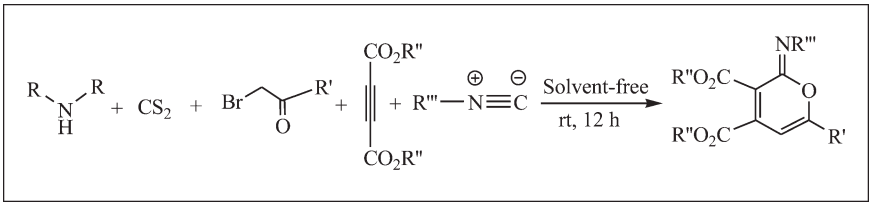
Faramarz Rostami-Charati,[a]* Zinatossadat Hossaini,[b]
Mohammad A. Khalilzadeh,[b] Mahboube Islami Moghaddam,[c]
and Vahid Babaei[d]

[a]Department of Chemistry, Faculty of Science, Gonbad Kavous University,
PO Box 163, Gonbad, Iran
[b]Department of Chemistry, Islamic Azad University, Qaemshahr Branch, Mazandaran, Iran
[c]Department of Chemistry, University of Payam Noor, Iran
[d]Department of Environmental and Civil Engineering, Babol Noshirvani University of Technology,
Babol, Iran
*E-mail: f_rostami_ch@yahoo.com or frostami@gau.ac.ir
Received September 3, 2010
DOI 10.1002/jhet.818
Published online 10 November 2011 in Wiley Online Library (wileyonlinelibrary.com).



A straightforward and an efficient method for the synthesis of 2*H*-pyrans *via* the one-pot, reaction of alkyl bromides, carbon disulfide, secondary amines, activated acetylenes and isocyanides under solvent-free conditions without using any catalyst at room temperature is reported. The method offers several advantages including high yields of products and an easy work-up procedure.

J. Heterocyclic Chem., **49**, 405 (2012).

INTRODUCTION

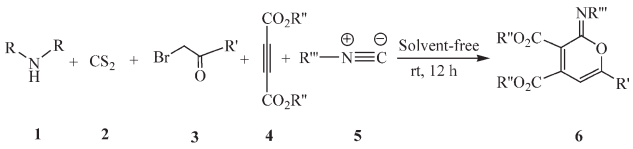
Within the last decade, the resurgence of interest in multicomponent reactions (MCRs) has been considered, not only because of their inherent atom efficiency and ease of implementation, but also because of their value to the pharmaceutical industry for construction of low molecular weight compound libraries through combinatorial strategies or parallel synthesis [1,2]. The capability of an isonitrile to undergo facile a-addition with a nucleophile and an electrophile under mild conditions made it a popular reactant for the development of novel MCRs [3–9]. For the purposes of eco-friendly ''green chemistry'', a reaction should ideally, be conducted under solvent-free conditions with minimal or no side-product formation and with utmost atom economy [10]. In the context of our on going studies on heterocyclic construction mediated by zwitterionic intermediates [11], the possibility of trapping the 1:1 intermediate formed between dialkyl acetylenedicarboxylate and isocyanides with suitable C—H acids appeared attractive from the viewpoint of devising a novel MCR [12]. Here, we describe an efficient synthesis of 2*H*-pyran derivatives *via* the reaction of alkyl bromides, carbon disulfide, secondary amines, and activated acetylenes and isocyanides under solvent-free conditions at room temperature (Scheme 1).

RESULTS AND DISCUSSION

As indicated in Scheme 1, secondary amines **1**, carbon disulfide **2**, alkyl bromides **3**, activated acetylenes **4**, and isocyanides **5** undergo a smooth 1:1:1:1:1 addition reaction under solvent-free conditions at room temperature (the secondary amines and carbon disulfide are mixed first and then alkyl bromides is added. subsequently, activated acetylenes and isocyanides are added) to produce 2*H*-pyran derivatives **6** in 90–97% yields (Scheme 1). Structures of compounds **6a–6l** were deduced from their IR, mass, ^{1}H NMR and ^{13}C NMR spectra. The mass spectra of these compounds displayed molecular ion peaks at the appropriate *m*/*z* values. The ^{1}H NMR spectrum of **6a** exhibited a triplet at $\delta = 1.29$ ($^3J = 7.6$ Hz) for the methyl group and four singlets for the *tert*-butyl (δ 1.27 ppm), methoxy (3.75 and 3.82 ppm), and olefinic (δ 6.57 ppm) protons. The proton-decoupled ^{13}C NMR spectrum of **6a** showed 16 distinct resonances in agreement with the proposed structure. Three single resonances at $\delta = 160.4$, 162.3 and 163.8 ppm are observed in the ^{13}C NMR spectrum of **6a**, which are attributed to the carbonyl groups.

Although we have not established the mechanism of the reactions in an experimental manner, a possible explanation is proposed in Scheme 2. Dithiocarbamates **7** were produced from the reaction of secondary amines **1**, carbon

Scheme 1



3, 4, 5, 6	R'	R''	R'''	Yield (%) of 6
a	CO_2Et	Me	$(CH_3)_3C$	95
b	CO_2Et	Et	$(CH_3)_3C$	93
c	CO_2Et	Me	cyclohexyl	92
d	CO_2Et	Me	CH_2CO_2Et	95
e	Ph	Me	cyclohexyl	97
f	4-Me-C_6H_4	Me	$(CH_3)_3C$	94
g	4-NO_2-C_6H_4	Me	$(CH_3)_3C$	90
h	4-NO_2-C_6H_4	Et	$(CH_3)_3C$	95
i	4-NO_2-C_6H_4	^{t}Bu	cyclohexyl	94
j	CO_2Et	Me	1,1,3,3-tetramethylbutyl	92
k	4-Me-C_6H_4	Me	CH_2CO_2Et	90
l	Ph	Et	CH_2CO_2Et	93

disulfide **2**, and alkyl bromides **3** under solvent-free conditions. On the basis of the well-established chemistry of isocyanides [12–17], it is reasonable to assume that the compound **8** apparently results from initial addition of the isocyanide to the activated acetylenes that adds to dithiocarbamate **7** resulting in the formation of **9**, which undergoes cyclization to generate the 2*H*-pyrans **6a–l**.

In conclusion, we have developed the most useful and dependable procedure currently available for the synthesis of 2*H*-pyrans by using low-cost and readily available starting materials in one-pot. This method represents a simple and green procedure, uses mild reaction conditions, and has general applicability.

EXPERIMENTAL

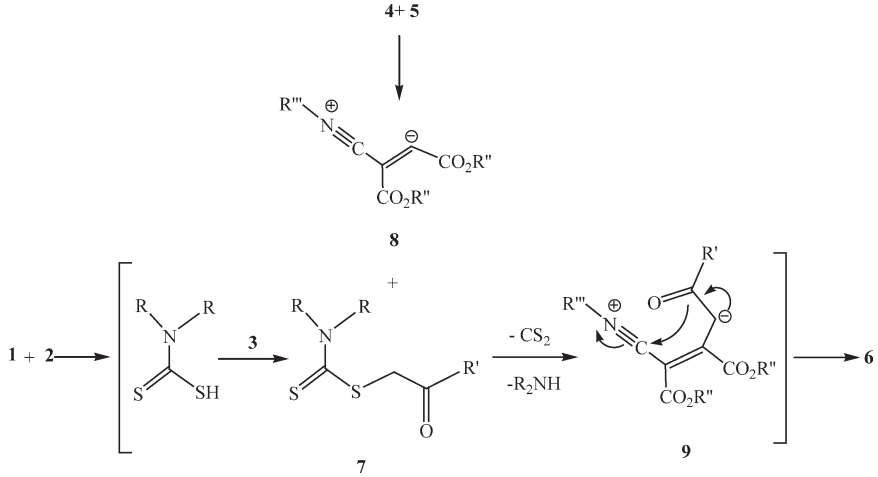
All chemicals were obtained from commercial sources. Melting points were measured on a Kofler hot stage apparatus and are uncorrected. ^{1}H NMR and ^{13}C NMR spectra were obtained with a Bruker FT-500 spectrometer in chloroform-d1, and tetramethylsilane (TMS) was used as an internal standard. Mass spectra were recorded with a Finnigan Mat TSQ-70 spectrometer. Infrared (IR) spectra were acquired on a Nicollet Magna 550-FT spectrometer. Elemental analyses were carried out with a Perkin-Elmer model 240-C apparatus. The results of elemental analyses (C, H, N) were within ±0.4% of the calculated values.

Typical procedure for the preparation of 2*H*-pyran derivatives 6. To a mixture of the alkyl bromides (2 mmol) and carbon disulfide (4 mmol) in the test tube, amine (4 mmol) was added and stirred at 0°C for 30 min, then warmed to room temperate and stirring was continued until the reaction was complete (reaction mixture solidified or monitored by TLC). To this mixture activated acetylenes (2 mmol) was added. Then, isocyanides (2 mmol) was added slowly and mixture of reaction was stirred for 12 hours. Upon completion that monitored by TLC, water (15 mL) was poured into the mixture of reaction. The resulting precipitate was separated by filtration and was purified with diethyl ether (Et_2O) to afford the pure title compounds.

6-Ethyl 3,4-dimethy-2-(*tert*-butylimino)-2*H*-pyran-3,4,6-tricarboxylate (6a). White powder, mp 124–127°C, yield: 0.64 g (95%). IR (KBr) (ν_{max}/cm^{-1}): 1738, 1732, 1723, 1547 and 1245 cm^{-1}. ^{1}H NMR: δ = 1.27 (9 H, s, *Me*$_3$C), 1.29 (3 H, t, ^3J = 7.6 Hz, Me), 3.75 (3 H, s, MeO), 3.82 (3 H, s, MeO), 4.32 (2 H, q, ^3J = 7.6 Hz, CH_2O), 6.57 (1 H, s, CH) ppm. ^{13}C NMR: δ = 14.0 (Me), 29.6 (*Me*$_3$C), 51.6 (MeO), 52.3 (MeO), 55.3 (C—N), 63.1 (CH_2O), 117.4 (CH), 135.4 (C), 143.2 (C), 145.4 (C), 149.8 (C=N), 160.4 (C=O), 162.3 (C=O), 163.8 (C=O) ppm. MS: *m*/*z* (%) = 339 (M^+, 15), 308 (45), 282 (85), 57 (100), 31(86). Anal. Calc. for $C_{16}H_{21}NO_7$ (339.34): C, 56.63; H, 6.24; N, 4.13 found: C, 56.58; H, 6.12; N, 4.02%.

Triethyl 2-(*tert*-butylimino)-2*H*-pyran-3,4,6-tricarboxylate (6b). Yellow powder, mp 130–132°C. yield: 0.68 g (93%). IR (KBr) (ν_{max}/cm^{-1}): 1738, 1735, 1724, 1582 and 1195 cm^{-1}. ^{1}H NMR: δ = 1.26 (9 H, s, *Me*$_3$C), 1.29 (3 H, t, ^3J = 7.3 Hz, Me), 1.32 (3 H, t, ^3J = 7.3 Hz, Me), 1.36 (3 H, t, ^3J = 7.4 Hz, Me), 4.13 (2 H, q, ^3J = 7.3 Hz, CH_2O), 4.15 (2 H, q, ^3J = 7.3 Hz, CH_2O), 4.24 (2 H, q, ^3J = 7.4 Hz, CH_2O), 6.64 (1

Scheme 2



H, s, CH) ppm. ^{13}C NMR: δ = 13.8 (Me), 14.1 (Me), 14.3 (Me), 30.2 (*Me*$_3$C), 55.6 (C—N), 62.3 (CH_2O), 62.6 (CH_2O), 63.0 (CH_2O), 119.2 (CH), 136.8 (C), 142.6 (C), 145.4 (C), 148.4 (C=N), 161.5 (C=O), 162.8 (C=O), 163.3 (C=O) ppm. MS: *m*/*z* (%) = 367 (M^+, 10), 339 (62), 310 (84), 57 (100), 45 (48). Anal. Calc. for $C_{18}H_{25}NO_7$ (367.39): C, 58.85; H, 6.86; N, 3.81 found: C, 58.78; H, 6.73; N, 3.75%.

6-Ethyl 3,4-dimethy-2-(cyclohexylimino)-2*H*-pyran-3,4,6-tricarboxylate (6c). White powder, mp 127–129°C, yield: 0.67 g (92%). IR (KBr) (ν_{max}/cm^{-1}): 1735, 1730, 1722, 1554 and 1215 cm^{-1}. ^{1}H NMR: δ = 1.25 (3 H, t, ^3J = 7.5 Hz, Me), 1.32 (2 H, m, CH_2), 1.38 (2 H, m, CH_2), 1.45 (2 H, m, CH_2), 1.67 (2 H, m, CH_2), 1.80 (2 H, m, CH_2), 3.74 (3 H, s, MeO), 3.77 (1 H, m, N-CH), 3.80 (3 H, s, MeO), 4.35 (2 H, q, ^3J = 7.5 Hz, CH_2O), 6.48 (1 H, s, CH) ppm. ^{13}C NMR: δ = 14.1 (Me), 24.2 (CH_2), 24.6 (CH_2), 25.8 (CH_2), 33.5 (CH_2), 33.9 (CH_2), 52.0 (MeO), 52.5 (MeO), 56.7 (C—N), 63.4 (CH_2O), 117.5 (CH), 135.6 (C), 143.4 (C), 145.5 (C), 148.7 (C=N), 161.2 (C=O), 162.4 (C=O), 164.2 (C=O) ppm. Anal. Calc. for $C_{18}H_{23}NO_7$ (365.38): C, 59.17; H, 6.34; N, 3.83 found: C, 59.02; H, 6.27; N, 3.72%.

6-Ethyl 3,4-dimethy-2-(2-ethoxy-2-oxoethylimino)-2*H*-pyran-3,4,6-tricarboxylate (6d). Pale yellow powder, mp 131–133°C. yield: 0.68 g (95%). IR (KBr) (ν_{max}/cm^{-1}): 1736, 1732, 1725, 1527 and 1159 cm^{-1}. ^{1}H NMR: δ = 1.32 (3 H, t, ^3J = 7.4 Hz, Me), 1.35 (3 H, t, ^3J = 7.5 Hz, Me), 3.76 (3 H, s. MeO), 3.82 (3 H, s. MeO), 4.15 (2 H, q, ^3J = 7.4 Hz, CH_2O), 4.18 (2 H, q, ^3J = 7.4 Hz, CH_2O), 4.22 (2 H, s, CH_2), 6.58 (1 H, s, CH) ppm. ^{13}C NMR: δ = 13.7 (Me), 14.0 (Me), 50.8 (CH_2-N), 51.7 (MeO), 52.3 (MeO), 61.4 (OCH_2), 62.3 (OCH_2), 118.7 (CH), 136.5 (C), 142.3 (C), 144.9 (C), 147.8 (C=N), 161.8 (C=O), 162.0 (C=O), 163.2 (C=O), 163.6 (C=O) ppm. Anal. Calc. for $C_{16}H_{19}NO_9$ (369.32): C, 52.03; H, 5.19; N, 3.79 found: C, 51.97; H, 5.07; N, 3.68%.

Dimethyl 2-[cyclohexylimino)-6-phenyl-2*H*-pyran-3,4-dicarboxylate (6e). Yellow powder, mp 135–137°C, yield: 0.72 g (97%). IR (KBr) (ν_{max}/cm^{-1}): 1730, 1725, 1548 and 1278 cm^{-1}. ^{1}H NMR: δ = 1.34 (2 H, m, CH_2), 1.40 (2 H, m, CH_2), 1.47 (2 H, m, CH_2), 1.66 (2 H, m, CH_2), 1.84 (2 H, m, CH_2), 3.75 (3 H, s, MeO), 3.79 (1 H, m, N-CH), 3.82 (3 H, s, MeO), 6.68 (1 H, s, CH), 7.16 (2 H, t, ^3J = 7.2 Hz, 2 CH), 7.30 (1 H, t, ^3J = 7.5 Hz, CH), 7.63 (2 H, d, ^3J = 7.4 Hz, 2 CH) ppm. ^{13}C NMR: δ = 24.3 (CH_2), 24.5 (CH_2), 25.5 (CH_2), 33.3 (CH_2), 33.5 (CH_2), 51.5 (3 H, s, Me), 52.4 (3 H, s, Me), 56.9 (C—N), 105.8 (CH), 128.3 (2 CH), 129.5 (2 CH), 131.5 (CH), 134.0 (C), 136.4 (C), 140.6 (C), 155.2 (C=N), 158.6 (C), 163.5 (C=O), 165.9 (C=O) ppm. Anal. Calc. for $C_{21}H_{23}NO_5$ (369.41): C, 68.28; H, 6.28; N, 3.79 found: C, 68.32; H, 6.37; N, 3.84%.

Dimethyl 2-(*tert*-butylimino)-6-(4-methylpheny)l-2*H*-pyran-3,4-dicarboxylate (6f). Pale yellow powder, mp 143–145°C, yield: 0.67 g (94%). IR (KBr) (ν_{max}/cm^{-1}): 1732, 1725, 1557, 1410 and 1127 cm^{-1}. ^{1}H NMR: δ = 1.25 (9 H, s, C*Me*$_3$), 2.35 (3 H, s, Me), 3.78 (3 H, s, MeO), 3.83 (3 H, s, MeO), 6.42 (1 H, s, CH), 7.42 (2 H, d, ^3J = 7.6 Hz, 2 CH), 7.84 (2 H, d, ^3J = 7.6 Hz, 2 CH) ppm. ^{13}C NMR: δ = 21.7 (Me), 30.6 (C*Me*$_3$), 51.6 (MeO), 52.4 (MeO), 57.2 (C—N), 108.6 (CH), 125.7 (2 CH), 129.3 (2 CH), 133.2 (C), 134.5 (C), 137.0 (C), 142.4 (C), 155.2 (C=N), 156.8 (C), 163.8 (C=O), 164.7 (C=O) ppm. Anal. Calc. for $C_{20}H_{23}NO_5$ (357.40): C, 67.21; H, 6.49; N, 3.92 found: C, 67.18; H, 6.53; N, 4.02%.

Dimethyl 2-(*tert*-butylimino)-6-(4-nitropheny)l-2*H*-pyran-3,4-dicarboxylate (6g). Yellow powder, mp 152–154°C, yield: 0.70 g (90%). IR (KBr) (ν_{max}/cm^{-1}): 1738, 1732, 1545, 1412 and 1175 cm^{-1}. ^{1}H NMR: δ = 1.27 (9 H, s, C*Me*$_3$), 3.80 (3 H, s, MeO), 3.85 (3 H, s, MeO), 6.48 (1 H, s, CH), 7.87 (2 H, d, ^3J = 8.2 Hz, 2 CH), 8.45 (2 H, d, ^3J = 8.2 Hz, 2 CH) ppm. ^{13}C NMR: δ = 29.8 (C*Me*$_3$), 52.0 (MeO), 52.6 (MeO), 56.8 (C—N), 107.6 (CH), 125.2 (2 CH), 129.8 (2 CH), 133.9 (C), 139.7 (C), 142.9 (C), 146.8 (C), 156.4 (C= C=N), 158.9 (C), 162.4 (C=O), 164.6 (C=O) ppm. MS: *m*/*z* (%) = 388 (M^+, 15), 357 (54), 266 (52), 120 (85), 57 (100), 31(78). Anal. Calc. for $C_{19}H_{20}N_2O_7$ (388.37): C, 58.76; H, 5.19; N, 7.21 found: C, 58.68; H, 5.04; N, 7.14%.

Diethyl 2-(*tert*-butylimino)-6-(4-nitropheny)l-2*H*-pyran-3,4-dicarboxylate (6h). Pale yellow powder, mp 154–156°C, yield: 0.79g (95%). IR (KBr) (ν_{max}/cm^{-1}): 1730, 1727, 1645, 1548 and 1232 cm^{-1}. ^{1}H NMR: δ = 1.27 (9 H, s, C*Me*$_3$), 1.31 (3 H, t, ^3J = 7.4 Hz, Me), 1.37 (3 H, t, ^3J = 7.3 Hz, Me), 4.12 (2 H, q, ^3J = 7.4 Hz, CH_2O), 4.25 (2 H, q, ^3J = 7.3 Hz, CH_2O), 6.45 (1 H, s, CH), 8.05 (2 H, d, ^3J = 8.0 Hz, 2 CH), 8.37 (2 H, d, ^3J = 8.0 Hz, 2 CH) ppm. ^{13}C NMR: δ = 13.8 (Me), 14.2 (Me), 30.4 (C*Me*$_3$), 57.0 (C—N), 61.4 (CH_2O), 62.6 (CH_2O), 107.5 (CH), 126.0 (2 CH), 130.1 (2 CH), 140.2 (C), 141.3 (C), 143.0 (C), 146.5 (C), 157.2 (C=N), 157.6 (C), 162.5 (C=O), 165.3 (C=O) ppm. Anal. Calc. for $C_{21}H_{24}N_2O_7$ (416.43): C, 60.57; H, 5.81; N, 6.73 found: C, 60.49; H, 5.75; N, 6.68%.

Di(*tert*-butyl) 2-[cyclohexylimino)-6-phenyl-2*H*-pyran-3,4-dicarboxylate (6i). Yellow powder, mp 147–149°C, yield: 0.85 g (94%). IR (KBr) (ν_{max}/cm^{-1}): 1728, 1718, 1654, 1568 and 1258 cm^{-1}. ^{1}H NMR: δ = 1.32 (2 H, m, CH_2), 1.38 (2 H, m, CH_2), 1.45 (2 H, m, CH_2), 1.65 (2 H, m, CH_2), 1.67 (9 H, s,C*Me*$_3$), 1.71 (9 H, s,C*Me*$_3$), 1.83 (2 H, m, CH_2), 3.75 (1 H, m, N-CH), 6.48 (1 H, s, CH), 7.68 (2 H, d, ^3J = 7.8 Hz, 2 CH), 7.92 (2 H, d, ^3J = 7.8 Hz, 2 CH) ppm. ^{13}C NMR: δ = 24.5 (CH_2), 24.7 (CH_2), 25.8 (CH_2), 28.2 (C*Me*$_3$), 29.4 (C*Me*$_3$), 34.3 (CH_2), 35.0 (CH_2), 57.0 (C—N), 79.2 (*C*Me$_3$), 82.3 (*C*Me$_3$), 108.3 (CH), 125.6 (2 CH), 129.8 (2 CH), 135.8 (C), 138.5 (C), 142.0 (C), 148.3 (C), 154.8 (C=N), 159.2 (C), 163.4 (C=O), 165.7 (C=O) ppm. Anal. Calc. for $C_{27}H_{33}NO_5$ (453.58): C, 71.50; H, 7.78; N, 3.09 found: C, 71.45; H, 7.70; N, 2.98%.

6-Ethyl 3,4-dimethy-2-(1,1,3,3-tetramethylbutylimino)-2*H*-pyran-3,4,6-tricarboxylate (6j). Yellow powder, mp 137–139°C, yield: 0.73 g (92%). IR (KBr) (ν_{max}/cm^{-1}): 1730, 1722, 1720, 1545, 1425 and 1167 cm^{-1}. ^{1}H NMR: δ = 1.03 (9 H, s, C*Me*$_3$), 1.24 (3 H, t, ^3J = 7.3 Hz, Me), 1.55 (3 H, s, Me), 1.56 (3 H, s, Me), 1.83 (2 H, s, CH_2), 3.74 (3 H, s, MeO), 3.82 (3 H, s, MeO), 4.23 (2 H, q, ^3J = 7.3 Hz, Me), 6.84 (1 H, s, CH) ppm. ^{13}C NMR: δ = 13.9 (Me), 29.7 (C), 29.8 (Me), 31.6 (C*Me*$_3$), 31.9 (Me), 51.5 (MeO), 52.7 (MeO), 55.0 (CH_2), 59.2 (C—N), 61.2 (CH_2O), 109.4 (CH), 139.0 (C), 141.3 (C), 144.5 (C), 158.4 (C), 160.2 (C=O), 161.3 (C=O), 163.5 (C=O) ppm. Anal. Calc. for $C_{20}H_{29}NO_7$ (395.45): C, 60.75; H, 7.39; N, 3.54 found: C, 60.68; H, 7.32; N, 3.43%.

Dimethyl 2-(2-ethoxy-2-oxoethylimino)-6-(4-methylphenyl)-2*H*-pyran-3,4-dicarboxylate (6k). White powder, mp 138–140°C, yield: 0.70 g (90%). IR (KBr) (ν_{max}/cm^{-1}): 1738, 1730, 1728, 1624, 1557, 1410 and 1127 cm^{-1}. ^{1}H NMR: δ = 1.30 (3 H, t, ^3J = 7.2 Hz, Me), 2.35 (3 H, s, Me), 3.76 (3 H,

s, MeO), 3.83 (3 H, s, MeO), 4.20 (2 H, s, CH_2), 4.28 (2 H, q, ^3J = 7.2 Hz, OCH_2), 6.47 (1 H, s, CH), 7.54 (2 H, d, ^3J = 7.5 Hz, 2 CH), 7.85 (2 H, d, ^3J = 7.6 Hz, 2 CH) ppm. ^{13}C NMR: δ = 14.1 (Me), 21.4 (Me), 50.3 (CH_2-N), 51.6 (MeO), 52.4 (MeO), 61.3 (OCH_2), 107.4 (CH), 126.3 (2 CH), 129.7 (2 CH), 134.8 (C), 135.2 (C), 137.3 (C), 142.8 (C), 153.6 (C), 156.5 (C=N), 160.2 (C=O), 162.7 (C=O), 164.3 (C=O) ppm. Anal. Calc. for $C_{20}H_{21}NO_7$ (387.39): C, 62.01; H, 5.46; N, 3.62 found: C, 61.96; H, 5.38; N, 3.54%.

Diethyl 2-(2-ethoxy-2-oxoethylimino)-6-phenyl-2*H*-pyran-3,4-dicarboxylate (6l). White powder, mp 144–146°C, yield: 0.75 g (93%). IR (KBr) (ν_{max}/cm^{-1}): 1732, 1728, 1725, 1612, 1547, 1385 and 1215 cm^{-1}. ^{1}H NMR: δ = 1.23 (3 H, t, ^3J = 7.4 Hz, Me), 1.26 (3 H, t, ^3J = 7.5 Hz, Me), 1.29 (3 H, t, ^3J = 7.3 Hz, Me), 4.12 (2 H, q, ^3J = 7.4 Hz, OCH_2), 4.15 (2 H, q, ^3J = 7.5 Hz, OCH_2), 4.18 (2 H, s, CH_2), 4.20 (2 H, q, ^3J = 7.5 Hz, OCH_2), 6.52 (1 H, s, CH), 7.21 (2 H, t, ^3J = 7.8 Hz, 2 CH), 7.42 (1 H, t, ^3J = 7.6 Hz, CH), 7.79 (2 H, t, ^3J = 7.6 Hz, 2 CH) ppm. ^{13}C NMR: δ = 13.8 (Me), 14.0 (Me), 14.3 (Me), 52.4 (CH_2-N), 60.3 (OCH_2), 61.5 (OCH_2), 62.0 (OCH_2), 108.0 (CH), 128.2 (2 CH), 129.5 (2 CH), 131.4 (CH), 135.0 (C), 135.8 (C), 142.3 (C), 154.8 (C), 158.5 (C=N), 162.3 (C=O), 163.5 (C=O), 164.8 (C=O) ppm. Anal. Calc. for $C_{21}H_{23}NO_7$ (401.41): C, 62.84; H, 5.78; N, 3.49 found: C, 62.75; H, 5.69; N, 3.38%.

REFERENCES AND NOTES

[1] Posner, G. H. Chem Rev 1986, 86, 831.
[2] Tietze, L. F.; Beifuss, U. Angew Chem Int Ed Engl 1993, 32, 131.
[3] Bienayme, H.; Hulme, C.; Oddon, G.; Schmitt, P. Chem Eur J 2000, 6, 3321.
[4] Dömling, A. Curr Opin Chem Biol 2002, 6, 306.
[5] Hulme, C.; Gore, V. Curr Med Chem 2003, 10, 51.
[6] Orru, R. V. A.; DeGreef, M. Synthesis 2003, 1471.
[7] Balme, G.; Bossharth, E.; Monteiro, N. Eur J Org Chem 2003, 4101.
[8] Jacobi von Wangelin, A.; Neumann, H.; Gördes, D.; Klaus, S.; Strübing, D.; Beller, M. Chem Eur J 2003, 9, 4286.
[9] Murakami, M. Angew Chem Int Ed Engl 2003, 42, 718.
[10] Nagendrappa, G. Resonance 2002, 59.
[11] Yavari, I.; Alizadeh, A.; Anary-Abbasinejad, M.; Bijanzadeh, H. R. Tetrahedron 2003, 59, 6083.
[12] Yavari, I.; Anary-Abbasinejad, M.; Alizadeh, A.; Hossaini, Z. Tetrahedron 2003, 59, 1289.