A Facile One-pot Synthesis of Novel Substituted 1,2,3,4-Tetrahydropyrimidine, Part 2[1]: Synthesis of 1-(Aralkyl/aryl)-3-(alkyl/aralkyl/aryl)-5-aroyl-1,2,3,4-tetrahydropyrimidines

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1-(Aralkyl/aryl)-3-(alkyl/aralkyl)-5-aroyl-1,2,3,4-tetrahydropyrimidines (**2a-c**) have been synthesized by dethiomethylation of 5-aroyl-6-methylthio-1,2,3,4-tetrahydropyrimidines (**1a-c**). An alternative one-pot synthetic strategy has been developed for the title compounds **2a-t** by the reaction of enaminones **3** with primary amine and formaldehyde in refluxing methanol in good yields.

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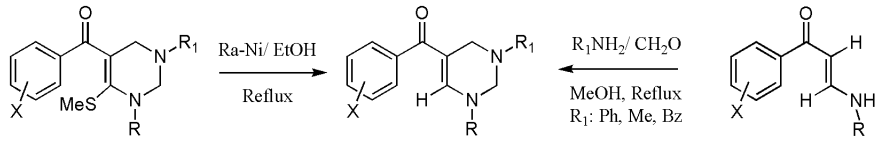
In connection with our on-going program on the development of synthetic strategies for 1,2,3,4-tetrahydropyrimidines [1,2], we have recently reported a facile one-pot method for the synthesis of 1-(aralkyl/aryl)-3-(alkyl/aralkyl/aryl)-5-benzoyl-6-methylthio-1,2,3,4-tetrahydropyrimidines (**1**) [1]. We envisaged that the absence of thiomethyl group in position 6 of the pyrimidine ring of **2** could have a positive impact on the biological properties of the molecules. Our literature survey at this stage revealed that 5-benzoyl-1,3-substituted-1,2,3,4-tetrahydropyrimidines are unknown in the literature. Thus, we undertook to develop methodologies for their synthesis and the results of our synthetic studies are reported herein.

In order to achieve the synthesis of 1-(aralkyl/aryl)-3-(alkyl/aralkyl/aryl)-5-aroyl-1,2,3,4-tetrahydropyrimidines (**2**), we planned to carry out dethiomethylation of **1** (Scheme). Thus, when an ethanolic solution of **1a** was refluxed with Raney-Nickel [3] for 20 hours, work-up of the reaction mixture followed by column chromatography yielded the corresponding desired product **2a** in 50% yields. The structure of **2a** was established on the basis of spectral and analytical data. Dethiomethylation of **1b** and **1c** proceeded smoothly under identical conditions giving corresponding **2b** and **2c** in 52 and 55% yields respectively.

Scheme



1

	X	R	R_1
a	4-Me	$PhCH_2$	$PhCH_2$
b	4-Cl	Ph	Et
c	4-MeO	Ph	$PhCH_2$

2

	X	R	R_1
a	4-Me	$PhCH_2$	$PhCH_2$
b	4-Cl	Ph	Et
c	4-MeO	Ph	$PhCH_2$
d	4-MeO	Ph	Ph
e	H	Ph	Ph
f	H	Ph	$PhCH_2$
g	H	Ph	Me
h	H	4-MePh	Ph
i	H	4-MePh	$PhCH_2$
j	H	$PhCH_2$	Ph
k	H	$PhCH_2$	$PhCH_2$
l	4-Me	Ph	Ph
m	4-Cl	Ph	Ph
n	4-Cl	Ph	Me
o	4-Cl	4-MePh	$PhCH_2$
p	4-Cl	4-MePh	Ph
q	4-Cl	Ph	$PhCH_2$
r	4-MeO	4-MePh	$PhCH_2$
s	3-MeO	Ph	Ph
t	3-MeO	Ph	$PhCH_2$

3

	X	R
a	4-Me	$PhCH_2$
b	4-Cl	Ph
c	4-MeO	Ph
d	3-MeO	Ph
e	H	Ph
f	H	4-MePh
g	H	$PhCH_2$
h	4-Me	Ph
i	4-Cl	4-MePh
j	4-MeO	4-MePh

Table 1

Preparation of 1-(Aralkyl/ aryl)-3-(alkyl/aralkyl/aryl)-5-aroyl-1,2,3,4-tetrahydropyrimidines (**2a-t**)

Comp.	X	R	R_1	Reaction time, hrs	Yield (%)	Mp °C	Molecular formula	Analysis (%) Calcd/Found C	H	N
2a	4-Me	$C_6H_5CH_2$	$C_6H_5CH_2$	5	79	114-115	$C_{26}H_{26}N_2O$	81.64	6.85	7.32
								81.43	6.78	7.40
2b	4-Cl	C_6H_5	C_2H_5	3.5	50	gum	$C_{19}H_{19}ClN_2O$	69.83	5.86	8.57
								70.03	5.92	8.51
2c	4-MeO	C_6H_5	$C_6H_5CH_2$	24	64	106-108	$C_{25}H_{24}N_2O_2$	78.09	6.29	7.28
								78.30	6.22	7.23
2d	4-MeO	C_6H_5	C_6H_5	53	41	110-111	$C_{24}H_{22}N_2O_2$	77.81	5.98	7.56
								77.60	5.92	7.63
2e	H	C_6H_5	C_6H_5	16	72	160-161	$C_{23}H_{20}N_2O$	81.15	5.92	8.22
								81.35	5.99	8.17
2f	H	C_6H_5	$C_6H_5CH_2$	24	83	122-123	$C_{24}H_{22}N_2O$	81.33	6.25	7.90
								81.48	6.20	7.97
2g	H	C_6H_5	Me	7	60	138-139	$C_{18}H_{18}N_2O$	77.67	6.51	10.06
								77.60	6.45	10.10
2h	H	4-MeC_6H_4	C_6H_5	7	50	125-126	$C_{24}H_{22}N_2O$	81.30	6.25	7.90
								81.11	6.31	7.98
2i	H	4-MeC_6H_4	$C_6H_5CH_2$	5	52	92-93	$C_{25}H_{24}N_2O$	81.49	6.56	7.60
								81.70	6.63	7.52
2j	H	$C_6H_5CH_2$	C_6H_5	29	60	151-152	$C_{24}H_{22}N_2O$	81.30	6.25	7.90
								81.06	6.16	7.98
2k	H	$C_6H_5CH_2$	$C_6H_5CH_2$	23	72	105-106	$C_{25}H_{24}N_2O$	81.49	6.56	7.60
								81.30	6.49	7.51
2l	4-Me	C_6H_5	C_6H_5	23	60	143-144	$C_{24}H_{22}N_2O$	81.30	6.25	7.90
								81.52	6.33	7.97
2m	4-Cl	C_6H_5	C_6H_5	20	60	139-141	$C_{23}H_{19}ClN_2O$	73.69	5.10	7.47
								73.50	5.03	7.40
2n	4-Cl	C_6H_5	Me	2	35	92-93	$C_{18}H_{17}ClN_2O$	69.12	5.48	8.96
								68.95	5.40	9.02
2o	4-Cl	4-MeC_6H_4	$C_6H_5CH_2$	6	72	114-116	$C_{25}H_{23}ClN_2O$	74.52	5.75	6.95
								74.77	5.66	7.01
2p	4-Cl	4-MeC_6H_4	C_6H_5	9	62	118-119	$C_{24}H_{21}ClN_2O$	74.12	5.44	7.20
								73.99	5.49	7.28
2q	4-Cl	C_6H_5	$C_6H_5CH_2$	27	51	124-126	$C_{24}H_{21}ClN_2O$	74.12	5.44	7.20
								73.90	5.52	7.13
2r	4-MeO	4-MeC_6H_4	$C_6H_5CH_2$	3.5	61	104-105	$C_{26}H_{26}N_2O_2$	78.36	6.58	7.03
								78.41	6.66	6.93
2s	3-MeO	C_6H_5	C_6H_5	6	68	112-114	$C_{24}H_{22}N_2O_2$	77.80	5.98	7.56
								78.02	5.90	7.64
2t	3-MeO	C_6H_5	$C_6H_5CH_2$	24	79	110-111	$C_{25}H_{24}N_2O_2$	78.09	6.29	7.29
								78.17	6.36	7.37

However, in order to achieve the synthesis of the tetrahydropyrimidines in a single step, we then planned to design an alternative synthetic strategy using enaminones of the type **3** as starting materials. To examine the efficacy of the strategy, enaminones **3a-j** were synthesized and then reacted with primary amines and formaldehyde. Thus, when a mixture of **3a**, formaldehyde and benzylamine (1:2:1) was refluxed in methanol for 6 hours, work-up of the reaction mixture yielded **2a** in 79% yields, which was characterized as 1,3-dibenzyl-5-(4-methylbenzoyl)-1,2,3,4-tetrahydropyrimidine. The reaction was found to be general with other alkyl, aralkyl and aryl amines and with corresponding **3b-j** to give the respective **2b-t** in 50-83% yields, except in **2d** and **2n** which were obtained in 41% and 35% yields respectively. The structures of the products were established on the basis of spectral and analytical data. Thus, the infrared spectra of **2a-t** showed a strong absorption band in the range 1491-1641cm^{-1} due to highly delocalized double bonds and carbonyl group stretching frequencies of enaminone functionalities. The ^{1}H nmr spectra of **2a** and **2k** exhibited a singlet due to the methylene protons at C-2 between 4.25-4.58 ppm while the methylene protons at C-4 appeared as singlet between 3.93-3.98 ppm. The methylene protons of benzylic groups in these molecules appeared as singlets in the range of 3.65-3.85 ppm. It is interesting to note that in the ^{1}H nmr

Table 2

Spectral Data for Compounds **2a-t**.

Comp.	IR (KBr) cm^{-1}	^1H NMR δ (ppm)	^{13}C NMR δ (ppm)	MS
2a	1583, 1607, 1624	2.40 (s, 3H), 3.65 (s, 2H), 3.83 (s, 2H), 3.93 (s, 2H), 4.25 (s, 2H), 7.13-7.76 (m, 15H)	-	383 (MH^+)
2b	1562, 1589, 1641	1.17 (t, 3H), 2.67 (q, 2H), 3.83 (s, 2H), 4.56 (s, 2H), 6.93-6.96 (m, 2H), 7.10-7.15 (m, 1H), 7.26-7.52 (m, 7H)		-
2c	1560, 1579, 1607	3.75 (s, 2H), 3.84 (s, 3H), 3.90 (s, 2H), 4.48 (s, 2H), 6.88-7.09 (m, 5H), 7.25-7.29 (m, 6H), 7.59-7.63 (m, 4H)	48.4, 55.3, 56.9, 66.5, 109.5, 113.4, 118.2, 123.9, 127.4, 128.4, 129.1, 129.6, 130.4, 132.3, 137.6, 144.2, 144.4, 161.4, 193.1	-
2d	1580, 1601, 1619	3.86 (s, 3H), 4.53 (s, 2H), 5.23 (s, 2H), 6.70-7.90 (m, 15H)	47.2, 55.3, 65.4, 110.9, 113.4, 117.8, 118.4, 121.1, 124.2, 129.3, 129.8, 130.5, 139.0, 144.0, 145.1, 149.1, 161.5, 193.4	-
2e	1576, 1594, 1619	4.50 (s, 2H), 5.15 (s, 2H), 6.88-6.99 (m, 5H), 7.10-7.26 (m,3H), 7.31-7.44 (m, 5H), 7.51-7.56 (m, 3H)	47.1, 65.5. 110.8, 117.8, 118.5, 121.2, 124.5, 128.2, 128.4129.3, 129.8, 130.3, 139.5, 143.9, 146.0, 148.4, 193.4	-
2f	1491, 1561, 1579, 1603	3.76 (s, 2H), 3.92 (s, 2H), 4.49 (s, 2H), 6.87-6.90 (m, 2H), 7.09-7.11 (m, 1H), 7.26-7.32 (m, 7H), 7.41-7.46 (m, 3H), 7.57-7.60 (m, 3H)	48.3, 57.0, 66.4, 109.4, 118.4, 119.6, 124.1, 127.5, 128.2, 128.4, 129.1, 129.6, 130.2, 137.6, 139.8, 144.4, 145.1, 194.0	-
2g	1582, 1607, 1618	2.52 (s, 3H), 3.78 (s, 2H), 4.48 (s, 2H), 6.93-6.96 (m, 2H), 7.08-7.13 (m, 1H), 7.26-7.48 (m, 5H), 7.55-7.58 (m, 3H)	-	279 (MH^+)
2h	1590**,** 1597, 1622	2.33 (s, 3H), 4.59 (s, 2H), 5.22 (s, 2H), 6.86-7.79 (m, 15H)	20.7, 47.0, 65.7, 110.3, 117.8, 118.8, 121.1, 126.6, 128.1, 128.4, 128.9, 129.3, 129.8, 130.2, 130.3, 146.4, 161.9, 193.0	-
2i	1560, 1587, 1618	2.33 (s, 3H), 3.83 (s, 2H), 3.96 (s, 2H), 4.53 (s, 2H), 6.70-7.06 (m, 3H), 7.13-7.90 (m, 12H)	20.7, 48.2, 57.0, 66.8, 108.8, 118.6, 127.4, 128.1, 128.4, 129.1, 130.0, 130.1, 134.1, 137.7, 139.9, 142.0, 145.5, 193.7	-
2j	1562, 1584, 1598, 1632	4.35 (s, 2H), 4.43 (s, 2H), 4.65 (s, 2H), 6.86-7.16 (m, 2H), 7.20-7.83 (m, 14H)	-	355 (MH^+)
2k	1567, 1589, 1600, 1617	3.70 (s, 2H), 3.85 (s, 2H), 3.98 (s, 2H), 4.28 (s, 2H), 7.15-7.80 (m, 16H)		
2l	1559, 1573, 1621	2.38 (s, 3H), 4.49 (s, 2H), 5.15 (s, 2H), 6.88-6.99 (m, 5H), 7.12-7.36 (m, 8H), 7.44-7.53 (m, 2H)	21.4, 47.1, 65.4, 110.6, 117.8, 118.5, 121.1, 124.3, 128.6, 128.8, 129.3, 129.8, 130.0, 139.3, 143.5, 145.6, 146.1, 193.2	-
2m	1556, 1572, 1589, 1619	4.48 (s, 2H), 5.16 (s, 2H), 6.88-6.98 (m, 5H), 7.12-7.26 (m, 4H), 7.33-7.38 (m, 3H), 7.46-7.50 (m, 3H)	47.0, 65.5, 110.6, 114.0, 117.8, 118.6, 121.2, 124.7, 128.4, 129.3, 129.8, 137.8, 143.8, 145.9, 147.9, 148.3, 191.9	376 (MH^+)
2n	1555, 1572, 1618	2.51(s, 3H), 3.76 (s, 2H), 4.48 (s, 2H), 6.94-6.96 (m, 2H), 7.11-7.16 (m, 1H), 7.32-7.40 (m, 4H), 7.50-7.53 (m, 3H)	41.0, 49.5, 69.5, 109.2, 118.7, 124.5, 128.4, 129.7, 129.8, 136.6, 138.0, 144.3. 144.7, 192.5	314 (MH^+)
2o	1516, 1553, 1572, 1586	2.36 (s, 3H), 3.83 (s, 2H), 3.98 (s, 2H), 4.56 (s, 2H), 6.76-7.06 (m, 2H), 7.10-7.90 (m, 12H)	20.7, 48.1, 57.0, 66.9, 108.6, 118.7, 127.5, 128.4, 128.5, 129.0, 129.8, 130.2, 134.3, 136.1, 137.5, 138.2, 141.9, 145.5, 192.2	-
2p	1512, 1559, 1573, 1584	2.36 (s, 3H), 4.53 (s, 2H), 5.20 (s, 2H), 6.83-7.80 (m, 14H)	20.7, 47.0, 65.7, 110.1, 116.1, 117.8, 118.6, 118.9, 121.2, 128.4, 129.3, 129.8, 130.4, 134.7, 141.5, 146.3, 148.4, 162.8, 192.8	-
2q	1563, 1580, 1607	3.80 (s, 2H), 3.93 (s, 2H), 4.53 (s, 2H), 6.90-7.10 (m, 2H), 7.20-7.86 (m, 13H)	48.1, 56.9, 66.7, 109.2, 118.4, 124.3, 127.5, 128.4, 129.0, 129.6, 129.8, 136.2, 137.4, 138.1, 144.2, 145.1, 192.4	-
2r	1513, 1557, 1587, 1602	2.35 (s, 3H), 3.80 (s, 2H), 3.92 (s, 3H), 4.00 (s, 2H), 4.55 (s, 2H), 6.78-7.60 (m, 11H), 7.70-7.96 (m, 3H)	20.6, 48.4, 55.3, 56.9, 66.7, 108.9, 113.4, 118.5, 127.4, 128.4, 129.1, 130.4, 131.1 132.4, 133.8, 137.7, 142.2, 144.7, 161.3, 193.0	-
2s	1562, 1584, 1596, 1622	3.82 (s, 3H), 4.49 (s, 2H), 5.15 (s, 2H), 6.88-6.99 (m, 5H), 7.08-7.16 (m, 3H), 7.21-7.37 (m, 6H), 7.55 (s, 1H)	47.0, 55.3, 65.5, 110.7, 113.3, 116.4, 117.8, 118.5, 120.8, 121.1, 124.5, 129.0, 129.3, 129.8, 140.9, 143.9, 145.9, 148.4, 159.5, 193.0	-
2t	1554, 1576, 1602, 1617	3.76 (s, 2H), 3.84 (s, 3H), 3.91 (s, 2H), 4.49 (s, 2H), 6.88-6.91 (m, 2H), 6.98-7.02 (m, 1H), 7.07-7.16 (m, 3H), 7.24-7.34 (m, 9H)	48.2, 55.3, 57.0, 66.7, 109.3, 113.3, 116.3, 118.4, 120.8, 124.8, 127.4, 128.1, 128.4, 129.0, 129.6, 137.6, 141.2, 144.3, 145.1, 159.5, 193.6	-

spectra of **2d, 2e, 2h, 2l, 2m, 2p** and **2s** the methylene protons at C-2 and C-4 were highly deshielded giving singlets between 5.15-5.23 ppm and 4.48-4.59 ppm respectively. This is due to delocalization of the lone pair of electrons of nitrogen atoms over benzene ring. In compounds **2c, 2f, 2i, 2o, 2q, 2r** and **2t** the singlets due to the protons at C-2 and C-4 appeared in the range of 4.48-4.53 ppm and 3.90-3.98 ppm respectively, while the benzylic methylene protons appeared between 3.75-3.83 ppm. Compounds **2b, 2g** and **2n** have their signals due to C-2 and C-4 protons appearing as singlets between 4.48-4.56 ppm and 3.76-3.83 ppm respectively. The signal due to the vinylic proton at C-6 of the ring was found mixed with those of the aromatic protons in the range of 6.70-7.96 ppm. The tetrahydropyrimidines **(2a-t)** were found to be stable and were crystallized from ether-hexane mixture. The synthesis of other tetrahydropyrimidines and studies of their biological properties are in progress.

EXPERIMENTAL

Melting points were recorded by open capillary method and are uncorrected. The infrared spectra were recorded on a Perkin-Elmer 983 spectrometer. ^{1}H nmr (90 MHz) spectra were recorded on a Varian EM-390 spectrometer. High-resolution ^{1}H nmr and ^{13}C nmr (300 MHz) spectra were recorded on Bruker ACF-300 spectrometer. The chemical shifts (δ ppm) and the coupling constants (Hz) are reported in the standard fashion with reference to TMS as internal reference. FAB-mass spectra (MS) were measured on JEOL SX 102/DA-6000 Mass spectrometer using Argon as the FAB gas and *m*-nitrobenzylalcohol as the matrix.

Starting materials **1a-c** [1], **3b** [4], **3c** [5], **3h** [5], **3e** [6], **3f** [6], **3g** [7] were prepared by reported procedures. The unknown starting materials **3a, 3d, 3i** and **3j** were prepared by the reported procedures [6,7] and their analytical and spectral data are given below.

3-(Benzylamino)-1-(4-methylphenyl)-prop-2-en-1-one (**3a**).

This compound was obtained as a pale yellow solid in 84% yield, mp 112-113 °C (hexane); ir (KBr): 1580, 1606, 1631, 3279, 3439 cm^{-1}; ^{1}H nmr ($CDCl_3$): δ 2.45 (s, 3H), 4.52 (d, 2H, J=5.4 Hz), 5.90 (d, 1H, C_2-H, J=8.1 Hz), 7.00-7.70 (m, 8H), 7.85-8.15 (m, 2H), 11.00 (broad m, 1H, exchangeable with D_2O); m/z: 252 (MH^+).

Anal. Calcd. for $C_{17}H_{17}NO$ (251.33): C, 81.24; H, 6.82; N, 5.57. Found: C, 81.45; H, 6.76; N, 5.65.

3-Anilino-1-(3-methoxyphenyl)-prop-2-en-1-one (**3d**).

This compound was obtained as a yellow solid in 80 % yield, mp 148-149 °C (MeOH); ir (KBr): 1577, 1591, 1633, 3418 cm^{-1}; ^{1}H nmr ($CDCl_3$): δ 3.88 (s, 3H), 6.38 (d, 1H, J=9.0 Hz, C_2-H), 7.04-7.13 (m, 4H), 7.26-7.38 (m, 3H), 7.45-7.57 (m, 3H), 12.14 (broad d, 1H, exchangeable with D_2O).

Anal. Calcd. for $C_{16}H_{15}NO_2$ (253.11): C, 75.87; H, 5.97; N, 5.53. Found: C, 75.71; H, 6.05; N, 5.60.

3-(4-Toluidino)-1-(4-chlorophenyl)-prop-2-en-1-one (**3i**).

This compound was obtained as a pale yellow solid in 92% yield, mp 202-203 °C (MeOH); ir (KBr): 1576, 1588, 1636, 3441 cm^{-1}; ^{1}H nmr ($CDCl_3$): δ 2.33(s, 3H), 5.93 (d, 1H, J=7.2 Hz, C_2-H), 7.02 (d, 2H, J=7.8 Hz), 7.16 (d, 2H, J=7.8 Hz), 7.41 (d, 2H, J=8.1 Hz), 7.60-7.70 (m, 1H), 7.83 (d, 2H, J=8.1 Hz), 12.13 (broad d, 1H, exchangeable with D_2O); m/z: 273 (MH^+).

Anal. Calcd. for $C_{16}H_{14}ClNO$ (271.75): C, 70.72; H, 5.19; N, 5.15. Found: C, 70.60; H, 5.13; N, 5.21.

3-(4-Toluidino)-1-(4-methoxyphenyl)-prop-2-en-1-one (**3j**).

This compound was obtained as a yellow solid in 91 % yield, mp 151-152 °C (MeOH), ir (KBr): 1575, 1602, 1616, 3430 cm^{-1}; ^{1}H nmr ($CDCl_3$): δ 2.32 (s, 3H), 3.86 (s, 3H), 5.96 (d, 1H, J=7.8 Hz, C_2-H), 6.93-7.01 (m, 3H), 7.12-7.15 (m, 2H), 7.45-7.49 (m, 1H), 7.90-7.94 (m, 2H), 12.07 (broad d, 1H, exchangeable with D_2O).

Anal. Calcd. for $C_{17}H_{17}NO_2$ (267.33): C, 76.38; H, 6.41; N, 5.24. Found: C, 76.50; H, 6.46; N, 5.32.

Dethiomethylation [8] of 1-(Aralkyl/aryl)-3-(alkyl/aralkyl)-5-aroyl-6-methylthio-1,2,3,4-tetrahydrpyrimidines (**1a-c**).

General Procedure.

A mixture of **1** (1 mmol) and Raney Ni (four times by weight) in 5 ml ethanol was refluxed for 20 hours and after the completion of the reaction (monitored by tlc), the reaction mixture was filtered and the residue was washed with ethanol (3x1 mL). Ethanol was then removed under reduced pressure to give a paste, which was dissolved in chloroform (5 mL). The solution was washed with water (2x5 mL), dried over sodium sulphate and the solvent was distilled off to give pure **2** in 50-55% yields. Compounds **2a** and **2c** were subjected to recrystallization using a hexane-ether mixture. The analytical and spectral data of **2a-c** are given in Tables 1 and 2.

1-(Aralkyl/aryl)-3-(alkyl/aralkyl/aryl)-5-aroyl-1,2,3,4-tetrahydropyrimidines (**2a-t**)**.**

General Procedure.

A mixture of primary amine (1 mmol) and formaldehyde (2 mmol, 40% solution) in methanol (2 mL) was stirred at room temperature for 5 minutes. To this was added a solution of enaminone **3** (1 mmol) in 4-5 mL of methanol and the resulting mixture was refluxed and the progress of the reaction was monitored by tlc. After completion of the reaction, solvent was removed under reduced pressure and chloroform (5 mL) was added, the mixture was then washed with water (2x5 mL) and then dried over sodium sulphate. Chloroform was distilled off giving a gum, which on trituration with hexane gave pure product **2**. These compounds were recrystallized from a hexane-ether mixture. The analytical and spectral data are given in Tables 1 and 2.

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