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Green methodologies in organic synthesis: Microwave assisted solvent- and catalyst-free synthesis of enaminones and their conversion into 1,3,5-trisubstituted benzenes as well as 3-aroyl-6-substituted pyridines

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ABSTRACT

Enaminones were obtained in good yields via condensing methyl ketones with (*N*,*N*-dimethylformamide dimethyl acetal) DMF-DMA under microwave irradiation in absence of solvent. These enaminones were readily converted into 1,3,5-trisubstituted benzenes. Reacting enaminones in presence of ammonium acetate has afforded pyridine derivatives.

1. Introduction

Enaminones are polydentate reagents that are finding plenty of neat utilities in synthetic organic chemistry [1-4] and/or dye intermediates [5-7]. Microwave irradiation has been frequently used in diverse organic transformations with a remarkable reduction in reaction times and, in many cases, improving the yields and selectivity of the processes. We have already previously reported on the utility of enaminones as precursors to polyfunctional aromatics and heteroaromatics [8,9].

Enaminones are obtained via condensing methyl ketones with DMF-DMA, through pathways employed benzene as solvent for this condensation. However several authors have reported that yields are quite low according to this protocol [10]. Domestic microwave irradiation has already been described in the preparation of enaminones by Braibante *et al.* [11,12] and by Hamelin *et al.* [13], in both cases, either a solid support or an acid catalyst was employed as reaction promoter. Lee *et al.* [14] reported a solvent-free microwave assisted preparation of enaminones in a microwave (MW) reactor, using catalytic HCl and an excess of the amines. Synthesis of enaminones in absence of solvent has been reported in domestic microwave oven to afford better yield, shorter reaction times [15], but under such conditions productions of hazardous vapours could not be avoided. In the present article, we report high yield synthesis of enaminones **2a-c** via reacting **1a-c** with DMF-DMA in a direct beam microwave reactor and report on conversion of the obtained enaminones as precursors for the title compounds which were needed in connection with a biological chemically programme in our laboratories.

2. Experimental

2.1. Instrumentation

Melting points are uncorrected. All the reactions were conducted under microwave irradiation in heavy-walled Pyrex tubes (capacity 10 mL). Microwave heating was carried out with a single mode cavity Explorer Microwave Synthesizer (CEM Corporation, NC, USA), producing continuous irradiation and equipped with simultaneous external air-cooling system. IR spectra were recorded in KBr disks using a Perkin-Elmer System 2000 FT-IR spectrophoto-meter. ^{1}H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were recorded on a Bruker DPX 400, super-conducting NMR spectrometer in $CDCl_3$ or DMSO-d_6 as solvent and TMS as internal standard; chemical shifts were reported in δ units (ppm). X-ray crystallography was carried out on a Kappa CCD Enraf Nonius FR 590 diffractometer, National Research Center, Dokki, Giza, Egypt (Table 1). Mass spectra were measured on a VG Autospec-Q (high resolution, high performance, tri-sector GC/MS/MS). Microanalyses were performed on a LECO CHNS-932 Elemental Analyzer.

2.2. Synthesis and purification of compounds (2a-c) and (3)

A mixtures of phthalimidoacetone (**1a**) or 2-acetylpyrrole (**1b**) or acetophenone (**1c**) (0.01 mol) and DMF-DMA (1.19 g, 0.01 mol) were irradiated by focused microwave at 180 °C for 20 min for product **2a**, 120 °C for 10 min for product **2b**, and 120 °C for 30 min for product **2c**. Completion of the reactions was monitored by TLC. The build-up of pressure in the closed

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reaction vessel was carefully monitored. After the irradiation, the reaction tube was cooled with high-pressure air through an inbuilt system in the instrument until the temperature had fallen below 50 °C. The reaction mixture left to cool to room temperature and then treated with a mixture of EtOH:dioxane (3:1). The solid product, so formed, was collected by filtration and crystallized from dioxane to afford compounds **2a-c**. The filtrate of product **2a** was dried over anhydrous sodium sulphate and concentrated on vacuum. The residue was purified by column chromatography on silica gel with ethyl acetate:EtOH (9:1) as then eluent to afford product **3** in 10 % yields.

2-(4-Dimethylamino-2-oxo-but-3-enyl)isoindole-1,3-dione, **2a:** Yellow crystals. Yield: 77%. M.p.: 162 °C (Lit. 159-162 °C [16]). FT-IR (KBr, cm^{-1}): 1769, 1714, 1660 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 2.72 (s, 3H, CH_3), 3.05 (s, 3H, CH_3), 4.40 (s, 2H, CH_2), 5.04 (d, 1H, *J*=12 Hz, CH), 7.61 (d, 1H, *J*=12 Hz, CH), 7.85-7.91 (m, 4H, phthalimidyl-H). MS (EI, m/z, %): 258 [M^+, 24%]. Anal. Calcd. for $C_{14}H_{14}N_2O_3$: C, 65.11; H, 5.46; N, 10.85. Found: C, 65.18; H, 5.44; N, 10.76.

3-Dimethylamino-1-(1H-pyrrol-2-yl)-propenone, **2b:** Orange crystals. Yield: 86%. M.p.: 199-200 °C. FT-IR (KBr, cm^{-1}): 3252 (NH), 1625 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 2.88 (s, 3H, CH_3), 3.03 (s, 3H, CH_3), 5.62 (d, 1H, *J*=12.4 Hz, CH), 6.08-6.10 (m, 1H, pyrrolyl-H), 6.76-6.78 (m, 1H, pyrrolyl-H), 6.86-6.88 (m, 1H, pyrrolyl-H), 7.68 (d, 1H, *J*=12.4 Hz, CH), 11.41 (s, 1H, NH). MS (EI, m/z, %): 164 [M^+, 100%]. Anal. Calcd. for $C_9H_{12}N_2O$: C, 65.83; H, 7.37; N, 17.06. Found: C, 65.82; H, 7.91; N, 17.06.

3-Dimethylamino-1-phenyl-propenone, 2c: Orange crystals. Yield: 87%. FT-IR (KBr, cm^{-1}): 1639 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 2.90 (s, 3H, CH_3), 3.14 (s, 3H, CH_3), 5.81 (d, 1H, *J*=12.4 Hz, CH), 7.41-7.50 (m, 3H, phenyl-H), 7.70 (d, 1H, *J*=12.4 Hz, CH), 7.88-7.90 (m, 2H, phenyl-H). MS (EI, m/z, %): 175 [M^+, 89%].

2-(1-Dimethylaminomethylene-2-oxo-propyl)isoindole-1,3-dione, **3:** Light yellow crystals. Yield: 15%. M.p.: 176-178 °C. FT-IR (KBr, cm^{-1}): 1771, 1715, 1653 (CO). ^{1}H NMR (400 MHz, $CDCl_3$): 2.91 (s, 3H, CH_3), 2.98 (s, 3H, NCH_3), 3.22 (s, 3H, NCH_3), 7.72-7.75 (m, 2H, phthalimidyl-H), 7.79 (s, 1H, CH), 7.87-7.90 (m, 2H, phthalimidyl-H). MS (EI, m/z, %): 258 [M^+, 8%]. Anal. Calcd. for $C_{14}H_{14}N_2O_3$: C, 65.11; H, 5.46; N, 10.85. Found: C, 65.01; H, 5.61; N, 11.05.

2.3. Synthesis of compounds (6a,b)

2.3.1. 2-(2-3,5-Di[2-(1,3-dioxo-2,3-dihydro-1H-2-isoindolyl) acetyl]phenyl-2-oxoethyl)-1,3-isoindolinedione (6a)

A mixture of compound **2a** (2.58 g, 0.01 mol), indium chloride (0.02 g) and acetic acid (0.5 mL) was irradiated in microwave at 140 °C for 30 min. The crude product was poured onto water, the solid product, so formed, was collected by filtration and crystallized from toluene. Light brown. Yield: 58 %. M.p.: 300-301 °C. FT-IR (KBr, cm^{-1}): 1776, 1721 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 5.57 (s, 6H, CH_2), 7.93-7.99 (m-12H, phthal-imidyl-H), 8.98 (s, 3H, benzyl-H). ^{13}C NMR (DMSO-d_6): 192.23, 167.98 (2 CO), 135.33, 135.20, 133.39, 132.02, 123.93, 45.50 (CH_2). MS (EI, m/z, %): 639 [M^+, 12%]. Anal. Calcd. for $C_{36}H_{21}N_3O_9$: C, 67.61; H, 3.31; N, 6.57. Found 67.46; H, 3.33; N, 6.81.

2.3.2. [3,5-di(1H-2-pyrrolylcarbonyl)phenyl](1H-2-pyrrolyl) methanone (6b)

A mixture of compound **2b** (1.64 g, 0.01 mol) and acetic acid (0.5 mL) was irradiated by focused microwave at 140 °C for 30 min. Completion of reaction was monitored by TLC. The solid product, so formed, was collected and crystallized from dioxane to afford compounds **6b**. Yield 93%. M.p.: 259-260 °C. FT-IR (KBr, cm^{-1}): 3290 (NH), 1626 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 6.130-6.30 (m, 1H, pyrrolyl-H), 6.91-6.93 (m, 1H, pyrrolyl-H), 7.28-7.30 (m, 1H, pyrrolyl-H), 8.37 (s, 1H, benzoyl-H), 12.22 (s, 1H, NH). ^{13}C NMR (DMSO-d_6, 100 MHz): 182.10 (CO), 138.84, 131.21, 130.17, 127.20, 119.81, 110.69. MS (EI, m/z, %): 357 [M^+, 100%]. Anal. Calcd. for $C_{21}H_{15}N_3O_3$: C, 70.58; H, 4.23; N, 11.76. Found: C, 69.77; H, 4.74; N, 11.30.

2.4. General procedure for the synthesis of compounds (8a,b)

A mixture of compound **2a** (0.02 mol), **2b** or **2c** (0.1 mol) and acetic acid (0.5 mL) was irradiated by focused microwave at 140 °C for 30 min. The solid product, so formed, was collected and crystallized from dioxane to afford compounds **8a**,**b**.

2-2-[3-[2-(1,3-dioxo-2,3-dihydro-1H-2-isoindolyl)acetyl]-5-(1H-2-pyrrolylcarbony)phenyl]-2-oxoethyl-1,3-isoindolinedione, **8a:** Buff powder. Yield: 52%. M.p.: 252-253 °C. FT-IR (KBr, cm^{-1}): 3343 (NH), 1717 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 5.54 (d, 4H, *J*=12.4 Hz, $2NCH_2$), 6.23-6.33 (m, 1H, pyrrolyl-H), 6.93-6.96 (m, 1H, pyrrolyl-H), 7.31-7.33 (m, 1H, pyrrolyl-H), 7.91-7.95 (m, 4H, Ar-H), 7.98-8.00 (m, 4H, Ar-H), 8.62 (d, 1H, *J*=1.2 Hz, CH), 8.98 (s, 1H, CH), 12.29 (s, 1H, NH). MS (EI, m/z, %): 545 [M^+, 18%]. Anal. Calcd. for $C_{31}H_{19}N_3O_7$: C, 68.26; H, 3.51; N, 7.70. Found: 68.37; H, 3.91; N, 7.74.

2-(2-3-benzoyl-5-[2-(1,3-dioxo-2,3-dihydro-1H-2-isoindolyl) acetyl]phenyl-2-oxoethyl)-1,3-isoindolinedione, **8b:** Brown powder. Yield: 68%. M.p.: 194-496 °C. FT-IR (KBr, cm^{-1}): 1718 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 5.55 (d, 4H, *J*=7.6 Hz, $2NCH_2$), 7.30-732 (m, 3H, Ar-H), 7.56-7.59 (m, 2H, Ar-H), 7.83-7.87 (m, 4H, Ar-H), 7.93-7.98 (m, 4H, Ar-H), 8.57 (s, 1H, CH), 8.98 (s, 1H, CH). MS (EI, m/z, %): 556 [M^+, 32%]. Anal. Calcd. for $C_{33}H_{20}N_2O_7$: C, 71.22; H, 3.62; N, 5.03. Found: C, 70.94; H, 3.96; N, 5.72.

2.5. General procedure for the synthesis of compounds (9a-c)

A mixture of compounds **2a-c** (0.01 mol), EtOH (1 mL) and aniline (0.93g, 0.01 mol) were irradiated by focused microwave at 140 °C for 5 min. The reaction mixture was poured into cold water, filtered and crystallized from ethanol to afford compounds **9a-c**.

2-(2-Oxo-4-phenylamino-but-3-enyl)isoindole-1,3-dione, **9a:** Yellow-orange crystals. Yield: 88%. M.p.: 167-169 °C. FT-IR (KBr, cm^{-1}): 3457 (NH), 1773, 1706, 1640 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 4.50 (s, 2H, NCH_2), 5.45 (d, 1H, *J=8.0* Hz, CH), 7.03 (t, 1H, *J=7.2* Hz, phenyl-H), 7.25 (d, 2H, *J*=8.0 Hz, phenyl-H), 7.30 (t, 2H, *J*=7.2 Hz, phenyl-H), 7.77 (dd, 1H, *J*=8.0 Hz, *J*=8.0 Hz CH), 7.87-7.90 (m, 2H, phthalimidyl-H), 7.91-7.95 (m, 2H, phthalimidyl-H), 11.15 (d, 1H, *J*=12.6 Hz, NH). ^{13}C NMR (DMSO-d_6, 100 MHz): 191.47, 168.22 (CO), 145.75, 140.38, 135.16, 132.14, 130.06, 123.96, 123.69, 116.66, 93.58, 45.79 (NCH_2). MS (EI, m/z, %): 306 [M^+, 20%]; 160 (25), 146 (100), 117 (8), 104 (14), 77 (12). Anal. Calcd. for $C_{18}H_{14}N_2O_3$: C, 70.58; H, 4.61, N, 9.15. Found: C, 70.55; H, 4.61, N, 9.27.

3-Phenylamino-1-(1H-pyrrol-2-yl)-propenone, **9b:** Golden-yellow crystals. Yield: 74%. M.p.: 217-218 °C. FT-IR (KBr, cm^{-1}): 3262 (NH), 1635 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 5.85 (d,1H, *J*=8.0 Hz, CH), 6.16-6.18 (m, 1H, pyrrolyl-H), 6.88-6.90 (m, 1H, pyrrolyl-H), 7.00-7.04 (m, 1H, pyrrolyl-H), 7.21 (d,2H, *J*=8.4 Hz, phenyl-H), 7.32 (t, 3H, *J*=8.4 Hz, phenyl-H), 7.69 (dd, 1H, *J*=8.0 Hz, *J*=8.0 Hz, CH), 11.63 (s, 1H, NH), 11.71 (s, 1H, NH). MS (EI, m/z, %): 212 [M^+, 100%]. Anal. Calcd. for $C_{13}H_{12}N_2O$: C, 73.56; H, 5.70; N, 13.20. Found: C, 73.42; H, 6.03; N, 13.07.

1-Phenyl-3-phenylamino-propenone, **9c:** Yellow crystals. Yield: 83%. All data agreed with the published one [17].

Table 1. Crystal data and structure refinement for compound **3**.

Chemical formula	$C_{14}H_{14}N_2O_3$
Formula weight	258.277
Crystal System	Triclinic
Space group	P-1
a	7.9478 (5) Å
b	8.4931 (6) Å
c	11.1333 (8) Å
α	88.231 (2) °
β	83.557 (2) °
γ	62.615 (2) °
V	662.90 (8) Å^3
Z	2
Temperature	298 K
Radiation type	Mo *Kα*
Measured reflections	3228
Independent reflections	2435
Observed reflections	1252
R_{int}	0.036
R(all)	0.111
R(gt)	0.064
wR(ref)	0.119
wR(all)	0.126
Parameters	172

2.6. General procedure for the synthesis of compounds (12a,b)

A mixture of compound **2a-b** (0.01 mol) and ammonium acetate (0.2 mol) was irradiated by focused microwave at 160 °C for 10 min for product **12a**, 140 °C for 10 min for product **12b**. Completion of reaction was monitored by TLC. The solid product, so formed, was collected and crystallized from dioxane:EtOH (1:3) to afford compounds **12a**,**b**.

2-(5-[2-(1,3-dioxo-2,3-dihydro-1H-2-isoindolyl)acetyl]-2-pyridylmethyl)-1,3-isoindolinedione, **12a:** Buff powder. Yield: 53%. M.p.: 235-236 °C. FT-IR (KBr, cm^{-1}): 1714 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 5.04 (s, 2H, CH_2), 5.29 (s, 2H, CH_2), 7.23 (d, 1H, *J*=7.6 Hz, Ar-H), 7.59-7.63 (m, 1H, Ar-H), 7.31 (d, 1H, *J*=8.4 Hz, Ar-H), 7.76-7.81 (m, 1H, Ar-H). 7.87 (s, 1H, Ar-H), 7.98-8.00 (m, 4H, Ar-H), 7.31 (dd, 1H, *J*=2.4 Hz, *J*=2.0 Hz, Ar-H), 9.15 (s, 1H, Ar-H). MS (EI, m/z, %): 425 [M$^+$, 10%]. Anal. Calcd. for $C_{24}H_{15}N_3O_5$: C, 67.76; H, 3.55; N, 9.88. Found: C, 67.70; H, 3.63; N, 9.77.

1H-2-pyrrolyl[6-(1H-2-pyrrolyl)-3-pyridyl]methanone, **12b:** Brown powder. Yield: 78%. M.p.: 136-137 °C. FT-IR (KBr, cm^{-1}): 3295 (NH), 1618 (CO). ^{1}H NMR (400 MHz, DMSO-d_6): 6.31 (t, 1H, *J*=4.0 Hz, pyrrolyl-H), 6.92 (t, 1H, *J*=2.8 Hz, pyrrolyl-H), 7.30 (d, 1H, *J*=9.2 Hz, pyrrolyl-H), 8.41(d, 2H, *J*=4.8 Hz, pyridinyl-H), 9.14 (s, 1H, pyridinyl-H), 12.28 (s, 2H, NH). MS (EI, m/z, %): 237 [M$^+$, 100%]. Anal. Calcd. for $C_{14}H_{11}N_3O$: C, 70.87; H, 4.67; N, 17.71. Found: C, 69.87; H, 4.66; N, 17.53.

3. Results and discussion

Condensation of phthalimidoacetone **1a** with dimethyl formamide dimethyl acetal (DMF-DMA) has afforded enaminone **2a** in focused microwave oven at 180 °C for 20 min in absence of solvent in 77 % yield. Also, a side product was isolated from this reaction. Isolation of the side product has not been observed earlier. The X-ray crystal structure for this product indicated that it resulted from condensation of methylene moiety in **1a** with DMF-DMA to yield 2-(1-dimethylamino methylene-2-oxo-propyl)isoindole-1,3-dione (**3**) (Figure 1).

In Table 2, we list the selected bond lengths and angles for compound **3**. It is clear from Figure 1 that the product adopted sterical strained form compound **3** rather than compound **3a**. This is a further example indicating that steroelectronic factors overweight steric factors. It has previously indicated that this is the case when steroelectronic factors and steric factors contradict [18]. Bond length for N2-C15 is typical for N-C single bond. The same applies for N5 and C7 indicating that charge separated resonance form **3a** does play significant role.

Heating of methyl ketones **1b**,**c** with (DMF-DMA) in MW with solvent-free at 120 °C for 10-30 min has afforded enaminones **2b**,**c** in 86% and 87% yields, respectively. Recently, it was reported that heating neat reactants in ionic liquid afforded compounds **2b**,**c** in 86% and 70% yields, respectively [19] (Scheme 1).

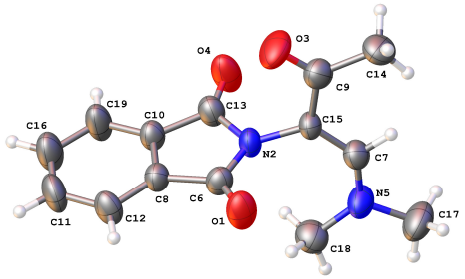
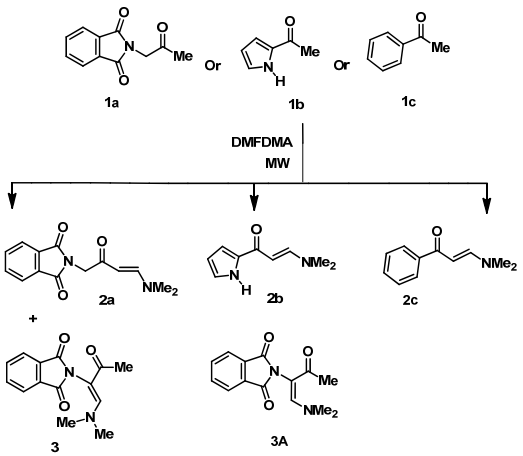


Figure 1. X-ray crystal structure of compound (**3**).

Table 2. Selected bond lengths and angles for compound **3**.

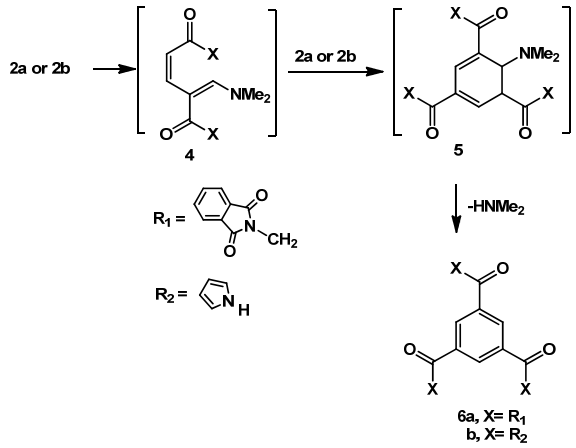
Bond	Bond length (Å)	Bond	Bond angle (°)
N2-C6	1.397 (6)	C7-N5-C17	120.0 (4)
N2-C13	1.414 (6)	N2-C15-C7	123.5 (3)
N2-C15	1.434 (5)	C7-C15-C9	121.8 (4)
N5-C7	1.339 (6)	N5-C7-C15	33.3 (4)
N5-C17	1.453 (6)	N2-C15-C9	114.7 (3)
C7-C15	1.349 (5)	N2-C15-C7	123.5 (3)
N5-C18	1.448 (6)	N5-C7-H7	120.3 (4)
		N5-C7-C15-C9	-179.4 (7)



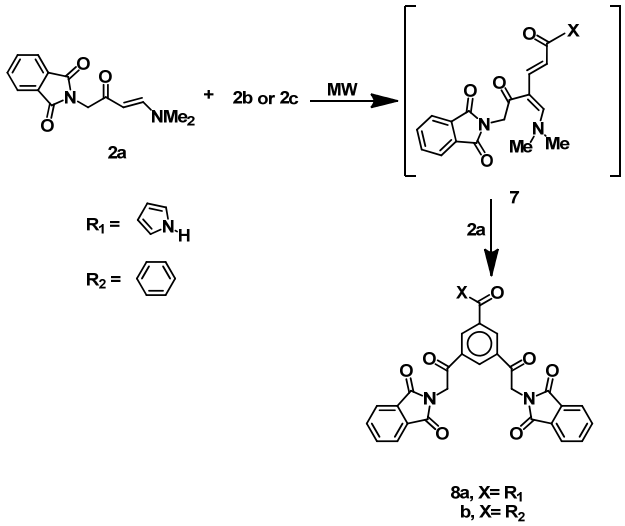
Scheme 1

Compound **2b** undergoes self-condensation on heating in focused microwave at 140 °C for 30 min in presence of a few drops of acetic acid to give 1,3,5-triacylbenzene **6b** in 93% yields. We believe that compound **2b** is initially converted to the open chain intermediate **4** that further adds to one molecule of enaminone **2b** to yield the intermediate **5**, which aromatize under these reaction conditions to give the final isolable product **6b**. In a trial to enhance the polycondensation of **2a** using $InCl_3$ as catalyst the reaction resulted only in the formation of **6a**, which has been recently reported by us [16] (Scheme 2).

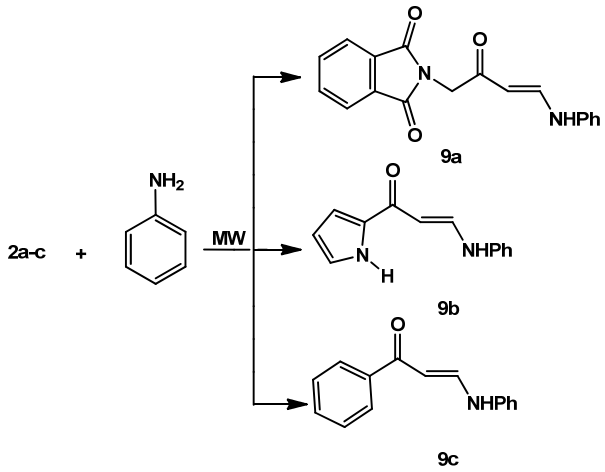
Compounds **2a-c**, have proved to be a versatile starting materials for a variety of otherwise not readily obtainable functionally substituted aromatics and heteroaromatics. Thus, heating a mixture of enaminones **2a** with **2b** or **2c** as 2:1 ratio in focused microwave oven at 140 °C for 30 min in presence of few drops of acetic acid afforded products that were identified as 1,3,5-trisubstituted benzenes **8a**,**b** in 52% and 68% yields, respectively. It is most likely that the initial step in this reaction is addition of **2a** to **2b** or **2c** to furnish an intermediate **7**, which then reacts further with **2a** to yield final isolable products **8a**,**b** (Scheme 3).



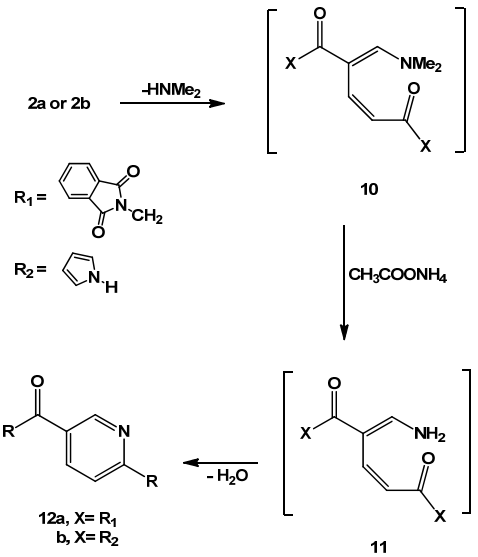
Scheme 2



Scheme 3



Scheme 4



Scheme 5

Compound **2a-c** reacted with aniline in focused microwave oven at 140 ºC for 5 min afforded the aniline derivatives **9a-c** in 88%, 74% and 83% yields, respectively (Scheme 4).

Heating enaminones **2a**,**b** with ammonium acetate, in focused microwave oven at 160-140 oC for 10 min afforded the 3-aroyl-6-substituted pyridines **12a**,**b** in 53% and 78% yields, respectively. Formation of **12a**,**b** may take place through an initial self condensation of **2** to afford the intermediate **10** which reacts directly with ammonium ion to give a further intermediate **11**, that undergoes intramolecular cycloconden-sation, *via* loss one molecule of water to give the final products (Scheme 5).

4. Conclusion

The studies described above clearly demonstrate that an efficient method was achieved for the preparation of enaminones, employing green methodologies. The simplicity of the reaction conditions, their efficacy and excellent yields obtained using MW irradiation, under solvent- and catalyst-free conditions. Also we could show that the enaminones **2a-c** is a valuable precursor to the triacylbenzene 1,3,5-trisubstituted benzenes as well as 3-aroyl-6-substituted pyridines.

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Supplementary material

CCDC-772198 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/data_request/cif, or by e-mailing data_request@ccdc.cam.ac.uk, or by contacting The Cambridge Crystallographic Data Centre, 12 Union Road, Cambridge CB2 1EZ, UK; fax: +44(0)1223-336033.

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