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Reaction of benzoyl chlorides with Huisgen's zwitterion: synthesis of functionalized 2,5-dihydro-1*H*-pyrroles and tetrasubstituted furans

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ABSTRACT

The 1:1 intermediate generated by the addition of alkyl(aryl) isocyanides to dimethyl acetylenedicarboxylate is trapped by benzoyl chloride to yield functionalized 2,5-dihydro-1*H*-pyrroles. The presence of electron-withdrawing groups at the *para* position of benzoyl chloride leads to tetrasubstituted furans. The structures of these products were confirmed by single-crystal X-ray diffraction studies.

1. Introduction

The rich chemistry that stems from the addition of nucleophiles to activated acetylenic compounds has evoked considerable interest. Isocyanides are known to form zwitterions with activated acetylene compounds such as dimethyl acetylenedicarboxylate (DMAD).[1–4] In recent years, work in our laboratory has shown that these types of zwitterions can be trapped by a variety of electrophiles and proton donors, for the synthesis of heterocyclic compounds.[5–9] Hence, it was of interest to investigate the reactivity of the isocyanide–DMAD zwitterion toward benzoyl chlorides.

2. Results and discussion

In this paper, we report the results of our studies involving the reactions of zwitterions derived from isocyanides **1** and dialkyl acetylenedicarboxylates **2** in the presence of benzoyl chlorides **3**, which constitute a synthesis of functionalized 2,5-dihydro-1*H*-pyrroles **4** and tetrasubstituted furans **5**.[10–14] Thus, the 1:1 intermediate generated by the addition of alkyl(aryl) isocyanides to DMAD was trapped by benzoyl chloride to yield dimethyl 1-alkyl-(aryl)-2-hydroxy-5-oxo-2-phenyl-2,5-dihydro-1*H*-pyrrole-3,4-dicarboxylates **4** (Scheme 1). The presence of electron-withdrawing groups at the *para* position of benzoyl chloride leads to dimethyl 2-alkyl(aryl)amino-5-aryl-3,4-furandicarboxylates **5** (Scheme 2). When these reactions were carried out with benzoyl chlorides having electron-donating groups (such as Me or OMe) at the *para* position, complex reaction mixtures were obtained, which was not investigated.

The structures of compounds **4a**–**4d** and **5a**–**5e** were deduced from their elemental analyses and their IR, ^{1}H NMR, and ^{13}C NMR spectroscopies and single-crystal X-ray analyses. For example, the ^{1}H NMR spectrum of **4a** exhibited five singlets identified as methyl (δ 1.12 and 2.43), methoxy (δ 3.86 and 4.03), and OH (δ 4.00) protons, along with multiplets for the aromatic protons. The ^{1}H-decoupled ^{13}C NMR spectrum of **4a** showed 20 distinct resonances that confirm the proposed structure. The presence of two Ar–Me signals in the ^{1}H and ^{13}C NMR spectra of **4a** can be explained in terms of restricted rotation in the dimethylphenyl group. The IR spectrum of **4a** displayed characteristic carbonyl (1740, 1700, 1686 cm^{-1}) and O–H (3365 cm^{-1}) stretching vibrations. The ^{1}H NMR spectrum of **5c** exhibited four singlets identified as *tert*-butyl (δ 1.55), methoxy (δ 3.82 and 4.00), and NH (δ 7.00) protons, along with two doublets (δ 7.64 and 8.25, ^3J=8.9 Hz) for the aromatic region. The ^{13}C NMR spectrum of **5c** showed 14 distinct resonances in agreement with the proposed structure.

Unambiguous evidences for the structures of **4a** and **5d** were obtained from single-crystal X-ray analyses. The ORTEP[15] diagrams of **4a** and **5d** are shown in Figures 1 and 2. The structures were deduced from the crystallographic data and the same were assumed for the other derivatives on account of their NMR spectroscopic similarities.

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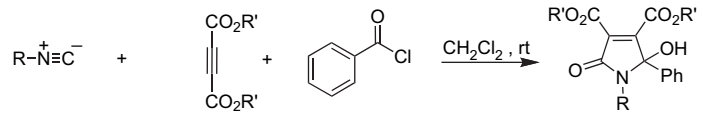


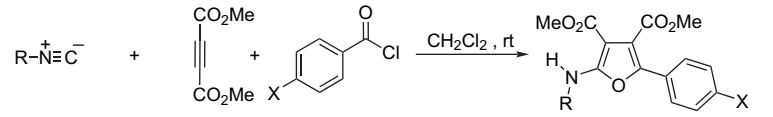
1	R
a	2,6-Dimethylphenyl
b	Cyclohexyl

2	R'
a	Me
b	Et
c	^{t}Bu

3a

4	R	R'	Yield/%
a	2,6-Dimethylphenyl	Me	82
b	Cyclohexyl	Me	75
c	Cyclohexyl	^{t}Bu	65
d	Cyclohexyl	Et	70

Scheme 1. Synthesis of compounds **4**.



1	R
a	^{t}Bu
b	Cyclohexyl
c	1,1,3,3-Tetramethylbutyl
d	2,6-Dimethyl phenyl

2a

3	X
b	Cl
c	NO_2

5	R	X	Yield/%
a	Cyclohexyl	Cl	75
b	2,6-Dimethylphenyl	Cl	78
c	^{t}Bu	NO_2	80
d	Cyclohexyl	NO_2	70
e	1,1,3,3-Tetramethylbutyl	NO_2	72

Scheme 2. Synthesis of compounds **5**.

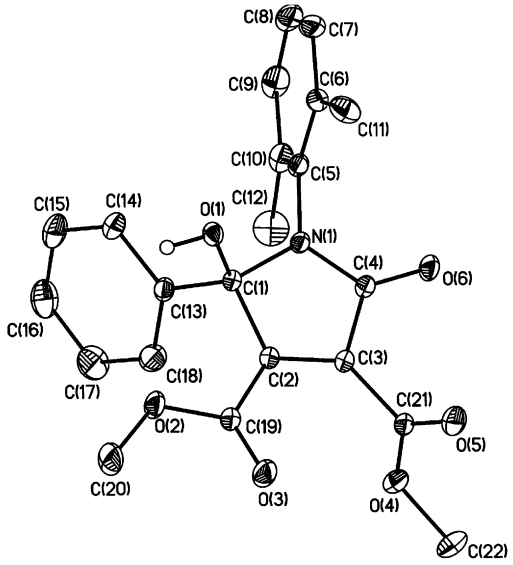


Figure 1. X-ray crystal structure (ORTEP) of **4a**.

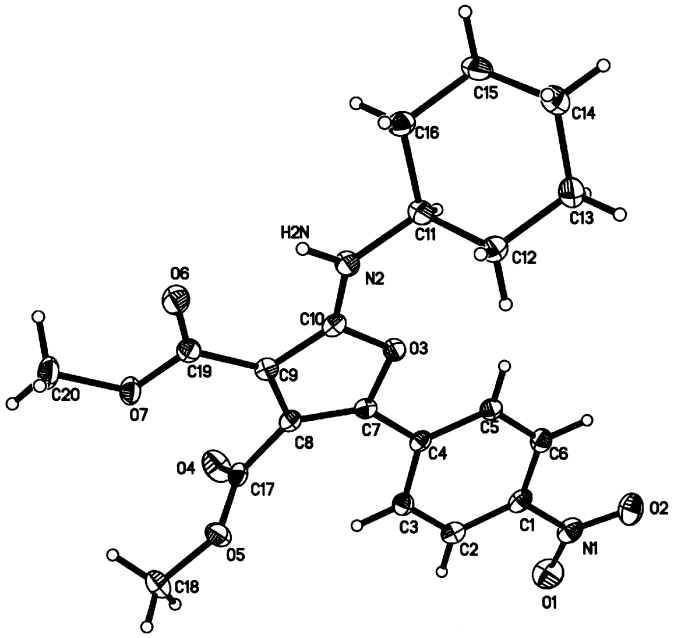


Figure 2. X-ray crystal structure (ORTEP) of **5d**.

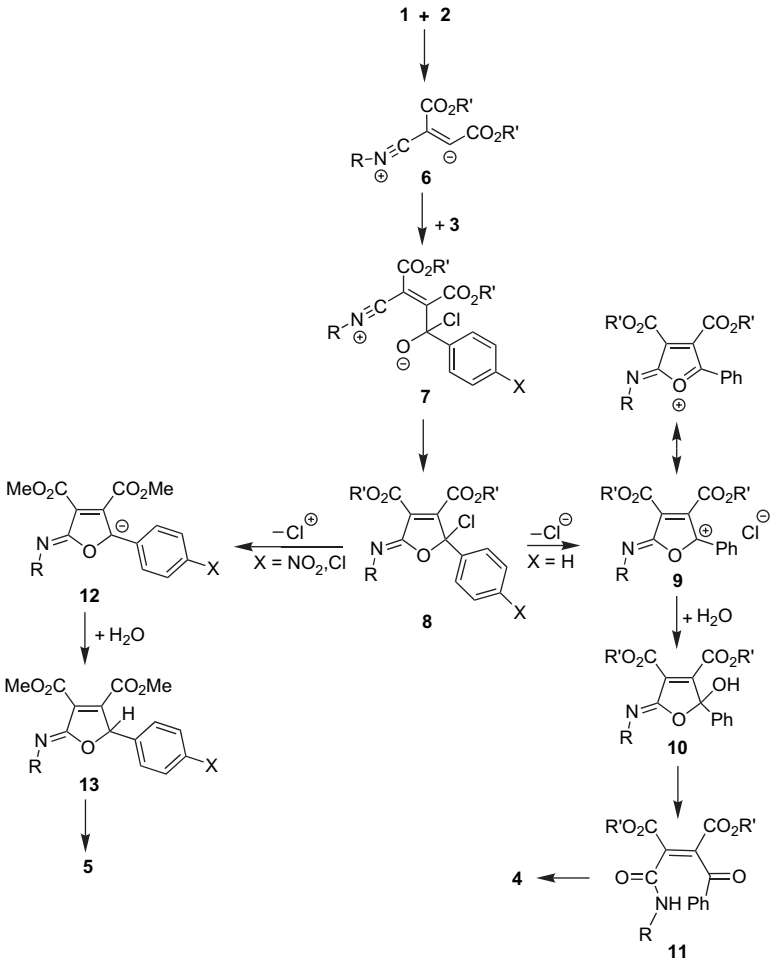
Although the mechanistic details of the reaction are not known, a plausible rationalization may be advanced to explain the product formation. Presumably, the zwitterionic intermediate[16,17] **6** formed from isocyanides and dialkyl acetylenedicarboxylates is attacked by benzoyl chloride to furnish intermediate **7**, which is converted to **8**. This intermediate can lose chloride ion to generate the stabilized cation **9**, which is converted to **10** by absorbing H_2O (presumably from moisture). Intermediate **10** is converted to **4** via the open-chain structure **11** (Scheme 3). However, when X=NO_2 or Cl, intermediate **8** eliminates Cl^+ ion to produce **12**. Nucleophilic attack of H_2O on this intermediate leads to **5** (Scheme 3). Thus, the presence of electron-withdrawing groups at the *para* position of benzoyl chloride changes the reaction pathway and leads to the formation of a different product.

In conclusion, we report novel transformations involving benzoyl chlorides and dialkyl acetylenedicarboxylates in the presence of isocyanides, which afford a new application of benzoyl chlorides for the synthesis of functionalized 2,5-dihydro-1*H*-pyrroles and tetrasubstituted furans. The present procedure has the advantage that not only the reaction is performed under neutral conditions but also the reactants can be mixed without any prior activation or modification. The simplicity of the present procedure makes it an interesting alternative to other approaches.

3. Experimental

3.1. General

The reagents and solvents used in this work were obtained from Fluka and were used without further purification. Mp: Electrothermal-9100 apparatus. IR spectra: Shimadzu IR-460 spectrometer, in



Scheme 3. Proposed mechanism for the formation of compounds **4** and **5**.

cm^{-1}. ^1H and ^{13}C NMR spectra: Bruker DRX-500 AVANCE instrument, in $CDCl_3$ at 500.1 and 125.7 MHz, respectively; δ in parts per million and J in hertz. EIMS: Finnigan-MAT-8430 mass spectrometer, at 70 eV, in m/z. Elemental analyses: Heraeus CHN-O-Rapid analyzer.

3.2. General procedure for the synthesis of functionalized 2,5-dihydro-1*H*-pyrroles (4) and tetrasubstituted furans (5)

To a stirred solution of dialkyl acetylenedicarboxylate (2 mmol) and benzoyl chloride (0.28 g, 2 mmol) in dry CH_2Cl_2 (10 mL) was added a solution of the isocyanide (2 mmol) in dry CH_2Cl_2 (5 mL) at room temperature. The reaction mixture was stirred for 24 h. The solvent was removed under reduced pressure and the residue was separated by silica gel (Merck 230–240 mesh) column chromatography using hexane–EtOAc mixtures as eluent.

*3.2.1. Dimethyl 1-(2,6-dimethylphenyl)-2-hydroxy-5-oxo-2-phenyl-2,5-dihydro-1H-pyrrole-3,4-dicarboxylate (**4a**)*

Colorless crystals, mp 193–195 °C, 0.65 g, yield 82%. IR (KBr) (ν_{max}/cm^{-1}): 3365 (OH), 1740, 1700 and 1686 (C=O), 1272, 1212. MS, m/z (%): 395 (M^+, 18), 120 (8), 105 (42), 77 (10), 59 (28), 58 (100), 44 (70). Anal. Calcd for $C_{22}H_{21}NO_6$ (395.39): C, 66.83; H, 5.35; N, 3.54%. Found: C, 65.95; H, 5.41; N, 3.45%. ^1H NMR: δ 1.12 (3H, s, Me), 2.43 (3H, s, Me), 3.86 (3H, s, OMe), 4.00 (1H, s, OH), 4.03 (3H, s, OMe), 6.83 (m, CH), 7.16 (2H, d, ^3J=7.0 Hz, CH), 7.20 (2H, d, ^3J=7.5 Hz, CH), 7.29 (2H, t, ^3J=8.0 Hz, CH), 7.37 (1H, t, ^3J=7.0 Hz, CH). ^{13}C NMR: δ 18.0 (Me), 20.0 (Me), 53.6 (OMe), 53.7 (OMe), 93.1 (C–N), 126.3 (2CH), 128.5 (CH), 129.0 (2CH), 129.1 (CH), 129.2 (CH), 130.0 (CH), 131.1 (C), 136.2 (C), 137.7 (C), 139.6 (C), 140.3 (C), 146.5 (C), 162.5 (C=O), 162.6 (C=O), 163.2 (C=O).

*3.2.1.1. X-ray crystal structural determination of **4a**.* Structural determination and refinement data: formula, $C_{22}H_{21}NO_6$, M_r 395.40; crystal size, $0.10\times0.10\times0.10$ mm^3; crystal system, monoclinic, a=8.2989(4), b=18.1793(8), c=13.7544(6) Å, β=94.6380(10); space group, $P2_1/c$; Z=4, V=2068.17(16) $Å^3$, D_{calcd}=1.270 g cm^{-3}; R=0.0412 (for 5179 reflections), R_w=0.1102; $-11\le h\le 11$, $-25\le k\le 25$, $-19\le l\le 19$; Mo Kα radiation (λ=0.71073 Å); T=100(2) K. The crystallographic data of **1a** have been deposited with the *Cambridge Crystallographic Data Centre* as supplementary publication number CCDC-647813. Copies of the data can be obtained, free of charge, via the internet (http://www.ccdc.cam.ac.uk/data_request/cif), e-mail (data_request@ccdc.cam.ac.uk) or fax (+44 1223 336033).

*3.2.2. Dimethyl 1-cyclohexyl-2-hydroxy-5-oxo-2-phenyl-2,5-dihydro-1H-pyrrole-3,4-dicarboxylate (**4b**)*

Colorless crystals, mp 167–170 °C, 0.56 g, yield 75%. IR (KBr) (ν_{max}/cm^{-1}): 3450 (OH), 1722, 1701 and 1681 (C=O), 1262, 1206. Anal. Calcd for $C_{20}H_{23}NO_6$ (373.38): C, 64.33; H, 6.21; N, 3.75%. Found: C, 64.48; H, 6.27; N, 3.83%. ^1H NMR: δ 0.88–0.96 (1H, m, CH), 1.06–1.24 (3H, m, CH), 1.50–1.52 (1H, m, CH), 1.58–1.61 (1H, m, CH), 1.71–1.75 (2H, m, CH), 1.99–2.10 (2H, m, CH), 3.09–3.14 (1H, m, CH), 3.64 (3H, s, OMe), 3.92 (3H, s, OMe), 4.98 (1H, s, OH), 7.35–7.39 (3H, m, CH), 7.50 (2H, d, ^3J=7.0 Hz, CH). ^{13}C NMR: δ 25.5

(CH_2), 26.4 (CH_2), 26.5 (CH_2), 29.4 (CH_2), 30.5 (CH_2), 53.1 (CH), 53.4 (OMe), 53.8 (OMe), 91.8 (C–N), 126.5 (2CH), 128.8 (2CH), 129.4 (CH), 134.8 (C), 135.8 (C), 148.3 (C), 161.8 (C=O), 162.7 (C=O), 163.8 (C=O).

*3.2.3. Di(tert-butyl) 1-cyclohexyl-2-hydroxy-5-oxo-2-phenyl-2,5-dihydro-1H-pyrrole-3,4-dicarboxylate (**4c**)*

White powder, mp 191–193 °C, 0.59 g, yield 65%. IR (KBr) (ν_{max}/cm^{-1}): 3390 (OH), 1705, 1700 and 1680 (C=O), 1290, 1163. Anal. Calcd for $C_{26}H_{35}NO_6$ (457.54): C, 68.25; H, 7.71; N, 3.06%. Found: C, 68.41; H, 7.79; N, 3.11%. ^{1}H NMR: δ 0.89–0.93 (1H, m, CH), 1.06–1.12 (2H, m, CH), 1.26 (9H, s, CMe_3), 1.34–1.51 (3H, m, CH), 1.57 (9H, s, CMe_3), 1.70–1.80 (2H, m, 2CH), 2.08–2.15 (2H, m, 2CH), 3.01–3.08 (m, 1H, CH), 6.07 (s, 1H, OH), 7.39 (1H, t, ^3J=7.0 Hz, CH), 7.43 (2H, t, ^3J=7.0 Hz, 2CH), 7.52 (2H, d, ^3J=7.5 Hz, 2CH). ^{13}C NMR: δ 24.5 (CH_2), 25.0 (CH_2), 25.5 (CH_2), 28.2 (CMe_3), 28.6 (CMe_3), 29.6 (CH_2), 31.4 (CH_2), 51.3 (CH), 84.3 (CMe_3), 84.5 (CMe_3), 91.6 (C–N), 126.4 (2CH), 128.8 (2CH), 129.2 (CH), 135.3 (C), 136.8 (C), 146.4 (C), 161.0 (C=O), 161.4 (C=O), 164.5 (C=O).

*3.2.4. Diethyl 1-cyclohexyl-2-hydroxy-5-oxo-2-phenyl-2,5-dihydro-1H-pyrrole-3,4-dicarboxylate (**4d**)*

Colorless crystals, mp 140–142 °C, 0.56 g, yield 70%. IR (KBr) (ν_{max}/cm^{-1}): 3440 (OH), 1720, 1703 and 1681 (C=O), 1260, 1201. Anal. Calcd for $C_{22}H_{27}NO_6$ (401.44): C, 65.82; H, 6.78; N, 3.49%. Found: C, 65.93; H, 6.82; N, 3.54%. ^{1}H NMR: δ 0.87–0.90 (1H, m, CH), 1.13–1.15 (1H, m, CH), 1.14 (3H, t, ^3J=7.2 Hz, 3CH), 1.37 (3H, t, ^3J=7.2 Hz, 3CH), 1.18–1.21 (2H, m, CH), 1.66–1.69 (2H, m, CH), 1.70–1.72 (2H, m, CH), 2.01–2.05 (2H, m, CH), 3.08–3.11 (1H, m, CH), 3.76 (1H, s, OH), 4.08–4.19 (2H, m, CH), 4.40 (2H, q, ^3J=7.2 Hz, CH), 7.35–7.40 (3H, m, CH), 7.48 (2H, d, ^3J=7.1 Hz, CH). ^{13}C NMR: δ 13.7 (CH_3), 14.1 (CH_3), 25.1 (CH_2), 26.0 (CH_2), 26.1 (CH_2), 29.1 (CH_2), 30.3 (CH_2), 53.6 (CH), 62.1 (OCH_2), 62.2 (OCH_2), 91.3 (C–N), 125.9 (2CH), 128.6 (2CH), 129.0 (CH), 135.4 (C), 135.8 (C), 145.6 (C), 161.1 (C=O), 161.7 (C=O), 163.5 (C=O).

3.3. Dimethyl 2-(4-chlorophenyl)-5-(cyclohexylamino)-3,4-furandicarboxylate (5a)

Light yellow crystals; mp 157–160 °C (lit. 18, reported as an oil), 0.59 g, yield 75%. IR (KBr) (ν_{max}/cm^{-1}): 3275 (NH), 1738 (C=O), 1685 (C=O), 1365, 1268, 1171, 742. Anal. Calcd for $C_{20}H_{22}ClNO_5$ (391.84): C, 61.30; H, 5.66; N, 3.57%. Found: C, 60.88; H, 5.60; N, 3.62%. ^{1}H NMR: δ 0.94–0.97 (1H, m, CH), 1.07–1.10 (1H, m, 1CH), 1.19–1.25 (2H, m, 2CH), 1.24–1.40 (1H, m, 1CH), 1.52–1.54 (1H, m, 1CH), 1.59–1.64 (1H, m, 1CH), 1.71–1.79 (2H, m, 2CH), 2.10–2.14 (1H, m, 1CH), 3.04–3.09 (1H, m, CH), 3.63 (3H, s, OMe), 3.87 (3H, s, OMe), 6.33 (1H, br, NH), 7.46 (2H, d, ^3J=8.5 Hz, 2CH), 7.55 (2H, d, ^3J=8.5 Hz, 2CH). ^{13}C NMR: δ 25.5 ($2CH_2$), 26.2 ($2CH_2$), 26.3 (CH_2), 52.3 (OMe), 52.5 (OMe), 53.2 (CH), 91.21 (N–C=C), 128.6 (2CH), 128.7 (2CH), 134.4 (C), 136.0 (CH), 137.3 (C), 146.2 (O–C=C), 160.7 (NCO), 162.6 (C=O), 163.1 (C=O).

*3.3.1. Dimethyl 2-(4-chlorophenyl)-5-[(2,6-dimethylphenyl)-amino]-3,4-furandicarboxylate (**5b**)*

Light brown crystals; mp 149–151 °C, 0.64 g, yield 78%. IR (KBr) (ν_{max}/cm^{-1}): 3149 (NH), 1725 (C=O), 1684 (C=O), 1339, 1250, 1090, 848. Anal. Calcd for $C_{22}H_{20}ClNO_5$ (413.85): C, 63.85; H, 4.87; N, 3.38%. Found: C, 63.92; H, 4.74; N, 3.42%. ^{1}H NMR: δ 1.20 (3H, s, CH_3), 2.40 (3H, s, CH_3), 3.75 (3H, s, OMe), 3.94 (3H, s, OMe), 6.48 (1H, br, NH), 6.86–6.87 (1H, m, CH), 7.11–7.17 (2H, m, 2CH), 7.25 (2H, d, ^3J=8.7 Hz, 2CH), 7.33 (2H, d, ^3J=8.7 Hz, 2CH). ^{13}C NMR: δ 18.3 (CH_3), 20.1 (CH_3), 53.1 (OMe), 53.2 (OMe), 93.4 (N–C=C), 128.8 (2CH), 128.9 (1CH), 129.0 (2CH), 129.4 (CH), 129.5 (CH), 132.4 (C), 135.2 (C), 137.4 (C), 138.2 (C), 139.8 (C), 140.6 (C), 146.8 (O–C=C), 161.6 (NCO), 163.0 (C=O), 163.1 (C=O).

*3.3.2. Dimethyl 2-(1,1-dimethylethyl)amino-5-(4-nitrophenyl)-3,4-furandicarboxylate (**5c**)*

Light brown crystals, mp 135–137 °C, 0.60 g, yield 80%. IR (KBr) (ν_{max}/cm^{-1}): 3165 (NH), 1728 (C=O), 1667 (C=O), 1327, 1245, 1091, 843. Anal. Calcd for $C_{18}H_{20}N_2O_7$ (376.34): C, 57.44; H, 5.36; N, 7.44%. Found: C, 57.51; H, 5.40; N, 7.48%. ^{1}H NMR: δ 1.55 (9H, s, CMe_3), 3.82 (3H, s, OMe), 4.00 (3H, s, OMe), 7.00 (br, NH), 7.64 (2H, d, ^3J=8.9 Hz, 2CH), 8.25 (2H,d, ^3J=8.9 Hz, 2CH). ^{13}C NMR: δ 30.1 (CMe_3), 51.8 (CMe_3), 53.5 (OMe), 53.6 (OMe), 90.0 (N–C=C), 117.9 (C), 124.3 (2CH), 124.8 (2CH), 135.4 (C), 138.7 (C), 146.4 (O–C=C), 162.3 (NCO), 165.0 (C=O), 165.9 (C=O).

*3.3.3. Dimethyl 2-(cyclohexylamino)-5-(4-nitrophenyl)-3,4-furandicarboxylate (**5d**)*

Light brown crystals; mp 176–178 °C, 0.56 g, yield 70%. IR (KBr) (ν_{max}/cm^{-1}): 3140 (NH), 1722 (C=O), 1671 (C=O), 1327, 1224, 1103, 841. Anal. Calcd for $C_{20}H_{22}N_2O_7$ (402.38): C, 59.69; H, 5.51; N, 6.96%. Found: C, 59.78; H, 5.60; N, 6.98%. ^{1}H NMR: δ 1.25–1.32 (1H, m, CH), 1.38–1.47 (4H, m, 4 CH), 1.65–1.68 (1H, m, CH), 1.79–1.82 (2H, m, 2CH), 2.05–2.07 (2H, m, 2CH), 3.75–3.77 (1H, m, CH), 3.79 (3H, s, OMe), 3.95 (3H, s, OMe), 6.73 (1H, d, ^3J=7.8 Hz, NH), 7.60 (2H, d, ^3J=8.8 Hz, 2CH), 8.20 (2H, d, ^3J=8.8 Hz, 2CH). ^{13}C NMR: δ 24.2 ($2CH_2$), 25.3 (CH_2), 33.4 ($2CH_2$), 51.3 (OMe), 51.7 (OMe), 52.9 (CH), 88.8 (N–C=C), 117.9 (C), 124.0 (2CH), 124.2 (2CH), 135.0 (C), 137.9 (C), 146.0 (O–C=C), 161.6 (NCO), 164.4 (C=O), 165.3 (C=O).

*3.3.3.1. X-ray crystal structural determination of **5d**.* Structural determination and refinement data: formula, $C_{20}H_{22}N_2O_7$, M_r 402.40; crystal size, 0.55×0.40×0.25 mm^3; crystal system, triclinic, a=6.8628(5), b=11.5872(9), c=12.6526(9) Å, β=80.4570(10); space group, $P\bar{1}$; Z=2, V=945.10(12) $Å^3$, D_{calcd}=1.414 g cm^{-3}; R=0.0412 (for 3857 reflections), R_w=0.0843; $-9\le h\le 9$, $-16\le k\le 16$, $-17\le l\le 17$; Mo Kα radiation (λ=0.71073 Å); T=100(2) K. The crystallographic data of **1a** have been deposited with the *Cambridge Crystallographic Data Centre* as supplementary publication number CCDC-628362. Copies of the data can be obtained, free of charge, via the internet (http://www.ccdc.cam.ac.uk/data_request/cif), e-mail (data_request@ccdc.cam.ac.uk) or fax (+44 1223 336033).

*3.3.4. Dimethyl 2-(4-chlorophenyl)-5-[(1,1,3,3-tetramethylbutyl)amino]-3,4-furandicarboxylate (**5e**)*

Light brown crystals, mp 98–100 °C, 0.62 g, yield 72%. IR (KBr) (ν_{max}/cm^{-1}): 3240 (NH), 1725 (C=O), 1669 (C=O), 1325, 1211, 1100, 839. Anal. Calcd for $C_{22}H_{28}N_2O_7$ (432.44): C, 59.69; H, 5.51; N, 6.96%. Found: C, 59.41; H, 5.56; N, 7.01%. ^{1}H NMR: δ 1.03 (9H, s, CMe_3), 1.25 (2H, s, CH_2), 1.56 (6H, s, CMe_2), 3.78 (3H, s, OMe), 3.95 (3H, s, OMe), 7.08 (1H, br, NH), 7.59 (2H, d, ^3J=8.8 Hz, 2CH), 8.21 (2H, d, ^3J=8.8 Hz, 2CH). ^{13}C NMR: δ 29.6 (CMe_2), 31.4 (CMe_3), 31.7 (CMe_3), 51.3 (OMe), 52.9 (OMe), 53.4 (CMe_2), 56.7 (CH_2), 89.4 (N–C=C), 117.5 (C), 123.7 (2CH), 124.3 (2CH), 134.9 (C), 138.1 (C), 145.8 (O–C=C), 161.7 (NCO), 164.5 (C=O), 165.4 (C=O).

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