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PASSERINI THREE-COMPONENT REACTION OF ANTHRACENE-9-CARBALDEHYDE IN WATER: GREEN SYNTHESIS OF ANTHRACENE- CONTAINING α-ACYLOXYCARBOXAMIDES

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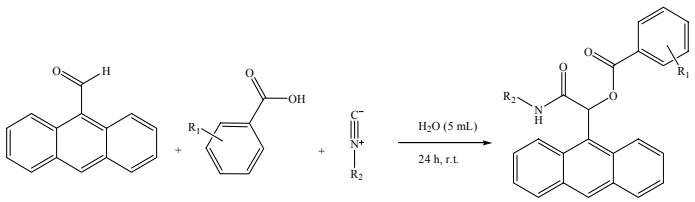
In the current study, we have performed the synthesis of novel α-acyloxycarboxamide derivatives *via* a passerini one-pot three component reactions of anthracene-9-carbaldehyde, an isocyanide and a benzoic acid derivative in aqueous media in 74-86% yields at room temperature. The remarkable features of this "green" protocol include; high product yield, inexpensive and non-toxic solvent, operational simplicity, environmentally benign and the absence of any volatile or hazardous organic solvents.



INTRODUCTION

In recent years, green chemistry has attracted considerable attention of organic and medicinal chemists. One of the most important roles of green chemistry is the invention, design and application of chemical reactions that reduce or eliminate the use of hazardous solvents.[1] In comparison with organic solvents, water is non-toxic, non-corrosive and non-explosive, and is readily available at low cost. These properties along with the network of hydrogen bonds, large surface tension, high polarity and high specific heat capacity make it both economical and environmentally friendly and thus suitable as a green solvent.[2,3]

Multicomponent reactions (MCRs) have become important tools in modern preparative synthetic chemistry because these reactions increase the efficiency by combining several operational steps without isolation of intermediates or changes of the conditions.[4-9] Hence, this principle is very efficient in terms of time as nice as resources.[10] Among the known multicomponent reactions to date, the most valuable reactions are those based on isocyanides. Isocyanide-based multicomponent reactions (abbreviated to IMCRs by Ugi and Dömling) by virtue of their synthetic potential, their inherent atom efficiency, convergent nature, ease of implementation, and the generation of molecular diversity, have attracted much attention because of the advantages that they offer to the field of combinatorial chemistry.[11]

IMCRs are particularly interesting because they are more versatile and diverse than the remaining MCRs. The great potential of isocyanides for development of multicomponent reactions lies in the diversity of bond-forming processes available,

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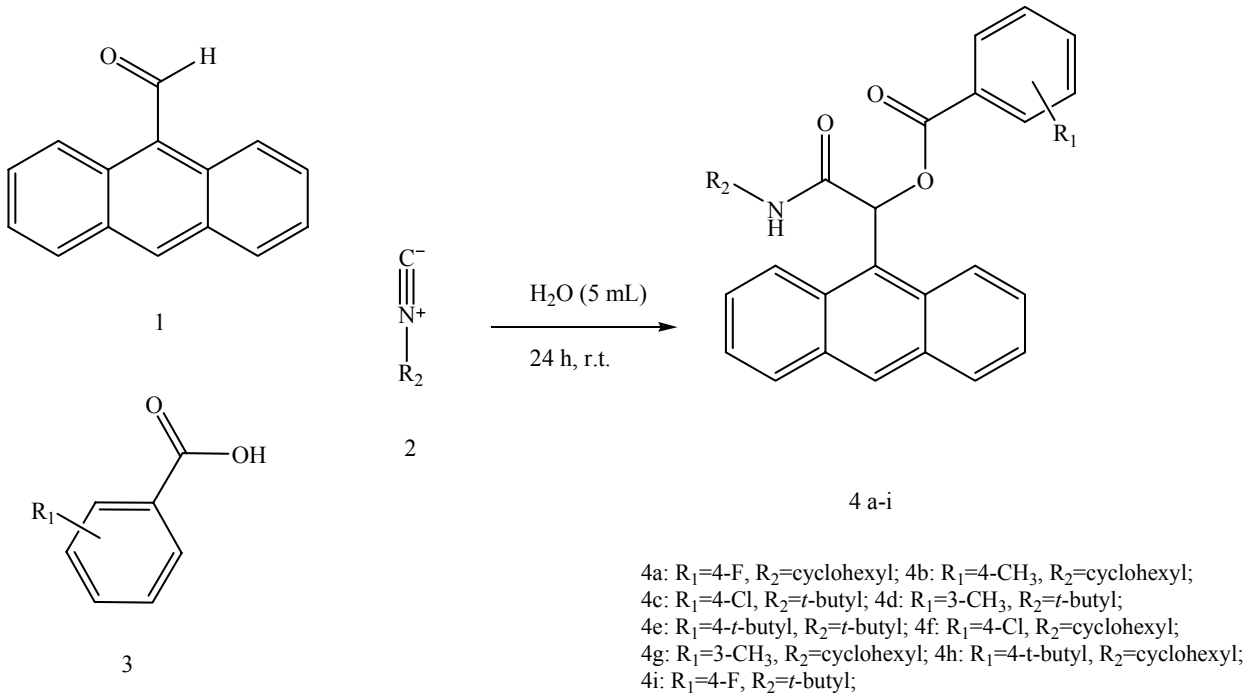
their functional group tolerance, and the high levels of chemo-, regio-, and stereoselectivity often observed.[5,12-19] In 1921, Passerini pioneered the use of isocyanides and successfully developed a three-component synthesis of a-acyloxycarboxamides by the reaction between carboxylic acid, an aldehyde, and an isocyanide.[20,21]

To the best of our knowledge, the present study is the first report for the synthesis of 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives **4a-i** *via* a passerini one-pot three component reaction of anthracene-9-carbaldehyde **1**, isocyanides **2** and benzoic acid derivatives **3** (Scheme 1).

RESULTS AND DISCUSSION

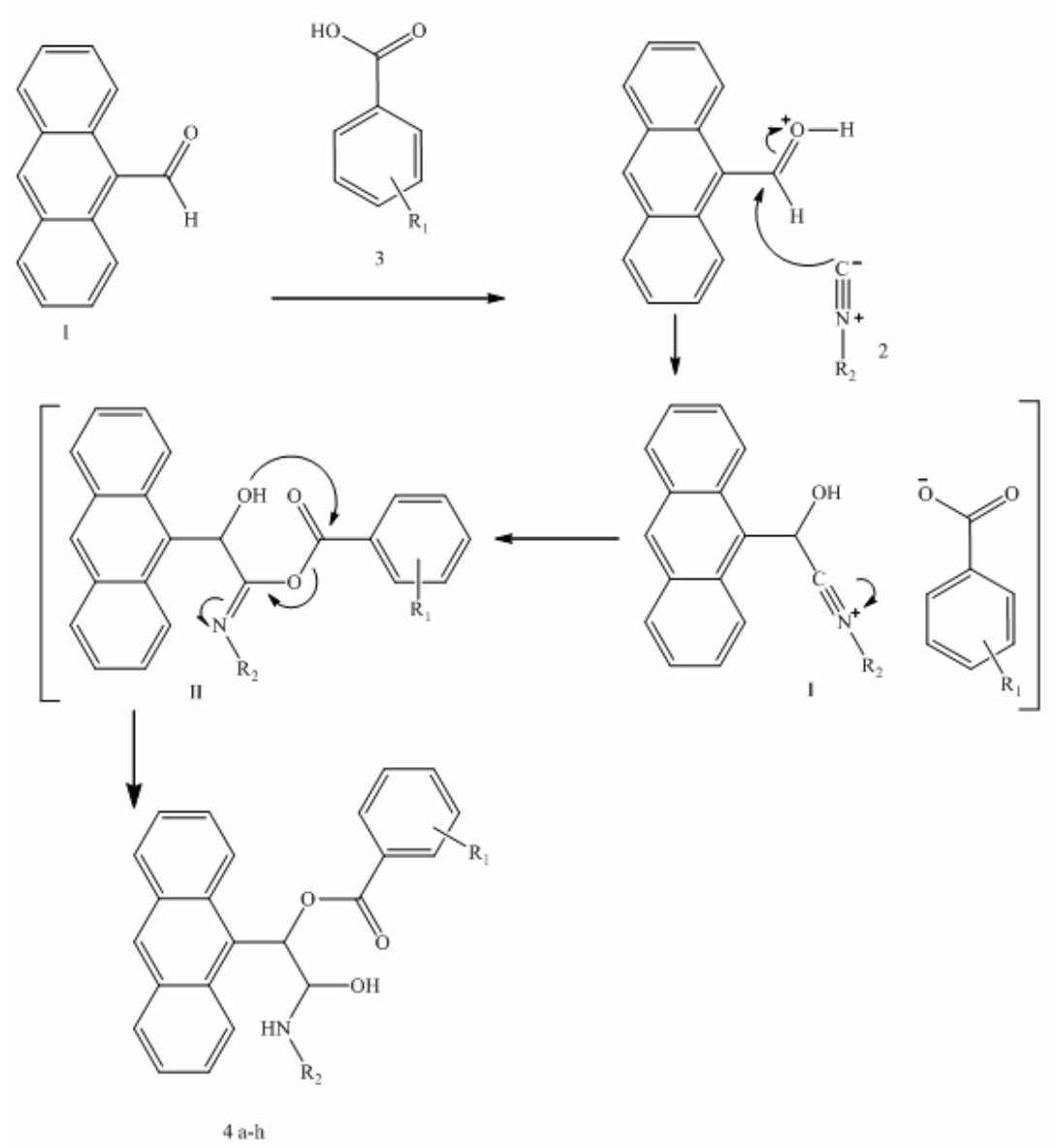
As part of a continuing effort in our laboratory towards the development of new methods in organic synthesis,[5a-k,22] here, a green route for efficient synthesis of 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives *via* a passerini one-pot three component reaction of anthracene-9-carbaldehyde **1**, isocyanides **2** and benzoic acid derivatives **3** in water is described. The anthracene-9-carbaldehyde **1**, isocyanides **2** and benzoic acid derivatives **3** in water react together in a 1:1:1 ratio at room temperature to produce α-acyloxycarboxamides **4a-i** (**Scheme 1**). The reaction proceeds smoothly and cleanly under mild conditions and no side reactions are observed. The pure products **4a-i** are stable at room temperature for several months. The structures of the products were deduced from IR, ^{1}H NMR, and ^{13}C NMR spectra. For example the ^{1}H NMR spectrum of **4a** exhibited distinct signals arising from δ= δ= 0.85-1.85 (m, 10H, 5CH_2), 3.79 (m, 1H, CH), 5.29 (d, $^{3}J_{HH}$ =8.0 Hz, 1H, NH), 7.06 (t, $^{3}J_{HF}$= 8.0 Hz, 2H, meta), 7.48 (t, $^{3}J_{HH}$ =8.5 Hz, 2H), 7.63 (t, $^{3}J_{HH}$ =8.5 Hz, 2H), 7.94 (s, 1H, CH-O), 8.04 (s, 1H), 8.07 (d, $^{3}J_{HH}$ =8.0 Hz, 2H), 8.09 (d, $^{3}J_{HH}$ =8.5 Hz, 2H), 8.56 (d, $^{3}J_{HH}$ =8.5 Hz, 2H, orto). The ^{13}C NMR spectrum of 4a showed 21 distinct resonances arising from δ= 24.52 (CH_2), 24.69 (CH_2), 25.26 (CH_2), 32.51 (CH_2), 32.66 (CH_2), 48.69 (CH), 69.70 (CH), 115.57 (d, $^{2}J_{CF}$=22.0 Hz, 2CH, meta), 124.03 (C), 125.30 (2CH), 125.88 (C), 127 (2CH) 129.35 (2CH), 130.28 (2CH), 131.03 (C), 131.56 (CH), 132.56 (d, $^{3}J_{CF}$=9.5 Hz, 2CH, orto), 134.08 (C), 162.20 (d, $^{1}J_{CF}$= 250.0 Hz, C, para), 164.84(C=O of ester), 168.15 (C=O of amide). The IR spectrum showed a N-H absorption at 3411cm^{-1}.

The suggested mechanism for the formation of products **4a-i** is illustrated in **Scheme 2**. On the basis of the chemistry of isocyanides, it is reasonable to assume that the first step may involve protonation of anthracene-9-carbaldehyde **1** by the carboxylic acid **3** to form an intermediate, which is then attacked by the alkyl isocyanide **2**, leading to the formation of **4**.[22]



4a: R_1=4-F, R_2=cyclohexyl; 4b: R_1=4-CH_3, R_2=cyclohexyl;
4c: R_1=4-Cl, R_2=*t*-butyl; 4d: R_1=3-CH_3, R_2=*t*-butyl;
4e: R_1=4-*t*-butyl, R_2=*t*-butyl; 4f: R_1=4-Cl, R_2=cyclohexyl;
4g: R_1=3-CH_3, R_2=cyclohexyl; 4h: R_1=4-t-butyl, R_2=cyclohexyl;
4i: R_1=4-F, R_2=*t*-butyl;

Scheme 1 – Synthesis of 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives (**4a-i**).



Scheme 2 – A proposed mechanism for the formation of produce 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives **4a-i**.

EXPERIMENTAL

Materials and Methods

All chemicals were purchased from Merck (Darmstadt, Germany) and Fluka (Switzerland), and used without further purification. Melting points were determined on an Electrothermal 9100 apparatus. The IR spectra were measured on a Jasco 6300 FT-IR spectrometer (KBr disks). ^{1}H- and ^{13}C-NMR spectra were measured ($CDCl_3$ solution) with a BRUKER DRX-250 AVANCE spectrometer at 250.0 and 62.9 MHz, respectively. The progress of reactions was monitored by thin layer chromatography (TLC) performed on F_{254} silica gel plates.

General procedure for the preparation of 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives 4a-i

A mixture of anthracene-9-carbaldehyde **1** (1 mmol, 0.203 g), an alkyl isocyanide **2** (1 mmol, cyclohexyl isocyanide, *t*-butyl isocyanide and *n*-butyl isocyanide) and a carboxylic acid **3** (1 mmol; 0.156 g (Ar=4-ClC_6H_4), 0.156 g (Ar=3-ClC_6H_4), 0.136 g (Ar=3-MeC_6H_4), 0.140 g (Ar=4-FC_6H_4), 0.136 g (Ar=4-MeC_6H_4), 0.178 g (Ar=4-*t*-BuC_6H_4) in water (5 mL) was stirred at room temperature for 24 h. Upon completion of the reaction, the solvent was removed under reduced pressure and the products were purified by Preparative-Layer Chromatography (PLC) plate (silica gel powder; petroleum ether: ethylacetate, 10 : 2). The characterization data of the compounds are given below:

1-(anthracen-9-yl)-2-(cyclohexylamino)-2-oxoethyl 4-fluorobenzoate **(4a)**

White powder, yield (84 %); m.p.: 189-191 °C; IR (KBr) (υ_{max}, cm^{-1}): 3420 (N-H of amide), 2926, 2853, 1729 (C=O of ester), 1684 (C=O of amide), 1603, 1507, 1449, 1260, 1088 (C-O), 891, 853, 763, 734 cm^{-1}; ^{1}H NMR (250 MHz, $CDCl_3$): δ= 0.85-1.85 (m, 10H, 5CH_2), 3.79 (m, 1H, CH), 5.29 (d, $^{3}J_{HH}$ =8.0 Hz, 1H, NH), 7.06 (t, $^{3}J_{HF}$= 8.0 Hz, 2H, meta), 7.48 (t, $^{3}J_{HH}$ =8.5 Hz, 2H), 7.63 (t, $^{3}J_{HH}$ =8.5 Hz, 2H), 7.94 (s, 1H,

CH-O), 8.04 (s, 1H), 8.07 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.09 (d, $^3J_{HH}$ =8.5 Hz, 2H), 8.56 (d, $^3J_{HH}$ =8.5 Hz, 2H, orto). ^{13}C NMR ($CDCl_3$): δ= 24.52 (CH_2), 24.69 (CH_2), 25.26 (CH_2), 32.51 (CH_2), 32.66 (CH_2), 48.69 (CH), 69.70 (CH), 115.57 (d, $^2J_{CF}$=22.0 Hz, 2CH, meta), 124.03 (C), 125.30 (2CH), 125.88 (C), 127 (2CH) 129.35 (2CH), 130.28 (2CH), 131.03 (C), 131.56 (CH), 132.56 (d, $^3J_{CF}$=9.5 Hz, 2CH, orto), 134.08 (C), 162.20 (d, $^1J_{CF}$= 250.0 Hz, C, para), 164.84(C=O of ester), 168.15 (C=O of amide).

***1-(anthracen-9-yl)-2-(cyclohexylamino)-2-oxoethyl 4-methylbenzoate* (4b)**

Yellow oil, yield (74 %); IR (neat) (υ_{max}, cm^{-1}): 3349 (N-H of amide), 2928, 2853, 1724(C=O of ester), 1674 (C=O of amide), 1611, 1525, 1449, 1315, 1269, 1093, 890, 790, 750, 733 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ= 0.96-2.15 (m, 10H, $5CH_2$), 2.35 (s, 3H, CH_3), 3.82 (m, 1H, CH), 5.52 (d, $^3J_{HH}$= 7.5Hz, 1H, NH), 7.18 (d, $^3J_{HH}$= 7.0 Hz, 2H, meta), 7.49 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.61 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.95 (s, 1H, CH-O), 8.03 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.52 (s, 1H), 8.60 (d, $^3J_{HH}$ =8.5 Hz, 2H), , 8.59 (d, $^3J_{HH}$= 8.5 Hz, 2H, orto). ^{13}C NMR ($CDCl_3$): δ= 21.64 (CH_3), 24.56 (CH_2), 24.70 (CH_2), 25.31 (CH_2), 32.31 (CH_2), 32.74 (CH_2), 48.57 (CH), 69.59 (CH), 124.20 (C), 125.18 (2CH), 126.41 (C), 126.79 (2C), 127.07 (2CH), 129.13 (2CH), 129.28 (2CH), 129.99 (2CH), 130.06 (2CH), 131.04 (2C), 131.56 (CH), 144.06 (C), 165.73 ((C=O of ester), 168.35 (C=O of amide).

***1-(anthracen-9-yl)-2-(tert-butylamino)-2-oxoethyl 4-chlorobenzoate* (4c)**

White powder, yield (82 %); m.p.: 186-187 °C; IR (KBr) (υ_{max}, cm^{-1}): 3411(N-H of amide), 2967, 1727 (C=O of ester), 1693 (C=O of amide), 1624, 1593, 1511, 1452, 1400, 1259, 1172, 1093, 922, 892, 781 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ=1.20 (s, 9H, $3CH_3$), 5.24 (s, 1H, NH), 7.52 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.64 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.89 (s, 1H, CH-O), 7.99 (s, 1H), 8.05 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.60 (d, $^3J_{HH}$ =8.5 Hz, 2H), 8.57 (d, $^3J_{HH}$= 8.5 Hz, 2H, orto). ^{13}C NMR ($CDCl_3$): δ= 28.47 ($3CH_3$), 51.85 (C), 69.83 (CH), 124.01 (C), 125.30 (2CH), 125.99 (2C), 127.27 (2CH), 128.06 (2C), 128.75 (2CH), 129.38 (2CH), 130.27 (2CH), 131.00 (C), 131.36 (2CH), 131.56 (CH), 139.73 (C), 164.97 (C=O of ester), 168.12 (C=O of amide).

***1-(anthracen-9-yl)-2-(tert-butylamino)-2-oxoethyl 4-methylbenzoate* (4d)**

Yellow oil, yield (74 %); IR (neat) (υ_{max}, cm^{-1}): 3412 (N-H of amide), 2965, 1723 (C=O of ester), 1694 (C=O of amide), 1611, 1514, 1454, 1365, 1259, 1177, 1101, 891, 841, 750, 733 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ=1.25 (s, 9H, $3CH_3$), 2.35 (s, 3H, CH_3), 5.52 (s, 1H, NH), 7.50 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.63 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.94 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.01 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.04 (s, 1H, CH-O), 8.52 (s, 1H), 8.62 (d, $^3J_{HH}$= 8.5 Hz, 2H, orto). ^{13}C NMR ($CDCl_3$): δ= 21.63 (CH_3), 28.54 ($3CH_3$), 51.78 (C), 69.69 (CH), 124.24 (2C), 125.17 (2CH), 126.67 (2C), 127.03 (2CH), 129.14 (2CH), 129.29 (2CH), 129.98 (2CH), 131.04 (2C), 131.59 (CH), 144.06 (C), 165.67 (C=O of ester), 168.20 (C=O of amide).

***1-(anthracen-9-yl)-2-(tert-butylamino)-2-oxoethyl 4-(tert-butyl)benzoate* (4e)**

Orange oil, yield (81 %); IR (neat) (υ_{max}, cm^{-1}): 3413 (N-H of amide), 2963, 2869, 1730 (C=O of ester) , 1694 (C=O of amide), 1624 , 1608, 1514, 1454, 1409, 1315, 1259, 1188, 1114, 1016, 923, 803, 772 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ=1.28 (s, 9H, $3CH_3$), 1.29 (s, 9H, $3CH_3$), 5.61 (s, 1H, NH), 7.42 (d, $^3J_{HH}$ =8.0 Hz, 2H), 7.51 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.63 (t, $^3J_{HH}$ =7.5 Hz, 2H), 7.93 (s, 1H), 8.03 (d, $^3J_{HH}$ =8.0 Hz, 2H), 8.52 (s, 1H, CH-O), 8.63 (d, $^3J_{HH}$= 8.5 Hz, 2H, orto). ^{13}C NMR ($CDCl_3$): δ= 28.57 ($3CH_3$), 31.02 ($3CH_3$), 35.06 ($C_{t\text{-}Bu}$), 51.79 ($C_{t\text{-}Bu\text{-}NH}$), 69.75 (CH), 124.25 (2C), 125.14 (2CH), 125.44 (2CH), 126.72 (C), 126.76 (C), 126.98 (2CH), 129.29 (2CH), 129.84 (2CH), 129.96 (2CH), 131.05 (2C), 131.60 (CH), 165.58 (C=O of ester) 168.43 (C=O of amide).

***1-(anthracen-9-yl)-2-(cyclohexylamino)-2-oxoethyl 4-chlorobenzoate* (4f)**

White powder, yield (81 %); m.p.: 193-194 °C; IR (KBr) (υ_{max}, cm^{-1}): 3411 (N-H of amide), 2967, 1727 (C=O of ester), 1693 (C=O of amide), 1593, 1511 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ=0.65-2.08 (m, 10H, $5CH_2$), 3.78 (m, 1H, CH), 5.26 (d, $^3J_{HH}$=8.5 Hz, 1H, NH), 7.93 (s, 1H, CH-O), 7.20-8.65 (m, 13H, arom). ^{13}C NMR ($CDCl_3$): δ=24.48 (CH_2), 24.67 (CH_2), 25.21 (CH_2), 29.60 (CH_2), 32.35 (CH_2), 48.62 (CH), 69.83 (CH), 123.97 (CH), 125.32 (CH), 125.75 (CH), 127.35 (2C), 128.10 (CH), 128.75 (2 CH), 129.34 (CH), 130.34 (2CH), 131.00 (CH), 131.40 (CH), 136.71 (2C), 139.67 (C), 164.72 (C=O of ester), 168.01 (C=O of amide).

***1-(anthracen-9-yl)-2-(cyclohexylamino)-2-oxoethyl 3-methylbenzoate* (4g)**

Yellow oil, yield (81 %); IR (neat) (υ_{max}, cm^{-1}): 3338 (N-H of amide), 2930, 2853, 1727 (C=O of ester), 1690 (C=O of amide), 1624, 1531 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ= 0.97-2.01 (m, 10H, $5CH_2$), 2.31 (s, 3H, CH_3), 3.84 (m, 1H, CH), 5.64 (d, $^3J_{HH}$= 7.5 Hz, 1H, NH), 7.91 (s, 1H, CH-O), 7.21-8.01 (m, 13H, arom). ^{13}C NMR ($CDCl_3$): δ= 21.22 (CH_3), 24.62 (CH_2), 24.75 (CH_2), 25.30 (CH_2), 32.53 (CH_2), 32.66 (CH_2), 48.66 (CH), 69.77 (CH), 124.25 (C), 125.22 (C), 125.54 (CH), 126.41 (C), 127.10 (CH), 128.31 (CH), 129.13 (CH), 129.35 (CH), 130.15 (CH), 130.54 (CH), 131.09 (C), 131.60 (C), 134.08 (C), 138.16 (C), 165.88 (C=O, of ester), 168.31 (C=O, of amide).

***1-(anthracen-9-yl)-2-(cyclohexylamino)-2-oxoethyl 4-(tert-butyl)benzoate* (4h)**

Yellow oil, yield (83 %); IR (neat) (υ_{max}, cm^{-1}): 3341 (N-H of amide), 3053, 2854, 1723 (C=O of ester), 1687 (C=O of amide), 1624, 1512 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ= 0.85-2.03 (m, 10H, $5CH_2$), 1.29 (s, 9H, $3CH_3$), 3.78 (m, 1H, CH), 5.71 (d, $^3J_{HH}$= 7.75 Hz, 1H, NH), 7.98 (s, 1H, CH-O), 7.42-8.63 (m, 13H, arom). ^{13}C NMR ($CDCl_3$): δ= 24.64 (CH_2), 24.76 (CH_2), 25.34 (CH_2), 31.03 ($3CH_3$), 32.60 (CH_2), 32.76 (CH_2), 35.06 (CH), 48.61 (CH), 69.71 (CH), 124.27 (C), 125.17 (CH), 125.44 (CH), 126.60 (C), 126.79 (C), 127.03 (CH), 129.31 (CH), 129.88 (CH), 130.04 (CH), 131.07 (C), 131.60 (CH), 157.07 (C), 165.88 (C=O, of ester), 168.31 (C=O, of amide).

***1-(anthracen-9-yl)-2-(tert-butylamino)-2-oxoethyl 4-fluorobenzoate* (4i)**

Yellow oil, yield (79 %); m.p.; 135-137 °C; IR (KBr) (υ_{max}, cm^{-1}): 3412 (N-H of amide), 2967, 1731(C=O of ester), 1694 (C=O of amide), 1603, 1506 cm^{-1}. ^{1}H NMR ($CDCl_3$): δ=1.20 (s , 9H, $3CH_3$), 5.26 (s, 1H, NH), 7.90 (s, 1H, CH-O), 7.06 (t, $^3J_{HH}$=8.5 Hz, 2H, meta,), 7.52 (t, $^3J_{HH}$= 7.5Hz, 2H), 7.64 (t, $^3J_{HH}$=7.5 Hz, 2H), 8.04 (s, 1H, CH-O), 8.08 (d, $^3J_{HH}$=8.5 Hz, 2H, orto), 8.11 (d, $^3J_{HH}$=10 Hz, 2H), 8.57 (d, $^3J_{HH}$=10 Hz, 2H). ^{13}C NMR ($CDCl_3$): δ= 28.47 ($3CH_3$), 51.85 (C), 69.83 (CH), 115.57(d, $^2J_{CF}$=22 Hz, 2CH, meta), 124.06 (2C), 125.28 (2CH), 125.88 (C), 126.12 (2C), 127.24 (2CH), 129.36 (2CH), 130.22 (CH), 131.01 (2C), 131.57 (CH),

132.57 (d, $^{3}J_{CF}$=9.5 Hz, 2CH, orto), 165.94 (d,$^{1}J_{CF}$=254.0 Hz, C, para), 163.92 (C=O, of ester) 168.20 (C=O, of ester).

CONCLUSION

We believe that the reported method offers a mild, simple, green, efficient and one-pot synthetic method for the preparation of 1-(anthracen-9-yl)-2-(alkylamino)-2-oxoethyl benzoate derivatives **4a-i** *via* a Passerini multicomponent reaction of anthracene-9-carbaldehyde **1** in water. The reaction proceeds smoothly and cleanly under mild conditions and no side reactions were observed. The products were obtained in excellent yields. The use of non-toxic and inexpensive solvent, simple procedure, good yields of the products, and mild reaction conditions are the advantages of this method.

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