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NEW β-TRICARBONYL COMPOUNDS: SYNTHESIS, REACTIONS WITH UREA AND SOME THIOUREAS

Mustafa Saçmacı,[a*] Abdulrezzak Alkan,[a] Şerife Saçmacı,[b] Emin Sarıpınar,[b] and Ertan Şahin[c]

[a] Department of Chemistry, Faculty of Arts and Sciences, Bozok University, Yozgat-Turkey

[b] Department of Chemistry, Faculty of Arts and Sciences, Erciyes University, Kayseri-Turkey

[c] Department of Chemistry, Faculty of Arts and Sciences, Atatürk University, Erzurum-Turkey

Corresponding author: Phone: 00 90 354 2421021 Fax: 00 90 354 2421022

E-Mail: msacmaci@hotmail.com

Abstract – 2,3-Dihydro-4-(4-methoxybenzoyl)-5-(4-methoxyphenyl)furan-2,3-dione **1** reacted with some alkyl carbamates (**3a-b**) *via p,p′*-dimethoxy-dibenzoylketene intermediate **2** giving new β-tricarbonyl compounds (**4a-b**). Then, these compounds were converted into 1,3-oxazine-2,4(3*H*)-dione **7** with urea and 2-thioxo-2,3-dihydropyrimidin-4(1*H*)-ones (**6a-c**) with some thiourea derivatives, respectively. Pyrimidin-4-ones (**6a-c**) and oxazine **7** were obtained in good yields. The structures and characterizations of new synthesized compounds were established by the ^{1}H and ^{13}C NMR, IR, UV-VIS spectroscopic data, elemental analysis, and X-ray diffraction method.

INTRODUCTION

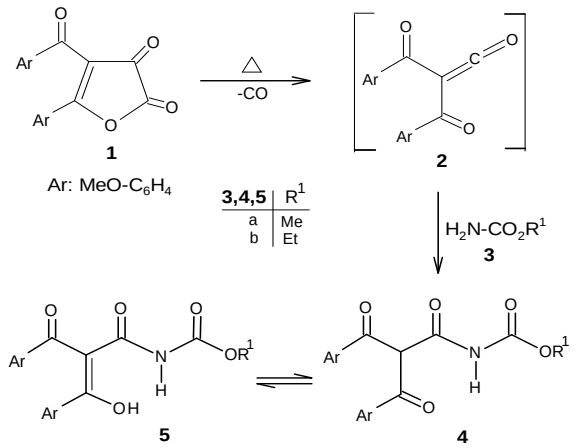
The cyclocondensation reaction of 1,3-diones with oxaly chloride represents a convenient synthesis of furan-2,3-dione systems.[1] These compounds have been demonstrated to be a versatile, convenient, multifunctional, synthetic building block for the construction of novel heterocyclic starting materials that has in general been widely explored during the last few decades.[2] The thermal decomposition of furan-2,3-diones leads to the diacylketene as intermediates which cannot be isolated.[3,4] These ketenes are currently of considerable interest, not only because of mechanistic and theoretical considerations,[5-8] but also because of their use as synthetic building blocks in organic synthesis.[9-11] Recently, 2,3-dihydro-4-(4-

methoxybenzoyl)-5-(4-methoxyphenyl)furan-2,3-dione **1** appeared to be an important starting compound in synthetic organic chemistry.[12-15] Also, a convenient preparation of functionalized pyrimidin-2-one, pyrimidinthione and oxazine derivatives from compound **1** and thiosemicarbazones have been reported.[16,17]

Pyrimidines generally have been found much interest for their widespread potential biological activities and medicinal applications, thus their chemistry has been investigated extensively.[18,19] In particular, various analogues of pyrimidinthiones possess effective antibacterial, antifungal, antiviral, anti-AIDS, insecticidal and miticidal activities.[20,21] Furthermore many condensed heterocyclic systems, especially when linked to a pyrimidine ring, play an important role as analgesic, antihypertensive,[22] antipyretic, and anti-inflammatory drugs,[23,24] also as pesticides, herbicides and plant growth regulators.[25] On the other hand, oxazine derivatives have been shown to be antimicrobial agents, fungicides and also exhibit some cytotoxic or as potential anti-tumor agents.[26-28] Since pyrimidines and oxazines in general have much more interest for biological and medicinal reasons, above mentioned, we have synthesized new 2-thioxo-2,3-dihydropyrimidin-4(1*H*)-ones and 1,3-oxazine-2,4(3*H*)-dione.

RESULTS AND DISCUSSION

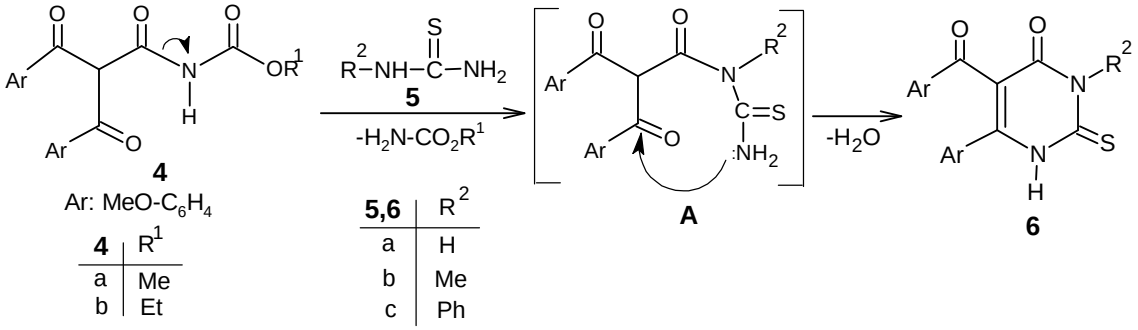
The starting compound (**1**) was prepared according to literature.[29] Since compound **1** readily forms *p,p′*-dimethoxydibenzoylketene **2** by thermal ring opening and decarbonylation,[30] the formation of the novel β-tricarbonyl compounds (**4a-b**) can be rationalized by nucleophilic addition of the alkyl carbamates (**3a-b**) to reactive intermediate **2**. Compound **1** with **3a-b** afforded β-tricarbonyl compounds in boiling toluene in good yields. The formation of **4a-b** were shown briefly in Scheme 1.



Scheme 1

Previously, the formation of a few β-tricarbonyl compounds, namely 2,2-dibenzoyl-*N*-alkoxycarbonylacetamides, from 4-benzoyl-5-phenylfuran-2,3-dione and urethanes were determined and

it was shown that β-tricarbonyl compounds were in enol form in solution, whereas keto forms were formed in solid phase.[31] Similarly, in this study, keto-enol tautomers (**4**, **5**) were determined for **4a-b**. The relative amounts of keto form of **4a** and **4b** were found from their ^{1}H NMR spectra as 83% and 75%, respectively. In the IR spectra of compound **4a**, the carbonyl absorption bands were found to be at about 1672, 1699 and 1754 cm^{-1}. Important structural information about **4a** was obtained from its ^{1}H NMR spectrum. The proton signal of **4a** which belongs to the -CH in the keto form was observed at 6.61 ppm. While proton signal of -CO_2CH_3 group of **4a** in the keto form was detected at 3.83 ppm, on the other hand, the proton signal of -CO_2CH_3 group belonging to **5a** in the enol form was determined at 3.79 ppm. The broad peak at 10.62 ppm represents the -NH and the peak at 8.89 ppm is thought to represent the -OH in the enol form. In the ^{13}C NMR spectrum of **4a**, the peaks at 57.55, 57.32 and 55.10 ppm are assigned to the methoxy groups. The peak at 66.31 ppm represents the -CH in the keto form. The formation of pyrimidin-4-ones (**6a-c**) is outlined briefly in Scheme 2.



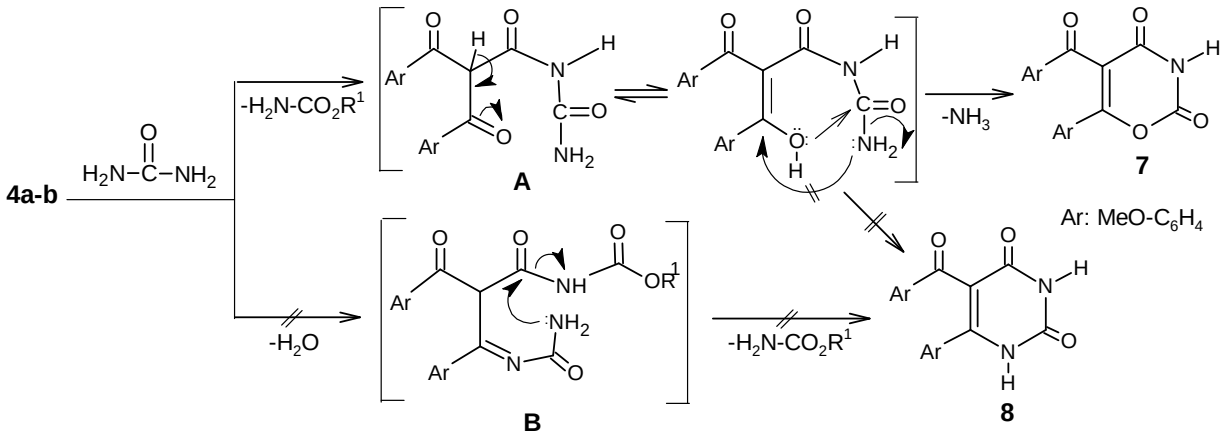
Scheme 2

In the reaction pathway, the formation of intermediate (**A**) may be initiated by a nucleophilic attack of -NH-R^2 on compound **5** to carbonyl group attached to -$NHCO_2R^1$ group of **4a-b**. By eliminating of -$H_2NCO_2R^1$ group, the product (**A**) which cannot be isolated is obtained and the -NH_2 group of intermediate **A** attacks to anisoyl carbonyl on the molecule.Then, intermediate **A** is cyclized to **6a-c** by eliminating H_2O. In the ^{1}H NMR spectrum of compound **6a** has one singlet signal at 9.42 ppm assignable to the -NH band on the pyrimidin-4-one moiety. In the ^{13}C NMR spectrum of compound **6a** carbonyl groups and thiocarbonyl group were observed at 189.42, 158.77 and 174.85 ppm, respectively.

The electronic spectra of the pyrimidin-4-ones (**6a-c**) recorded in the 200-700 nm region were obtained in DMF solution and their UV spectra were compaired with each others. The similarity of UV spectra of **6a** and **6b** were observed but **6c** pattern was determined quite different. The peaks in the UV absorption maxima of **6a**, **6b** and **6c** were observed at 297.0, 298.3 and 283.5 nm, respectively. Methyl group at the compound **6b** gives electron as an inductive effect to pyrimidine-4-one cycle. The giving of electron as an

inductive effect is effected to π electron in the cycle.[32] As a result, the energy of π* molecular orbital is decreased and the absorption band of **6b** shifts longer to λ_{max} (298.3 nm). For compounds (**6a-c**) UV bands with a bathochromic effect resulting from H, Me and Ph attached to N are observed at resulting from the π-π* transition. Also, for compounds **4a** and **4b** UV bands with a bathochromic effect resulting from the -OMe and -OEt attached to carbonyl were observed at 289.0 and 295.0 nm.

A reasonable proposal for different reaction pathway from **4a-b** to **7** is also outlined briefly in Scheme 3. It is assumed that the $-NH_2$ group of urea attacks to carbonyl group attached to $-NHCO_2R^1$ group of **4a-b** but not the carbonyl group Ar-C=O. Because, $-NHCO_2R^1$ group can be easily eliminated from **4a-b**. The intermediate (**A**) is also obtained by eliminating of $-H_2NCO_2R^1$ group. The reaction of **4a-b** with urea did not produce to compound **8** *via* intermediate **B**. Instead of **8**, compound **7** was isolated *via* intermediate **A**. When urea and thiourea compounds are compared, the nucleophilic effect of $-NH_2$ group of urea is less than $-NH_2$ group of thiourea compounds. For this reason, the formation of compound **8** cannot be occured by attacking $-NH_2$ group of intermediate **A** in the enol form. With rearrangement of intermediate **A** in the enol form, -OH nucleophile attacks to carbonyl group attached to $-NH_2$ on the molecule. Then, the elimination of ammonia leads to the formation of 1,3-oxazine moiety **7**.



Scheme 3

The IR spectrum of **7** showed three carbonyl bands 1789, 1692, 1649 cm^{-1}. The ^1H NMR spectrum of **7** showed an -NH absorption band at 8.92 ppm and the ^{13}C NMR spectrum of **7** showed three carbonyl carbons at 188.94, 161.26, 146.81 ppm.

The ORTEP of compound **7** shown in Figure 1, the molecule involves two 4-methoxyphenyl rings A (C5-C7) and B (C3-C15) connected to the 1,3-oxazine-2,4-dione ring C (C3-C5, O6). The rings (A, B, and C) are each essentially planar and the dihedral angles, torsion angles, bond lengths and bond angles values of very close analog 1,3-oxazine derivative are in agreement with reported literature values.[33,34] The dihedral angles between the planes as follows: A/B = 63.85 (9)° A/C =19.10 (16)° B/C = 62.05 (9)°. The molecules in the crystal structure are connected by van der Waals interaction.

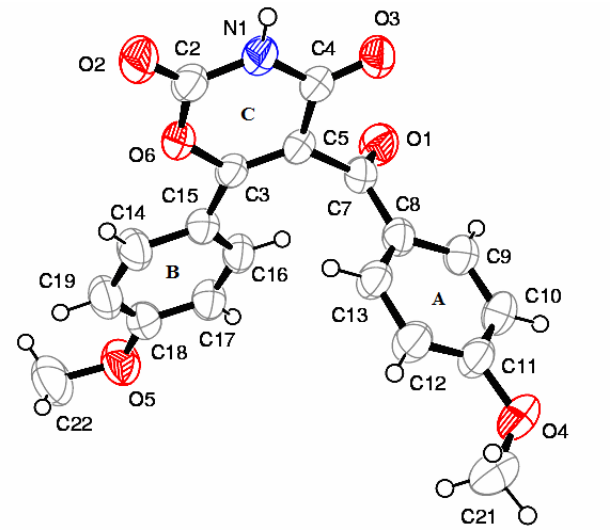


Figure 1. ORTEP drawing of molecular structure of **7**.
Displacement ellipsoids are drawn at the 50% probability level.

EXPERIMENTAL

Melting points were determined on an electrothermal 9200 apparatus and are uncorrected. Elemental analysis were carried out using LECO-932 CHNSO analyzer, IR spectra were recorded on a Jasco Plus Model 460 FT IR spectrometer as KBr pellets. The ^{1}H and ^{13}C NMR spectra were acquired from a Gemini-Varian 200 (50) MHz spectrometer (in deuteriochloroform solution containing tetramethyl silane as the internal standart). All experiments were followed by tlc using DC Alufolien Kieselgel 60 F 254 Merck and Camag TLC lamp (254/366 nm). Electronic spectra of compounds were measured on a Hitachi (150-20) Model UV-VIS spectrometer. For the crystal structure determination, the single-crystal of the compound **7** was used for data collection on a four-circle Rigaku R-AXIS RAPID-S diffractometer (equipped with a two-dimensional area IP detector). The cylindrically shaped imaging plate covers the two-theta angular range between -60 and 140 ° with a crystal-film distance of 127.4 mm. The graphite-monochromatized Mo Kα radiation (λ = 0.71073 Å) and oscillation scans technique with Δω = 5 ° for one image were used for data collection. Images for **7** was taken successfully by varying ω with three sets of different χ and φ values. For each compounds the 216 images for six different runs covering about 99.7% of the Ewald spheres were performed. The lattice parameters were determined by the least-squares methods on the basis of all reflections with $F^2>2\sigma(F^2)$. Integration of the intensities, correction for Lorentz and polarization effects and cell refinement was performed using Crystal Clear (Rigaku/MSC Inc., 2005) software.[35] The structures were solved by the direct method using SHELXS.[36] The positional and atomic displacement parameters (ADPs) were refined by the full-matrix least-squares method using SHELXL[36] and SIR2002.[37]

Methyl 2-(4-methoxybenzoyl)-3-(4-methoxyphenyl)-3-oxopropanoylcarbamate (4a). Compound **1** (1.0 g, 2.96 mmol) and methyl carbamate **3a** (0.22 g, 2.96 mmol) were refluxed for 3 h in toluene (50 mL). The toluene was evaporated and the remaining oily residue was triturated with anhydrous Et_2O. The white crude product was recrystallized from MeOH and dried on P_2O_5. Mp 165 ^oC, yield 0.80 g (70%), IR (KBr): υ = 3264 (N-H), 1754, 1699, 1672 cm^{-1} (C=O). ^1H NMR (200 MHz, $CDCl_3$): δ = 10.62 (-NH), 8.89 (s, 1H, enol-OH), 7.97-6.57 (m, 8H, Ar-H), 6.61 (s, 1H, keto-CH), 3.83, 3.80, 3.72 ppm (s, 9H, $-OCH_3$); ^{13}C NMR ($CDCl_3$, 50 MHz): δ = 197.97, 185.72, 172.09 (C=O), 105.07 (enol-CH), 134.23-115.30 (m, Ar-C), 66.31 (keto-CH), 57.55, 57.32, 55.10 ppm (OCH_3). *Anal.* Calcd for $C_{20}H_{19}NO_7$: C, 62.33; H, 4.97; N, 3.63. Found: C, 62.54; H, 5.04; N, 3.76.

Ethyl 2-(4-methoxybenzoyl)-3-(4-methoxyphenyl)-3-oxopropanoylcarbamate (4b). Compound **1** (1.0 g, 2.96 mmol) and ethyl carbamate **3b** (0.26 g, 2.96 mmol) were refluxed for 4 h in toluene (50 mL). The toluene was evaporated and the remaining oily residue was triturated with anhydrous Et_2O. The white crude product was recrystallized from EtOH and dried on P_2O_5. mp 155 ^oC, yield 0.95 g (81%), IR (KBr): υ = 3265 (N-H), 1752, 1697, 1671 cm^{-1} (C=O). ^1H NMR ($CDCl_3$): δ = 10.56 (s, keto-NH), 8.92 (s, enol-OH), 7.92-7.29 (m, 8H, Ar-H), 7.26 (s, keto-CH), 4.14 (q, *J* = 6.9 Hz, 2H, $-OCH_2$), 3.77, 3.66 (s, 6H, $-OCH_3$), 1.29, 1.26, 1.22 ppm (t, 3H, $-CH_3$). ^{13}C NMR ($CDCl_3$, 50 MHz): δ = 196.87, 184.75, 174.02 (C=O), 103.02 (enol-CH), 135.83-117.40 (m, Ar-C), 66.11 (keto-CH), 61.05 (OCH_2) 57.55, 57.32, 55.10 (OCH_3), 15.10 ppm (CH_3). *Anal.* Calcd for $C_{21}H_{21}NO_7$: C, 63.15; H, 5.30; N, 3.51. Found: C, 62.94; H, 5.10; N, 3.41.

5-(4-Methoxybenzoyl)-6-(4-methoxyphenyl)-2-thioxo-2,3-dihydropyrimidin-4(1*H*)-one (6a).
Compound **4a** (1.0 g, 2.60 mmol) or compound **4b** (1.0 g, 2.50 mmol) and thiourea (0.19 g, 2.50 mmol) were homogeneously mixed. The mixture was heated at 160 ^oC for 1 h without any solvent in a 50 mL round bottomed flask equipped with a calcium chloride guard tube. After cooling to rt the residue was triturated with anhydrous Et_2O and the crude product recrystallized from EtOH and dried on P_2O_5. Mp 225 ^oC, yields: 0.65 or 0.78 (71% or 84%), IR (KBr): 3312, 3164 (N-H), 1677, 1661 (C=O), 1605 (C=C), 1467 cm^{-1} (C=S). ^1H NMR (200 MHz, $CDCl_3$): δ = 9.42 (N-H), 9.14 (N-H), 7.82-6.83 (m, 8H, Ar-H), 3.87, 3.84 ppm (s, 6H, $-OCH_3$). ^{13}C NMR (50 MHz, $CDCl_3$): 189.42, 158.77 (C=O), 174.85 (C=S), 151.04 (C-NH), 132.09-114.30 (Ar-C), 55.78, 55.69 ppm (OCH_3). *Anal.* Calcd for $C_{19}H_{16}N_2O_4S$: C, 61.94; H, 4.38; N, 7.60; S, 8.70. Found: C, 61.75; H, 4.15; N, 7.51; S, 8.50.

5-(4-Methoxybenzoyl)-6-(4-methoxyphenyl)-3-methyl-2-thioxo-2,3-dihydropyrimidin-4(1*H*)-one (6b). Compound **4a** (1.0 g, 2.60 mmol) or compound **4b** (1.0 g, 2.50 mmol) and *N*-methylthiourea (0.22 g, 2.51 mmol) were homogeneously mixed. The mixture was heated at 160 ^oC for 1 h without any solvent in a 50

mL round bottomed flask equipped with a calcium chloride guard tube. After cooling to rt the residue was triturated with anhydrous Et_2O and the crude product recrystallized from EtOH and dried on P_2O_5. Mp 285 ^oC, yields: 0.80 g or 0.76 g (84% or 79%), IR (KBr): 3240 (N-H), 1663, 1644 (C=O), 1598 (C=C), 1492 cm^{-1} (C=S). ^1H NMR (200 MHz, $CDCl_3$): δ = 9.63 (N-H), 7.74-6.82 (m, 8H, Ar-H), 3.84, 3.76 (s, 6H, -OCH_3), 3.51 ppm (s, 3H, -CH_3). ^{13}C NMR (50 MHz, $CDCl_3$): 188.95, 157.36 (C=O), 177.14 (C=S), 154.91 (C-NH), 131.99-114.14 (Ar-C), 55.78, 55.56 (OCH_3), 41.08 ppm (N-CH_3). *Anal.* Calcd for $C_{20}H_{18}N_2O_4S$: C, 62.81; H, 4.74; N, 7.33; S, 8.38. Found: C, 62.60; H, 4.50; N, 7.10; S, 8.25.

5-(4-Methoxybenzoyl)-6-(4-methoxyphenyl)-3-phenyl-2-thioxo-2,3-dihydropyrimidin-4(1*H*)-one (6c). Compound **4a** (1.0 g, 2.60 mmol) or compound **4b** (1.0 g, 2.50 mmol) and *N*-phenylthiourea (0.38 g, 2.50 mmol) were homogeneously mixed. The mixture was heated at 160 ^oC for 1 h without any solvent in a 50 mL round bottomed flask equipped with a calcium chloride guard tube. After cooling to rt the residue was triturated with anhydrous Et_2O and the crude product recrystallized from EtOH and dried on P_2O_5. Mp 195 ^oC, yields: 0.67 g or 0.75 g (60% or 68%), IR (KBr): 3204 (N-H), 1680, 1646 (C=O), 1591 (C=C), 1467 cm^{-1} (C=S). ^1H NMR (200 MHz, $CDCl_3$): δ = 12.87 (N-H), 7.74-6.36 (m, 13H, Ar-H), 3.76, 3.74 ppm (s, 6H, OCH_3). ^{13}C NMR (50 MHz, $CDCl_3$): 194.23, 158.56 (C=O), 183.07 (C=S), 140.99-117.88 (Ar-C), 60.32, 59.78 ppm (OCH_3). *Anal*. Calcd for $C_{25}H_{20}N_2O_4S$: C, 67.55; H, 4.54; N, 6.30; S, 7.21. Found: C, 67.28; H, 4.45; N, 6.10; S, 7.10.

5-(4-Methoxybenzoyl)-6-(4-methoxyphenyl)-2*H*-1,3-oxazine-2,4(3*H*)-dione (7). Compound **4a** (1.0 g, 2.60 mmol) or compound **4b** (1.0 g, 2.50 mmol) and urea (0.15 g, 2.50 mmol) were mixed in 30 mL distilled xylene in a one-necked pear-sahped flask at rt and then refluxed for 5 h. After evaporation of the solvent under reduced pressure, the oily residue was stirred with anhydrous Et_2O. The precipitated crude white product was separated of Et_2O by filtering and recrystallized from EtOH to give **7** as colourless powder, yields: 0.60 g or 0.67 g (65% or 75%), mp 182 ^oC, IR (KBr): 3204 (N-H), 1789, 1692, 1649 (CO), 1600 cm^{-1} (C=C). ^1H NMR (200 MHz, $CDCl_3$): δ = 8.92 (N-H), 7.89-6.80 (Ar-H), 3.84, 3.79 ppm (OCH_3). ^{13}C NMR (50 MHz, $CDCl_3$): δ = 188.94, 161.26, 146.81 (CO), 164.91 (C-O), 132.29-111.25 (Ar-C), 55.84-59.71 ppm (OCH_3). *Anal.* Calcd for $C_{19}H_{15}NO_6$: C, 64.59; H, 4.28; N, 7.95. Found: C, 64.50; H, 4.40; N, 7.80.

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