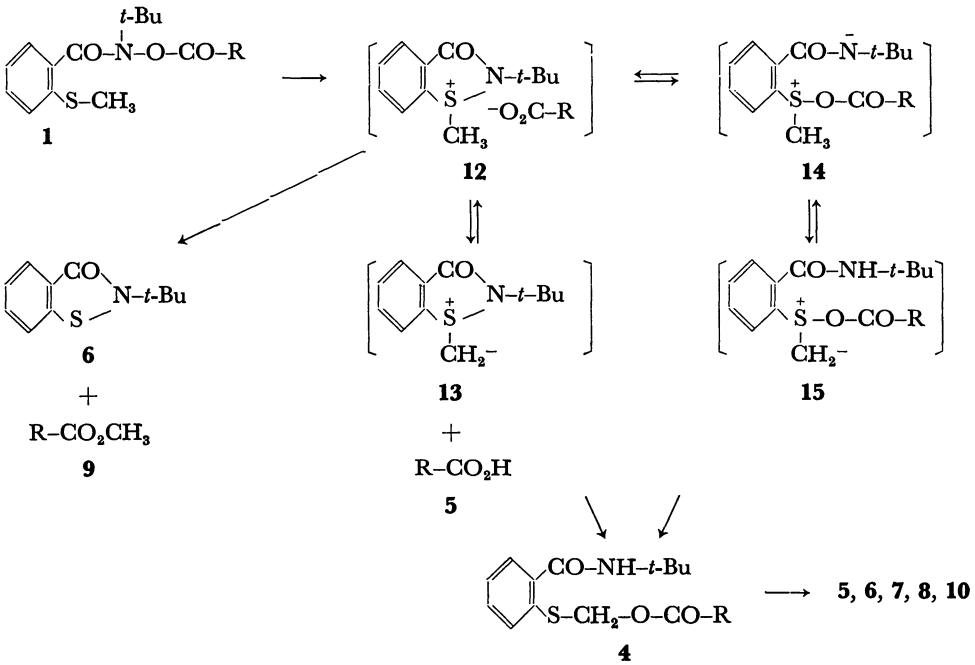
Compd	Mp °C	IR (KBr) $\tilde{\nu}$/cm^{-1} (N–H)	(Ester C=O)	(Amide C=O)	Found (Calcd) (%) C	H	N
4a	85.0— 86.0	3290	1720	1630	66.47 (66.45)	6.26 (6.16)	4.02 (4.08)
4b	91.0— 92.0	3300	1720	1640	67.05 (67.20)	6.44 (6.49)	4.00 (3.92)
4c	120.5—121.5	3310	1720	1640	58.94 (58.75)	5.19 (5.19)	7.24 (7.21)
4d	83.0— 84.0	3290	1720	1630	60.38 (60.39)	5.29 (5.34)	3.77 (3.71)
4e	69.5— 70.5	3350	1730	1660	60.00 (59.76)	6.95 (6.81)	5.01 (4.98)

the rates of the decompositions of **1** were affected by the substituent of the *O*-acyl group. Electron-withdrawing groups accelerated the reaction. Namely, the thermal decomposition of **1c** which has 4-nitrobenzoyl group, completed within 7 h on heating at 200 °C, while, 33 and 44% of the starting materials were recovered by the decompositions of **1b** and **1e**, respectively, under the same reaction conditions. The yields of **4** were found to be depressed by prolonged heating as shown in Table 2. In a control experiment, **4a** was found to be decomposed under the same reaction conditions (200 °C, 25 h) giving **5a**, **6**, **7**, **8**, and **10a** in 46, 6, 7, 10, and 5% yields, respectively, with 60% decomposition of the starting material. These results suggest a successive decomposition of **4** giving the products, **5**—**8** and **10**, for the decomposition of **1**, although the yield of **6** from **4** (6%) could not account for that from **1** (21%).

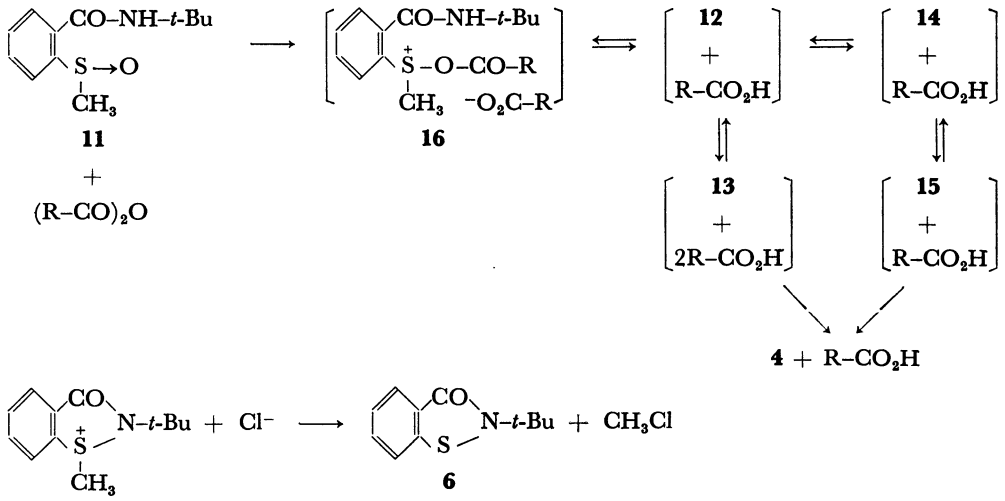
A probable mechanism leading to the rearranged product, **4**, from **1** would involve an intramolecular nucleophilic attack of sulfur atom on the nitrogen atom giving acylaminosulfonium salt, **12**, as the intermediate. The salt **12** would afford **4** by subsequent Pummerer rearrangement *via* ylid **13**. An alternative possible pathway leading to **4** from **12** may involve attack of the anion on the sulfur giving sulfonium salt **14** and proton migration would give the ylid **15**. Pummerer rearrangement of **15** may afford **4** as shown in Scheme 3. Direct formation of the sulfonium amide **14** by the attack of the sulfur on the ester oxygen of **1** seems to be unlikely by considering differences in leaving abilities of carboxylate and amide anions, and also in electronegativities of oxygen and nitrogen atoms. In fact, acceleration of the rate by the electron-withdrawing substituent on the *O*-acyl group is in accordance with the proposed mechanism involving the carbooxylate ion as the leaving group.

As mentioned above, another pathway would be required to account for the yield of **6**. This may be a demethylation reaction of the acylamisosulfonium ion (**12**) by a nucleophile. The formation of methyl ester (**9**) by the decomposition of **1** would support this prosess.

Pummerer Reactions of N-t-*Butyl-2-(methylsulfinyl)benzamide (11).* In order to prove the intermediacy of the acylaminosulfonium ion (**12**) for the thermolysis



Scheme 3.



Scheme 4.

of **1**, Pummerer reaction of **11** with acylating reagents, such as benzoic anhydride, acetic anhydride, acetyl chloride, and thionyl chloride were examined. In Pummerer reaction of **11**, the acylaminosulfonium ion **12** and/or the sulfonium salt **14** would be the intermediate. The sulfoxide **11** was heated at 180 °C for 10 h with 1.2 equiv. of benzoic anhydride in *o*-dichlorobenzene. The product **4a** (55%) was obtained together with small amount of **6** as was expected. The reaction of **11** with acetic anhydride gave **4e** and **6** similarly. On the other hand, the reaction of **11** with acetyl chloride or thionyl chloride proceeded at room temperature affording **6** in good yields. When excess of thionyl chloride was added to carbon tetrachloride solution of **11**, the NMR signals of *t*-butyl protons (δ 1.47, s) and *S*-methyl protons (δ 2.73, s) disappeared and new signals appeared at δ 1.73 (s), 1.80 (s), 2.99 (s), and 3.70 (s). During the reaction, the intensities of the signals at δ 1.80 and 3.70 decreased and the signals at δ 1.73 and 2.99 increased. The down field shift to δ 1.80 and 3.70 of the *t*-butyl and *S*-methyl signals, respectively, would apparently due to a positive charge which in turn suggest the formation of acylaminosulfonium ion (**12**) as the intermediate. After about 20 min, the signals at δ 1.80 and 3.70 disappeared and the ratio of the intensities of the signals at δ 1.73 and 2.99 was about 3 : 1. These signals were assigned to be *t*-butyl protons of **6** and methyl protons of chloromethane, respectively.

The reaction of **11** with acylating reagents can be explained by the mechanism shown in Scheme 4. The sulfonium salt **16**, initially formed by the reaction of **11** with acylating reagents gives **12** and/or **14**,[7] then rearranges to **4** by Pummerer reaction.[8] In the reactions of **11** with acetyl and thionyl chlorides, the acylaminosulfonium ion (**12**) is attacked by chloride ion giving **6** and chloromethane. Thus, the intermediacy of the acylaminosulfonium ion (**12**) for the thermal decomposition of **1** has been confirmed by the Pummerer reaction of the corresponding sulfoxide.

Experimental

All the melting points and the bioling points are uncorrected. The IR spectra were recorded on a Shimazdu IR-430 infrared spectrometer. The NMR spectra were recorded on a Varian EM-360 spectrometer using TMS as the internal standard.

Preparation of O-*Acyl*-N-[*2-(methylthio)benzoyl*]-N-t-*butylhydroxylamines (1).* *Typical Procedure (a)*: A solution of 2-(methylthio)benzoyl chloride (58.0 g, 300 mmol), *O*-benzoyl-*N*-*t*-butylhydroxylamine (58.0 g, 310 mmol),[1] and pyridine (25.0 g, 320 mmol) in dry benzene (200 cm^3) was stirred at 75 °C for 20 h. The reaction mixture was washed with aq NH_3, dil NaOH, dil HCl, and water, and then dried over Na_2SO_4. The solvent was evaporated *in vacuo*, and the residue was recrystallized from benzene giving **1a** (86.0 g, 83%).

By the reaction of *O*-(4-methylbenzoyl)-*N*-*t*-butylhydroxylamine[1] with 2-(methylthio)benzoyl chloride, **1b** was prepared similarly.

(b): A solution of *N*-[2-(methylthio)benzoyl]-*N*-*t*-butylhydroxylamine (**3**) (12.0 g, 50 mmol), 4-chlorobenzoyl chloride (10.5 g, 60 mmol), and pyridine (4.7 g, 59 mmol) in dry benzene (150 cm^3) was stirred at 75 °C for 20 h. The reaction mixture was washed with aq NH_3, dil NaOH, dil HCl, and water, and then dried over Na_2SO_4. The solvent was evaporated *in vacuo* and the residue was recrystallized from CH_2Cl_2–petroleum ether giving **1d** (15.2 g, 80%).

By the same procedure, **1c** and **1e** were prepared by the reaction of **3** with 4-nitrobenzoyl chloride and acetyl chloride, respectively. The yields and physical properties are summarized in Table 1.

Preparation of N-[*2-(Methylthio)benzoyl*]-N-t-*butylhydroxylamine (3).* A solution of NaOH (10.0 g, 250 mmol) in water (40 cm^3) was added to a solution of **1a** (34.3 g, 100 mmol) in methanol (200 cm^3). The mixture was stirred at room temperature for 3 h. After evaporation of the solvent, the residue was dissolved in water (200 cm^3), washed with diethyl ether, and acidified with HCl. The separated crystals were extracted with CH_2Cl_2, and the organic layer was washed with aq $NaHCO_3$ and water, and then dried over Na_2SO_4. The solvent was evaporated and the residue was recrystallized from benzene to give **3** (19.7 g, 82%); mp 145.0—146.0 °C. Found: C, 60.36; H, 7.31; N, 5.78%. Calcd for $C_{12}H_{17}O_2S$: C, 60.22; H, 7.16; N, 5.85%. IR: (KBr) 3150 (O–H) and 1600 cm^{-1} (C=O). NMR: ($CDCl_3$, δ) 1.38 (9H, s), 2.48 (3H,

s), 7.1—7.4 (4H, m), and 8.33 (1H, s).

Thermal Decomposition of **1**. A solution of **1** in *o*-dichlorobenzene (0.1 mol/dm³, 10 cm³) was sealed in a degassed tube and then heated in a constant-temperature bath at 200 °C. The tube was opened and 60 mg of biphenyl was added to the reaction mixture as an internal standard. The mixture was subjected to GLC analysis. In order to estimate the yield of carboxylic acid, an excess of an diethyl ether solution of diazomethane was added to a portion of the sample, and the mixture was allowed to stand for 1 h. After the evaporation of the ether *in vacuo*, the reaction mixture was subjected to GLC analysis. The following five columns were used; a 45 cm column packed with Silicone GE SE-30 (5%) on Shimalite W to analyze **1a**, **1**, **1d**, **4a**, **4b**, and **4d**; a 1 m column packed with Silicone GE SE-30 (5%) on Shimalite W to analyze **1e**, **4e**, **6**, and **8**; a 2 m column packed with Apiezon Grease L (30%) on Cerite 545 to analyze **6**, **7**, **8**, **9b**, **10a**, **10c**, and **10d**; a 2 m column packed with Thermon 1000+H_3PO_4 (5+0.5%) on Chromosorb W to analyze **9c**, and a 5 m column packed with polyethylene glycol 20 M (25%) on Shimalite 101 to analyze **9a** and **9d**.

For the isolation of the products, the thermal decompositions of **1** were carried out in a large scale. Typical procedure was as follows: A solution of **1c** (2.000 g, 5.15 mmol) in *o*-dichlorobenzene (40 cm³) was sealed in a degassed tube and heated at 200 °C for 7 h. The reaction mixture was washed with aq Na_2CO_3 and water. The alkaline solution was acidified with HCl to give **5c** (65 mg, 8%). The organic layer was dried over Na_2SO_4 and the solvent was evaporated *in vacuo*. The residue was chromatographed on silica gel with CH_2Cl_2 to give crude **4c**, **6**, and **7**. The crude **4c** was purified by recrystallization from ethanol (820 mg, 41%). The crude **6** and **7** were purified by preparative GLC.

Thermal Decomposition of **4a**. A solution of **4a** (343.4 mg, 1 mmol) in *o*-dichlorobenzene (10 cm³) was sealed in a deassed tube and then heated in a constant-temperature bath at 200 °C for 25 h. Biphenyl (60 mg) was added to the reaction mixture as an internal standard. The mixture was subjected to GLC analysis.

Preparation of N-t-*Butyl-2-(methylthio)benzamide (8)*. A solution of 2-(methylthio)benzoyl chloride (37.3 g, 200 mmol) in dry benzene (100 cm³) was added to a solution of *t*-butylamine (30.7 g, 420 mmol) in dry benzene (200 cm³) with stirring. The mixture was refluxed for 1 h, washed with dil HCl, aq $NaHCO_3$, and water, and then dried over Na_2SO_4. The benzene was evaporated, and the residue was recrystallized from benzene–petroleum ether giving **8** (38.1 g, 85%); mp 133.0—134.0 °C.

Preparation of N-t-*Butyl-2-(methylsulfinyl)benzamide (11)*. A solution of sodium metaperiodate (21.4 g, 100 mmol) in water (80 cm³) was added to a solution of *N-t*-butyl-2-(methylthio)benzamide (**8**) (22.3 g, 100 mmol) in methanol (200 cm³). The mixture was stirred for 3 h at room temperature. The reaction mixture was filtered and the filtrate was condensed to dryness. Water (50 cm³) and CH_2Cl_2 (150 cm³) was added to the residue, and the organic layer was separated. The extract was dried over Na_2SO_4 and the solvent was evaporated. The residue was recrystallized from CH_2Cl_2–petroleum ether to give **11** (22.1 g, 92%); mp 150.0—151.0 °C. Found: C, 59.93; H, 7.34; N, 5.78%. Calcd for $C_{12}H_{17}NO_2S$: C, 60.22; H, 7.16; N, 5.85%. IR (KBr): 3250 (N–H) and 1640 cm⁻¹ (C=O). NMR: ($CDCl_3$, δ) 1.48 (9H, s), 2.86 (3H, s), 6.57 (1H, broad s), and 7.4—8.2 (4H, m).

Reaction of **11** *with Acid Anhydrides*. A mixture of **11** (2.4 g, 10 mmol) and benzoic anhydride (2.7 g, 12 mmol) in *o*-dichlorobenzene (20 cm³) was refluxed for 10 h. The solvent was removed *in vacuo*, and the residue was dissolved in diethyl ether (50 cm³), washed with aq NH_3, Na_2CO_3, and water. The ether was evaporated after being dried over Na_2SO_4 and the residue was chromatographed on silica gel with CH_2Cl_2 as the eluant to give a mixture of **4a** and **6**. The mixture was recrystallized from CH_2Cl_2–petroleum ether giving **4a** (1.9 g, 55%).

The reaction of **11** with acetic anhydride was carried out in the same manner as mentioned above giving **4e** (1.7 g, 60%).

Reaction of **11** *with Acetyl Chloride*. Acetyl chloride (0.9 g, 10 mmol) was added to a solution of **11** (2.4 g, 10 mmol) in CH_2Cl_2 (20 cm³). The mixture was allowed to stand over night at room temperature, washed with aq NH_3 and water, and dried over Na_2SO_4. The solvent was removed and the residue was distilled under reduced pressure to give **6** (1.6 g, 77%); bp 165—166 °C/2.5 mmHg (lit,[9] bp 142 °C/0.5 mmHg). The distillate was recrystallized from petroleum ether; mp 57.0—58.0 °C. Found: C, 63.66; H, 6.55; N, 6.66%. Calcd for $C_{11}H_{13}NOS$: C, 63.74; H, 6.32; N, 6.76%. IR (KBr): 1650 cm⁻¹ (C=O). NMR (CCl_4, δ): 1.67 (9H, s) and 7.2—8.0 (4H, m).

Reaction of **11** *with Thionyl Chloride*. Thionyl chloride (1.8 g, 15 mmol) was added to a solution of **11** (2.4 g, 10 mmol) in CH_2Cl_2 (20 cm³). The mixture was allowed to stand for 1 h at room temperature, washed with aq Na_2CO_3 and water, and then dried over Na_2SO_4. The solvent was removed and the residue was distilled under reduced pressure to give **6** (1.6 g, 77%).

Preparation of 2-(Methylsulfinyl)benzoic Acid. Sodium metaperiodate (25.7 g, 120 mmol) in water (80 cm³) was added to a solution of 2-(methylthio)benzoic acid (20.0 g, 119 mmol) in methanol (250 cm³). The mixture was stirred for 4.5 h at room temperature and filtered. The filtrate was condensed to dryness and the residue was recrystallized from methanol–water to give 2-(methylsulfinyl)benzoic acid (18.4 g, 84%); mp 170.0—171.0 °C (dec) [lit,[10] mp 172 °C (dec)].

Preparation of 4H-*3,1-Benzoxathiin-4-one (7)*. A mixture of 2-(methylsulfinyl)benzoic acid (5.0 g, 27 mmol) and acetic anhydride (4.1 g, 40 mmol) was refluxed for 15 h. The excess acetic anhydride was removed by distillation. The residue was dissolved in diethyl ether and washed with aq $NaHCO_3$ and water. The solvent was removed after being dried over Na_2SO_4, and the residue was recrystallized from CCl_4–petroleum ether giving **7** (3.9 g, 87%); mp 47.0—48.0 °C (lit,[11] 47 °C). IR (KBr): 1720 cm⁻¹ (C=O). NMR (CCl_4, δ): 5.38 (2H, s), 7.1—7.5 (3H, m), and 8.0—8.3 (1H, m).

Preparation of N-t-*Butylbenzamide (10a)*. A solution of benzoyl chloride (14.0 g, 100 mmol) in dry benzene (50 cm³) was added to a solution of *t*-butylamine (18.0 g, 246 mmol) in dry benzene (150 cm³) with stirring. The reaction mixture was refluxed for 1 h, washed with aq NH_3, dil HCl, aq Na_2CO_3, and water, and then dried over Na_2SO_4. The solvent was removed, and the residue was recrystallized from CH_2Cl_2–petroleum ether giving **10a** (17.2 g, 97%); mp 134.0—135.0 °C (lit,[12] mp 134.0—134.5 °C).

By the same procedures. **10b—d** were prepared by the reactions of *t*-butylamine and the corresponding acyl chlorides. **10b**: mp 115.5—116.5 °C. **10c**: mp 159.0—160.0 °C. **10d**: mp 137.0—138.0 °C.

References

1) Part I of this series: Y. Uchida, Y. Hashimoto, and S. Kozuka. *Bull. Chem. Soc. Jpn.*, **53**, 2309 (1980).

2) P. G. Sammes, *J. Chem. Soc.*, **1965**, 6608; D. Sarantakis, J. K. Sutherland, C. Tortorella, and V. Tortorella, *J. Chem. Soc., C*, **1968**, 72.

3) S. Oae and T. Sakurai, *Bull. Chem. Soc. Jpn.*, **48**, 1075

(1975).
4) T. Ohta, K. Shudo, and T. Okamoto, *Tetrahedron Lett.*, **1978**, 1983.
5) H. Kise, G. F. Whitfield, and D. Swern, *J. Org. Chem.*, **37**, 1121 (1972); P. G. Gassman and R. J. Balchunis, *Tetrahedron Lett.*, **1977**, 2235.
6) T. Numata and S. Oae, *Chem. Ind. (London)*, **1972**, 726.
7) T. E. Varkey, G. E. Whitfield, and D. Swern, *J. Org. Chem.*, **39**, 3365 (1974); A. K. Sharma, T. Ku, A. D. Dawson, and D. Swern, *J. Org. Chem.*, **40**, 2758 (1975).
8) H. Kise, G. F. whitfield, and D. Swern, *J. Org. Chem.*, **37**, 1125 (1972).
9) J. S. Morley, Brit. Patent 848139 (1960); *Chem. Abstr.*, **55**, 9430f (1961).
10) A. Kucsman, I. Kapovits, and B. Tanacs, *Tetrahedron*, **18**, 79 (1962).
11) W. G. Bentrude and J. C. Martin, *J. Am. Chem. Soc.*, **84**, 1561 (1962).
12) J. J. Ritter and P. P. Minieri, *J. Am. Chem. Soc.*, **70**, 4045 (1948).