Electrochemical Synthesis of Semicyclic and Cyclic *N*-Acyl *N,O*-Acetals[1]

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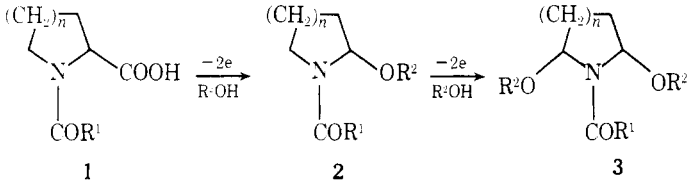
α-Amidoalkylation[2] which takes place via *N*-acylimmonium ion and/or *N*-acylimine[3,4] as transient intermediates has recently been reported to provide an elegant entry into amino acid or alkaloid syntheses. Of the reactions via linear *N*-acylimmonium ion, the introduction of amino acid skeletons is of current interest and has been demonstrated by us[5] and other workers.[6] The growing potential value of the semicyclic and cyclic *N*-acylimmonium ions has, most recently, been documented to allow nonenzymic biogenetic-type syntheses of several categories of alkaloids.[7] In spite of the copious literature dealing with the chemistry of semicyclic and cyclic *N*-acylimmonium ions, the general synthetic method of the precursors, *N*-acyl *N,O*-acetals, from which the *N*-acylimmonium ions can be generated invariably under Lewis acid catalyzed conditions, has received only minimum attention.

Semicyclic *N,O*-acetals have been synthesized mainly in connection with methods for biologically important nucleoside analogues[8] which contain a nitrogen atom in the hemiacetal ring. These semicyclic *N,O*-acetals have been prepared via acid-catalyzed skeletal rearrangement of 1,2-*O*-isopropylidene-5-acetamidotetrahydrofuran[9] and by electrochemical[10] or direct chemical oxidation[11] of *N*-acyl cyclic amines. These methods lack general acceptance because of the formation of complicated products and/or the tedious procedure in which the products are separated from the starting materials with difficulty. Methods for preparing cyclic *N,O*-acetals include cyclization of α-aminovinyl acetate,[12] photooxidation[13] or hydrogen peroxide oxidation[14] of pyrroles, ozonolysis of 4-pentenoamide,[15] and pH-controlled sodium borohydride reduction of succimides.[16]

We have previously reported that Kolbe-type electrochemical oxidation is effective for preparation of the linear *N*-acyl *N,O*-acetals such as α-methoxy-[17] or α-acetoxy-α-amino acid[18] derivatives. We now described a convenient synthesis of five- and six-membered semicyclic *N*-acyl *N,O*-acetals by anodic oxidation of *N*-acylprolines and *N*-acylpipecolic acids, respectively. This electrochemical method has been extended to a preparation of cyclic *N*-acyl *N,O*-acetals.

Semicyclic *N,O*-Acetals. Anodic oxidation of *N*-acylprolines and *N*-acylpipecolic acids was carried out at 10–15°C using graphite anode–graphite cathode in a nondivided cell. On electrolysis of *N*-formylprolines (**1a**) in methanol containing 1/40 molar equiv of sodium methoxide, *N*-formyl-2-methoxypyrrolidine (**2a**) was obtained in 96% yield. Other *N*-acylprolines such as *N*-acetyl- (**1b**), *N*-butyroyl- (**1c**), *N*-benzoyl- (**1d**), *N*-(ethoxycarbonyl)- (**1e**), *N*-(benzyloxycarbonyl)- (**1f**), and *N*-(benzyloxycarbonylglycyl)prolines (**1g**) were also electrolyzed to afford the corresponding semicyclic *N*-acyl *N,O*-acetals (**2b–g**) in 80–96% yields with quantitative current efficiency (Table I). A six-membered semicyclic *N*-acyl *N,O*-acetal (**2h**) was similarly prepared in good yield. The ethoxylation and isopropoxylation of *N*-acylprolines were also carried out to afford *N*-acyl-2-ethoxypyrrolidine (**2i**) and *N*-acyl-2-isopropoxypyrrolidine (**2j**), respectively. In the ethoxylation and isopropoxylation, however, the current efficiency was poor despite the good product yields. In addition, in order to obtain *N*-acyl-2-hydroxypyrrolidines or -piperidines, *N*-acylprolines or -pipecolic acids were electrolyzed in aqueous tetrahydrofuran containing 1/10 molar equiv of potassium hydroxide. In these electrolyses, the 2-hydroxy compounds (**2k–m**) were obtained in excellent yields. The presence of tetrahydrofuran[19] in this electrolysis system enabled the reactions to proceed smoothly with good current efficiency. Electrolysis in water or aqueous acetonitrile on the contrary led to greatly decreased current efficiency, and the electrolyzed solution became brownish, presumably because of the concurrent oxidation of the electrolysis products, 2-hydroxy compounds. In case of hydroxylation of *N*-acylprolines having a bulky substituent at the 2 positions, hydrolysis of the intermediates, 2-hydroxy derivatives, occurred; e.g., the

Table I. Electrolysis Products 2 and 3[a]



1 2 3

compd	*n*	R^1	R^2	bp (mmHg), °C	yield, %
2a	1	H	CH_3	135–137 (48)	96
2b	1	CH_3	CH_3	61–62 (8)	95
2c	1	n-C_3H_7	CH_3	109–110 (5)	94
2d	1	Ph	CH_3	103–104 (0.3)	86
2e	1	OC_2H_5	CH_3	65–67 (4)	94
2f	1	OCH_2Ph	CH_3	106–108 (0.4)	93
2g	1	$CH_2NHCOOCH_2Ph$	CH_3	syrup	80
2h	2	OCH_2Ph	CH_3	125–127 (0.6)	88
2i	1	Ph	C_2H_5	114–116 (0.2)	80
2j	1	OC_2H_5	i-C_3H_9	95–96 (3.5)	75
2k	1	H	H	122–125 (1.5)	86
2l	1	OC_2H_5	H	71–73 (0.35)	83
2m	2	OCH_2Ph	H	syrup	93
3	1	OC_2H_5	CH_3	66–67 (0.5)	98

[a] Satisfactory elemental analyses were obtained for compounds **2f, 2m,** and **3.** The elemental analyses of the other compounds showed the following limit of error due to their hygroscopic or volatile properties: C, ±1.50%; H, ±0.41%; N, ±0.38%.

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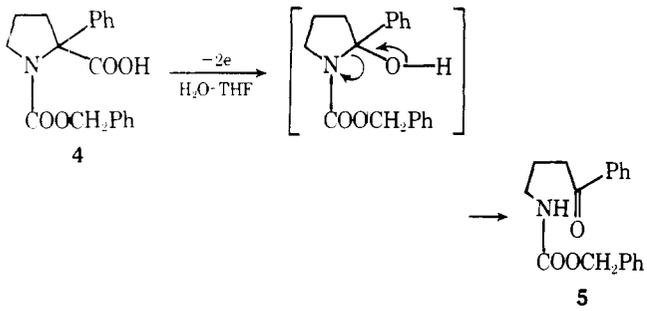
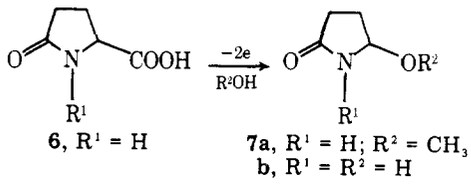
Scheme I



Scheme III



electrolysis of *N*-(benzyloxycarbonyl)-2-phenylproline (**4**) afforded the amino ketone **5** in 81% yield (Scheme I).[20]

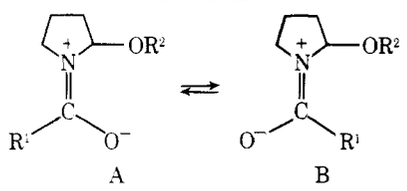
In order to obtain a dimethoxylated compound, the semicyclic *N*-acyl *N,O*-acetals obtained above have been further oxidized in methanol (Table I). Electrolysis of *N*-(ethoxycarbonyl)-2-methoxypyrrolidine (**2e**) in methanol–tetraethylammonium tosylate[10a] with 4 times the theoretical amount of current gave a mixture of *cis*- and *trans*-*N*-(ethoxycarbonyl)-2,5-dimethoxypyrrolidine (**3**) in quantitative yield; *N*-(ethoxycarbonyl)-2,2-dimethoxypyrrolidine was not detected. The exclusive formation of the 2,5-disubstituted pyrrolidine suggests that the positive charge density of the cation radical generated by an electron transfer from the substrate[10a] would be greater on the methylene group at the C-5 position than on the methine group at the C-2 position.[21]

The yields and boiling points of the semicyclic *N,O*-acetals obtained here are summarized in Table I.

Ring–chain tautomerism is frequently observed in cyclic hemiacetals, especially in carbohydrate chemistry.[22] The 2-hydroxy compounds **2k–m** obtained here did not show ring–chain tautomerism with the open-chain acylamino aldehydes. Proof of the cyclic structure was provided by the NMR spectra in $CDCl_3$, in which no signal of the aldehyde protons was observed. However, NMR spectra of the compounds possessing a bulky *N*-acyl group[23] such as the *N*-benzoyl group clearly gave an indication of the ring–chain tautomerism.

NMR spectroscopy of the semicyclic *N*-acyl *N,O*-acetals described above exhibited an interesting aspect due to the hindered rotation about the amide bonds.[24] Two rotational isomers, A and B (Scheme II), were observed in $CDCl_3$ at ambient probe temperature.[9,25] The conformational assignment of the isomers, A and B, was carried out on the basis of the relative difference of the chemical shifts of the C-2 protons in the NMR spectra. In 2-substituted *N*-acylpiperidines or -pyrrolidines,[25a,26] the C-2 proton cis to the carbonyl oxygen of the amido group is deshielded relative to the proton trans to the carbonyl oxygen due to the anisotropic effect[27] of the carbonyl bond. For example, *N*-acetyl-2-methoxypyrrolidine (**2b**) contains two multiplets at 5.00 and 5.45 ppm in the region characteristic of the C-2 protons in B and A, respectively. The resonances due to the methoxyl protons of B and A were observed as two singlets at 3.32 and 3.34 ppm, respectively. In addition, there are two singlets at 2.11 and 2.19 ppm which are assigned to be the *N*-acetyl protons of A and B, respectively. The ratio of A/B in *N*-acetyl-2-methoxypyrrolidine was determined to be 4.3:5.7 by integration of the spectrum. The conformational equilibrium[28] of the two isomers, A and B, lies more to the left with increasing bulkiness of R^1 groups; e.g., the ratios of A/B in compounds **2a** (R^1 = H) and **2c** (R^1 = n-C_3H_7) are 2.0:8.0 and 5.3:4.7, respectively.

Scheme II



Cyclic *N*-Acyl *N,O*-Acetals. This electrochemical method was further applied to the preparation of cyclic *N*-acyl *N,O*-acetals, which results in highly reactive cyclic *N*-acylimmonium ions under Lewis acid catalyzed conditions. Compound **6** (Scheme III) was electrolyzed in methanol under the same conditions as above to afford the corresponding cyclic *N*-acyl *N,O*-acetal **7a** in quantitative yield and nearly 100% current efficiency. The anodic oxidation of compound **6** was also carried out in aqueous tetrahydrofuran to afford compound **7b**. The NMR spectrum of **7b** in Me_2SO-d_6 supported the cyclic structure and excluded the open-chain one.[24] These cyclic *N*-acyl *N,O*-acetals are unstable to heat, and accordingly the electrolysis as well as the workup procedure should be carried out carefully at least below 20 °C.

The application of this electrochemical method are currently under investigation.

Experimental Section

Equipment. Melting points were measured using a Yamato melting point apparatus and were uncorrected. IR spectra were recorded on a Shimadzu IR-27G infrared spectrometer. NMR spectra were obtained using a Hitachi Perkin-Elmer R-20 high-resolution NMR spectrometer with tetramethylsilane as an internal standard. Electrolyses were carried out using a Hokuto 104 (1A-55V) or PGS 2500 (2.5A-60V) attached to a Hokuto HA 108A coulometer.

Starting Materials. *N*-Acyl-(*S*)-prolines and *N*-acyl-(*R*)-pipecolic acids were prepared from (*S*)-proline and (*R*)-pipecolic acid, respectively, by the usual Schotten–Baumann procedure. *N*-(Benzyloxycarbonylglycyl)proline (**1g**) was synthesized by the condensation of *N*-(benzyloxycarbonyl)glycine and proline ethyl ester with dicyclohexylcarbodiimide, followed by saponification with potassium hydroxide. The physical constants of the *N*-acyl compounds are described elsewhere.[29]

General Electrolysis Procedure. Alkoxylation was carried out on 0.1-mol scale in the same manner as that described in the previous reports.[17,18] In ethoxylation and isopropoxylation, 1.5 times the theoretical amount of current was passed. In addition, hydroxylation was carried out as follows. The substrate 1 (0.1 mol) was dissolved in 10 times the quantity of aqueous tetrahydrofuran (water/tetrahydrofuran = 3:1) containing 0.01 mol of potassium hydroxide. The electrolysis was carried out at a constant current of 200 mA/cm^2 and discontinued when 2 faraday/mol of electricity was passed. The electrolyzed solution was evaporated to dryness in vacuo below 20 °C. The resulting residue was dissolved in ethyl acetate, washed with water, dried over magnesium sulfate, and then evaporated to dryness in vacuo to afford the crude hydroxylated product. All alkoxylated and hydroxylated products were purified[30] by distillation under reduced pressure.

The yields and boiling points of the electrolysis products are listed in Table I.

Preparation of Compound 3. Compound **2e** (17.3 g, 0.1 mol) was dissolved in 200 mL of methanol containing 1 g of tetraethylammonium tosylate. The electrolysis was carried out at a constant current of 2.5 A with 4 times the theoretical amount of current. The electrolyzed solution was evaporated to dryness in vacuo, and the resulting residue was dissolved in ethyl acetate. The solution was washed once with water, dried over magnesium sulfate, and then evaporated to dryness in vacuo. The resulting syrup was purified by distillation under reduced pressure to afford 19.9 g (98%) of compound **3**. The boiling point is shown in Table I. The ratio of cis/trans is approximately 1.

Electrolysis of *N*-(Benzyloxycarbonyl)-2-phenylproline (4). The proline (1.63 g, 5 mmol) was electrolyzed in 20 mL of aqueous tetrahydrofuran (water/tetrahydrofuran = 3:1) containing 0.25 mL of 1 N potassium hydroxide at 8–10 °C at a constant current of 0.5 A. The electrolysis was discontinued when the theoretical amount of current was passed. The electrolyzed solution was concentrated to about 15 mL under reduced pressure. The solution was shaken with two 50-mL portions of ethyl acetate. The combined ethyl acetate layers were dried over magnesium sulfate and then evaporated to dryness in vacuo to afford 1.2 g (81%) of colorless crystals of 4-(benzyloxycarbonylamino)butyrophenone (**5**). Recrystallization from ethyl acetate–hexane gave the pure compound: mp 86–87 °C; IR (Nujol) 3350, 1710, 1670 cm^{-1}; NMR ($CDCl_3$) δ 1.8–2.3 (m, 2 H), 2.9–3.5 (m, 4 H), 4.8–5.2 (m, 1 H), 5.10 (s, 2 H), 7.30 (s, 5 H), 7.1–8.1 (m, 5 H). Anal. Calcd for $C_{18}H_{19}O_3N$: C, 72.70; H, 6.44; N, 4.71. Found: C, 72.23; H, 6.43; N, 4.64.

Compound 7a. Compound **6** (5.16 g, 0.04 mol) was electrolyzed according to the general electrolysis procedure. The electrolyzed solution was evaporated to dryness in vacuo below 30 °C. The residue was extracted with ethyl acetate, and the solution was treated with activated charcoal and filtered. The filtrate was evaporated to dryness in vacuo below 30 °C to afford 4.5 g (98%) of compound **7a**, which was recrystallized from ethyl acetate–hexane: mp 59–60.5 °C; IR (Nujol) 3180, 1710–1660 (broad) cm^{-1}; NMR ($CDCl_3$) δ 1.8–2.6 (m, 4 H), 3.30 (s, 3 H), 4.8–5.0 (m, 1 H), 8.70 (broad s, 1 H). Anal. Calcd for $C_5H_9O_2N$: C, 52.16; H, 7.88; N, 12.17. Found: C, 52.17; H, 7.89; N, 12.21.

Compound 7b. Compound **6** (5.16 g, 0.04 mol) was oxidized in aqueous tetrahydrofuran. After the theoretical amount of current was passed, the electrolyzed solution was evaporated to dryness in vacuo below 30 °C. The residue was dissolved in a mixture of acetonitrile (100 mL) and tetrahydrofuran (100 mL). The solution was dried over magnesium sulfate and then evaporated to dryness in vacuo below 30 °C. The resulting syrup (4.2 g) was washed with ether and ethyl acetate. The resulting crystals were recrystallized from ethanol–tetrahydrofuran to afford 0.5 g (13%) of compound **7b**: mp 146–148 °C (prism); IR (Nujol) 3180, 3110, 1690 cm^{-1}; NMR (Me_2SO-d_6) δ 1.5–2.6 (m, 4 H), 3.4 (broad s, 1 H), 4.9–5.2 (m, 1 H), 8.56 (broad s, 1 H). Anal. Calcd for $C_4H_7NO_2$: C, 47.52; H, 6.98; N, 13.86. Found: C, 47.66; H, 7.11; N, 13.59. From the filtrate of the recrystallized solution was recovered 0.8 g of the starting material.

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Registry No.—**1a**, 13200-83-4; **1b**, 68-95-1; **1c**, 23500-13-2; **1d**, 5874-58-8; **1e**, 5700-74-3; **1f**, 69352-26-7; **1g**, 1160-54-9; **1h**, 28697-09-8; **2a**, 61020-06-2; **2b**, 63050-21-5; **2c**, 68471-61-4; **2d**, 69001-12-3; **2e**, 69352-20-1; **2f**, 69352-21-2; **2g**, 69352-22-3; **2h**, 66893-75-2; **2i**, 69001-13-4; **2j**, 69352-23-4; **2k**, 69352-24-5; **2l**, 69352-25-6; **2m**, 69622-67-9; *cis*-**3**, 69352-27-8; *trans*-**3**, 69352-28-9; **4**, 69352-29-0; **5**, 69352-30-3; **6**, 98-79-3; **7a**, 63853-74-7; **7b**, 62312-55-4; (*S*)-proline, 147-85-3; (*R*)-pipecolic acid, 1723-00-8; *N*-(benzyloxycarbonyl)glycine, 1138-80-3; proline ethyl ester, 5817-26-5.

Supplementary Material Available: IR, NMR, and mass spectral data for compounds **2a–m** and **3** (4 pages). Ordering information is given on any current masthead page.

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Electrosynthesis of Hetero–Hetero Atom Bonds. 3. Sodium Bromide Promoted Electrolytic Cross-Coupling Reaction of Imides with Disulfides. Convenient Synthesis of *N*-(Cyclohexylthio)phthalimide, an Important Prevulcanization Inhibitor

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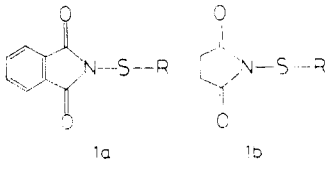
In our preceding paper, we reported the synthesis of 2-benzothiazolyl- and thiocarbamylsulfenamides from the corresponding disulfides and amines by electrolytic cross-coupling.[1] However, extension to the preparation of thiophthalimides (**1a**) and thiosuccinimides (**1b**) (sulfenimides)[2] by cross-coupling of imides **2** with disulfides **3** under similar conditions gave only ~20% yields of **1** even after passage of 5–10 equiv of electricity. This result can be ascribed to lack of an equilibrium reaction between imides **2**–disulfides **3** and sulfenimides **1**.[1] In an effort to find a more suitable electrochemical procedure for the cross-coupling of **2** and **3**,[3]



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