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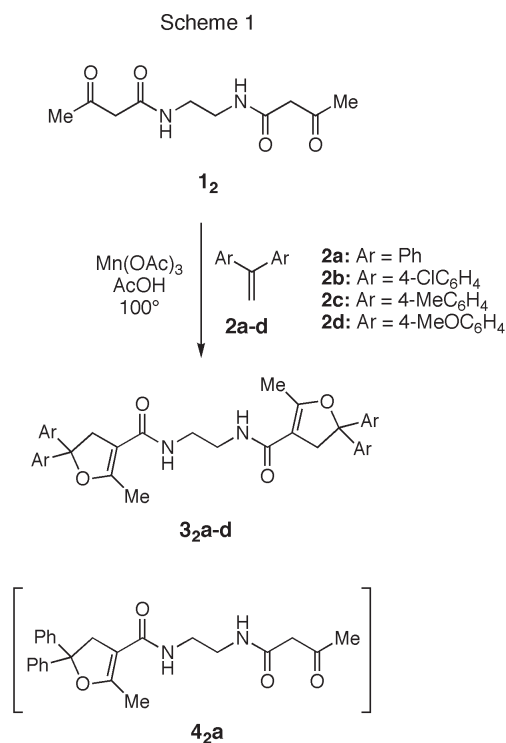
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The reaction of *N,N'*-oligomethylenebis(3-oxobutanamide)s with 1,1-diarylethenes in the presence of manganese(III) acetate in acetic acid at 100° produced *N,N'*-oligomethylenebis(2-methyl-5,5-diaryl-4,5-dihydrofuran-3-carboxamide)s. Similarly, the reaction of 3-oxobutanamidoethyl 3-oxobutanoate or *N,N'*-(3,6-dioxaoctamethylene)bis(3-oxobutanamide) with 1,1-diphenylethene gave (2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-amido)ethyl 2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxylate or *N,N'*-(3,6-dioxaoctamethylene)bis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) in moderate yields.

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Introduction.

During the past decades, manganese(III)-based oxidative free-radical cyclizations have been developed into a versatile protocol for the formation of highly functionalized products from simple precursors. Manganese(III) acetate is well-known as a mild one-electron oxidant which leads to formal dicarbonylmethyl radicals, $\bullet\text{CH}(\text{COR})\text{COR}'$, from 1,3-dicarbonyl compounds [1-11]. Reactions involving such species with alkenes or alkynes have attracted considerable attention. The works of Heiba [12,13], Fristad [14-16], Corey [17-20], Snider [7], and others [21-33] are noteworthy, since they provided mechanistic descriptions and synthetic applications of this chemistry that include inter- and intra-molecular cyclization processes. In our previous papers [34-45], we have described that the reactions of 1,3-dicarbonyl compounds with monoalkenes and terminal dienes in the presence of tris(2,4-pentanedionato)manganese(III), manganese(III) acetate, or tris(2,4-pentanedionato)cobalt(III) at elevated temperature produced dihydrofurans in good yields. Heiba and Dessau reported that the reaction of alkenes with β -diketones and β -keto esters in the presence of manganese(III) acetate also gave the corresponding dihydrofurans [13]. Corey applied these methods to enol ethers and obtained 2-alkoxy-2,3-dihydrofurans which were transformed into fused or spiro 2-cyclopentenones or converted into the corresponding furans in good yields [19,20]. The reaction of 1,3-dicarbonyl compounds with alkenes in the presence of thallium(III) acetate [46], lead(IV) acetate [47], silver oxide, and lead(IV) oxide [48] have also been reported to give



dihydrofurans. It was expected that *N,N'*-oligomethylenebis(3-oxobutanamide)s would be oxidized by manganese(III) acetate *via* a manner similar to the oxidation of 1,3-dicarbonyl compounds to give the corresponding formal biradicals, which would attack alkenic double bonds to produce the corresponding bis(dihydrofuran)s. Thus, we examined the reaction of various *N,N'*-oligomethylenebis(3-oxobutanamide)s with 1,1-diaryl-

ethenes in the presence of manganese(III) acetate, and found that they provided a convenient route for the synthesis of bis(dihydrofuran)s. The results of the reactions and the reaction mechanism are presented in this paper.

Table 1

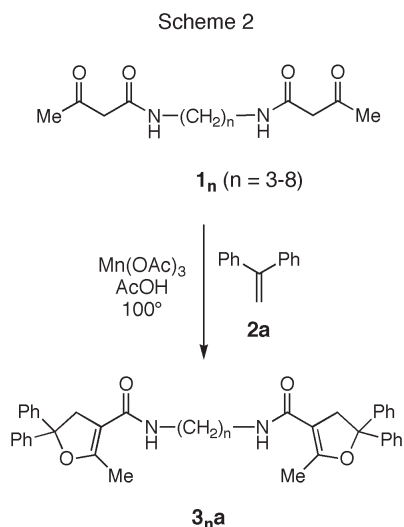
Reaction of *N,N'*-Ethylenebis(3-oxobutanamide) (**1₂**) with 1,1-Diarylethenes **2a-d** in the Presence of Manganese(III) Acetate in Acetic Acid at 100° [a]

| Entry | Diamine | Alkene | 1₂ : 2 :Mn(III) | Time min | Product Yield/% [b] |
|-------|-----------------------|-----------|--|----------|--|
| 1 | 1₂ | 2a | 2:1:2.5 | 5 | 3_{2a} (8) 4_{2a} (31) |
| 2 | 1₂ | 2a | 0.5:1:3 | 23 | 3_{2a} (34) |
| 3 | 1₂ | 2a | 0.5:1:3.5 | 20 | 3_{2a} (38) |
| 4 | 1₂ | 2a | 0.5:1.2:4 | 25 | 3_{2a} (45) |
| 5 | 1₂ | 2a | 0.5:1.2:4 [c] | 30 | 3_{2a} (30) |
| 6 | 1₂ | 2b | 0.5:1.2:4 | 20 | 3_{2b} (37) |
| 7 | 1₂ | 2c | 0.5:1.2:4 | 16 | 3_{2c} (52) |
| 8 | 1₂ | 2d | 0.5:1.2:4 | 10 | 3_{2d} (31) |
| 9 | 4_{2a} | 2a | 0.5:0.6:2 | 15 | 3_{2a} (63) |

[a] The reactions were carried out in acetic acid (30 mL) at 100° under an argon atmosphere. [b] The yields were based on the amount of **1₂** used except for entries 1 and 9 of which the yields were based on **2a** and **4_{2a}**, respectively. [c] The reaction was conducted in air.

Results and Discussion.

Reaction of *N,N'*-Ethylenebis(3-oxobutanamide) **1₂** with 1,1-Diarylethenes **2a-d** in the Presence of Manganese(III) Acetate at 100°.



When the reaction of **1₂** with **2a** was carried out in the presence of manganese(III) acetate under an argon atmosphere at 100°, the major products were found to be *N,N'*-ethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{2a}**) and *N*-(3-oxobutanamidoethyl)-2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide

Table 2

Reaction of *N,N'*-Oligomethylenebis(3-oxobutanamide)s **1_n** (n = 3-8) with 1,1-Diphenylethene (**2a**) in the Presence of Manganese(III) Acetate in Acetic Acid at 100° [a]

| Entry | Diamine | Alkene | 1_n : 2a :Mn(III) | Time min | Product Yield/% [b] |
|-------|----------------------|-----------|---|----------|----------------------------|
| 1 | 1₃ | 2a | 0.5:1.2:4 | 20 | 3_{3a} (43) |
| 2 | 1₄ | 2a | 0.5:1.2:4 | 20 | 3_{4a} (40) |
| 3 | 1₅ | 2a | 0.5:1.2:4 | 14 | 3_{5a} (35) |
| 4 | 1₆ | 2a | 0.5:1.2:4 | 16 | 3_{6a} (37) |
| 5 | 1₇ | 2a | 0.5:1.2:4 | 20 | 3_{7a} (35) |
| 6 | 1₈ | 2a | 0.5:1.2:4 | 23 | 3_{8a} (32) |

[a] The reactions were carried out in acetic acid (30 mL) at 100° under an argon atmosphere. [b] The yields were based on the amount of **1_n** used.

(**4_{2a}**), together with a small amount of benzophenone (Scheme 1). The yields also significantly varied depending on the molar ratio of **1₂**:**2a**:manganese(III) acetate (Table 1).

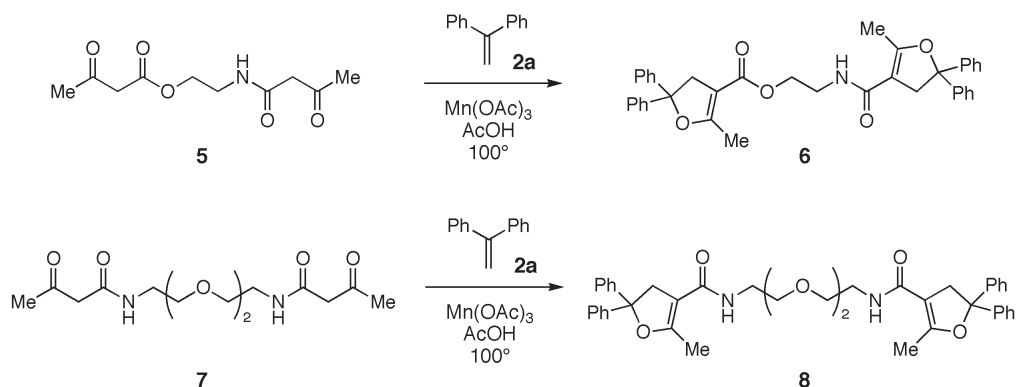
In the reaction of **1₂** with **2a** at a higher molar ratio, the mono(dihydrofuran) **4_{2a}** was the predominant product (Table 1, Entry 1). The expected bis(dihydrofuran) **3_{2a}**, on the other hand, was optimized in the reaction at a molar ratio of 0.5:1.2:4 for **1₂**:**2a**:manganese(III) acetate up to a 45% yield (Entry 4). Similar results for the synthesis of the bis(dihydrofuran)s were obtained by the reaction of **1₂** with 1,1-diarylethenes **2b-d** in the presence of manganese(III) acetate, and the yields are listed in Table 1 (Entries 6-8). The best yield of the bis(dihydrofuran) **3₂** was achieved for the reaction of **1c** having a methyl group on the aromatic ring with **1₂** (Entry 7). The presence of a remaining active methylene in compound **4_{2a}** indicated the possibility of forming another 4,5-dihydrofuran ring by the reaction of the alkene in the presence of manganese(III) acetate. The yield of **3_{2a}** was increased up to 63% when a mixture of **4_{2a}** and **2a** was allowed to react with manganese(III) acetate in acetic acid at 100° (Entry 9).

Reaction of *N,N'*-Oligomethylenebis(3-oxobutanamide)s **1_n** (n = 3-8) with 1,1-Diphenyl-ethene (**2a**) in the Presence of Manganese(III) Acetate in Acetic Acid at 100°.

A similar reaction with **1_n** (n = 3-8) was applied to **2a** using manganese(III) acetate (Scheme 2), and the yields are listed in Table 2 (Entries 1-6). It was found that the longer the methylene chain length of **1_n**, the lower the yield of **3_na**. Similar reactions of 3-oxobutanamidoethyl 3-oxobutanoate **5** and *N,N'*-(3,6-dioxaoctamethylene)-bis(3-oxobutanamide) **7** with **2a** were carried out, and the corresponding bis(dihydrofuran)s **6** and **8** were obtained in 61% and 33% yields, respectively (Scheme 3).

It was found that the replacement of one nitrogen atom by oxygen in the case of **5** increased the yield of bis(dihydrofuran). On the other hand, the yields of bis(dihydrofuran)s **3** and **8** were medium or low in both cases since the

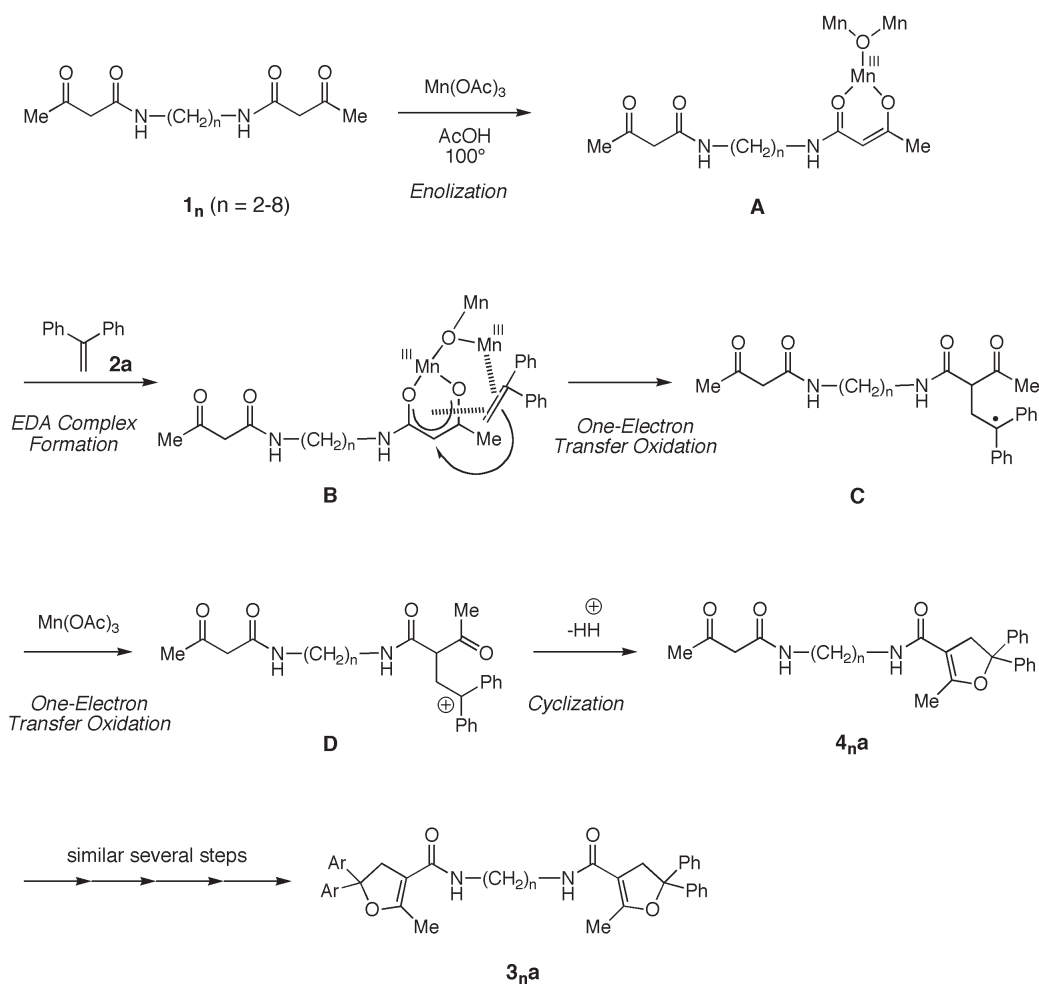
Scheme 3



amido hydrogen might be sensitive to the manganese(III) oxidant under these conditions [6,8,39,45,49]. In order to synthesize the compounds bearing two 1,2-dioxane skeletons [36], a mixture of *N,N'*-ethylenebis(3-oxobutanamide)

1₂ and **2a** was oxidized with manganese(III) acetate under a dry air stream at 23° , but our attempt failed. After work-up, benzophenone (12%) and a complex mixture were obtained.

Scheme 4



Reaction Pathway.

The formation of bis(dihydrofuran)s **3_na** could be explained on the basis of the mechanism outlined in Scheme 4. It seemed that the formation of a new complex **A** initiated the present reaction according to a similar mechanism for the reaction with the 1,3-dicarbonyl compounds [50-53]. We failed to isolate complex **A** from the solution, however, manganese(III) acetate readily reacted with 4,4,4-trifluoro-1-phenyl-1,3-butanedione to give tris(4,4,4-trifluoro-1-phenyl-1,3-butanedionato)-manganese(III) which was isolated and characterized [52]. Several alkenes were oxidized with the complex at reflux temperature to give the corresponding 3-benzoyl-2-trifluoromethyl-4,5-dihydrofurans [52]. Based on the isolated products, it was strongly suggested that complex **A** should be formed during the first stage. The weak interaction between complex **A** and alkene **2a**, such as an electron donor-acceptor (EDA) complex **B** [9], would induce a concerted one-electron transfer from the alkene to metal center *via* the coordinated 1,3-dicarbonyl ligand to form the tertiary radical **C** during the second stage [7,9,11,15,16,22]. Under the reaction conditions, further oxidation occurred and the mono(dihydrofuran) **4_na** was obtained *via* cyclization of the tertiary cation **D**. A similar reaction of **4_na** with manganese(III) acetate was repeated to produce bis(dihydrofuran) **3_na**.

Conclusion.

A simple one-step synthesis of bis(dihydrofuran)s was achieved by the reaction of *N,N'*-oligomethylenebis(3-oxobutanamide)s **1_n** and **7** with 1,1-diarylethenes **2** in the presence of manganese(III) acetate. Butanamidoethyl 3-oxobutanoate **5** has a higher selectivity for the formation of bis(dihydrofuran) than **1_n** and **7**. An increase in the methylene chain length of **1_n** does not produce any dramatic changes in the yields of the bis(4,5-dihydrofuran-3-carboxamide)s **3_n**, however, the present method would be convenient and important since bis(dihydrofuran)s could be converted into the corresponding synthons of the 1,3-dicarbonyl compounds [54] and bis(tetrahydrofuran)s which construct part of the structure of many natural products.

EXPERIMENTAL

Measurements.

All of the nmr spectra were recorded using a JNM-AL 300 FT nmr spectrometer at 300 MHz for ¹H and at 75 MHz for ¹³C with tetramethylsilane as the internal standard. The chemical shifts are shown in δ₋ and coupling constants in Hz. The IR spectra were measured using a Paragon 1000 FT ir spectrometer. The IR spectral data are expressed in cm⁻¹. All of the melting points were determined using a Yanaco micromelting-point apparatus MP-J3 and are uncorrected. Elemental analysis was performed by the Center of Instrumental Analysis, Kumamoto University, Japan.

Materials.

Manganese(II) acetate tetrahydrate, Mn(OAc)₂•4H₂O, was purchased from Wako Pure Chemical Ind., Ltd. Manganese(III) acetate dihydrate, Mn(OAc)₃•2H₂O, was prepared according to the method described in the literature [55]. 1,1-Diarylethenes **2a-d** were prepared by dehydration of the corresponding alcohols which were synthesized from substituted acetophenones and arylmagnesium bromides.

Preparation of *N,N'*-Oligomethylenebis(3-oxobutanamide) Derivatives **1_n** (n = 2-8), **5**, and **7** [56-58].

Ethylenediamine (4g; 0.06 mmol) was placed in a 100 mL flask equipped with a magnetic stirrer and a reflux condenser. Ethanol (50 mL) was added to the flask, which was placed in an ice bath, and the mixture was stirred. Diketene (9.2 mL; 0.12 mmol) was added dropwise with continuous stirring, and the reaction mixture was allowed to stand for a few minutes at room temperature. Needle-like crystals were immediately formed. The crude crystals were then collected by filtration and washed with cold ethanol. The crystals were then recrystallized from ethanol. *N,N'*-Oligomethylenebis(3-oxobutanamide)s **1_n** (n = 2-8) and *N,N'*-(3,6-dioxaoctamethylene)bis(3-oxobutanamide) (**7**) were prepared using the corresponding oligomethylenediamines and 1,2-bis(2-aminoethoxy)ethane according to a method similar to that mentioned above. However, in the case of 3-oxobutanamidoethyl 3-oxobutanoate (**5**), acetonitrile instead of ethanol was used as the solvent, and the reaction of 2-aminoethanol with diketene was carried out at reflux temperature, producing **5** as a liquid. The physical data of the compounds **1_n** (n = 2-8), **5**, and **7** are given below.

N,N'-Ethylenebis(3-oxobutanamide) (**1₂**).

This compound was obtained as colorless microcrystals (from EtOH), mp 165°; ir (KBr) 3600-3100 (NH), 1730, 1645 (C=O); ¹H nmr (DMSO-*d*₆): δ 8.08 (2H, br t, *J* = 6.16, NH x 2), 3.28 (4H, s, CH₂ x 2), 3.13 (4H, m, NHCH₂ x 2), 2.14 (6H, s, CH₃ x 2); ¹³C NMR (DMSO-*d*₆): δ 202.9 (C=O x 2), 166.2 (CON x 2), 51.2 (CH₂ x 2), 38.3 (CH₂ x 2), 29.9 (CH₃ x 2).

Anal. Calcd. for C₁₀H₁₆N₂O₄: C, 52.62; H, 7.07; N, 12.27. Found: C, 52.58; H, 7.24; N, 12.30.

N,N'-Trimethylenebis(3-oxobutanamide) (**1₃**).

This compound was obtained as colorless microcrystals (from EtOH), mp 138°; ir (KBr) 3600-3100 (NH), 1730, 1643 (C=O); ¹H nmr (DMSO-*d*₆) 8.02 (2H, br t, *J* = 6.16, NH x 2), 3.29 (4H, s, CH₂ x 2), 3.08 (4H, m, NHCH₂ x 2), 2.14 (6H, s, CH₃ x 2), 1.54 (2H, m, -CH₂CH₂CH₂-); ¹³C nmr (DMSO-*d*₆) 203.0 (C=O x 2), 165.9 (CON x 2), 51.2 (CH₂ x 2), 36.4 (CH₂ x 2), 29.9 (CH₃ x 2), 28.9 (CH₂).

Anal. Calcd. for C₁₁H₁₈N₂O₄: C, 54.53; H, 7.49; N, 11.56. Found: C, 54.27; H, 7.72; N, 11.54.

N,N'-Tetramethylenebis(3-oxobutanamide) (**1₄**).

This compound was obtained as colorless microcrystals (from EtOH), mp 145°; ir (KBr) 3600-3102 (NH), 1730, 1650 (C=O); ¹H nmr (DMSO-*d*₆) 8.03 (2H, br t, *J* = 6.16, NH x 2), 3.28 (4H, s, CH₂ x 2), 3.06 (4H, m, NHCH₂ x 2), 2.14 (6H, s, CH₃ x 2), 1.52-1.30 (4H, m, CH₂ x 2); ¹³C nmr (DMSO-*d*₆) 203.0 (C=O x 2), 165.8 (CON x 2), 51.3 (CH₂ x 2), 38.2 (CH₂ x 2), 29.9 (CH₃ x 2), 26.4 (CH₂ x 2).

Anal. Calcd. for C₁₂H₂₀N₂O₄: C, 56.23; H, 7.87; N, 10.93. Found: C, 56.05; H, 8.09; N, 11.00.

N,N'-Pentamethylenebis(3-oxobutanamide) (**1₅**).

This compound was obtained as colorless microcrystals (from EtOH), mp 130°; ir (KBr) 3600-3104 (NH), 1729, 1646 (C=O); ¹H nmr (DMSO-*d*₆) 8.00 (2H, br t, *J* = 6.80, NH x 2), 3.28 (4H, s, CH₂ x 2), 3.04 (4H, m, NHCH₂ x 2), 2.13 (6H, s, CH₃ x 2), 1.56-1.14 (6H, m, CH₂ x 3); ¹³C nmr (DMSO-*d*₆) 203.0 (C=O x 2), 165.7 (CON x 2), 51.3 (CH₂ x 2), 38.4 (CH₂ x 2), 29.9 (CH₃ x 2), 28.5 (CH₂ x 2), 23.6 (CH₂).

Anal. Calcd. for C₁₃H₂₂N₂O₄: C, 57.76; H, 8.20; N, 10.36. Found: C, 57.64; H, 8.56; N, 10.37.

N,N'-Hexamethylenebis(3-oxobutanamide) (**1₆**).

This compound was obtained as colorless microcrystals (from EtOH), mp 141°; ir (KBr) 3500-3100 (NH), 1729, 1650 (C=O); ¹H nmr (DMSO-*d*₆) 8.01 (2H, br t, *J* = 6.71, NH x 2), 3.28 (4H, s, CH₂ x 2), 3.05 (4H, m, NHCH₂ x 2), 2.14 (6H, s, CH₃ x 2), 1.56-1.15 (8H, m, CH₂ x 4); ¹³C nmr (DMSO-*d*₆) 203.0 (C=O x 2), 165.8 (CON x 2), 51.3 (CH₂ x 2), 38.5 (CH₂ x 2), 29.8 (CH₃ x 2), 28.9 (CH₂ x 2), 26.0 (CH₂ x 2).

Anal. Calcd. for C₁₄H₂₄N₂O₄: C, 59.13; H, 8.51; N, 9.85. Found: C, 59.07; H, 8.59; N, 9.97.

N,N'-Heptamethylenebis(3-oxobutanamide) (**1₇**).

This compound was obtained as colorless microcrystals (from EtOH), mp 138°; ir (KBr) 3500-3100 (NH), 1729, 1649 (C=O); ¹H nmr (CDCl₃) 7.03 (2H, br t, *J* = 6.71, NH x 2), 3.41 (4H, s, CH₂ x 2), 3.05 (4H, m, NHCH₂ x 2), 2.27 (6H, s, CH₃ x 2), 1.53-1.32 (10H, m, CH₂ x 5); ¹³C nmr (CDCl₃) 204.9 (C=O x 2), 165.4 (CON x 2), 49.6 (CH₂ x 2), 39.3 (CH₂ x 2), 31.0 (CH₃ x 2), 29.1 (CH₂ x 2), 28.5 (CH₂), 26.5 (CH₂ x 2).

Anal. Calcd. for C₁₅H₂₆N₂O₄: C, 60.38; H, 8.78; N, 9.39. Found: C, 60.29; H, 9.15; N, 9.36.

N,N'-Octamethylenebis(3-oxobutanamide) (**1₈**).

This compound was obtained as colorless microcrystals (from EtOH), mp 144°; ir (KBr) 3600-3100 (NH), 1729, 1644 (C=O); ¹H nmr (DMSO-*d*₆) 8.00 (2H, br t, *J* = 6.71, NH x 2), 3.27 (4H, s, CH₂ x 2), 3.05 (4H, m, NHCH₂ x 2), 2.13 (6H, s, CH₃ x 2), 1.39-1.25 (12H, m, CH₂ x 6); ¹³C NMR (DMSO-*d*₆) 203.0 (C=O x 2), 165.7 (CON x 2), 51.3 (CH₂ x 2), 38.5 (CH₂ x 2), 29.8 (CH₃ x 2), 28.9 (CH₂ x 2), 28.6 (CH₂ x 2), 26.2 (CH₂ x 2).

Anal. Calcd. for C₁₆H₂₈N₂O₄: C, 61.51; H, 9.03; N, 8.97. Found: C, 61.33; H, 8.83; N, 8.98.

3-Oxobutanamidoethyl 3-Oxobutanoate (**5**).

This compound was obtained as yellow liquid; ir (CDCl₃) 3600-3100 (NH), 1746, 1669 (C=O); ¹H nmr (CDCl₃) 7.44 (1H, br t, *J* = 5.37, NH), 4.25 (2H, t, *J* = 5.37, OCH₂), 3.55 (2H, m, NHCH₂), 3.54 (2H, s, CH₂), 3.43 (2H, s, CH₂), 2.28 (3H, s, CH₃), 2.26 (3H, s, CH₃); ¹³C nmr (CDCl₃) 203.8, 201.4 (C=O x 2), 167.1, 166.6 (C=O), 63.5, 50.4, 49.9, 38.3 (CH₂), 30.6 (CH₃), 30.3 (CH₃).

Anal. Calcd. for C₁₀H₁₅NO₅•1/4H₂O: C, 51.39; H, 6.47; N, 5.99. Found: C, 51.07; H, 6.65; N, 6.11.

N,N'-(3,6-Dioxaoctamethylene)bis(3-oxobutanamide) (**7**).

This compound was obtained as colorless microcrystals (from EtOH), mp 84°; ir (KBr) 3600-3000 (NH), 1729, 1644 (C=O); ¹H NMR (DMSO-*d*₆) 8.12 (2H, br t, *J* = 5.86, NH x 2), 3.53 (4H, s, CH₂ x 2), 3.44 (4H, t, *J* = 5.86, CH₂ x 2), 3.33 (4H, s, CH₂ x 2), 2.26 (4H, t, *J* = 5.86, CH₂ x 2), 2.15 (6H, s, CH₃ x 2); ¹³C nmr

(DMSO-*d*₆) 203.9 (C=O x 2), 167.0 (CON x 2), 70.3 (CH₂ x 2), 69.8 (CH₂ x 2), 51.9 (CH₂ x 2), 39.5 (CH₂ x 2), 30.6 (CH₃ x 2).

Anal. Calcd. for C₁₄H₂₄N₂O₆: C, 53.15; H, 7.65; N, 8.86. Found: C, 52.93; H, 7.96; N, 8.83.

Manganese(III)-Based Reaction of *N,N'*-Oligomethylenebis(3-oxobutanamide)s **1_n** (*n* = 2-8) with 1,1-Diarylethenes **2a-d** at Reflux Temperature.

N,N'-Oligomethylenebis(3-oxobutanamide) **1_n** (*n* = 2-8; 0.5-2.0 mmol) and 1,1-diarylethene **2** (1.0-1.5 mmol) were placed in a 50 mL flask equipped with a magnetic stirrer. Glacial acetic acid (25 mL) and manganese(III) acetate (2.5-4 mmol) were added to the flask. The flask was fitted with a reflux condenser and a gas inlet tube. The mixture was sufficiently degassed under reduced pressure using an ultrasonicator in order to exchange with an argon atmosphere, and then heated at 100° using an oil bath with continuous stirring until the brown color of the Mn(III) disappeared. The solvent was removed *in vacuo* and the residue was triturated with water (50 mL). The aqueous reaction mixture was next extracted with chloroform (20 mL x 3). The chloroform extract was washed with saturated sodium hydrogen carbonate solution (30 mL) and water (30 mL), dried over anhydrous sodium sulfate, filtered, and concentrated to dryness. The products were separated by silica gel TLC (Wakogel B-10) using 2% MeOH-CHCl₃ as the developing solvent. The yields are listed in Tables 1 and 2. The products were further purified by recrystallization to obtain the analytical samples. The physical properties, which were obtained in the pure state, are listed below.

N,N'-Ethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{2a}**).

This compound was obtained as colorless microcrystals (from CHCl₃-hexane); mp 246°; ir (KBr) 3600-3100 (NH), 1667 (C=O); ¹H nmr (CDCl₃) 7.36-7.23 (20H, m, arom H), 6.48 (2H, br t, *J* = 6.16, 2 x NH), 3.53 (4H, s, CH₂ x 2), 3.37 (4H, m, NHCH₂ x 2), 2.31 (6H, s, CH₃ x 2); ¹³C nmr (CDCl₃) 167.0 (C=O x 2), 163.9 (O-C= x 2), 145.1 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.5 (>C= x 2), 90.8 (>C-O x 2), 44.2 (CH₂ x 2), 40.3 (CH₂ x 2), 13.9 (CH₃ x 2).

Anal. Calcd. for C₃₈H₃₆N₂O₄: C, 78.06; H, 6.21; N, 4.79. Found: C, 77.76; H, 6.28; N, 4.84.

N,N'-Ethylenebis[5,5-bis(4-chlorophenyl)-2-methyl-4,5-dihydrofuran-3-carboxamide] (**3_{2b}**).

This compound was obtained as colorless microcrystals (from CHCl₃-hexane); mp 256°; ir (CHCl₃) 3600-3100 (NH), 1668 (C=O); ¹H nmr (CDCl₃) 7.40-7.20 (16H, m, arom H), 6.57 (2H, br t, *J* = 6.16, NH x 2), 3.46 (4H, m, CH₂ x 2), 3.34 (4H, m, NHCH₂ x 2), 2.29 (6H, s, CH₃ x 2); ¹³C nmr (CDCl₃) 166.7 (C=O x 2), 163.5 (O-C= x 2), 143.1 (4C), 133.7 (4C) (arom C), 128.6 (8C), 127.0 (8C) (arom CH), 102.6 (>C= x 2), 89.8 (>C-O x 2), 44.0 (CH₂ x 2), 40.5 (CH₂ x 2), 13.9 (CH₃ x 2).

Anal. Calcd. for C₃₈H₃₂N₂O₄Cl₄: C, 63.17; H, 4.46; N, 3.88. Found: C, 62.87; H, 4.51; N, 4.01.

N,N'-Ethylenebis[5,5-bis(4-methylphenyl)-2-methyl-4,5-dihydrofuran-3-carboxamide] (**3_{2c}**).

This compound was obtained as colorless microcrystals (from CH₂Cl₂-hexane); mp 270°; ir (CHCl₃) 3700-3000 (NH), 1665 (C=O); ¹H nmr (CDCl₃) 7.23-7.07 (16H, m, arom H), 6.53 (2H, br t, *J* = 6.16, NH x 2), 3.49 (4H, m, CH₂ x 2), 3.33

(4H, m, NHCH_2 x 2), 2.30 (6H, s, CH_3 x 2), 2.29 (12H, s, CH_3 x 4); ^{13}C nmr (CDCl_3) 167.1 ($\text{C}=\text{O}$ x 2), 163.8 ($\text{O}-\text{C}=\text{C}$ x 2), 142.4 (4C), 137.1 (4C) (arom C), 128.9 (8C), 125.6 (8C) (arom CH), 102.4 ($>\text{C}=\text{C}$ x 2), 90.7 ($>\text{C}-\text{O}$ x 2), 44.1 (CH_2 x 2), 40.4 (CH_2 x 2), 21.0 (CH_3 x 4), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{42}\text{H}_{44}\text{N}_2\text{O}_4$: C, 78.72; H, 6.92; N, 4.37. Found: C, 78.45; H, 7.10; N, 4.45.

N,N'-Ethylenebis[5,5-bis(4-methoxyphenyl)-2-methyl-4,5-dihydrofuran-3-carboxamide] (**3_{2d}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 281°; ir (CHCl_3) 3700-3000 (NH), 1665 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.25-6.80 (16H, m, arom H), 6.63 (2H, br t, $J = 6.16$, NH x 2), 3.75 (12H, s, OCH_3 x 2), 3.48 (4H, m, CH_2 x 2), 3.33 (4H, m, NHCH_2 x 2), 2.30 (6H, s, CH_3 x 2); ^{13}C nmr (CDCl_3) 167.1 ($\text{C}=\text{O}$ x 2), 163.7 ($\text{O}-\text{C}=\text{C}$ x 2), 158.8 (4C), 137.5 (4C) (arom C), 127.0 (8C), 113.6 (8C) (arom CH), 102.4 ($>\text{C}=\text{C}$ x 2), 90.5 ($>\text{C}-\text{O}$ x 2), 55.2 (OCH_3 x 4), 44.3 (CH_2 x 2), 40.4 (CH_2 x 2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{42}\text{H}_{44}\text{N}_2\text{O}_8$: C, 71.57; H, 6.29; N, 3.97. Found: C, 71.49; H, 6.37; N, 3.95.

N,N'-Trimethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{3a}**).

This compound was obtained as colorless microcrystals (from CHCl_3 -hexane); mp 268°; ir (KBr) 3600-3100 (NH), 1667 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.38-7.19 (20H, m, arom H), 6.42 (2H, br t, $J = 6.16$, NH x 2), 3.59 (4H, m, CH_2 x 2), 3.26 (4H, m, NHCH_2 x 2), 2.37 (6H, s, CH_3 x 2), 1.54 (2H, m, $-\text{CH}_2\text{CH}_2\text{CH}_2-$); ^{13}C nmr (CDCl_3) 166.1 ($\text{C}=\text{O}$ x 2), 163.4 ($\text{O}-\text{C}=\text{C}$ x 2), 145.2 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.8 ($>\text{C}=\text{C}$ x 2), 90.6 ($>\text{C}-\text{O}$ x 2), 44.3 (CH_2 x 2), 35.2 (CH_2 x 2), 29.9 (CH_2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{39}\text{H}_{38}\text{N}_2\text{O}_4$: C, 78.24; H, 6.40; N, 4.68. Found: C, 78.02; H, 6.55; N, 4.79.

N,N'-Tetramethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{4a}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 237-240°; ir (KBr) 3600-3100 (NH), 1668 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.38-7.22 (20H, m, arom H), 5.74 (2H, br t, $J = 6.16$, NH x 2), 3.54 (4H, m, CH_2 x 2), 3.25 (4H, m, NHCH_2 x 2), 2.35 (6H, s, CH_3 x 2), 1.49 (4H, m, CH_2 x 2); ^{13}C nmr (CDCl_3) 165.7 ($\text{C}=\text{O}$ x 2), 163.4 ($\text{O}-\text{C}=\text{C}$ x 2), 145.2 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.6 ($>\text{C}=\text{C}$ x 2), 90.6 ($>\text{C}-\text{O}$ x 2), 44.5 (CH_2 x 2), 38.7 (CH_2 x 2), 27.0 (CH_2 x 2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{40}\text{H}_{40}\text{N}_2\text{O}_4$: C, 78.40; H, 6.58; N, 4.57. Found: C, 78.42; H, 6.65; N, 4.73.

N,N'-Pentamethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{5a}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 225°; ir (KBr) 3600-3100 (NH), 1668 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.38-7.24 (20H, m, arom H), 5.33 (2H, br t, $J = 6.80$, NH x 2), 3.51 (4H, s, 2 x CH_2), 3.23 (4H, m, NHCH_2 x 2), 2.36 (6H, s, CH_3 x 2), 1.54-1.44 (4H, m, CH_2 x 2), 1.32-1.30 (2H, m, CH_2); ^{13}C nmr (CDCl_3) 165.6 ($\text{C}=\text{O}$ x 2), 163.4 ($\text{O}-\text{C}=\text{C}$ x 2), 145.1 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.5 ($>\text{C}=\text{C}$ x 2), 90.5 ($>\text{C}-\text{O}$ x 2), 44.6 (CH_2 x 2), 39.0 (CH_2 x 2), 29.5 (CH_2 x 2), 24.3 (CH_2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{41}\text{H}_{42}\text{N}_2\text{O}_4$: C, 78.57; H, 6.75; N, 4.47. Found: C, 78.37; H, 6.84; N, 4.58.

N,N'-Hexamethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{6a}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 218-220°; ir (KBr) 3700-3100 (NH), 1669 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.39-7.21 (20H, m, arom H), 5.50 (2H, br t, $J = 6.71$, NH x 2), 3.53 (4H, m, CH_2 x 2), 3.22 (4H, m, NHCH_2 x 2), 2.36 (6H, s, CH_3 x 2), 1.48-1.43 (4H, m, CH_2 x 2), 1.30-1.28 (4H, m, CH_2 x 2); ^{13}C NMR (CDCl_3) 165.5 ($\text{C}=\text{O}$ x 2), 163.3 ($\text{O}-\text{C}=\text{C}$ x 2), 145.2 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.6 ($>\text{C}=\text{C}$ x 2), 90.5 ($>\text{C}-\text{O}$ x 2), 44.6 (CH_2 x 2), 38.8 (CH_2 x 2), 29.6 (CH_2 x 2), 26.1 (CH_2 x 2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{42}\text{H}_{44}\text{N}_2\text{O}_4$: C, 78.72; H, 6.92; N, 4.37. Found: C, 78.61; H, 7.06; N, 4.29.

N,N'-Heptamethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{7a}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 234-236°; ir (KBr) 3600-3000 (NH), 1669 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.39-7.21 (20H, m, arom H), 5.34 (2H, br t, $J = 6.71$, NH x 2), 3.52 (4H, m, CH_2 x 2), 3.22 (4H, m, NHCH_2 x 2), 2.37 (6H, s, CH_3 x 2), 1.48-1.43 (4H, m, CH_2 x 2), 1.27 (6H, s, CH_2 x 3); ^{13}C nmr (CDCl_3) 165.5 ($\text{C}=\text{O}$ x 2), 163.3 ($\text{O}-\text{C}=\text{C}$ x 2), 145.2 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.6 ($>\text{C}=\text{C}$ x 2), 90.5 ($>\text{C}-\text{O}$ x 2), 44.6 (CH_2 x 2), 39.2 (CH_2 x 2), 29.7 (CH_2 x 2), 28.9 (CH_2), 26.8 (CH_2 x 2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{43}\text{H}_{46}\text{N}_2\text{O}_4$: C, 78.87; H, 7.08; N, 4.28. Found: C, 78.93; H, 7.11; N, 4.40.

N,N'-Octamethylenebis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**3_{8a}**).

This compound was obtained as colorless microcrystals (from CH_2Cl_2 -hexane); mp 218°; ir (KBr) 3600-3000 (NH), 1667 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.39-7.25 (20H, m, arom H), 5.25 (2H, br t, $J = 6.71$, NH x 2), 3.52 (4H, m, CH_2 x 2), 3.24 (4H, m, NHCH_2 x 2), 2.38 (6H, s, CH_3 x 2), 1.49-1.44 (4H, m, CH_2 x 2), 1.27 (8H, s, CH_2 x 4); ^{13}C nmr (CDCl_3) 165.5 ($\text{C}=\text{O}$ x 2), 163.3 ($\text{O}-\text{C}=\text{C}$ x 2), 145.2 (4C) (arom C), 128.3 (8C), 127.6 (4C), 125.7 (8C) (arom CH), 102.5 ($>\text{C}=\text{C}$ x 2), 90.5 ($>\text{C}-\text{O}$ x 2), 44.7 (CH_2 x 2), 39.3 (CH_2 x 2), 29.8 (CH_2 x 2), 29.1 (CH_2 x 2), 26.9 (CH_2 x 2), 14.0 (CH_3 x 2).

Anal. Calcd. for $\text{C}_{44}\text{H}_{48}\text{N}_2\text{O}_4$: C, 79.01; H, 7.23; N, 4.19. Found: C, 78.94; H, 7.27; N, 4.36.

N-3-Oxobutanamidoethyl-2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide (**4_{2a}**).

This compound was obtained as colorless microcrystals (from CHCl_3 -hexane); mp 139°; ir (CDCl_3) 3600-3100 (NH), 1715, 1666 ($\text{C}=\text{O}$); ^1H nmr (CDCl_3) 7.60 (1H, br t, $J = 6.16$, NH), 7.39-7.23 (10H, m, arom H), 6.38 (1H, br t, $J = 6.16$, NH), 3.55 (2H, m, CH_2), 3.36-3.33 (6H, m, CH_2 , NHCH_2 x 2), 2.35 (3H, s, CH_3), 2.14 (3H, s, CH_3); ^{13}C nmr (CDCl_3) 203.5, 167.5, 166.4 ($\text{C}=\text{O}$), 163.7 ($\text{O}-\text{C}=\text{C}$), 145.1 (2C) (arom C), 128.3 (4C), 127.5 (2C), 125.7 (4C) (arom CH), 102.6 ($>\text{C}=\text{C}$), 90.8 ($>\text{C}-\text{O}$), 50.3, 44.1 40.0, 39.6 (CH_2), 30.5, 14.0 (CH_3).

Anal. Calcd. for $\text{C}_{24}\text{H}_{26}\text{N}_2\text{O}_4$: C, 70.92; H, 6.45; N, 6.89. Found: C, 70.63; H, 6.60; N, 6.89.

(2-Methyl-5,5-diphenyl-4,5-dihydrofuran-3-amido)ethyl 2-Methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxylate (**6**).

This compound was obtained as colorless microcrystals (from CH₂Cl₂-hexane); mp 99°; ir (KBr) 3500-3000 (NH), 1671, 1647 (C=O); ¹H nmr (CDCl₃) 7.36-7.21 (20H, m, arom H), 5.88 (1H, br t, *J* = 5.37, NH), 4.21 (2H, t, *J* = 5.37, OCH₂), 3.58-3.50 (6H, m, CH₂ x 3), 2.35 (3H, s, CH₃), 2.30 (3H, s, CH₃); ¹³C nmr (CDCl₃) 167.4, 165.5 (C=O), 166.1, 163.5 (O-C=), 145.1 (2C), 144.9 (2C) (arom C), 128.3 (8C), 127.6 (2C), 127.5 (2C), 125.6 (4C), 125.5 (4C) (arom CH), 102.5, 101.2 (>C=), 91.8, 90.6 (>C-O), 62.3, 44.4, 44.0, 39.2 (CH₂), 14.3, 13.9 (CH₃).

Anal. Calcd. for C₃₈H₃₅NO₅•1/4H₂O: C, 77.33; H, 5.98; N, 2.37. Found: C, 77.33; H, 6.20; N, 2.68.

N,N'-(3,6-Dioxaoctamethylene)bis(2-methyl-5,5-diphenyl-4,5-dihydrofuran-3-carboxamide) (**8**).

This compound was obtained as colorless microcrystals (from CH₂Cl₂-hexane); mp 166°; ir (KBr) 3600-3000 (NH), 1669 (C=O); ¹H nmr (CDCl₃) 7.38-7.22 (20H, m, arom H), 5.83 (2H, br t, *J* = 5.86, NH x 2), 3.54 (4H, s, CH₂ x 2), 3.51 (4H, t, *J* = 5.86, CH₂ x 2), 3.44 (4H, t, *J* = 5.86, CH₂ x 2), 3.39 (4H, s, CH₂ x 2), 2.38 (6H, s, CH₃ x 2); ¹³C nmr (CDCl₃) 165.6 (C=O x 2), 163.6 (O-C= x 2), 145.1 (4C) (arom C), 128.3 (8C), 127.5 (4C), 125.6 (8C) (arom CH), 102.6 (>C= x 2), 90.6 (>C-O x 2), 70.0 (CH₂ x 2), 44.4 (CH₂ x 2), 38.9 (CH₂ x 2), 14.0 (CH₃ x 2).

Anal. Calcd. for C₄₂H₄₄N₂O₆•1/4H₂O: C, 74.49; H, 6.55; N, 4.14. Found: C, 74.43; H, 6.60; N, 4.08.

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