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Dedicated to the memory of Professor Margareta Avram on the remembrance of her 100th anniversary

MICROWAVE-ASSISTED MULTICOMPONENT SYNTHESIS OF BENZO[*f*]PYRROLO[1,2-*a*]QUINOLINE DERIVATIVES

Emilian GEORGESCU[a], Florentina GEORGESCU,[b] Florea DUMITRASCU,[c] Constantin DRAGHICI,[c] Alina NICOLESCU,[c,d] Daniela MARINESCU[a] and Calin DELEANU[c,d,*]

[a] Chimcomplex Research Center, Str. Uzinei 1, RO-240050 Râmnicu Vâlcea, Roumania
[b] Enpro Soctech Com srl, RO-050524 Bucharest, Roumania
[c] "C. D. Nenitescu" Centre of Organic Chemistry of the Roumanian Academy, Spl. Independenței 202-B, RO-060023 Bucharest, Roumania
[d] "Petru Poni" Institute of Macromolecular Chemistry, Roumanian Academy, Aleea Grigore Ghica Vodă 41-A, RO-700487 Iași, Roumania

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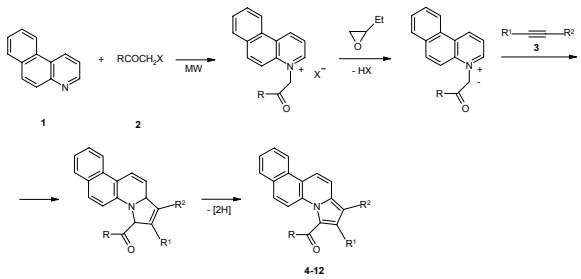
We present an efficient one-pot, three component microwave-assisted synthesis of benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives starting from benzo[*f*]quinoline, 2-bromo-acetophenones or 2-chloro-(*N*-phenyl)acetamides and electron-deficient alkynes. This synthetic strategy provides a direct and easy access to a range of novel benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives. The method has the advantages of considerable shorter reaction time, reduced solvent consumption, operational simplicity and minimal impact on the environment. Nine new benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives have been synthesized with the new method and they are fully characterized.



INTRODUCTION

Heterosteroids are synthetic steroidal systems in which at least one carbon atom is replaced by a heteroatom. Benzo[*f*]pyrrolo[1,2-*a*]quinoline is a fused ring system with a bridgehead nitrogen atom structurally similar to the steroid skeleton[1–4] and its derivatives show potential biological activity and interesting features as dyes.[5,6] Benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives have been prepared *via* multistep synthesis from 1-hydroxy-8-methoxy-benzo[*f*]quinoline,[2] by 1,3-dipolar cycloaddition reactions of benzo[*f*]quinolinium *N*-ylides with acetylenic dipolarophiles,[3] or by the cascade reactions of *N*-(naphthalen-2-yl)pent-4-ynamides with aryl-acetylenes under microwave irradiation in the presence of $AuBr_3$-$AgSbF_6$ catalyst system.[4] The 1,3-dipolar cycloaddition reactions of quinolinium *N*-ylides with acetylenic dipolarophiles is the most versatile method for the synthesis of a large range of related pyrrolo[1,2-*a*]quinoline compounds.[7-10] The reported synthetic route toward benzo[*f*]pyrrolo[1,2-*a*]quinolines implies the intermediate preparation of benzo[*f*]quinolinium salts from benzo[*f*]quinoline and

* Corresponding author: calin.deleanu@yahoo.com

2-bromoacetophenones, followed by 1,3-dipolar cycloaddition reactions of benzo[*f*]quinolinium *N*-ylides, generated *in situ* from the corresponding salts in the presence of an epoxide, with acetylenic dipolarophiles and the aromatization of intermediate primary cycloadducts.[3] The main disadvantage of this synthetic pathway was the long reaction time for both quaternization and 1,3-dipolar cycloaddition steps. Therefore, an efficient, rapid, and clean synthetic procedure towards benzo[*f*]pyrrolo[1,2-*a*]quinoline compounds remain an important subject.

Multicomponent reactions offer a convenient single step procedure toward structurally diverse heterocyclic compounds in a straight manner.[11–13] Multicomponent reactions combined with fast reaction kinetic of microwave-assisted synthesis offer a facile, efficient and versatile method toward novel heterocyclic compounds. The microwave-assisted multicomponent synthesis method substantially decreases the reaction time, is less solvent and energy consuming and it improves the yields.[14–21] Our interest in the synthesis of heterocyclic bioactive compounds[22–26] and extensive expertise in 1,3-dipolar cycloadditions,[27–29] prompted us to investigate an alternative route for preparation of novel benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives. Herein we present an efficient one-pot, three component microwave-assisted synthesis of a range of novel benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives which has the advantages of considerable shorter reaction time, reduced solvent consumption, operational simplicity and minimal impact on the environment.

RESULTS AND DISCUSSION

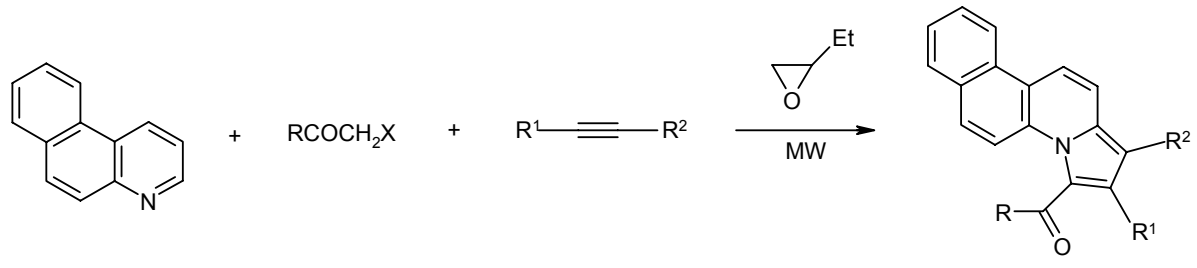
The one-pot, three components microwave-assisted synthesis of novel benzo[*f*]pyrrolo[1,2-*a*]quinolines starts from benzo[*f*]quinoline, 2-bromo-acetophenones or 2-chloro-(*N*-phenyl) acetamides and electron-deficient alkynes in 1,2-epoxybutane which acts both as reaction medium and acid scavenger. To choose the optimal conditions for this multicomponent reactions of benzo[*f*]quinoline with 2-bromo-acetophenones and activated alkynes we examined firstly the reaction of benzo[*f*]quinoline **1** with 2-bromo-3-methoxy-benzophenone **2a** and 1-phenyl-2-propyn-1-one **3a** in 1,2-epoxybutane under microwave irradiation at temperatures ranging from 80 to 120°C. By the end of each experiment, 1,2-epoxybutane was partly evaporated and the crystallized benzo[*f*]pyrrolo [1,2-*a*]quinoline **4** was filtered off and weighed. The best reaction conditions were reached under microwave irradiation of reaction mixture at 120°C for 30 minutes.

To find the optimal conditions for the successful multicomponent reactions of benzo[*f*]quinoline with 2-chloro-(*N*-phenyl)acetamides and activated alkynes we examined the reaction of benzo[*f*]quinoline **1** with 2-chloro-*N*-(3-chlorophenyl)acetamide **2e** and ethyl propiolate **3d** in 1,2-epoxybutane under microwave irradiation at different temperatures ranging from 90 to 130°C. By the end of each experiment, 1,2-epoxybutane was partly removed and the crystallized benzo[*f*]pyrrolo[1,2-*a*]quinoline **9** was filtered off and weighed. The best conditions were achieved under microwave irradiation of reaction mixture at 120°C for 120 minutes. It worth mentioning that in the same reaction carried out under classical heating for 12 days the yield of the isolated product **9** was under 2%.

All new benzo[*f*]pyrrolo[1,2-*a*]quinolines were synthesized under the optimal conditions by the one-pot, three-component reactions of benzo[*f*]quinolines **1**, with 2-bromo-acetophenones **2a-d** or 2-chloro-(*N*-phenyl)acetamides **2e-g** and electron-deficient alkynes **3a-d** in 1,2-epoxybutane (Scheme 1).

A wide range of 2-bromoacetyl derivatives or 2-chloro-(*N*-phenyl)acetamides which can stabilize the intermediate benzo[*f*]quinolinium *N*-ylides can be used as quaternizing reagents and any available electron-deficient alkynes can be used as dipolarophiles (Table 1).

The microwave-assisted multicomponent synthesis involves a sequence of steps starting with the quaternization of benzo*[f]*quinoline **1** with 2-haloacetyl derivatives **2** giving the benzo[*f*]quinolinium salts. In the next step, the benzo[*f*]quinolinium *N*-ylides are generated from the benzo[*f*]quinolinium salts under the action of 1,2-epoxypropane. Finally, the 1,3-dipolar cycloaddition reactions between benzo[*f*]quinolinium *N*-ylides and acetylenic dipolarophiles **3** lead to benzo[*f*]pyrrolo[1,2-*a*]quinolines **4–12** by the aromatization of the intermediate primary adduct (Scheme 2).



1 **2** **3** **4-12**

a: R=3-MeOC$_6$H$_4$, X=Br
b: R=3,4-(MeO)$_2$C$_6$H$_3$, X=Br
c: R=4-MeC$_6$H$_4$, X=Br
d: R=4-NO$_2$C$_6$H$_4$, X=Br
e: R=3-ClC$_6$H$_4$NH, X=Cl
f: R=2,6-Et$_2$C$_6$H$_3$NH, X=Cl
g: R=4-i-PrC$_6$H$_4$NH, X=Cl

a: R^1=H, R^2=COPh
b: R^1=H, R^2=COMe
c: R^1=R^2=CO$_2$Me
d: R^1=H, R^2=CO$_2$Et

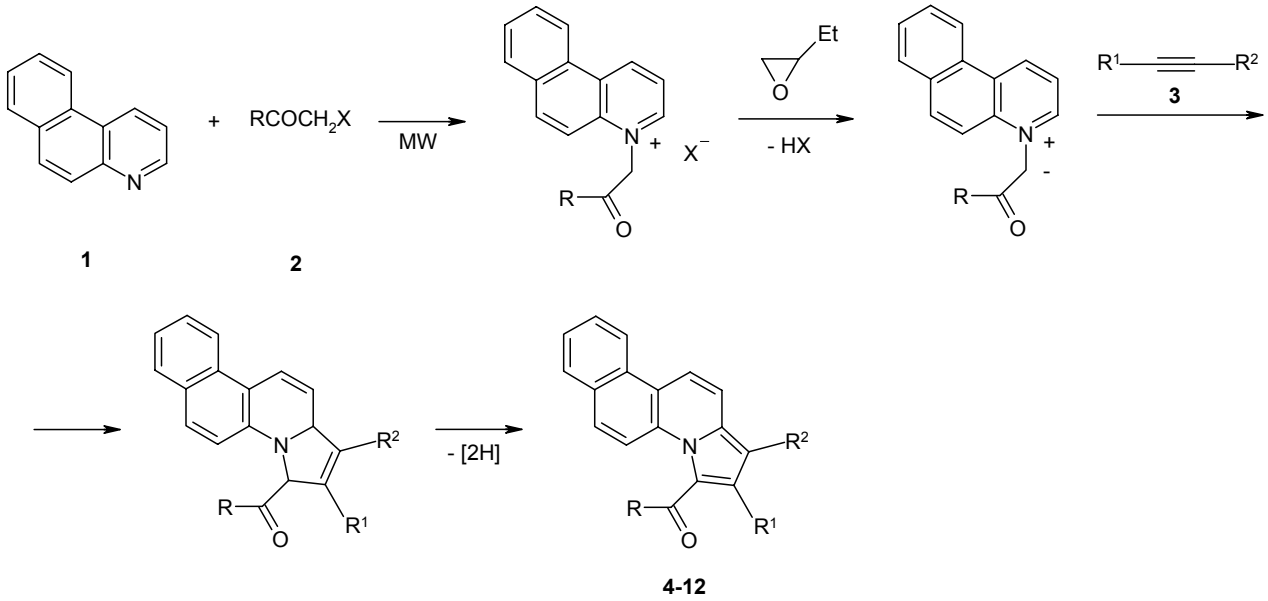
Scheme 1 – The microwave-assisted multicomponent synthesis of benzo[*f*]pyrrolo[1,2-*a*]quinolines.

Table 1

Melting points and yields of newly synthesized benzo[*f*]pyrrolo[1,2-*a*]quinolines

Comp.	R	R^1	R^2	mp (^{o}C)	Yield* (%)
4	3-MeOC$_6$H$_4$	H	COPh	154-155	48
5	3,4-(MeO)$_2$C$_6$H$_3$	H	COMe	250-252	47
6	3-MeOC$_6$H$_4$	CO$_2$Me	CO$_2$Me	231-232	36
7	4-MeC$_6$H$_4$	CO$_2$Me	CO$_2$Me	253-255	53
8	4-NO$_2$C$_6$H$_4$	CO$_2$Me	CO$_2$Me	289-291	40
9	3-ClC$_6$H$_4$NH	H	CO$_2$Et	284-286	42
10	2,6-Et$_2$C$_6$H$_3$NH	H	CO$_2$Et	303-305	39
11	4-*i*-PrC$_6$H$_4$NH	H	CO$_2$Et	295-297	38
12	3-ClC$_6$H$_4$NH	CO$_2$Me	CO$_2$Me	254-256	40

*isolated yields.



1 **2** **4-12**

Scheme 2 – The synthetic pathway toward benzo[*f*]pyrrolo[1,2-*a*]quinolines.

The IR spectra of compounds **4–8** show characteristic absorption bands of benzoyl group at about 1624–1652 cm^{-1} and carbonyl absorption bands at about 1732–1739 cm^{-1} and 1696–1706

cm^{-1} for the two carbomethoxy groups. The IR spectra of compounds **9-12** exhibit characteristic NH absorption bands at about 3217–3315 and 2948–2967 cm^{-1}, carbonyl absorption bands at about 1639–1666 cm^{-1} for the amido group, carbonyl absorption bands at 1693–1694 cm^{-1} for carbethoxy group and two carbonyl absorption bands at 1704 and 1696 cm^{-1} for the carbomethoxy group.

This synthetic procedure has the advantages of considerable shorter reaction time, reduced solvent spending and minimal environment impact.

EXPERIMENTAL

General information

Microwave-assisted syntheses were carried out using a Biotage Initiator 2.0 EXP – ED instrument**.** Melting points were determined on a Boëtius hot plate microscope. The IR spectra were recorded on a Nicolet Impact 410 spectrometer, in KBr pellets. The NMR spectra were recorded either on a Varian Gemini 300 BB instrument, operating at 300.1 MHz and 75.5 MHz for ^1H and ^{13}C nuclei respectively, or on a Bruker Avance NEO 600 instrument operating at 600.1 and 150.9 MHz for ^1H and ^{13}C nuclei respectively. Chemical shifts are reported as δ (ppm) and were referenced to internal TMS for ^1H chemical shifts and to the internal deuterated solvent for ^{13}C chemical shifts ($CDCl_3$ referenced at 77.0 ppm). Signals have been unambiguously assigned based on additional COSY, HSQC/HETCOR and HMBC experiments. The elemental analysis was carried out on a COSTECH Instruments EAS32 apparatus. Satisfactory microanalyses for all new compounds were obtained.

2-Chloro-(*N*-phenyl)acetamides were obtained from the corresponding anilines and chloroacetyl chloride in toluene in the presence of NaOH 25%. 3-Methoxyphenacyl and 3,4-dimethoxyphenacyl bromides were prepared by the bromination of corresponding acetophenones with bromine in diethyl ether. 1-Phenyl-2-propyn-1-one was obtained by the oxidation of 1-phenyl-2-propyn-1-ol with CrO_3 in acidic media. Benzo[*f*]quinoline was obtained in good yield by Skraup synthesis starting from 2-naphthylamine.[30] The other 2-bromoacetophenones, electron-deficient alkynes and 1,2-epoxybutane were commercially products and used without supplementary purifications.

General procedure for multicomponent microwave-assisted syntheses of benzo[*f*]pyrrolo[1,2-*a*[quinolines (4–8)

A mixture of benzo[*f*]quinoline **1** (0.27 g, 1.5 mmol), 2-bromoacetophenone **2a–d** (1.5 mmol) and activated alkyne **3** (1.75 mmol) in 18 mL 1,2-epoxybutane was placed into a sealed microwave reactor at 120°C for 30 min. The reaction mixture was cooled to room temperature, the solvent was partially removed in vacuum, 5 mL of MeOH was added under gentle stirring and the reaction mixture was left overnight at 5-10°C. The solid formed was filtered-off, washed on the filter with cold MeOH and then with diethyl ether. The crude products were recrystallized from $CHCl_3/Et_2O$.

1-Benzoyl-3-(3-methoxybenzoyl)benzo[f]pyrrolo[1,2-a]quinoline **(4).** Yellow crystals (0.33 g, 48 %). ^1H NMR (300.1 MHz, CDCl3) δ (ppm): 3.89 (s, 3H, OMe), 7.18 (dd, 1H, J = 8.2, 2.7 Hz, H-4, methoxybenzoyl), 7.42-7.74 (m, 8H, H-6, H-8, H-9; 3H-phenyl, 2H-methoxybenzoyl), 7.56 (s, 1H, H-2), 7.83-7.86 (m, 2H, H-2, H-6, phenyl), 7.93-7.97 (m, 2H, H-5, H-7), 7.96 (d, 1H, J = 1.9, H-2, methoxybenzoyl), 8.61 (d, 1H, J = 8.5, H-10), 8.65 (d, 1H, J = 9.3, H-12), 8.71 (d, 1H, J = 9.3 Hz, H-11). ^{13}C NMR (75.5 MHz, CDCl3) δ (ppm): 55.6 (MeO), 104.7, 114.1 (CH, methoxybenzoyl), 114.5, 118.4 (CH), 119.7 (CH), 119.8 (CH), 121.6 (CH), 122.9 (CH), 123.1 (CH), 124.9, 126.9 (CH), 127.6 (CH), 127.9 (CH), 128.5 (CH, C-3, C-5, phenyl), 128.9 (CH), 129.1 (CH, C-2, C-6, phenyl), 129.6 (CH), 129.7, 129.9 (CH), 130.9, 131.5 (CH), 131.7 (CH), 131.8, 139.6, 140.0, 141.1, 159.9 (C-3, methoxybenzoyl), 184.3, 190.6 (2CO). IR (KBr, cm^{-1}): 3064, 2834, 1624, 1608, 1578, 1537, 1489, 1414, 1349, 1292, 1252, 1166, 1048. Anal. Calcd. for $C_{31}H_{21}NO_3$ (455.52): C, 81.74; H, 4.65; N, 3.07%. Found: C, 81.91; H, 4.74; N, 2.96 %.

1-Acetyl-3-(3,4-dimethoxybenzoyl)benzo[f]pyrrolo[1,2-a]quinoline ***(5).*** Brown crystals (0.30 g, 47 %). ^1H NMR (600.1 MHz, $CDCl_3$) δ (ppm): 2.56 (s, 3H, Me), 4.00, 4.03 (2s, 6H, OMe), 7.03 (d, 1H, *J* = 8.3 Hz, H-5 from benzoyl), 7.63-7.66 (m, 2H, H-2, H-8), 7.70 (d, 1H, *J* = 1.8 Hz, H-2 from benzoyl), 7.75 (t, 1H, *J* = 8.0 Hz, H-9), 7.82 (dd, 1H, *J* = 8.3, 1.8 Hz, H-6 from benzoyl), 7.92-7.97 (m, 3H, H-5, H-6, H-7), 8.64-8.66 (m, 2H, H-10, H-11), 8.78 (d, 1H, *J* = 9.5 Hz, H-12). ^{13}C NMR (150.9 MHz, $CDCl_3$) δ (ppm): 28.2 (Me), 56.1, 56.2 (2xOMe), 110.0 (CH-5 benzoyl), 111.9 (CH-2 from benzoyl), 115.1 (C-1), 118.5 (CH-12), 119.5 (CH-5), 121.3 (C-4a), 122.9 (CH-10), 124.5 (CH-11), 125.1 (CH-6 from benzoyl), 126.8 (CH-8), 127.3 (C-3), 127.8 (CH-9), 128.81, 128.84 (CH-6, CH-2), 129.7 (CH-7and C-10a), 130.7 (C-7a), 131.0 (C-1 from benzoyl), 131.6 (C-11a), 139.3 (C-1a), 149.3 (C-3 from benzoyl), 153.5 (C-4 from benzoyl), 183.9 (CO), 193.4 (COMe). IR (KBr, cm^{-1}): 2927, 1652, 1599, 1506, 1450, 1406, 1341, 1258, 1235, 1169, 1139, 1024. Anal. Calcd. for $C_{27}H_{21}NO_4$ (423.47): C, 76.58; H, 4.53; N, 3.00%. Found: C, 76.72; H, 4.71; N, 2.88%.

Dimethyl 3-(3-methoxybenzoyl)benzo[f]pyrrolo[1,2-a]quinoline-1,2-dicarboxylate **(6).** Yellow crystals (0.25 g, 36 %). ^1H NMR (300.1 MHz, $CDCl_3$) δ (ppm): 3.47, 3.87, 3.91 (3s, 9H, 3MeO), 7.18 (dd, 1H, *J* = 8.2, 2.7 Hz, H-4, methoxybenzoyl), 7.38 (t, 1H, *J* = 7.4, H-5, methoxybenzoyl), 7.53-7.67 (m, 5H, H-6, H-8, H-9; 2H-methoxybenzoyl), 7.79 (d, 1H, *J* = 9.3 Hz, H-5), 7.84 (d, 1H, *J* = 8.0 Hz, H-7), 8.34 (d, 1H, *J* = 9.6 Hz, H-12). 8.41 (d, 1H, *J* = 9.6 Hz, H-12), 8.47 (d, 1H, *J* = 8.5 Hz, H-10). ^{13}C NMR (75.5 MHz, $CDCl_3$) δ (ppm): 51.8, 52.5, 55.6 (3MeO), 104.7, 113.4 (CH-4, methoxybenzoyl), 117.8 (CH-2), 118.5 (CH-6), 121.0 (CH-5), 121.1 (CH), 122.8 (CH), 123.1 (CH), 125.7, 126.8 (CH), 127.9 (CH), 128.8 (CH), 129.6 (CH), 129.7 (CH), 129.8, 130.0 (CH), 130.7, 131.1, 137.5, 139.1, 145.8, 159.8 (C-3, methoxybenzoyl), 163.6, 165.5 (2COO), 186.6 (CO). IR (KBr, cm^{-1}): 2997, 2940, 1732, 1706, 1633, 1581, 1540, 1466, 1435, 1352, 1288, 1213, 1088, 1041. Anal. Calcd. for $C_{28}H_{21}NO_6$ (467.48): C, 71.94; H, 5.46; N, 6.89%. Found: C, 71.75; H, 5.33; N, 7.02%.

Dimethyl 3-(4-methylbenzoyl)benzo[f]pyrrolo[1,2-a]quinoline-1,2-dicarboxylate **(7).** Orange crystals (0.36 g, 53 %). ^1H NMR (300.1 MHz, $CDCl_3$) δ (ppm): 2.44 (s, 3H, Me), 3.48, 3.92 (2s, 6H, 2MeO), 7.29 (d, 1H, *J* = 8.5 Hz, H-3, H-5,

methylbenzoyl), 7.58 (t, 1H, *J* = 7.4 Hz, H-9), 7.65 (d, 1H, *J* = 9.3 Hz, H-6), 7.69 (t, 1H, *J* = 7.7 Hz, H-9), 7.81 (d, 1H, *J* = 9.3 Hz, H-5), 7.86-7.91 (m, 3H, H-7, 2H-methylbenzoyl), 8.42 (d, 1H, *J* = 9.8 Hz, H-11), 8.47 (d, 1H, *J* = 9.8 Hz, H-12), 8.58 (d, 1H, *J* = 8.2 Hz, H-10). ^{13}C NMR (75.5 MHz, $CDCl_3$) δ (ppm): 22.0 (Me), 51.8, 52.5 (2MeO), 104.6, 118.1 (CH), 118.5 (CH), 121.1, 122.8 (CH), 122.9 (CH), 126.1, 126.8 (CH), 127.9 (CH), 128.8 (CH), 129.0, 129.5 (C-3, C-5, methylbenzoyl), 129.8, 129.9 (CH), 130.1 (C-2, C-6, methylbenzoyl), 130.7, 131.2, 135.2, 137.3, 144.9, 163.7, 165.5 (2COO), 187.0 (CO). IR (KBr, cm^{-1}): 2992, 1740, 1698, 1623, 1601, 1549, 1490, 1466, 1400, 1339, 1253, 1205, 1174, 1093. Anal. Calcd. for $C_{28}H_{21}NO_5$ (451.48): C, 74.49; H, 4.69; N, 3.10%. Found: C, 74.60; H, 4.84; N, 2.95%.

Dimethyl 3-(4-nitrobenzoyl)benzo[f]pyrrolo[1,2-a]quinoline-1,2-dicarboxylate **(8).** Pale brown crystals (0.29 g, 40 %). ^{1}H NMR (300.1 MHz, $CDCl_3$+TFA) δ (ppm): 3.60, 4.03 (2s, 6H, 2MeO), 7.52 (d, 1H, *J* = 9.3 Hz, H-6), 7.68 (t, 1H, *J* = 7.4 Hz, H-9), 7.78 (t, 1H, *J* = 7.7 Hz, H-8), 7.88 (d, 1H, *J* = 9.3 Hz, H-5), 7.93 (d, 1H, *J* = 8.2 Hz, H-7), 8.10 (d, 2H, *J* = 8.6 Hz, H-2, H-6, nitrobenzoyl), 8.35 (d, 1H, *J* = 8.6 Hz, H-3, H-5, nitrobenzoyl), 8.43 (d, 1H, *J* = 9.6 Hz, H-11), 8.58 (d, 1H, *J* = 8.2 Hz, H-10), 8.71 (d, 1H, *J* = 9.6 Hz, H-12). ^{13}C NMR (75.5 MHz, $CDCl_3$+TFA) δ (ppm): 53.1, 53.9 (2MeO), 105.0 (C-1),117.3 (CH), 118.0 (CH), 121.9, 122.9 (CH), 124.1 (C-2, C-6, nitrobenzoyl) 124.8, 125.9 (CH), 127.7 (CH), 128.7 (CH), 129.1 (CH), 129.5, 130.8, 131.0 (C-3, C-5, nitrobenzoyl), 131.2 (CH), 131.5, 139.0, 142.0, 150.8 (C-4, nitrobenzoyl), 165.1, 167.7 (2COO), 185.5 (CO). IR (KBr, cm^{-1}): 2953, 1739, 1696, 1626, 1597, 1524, 1491, 1469, 1424, 1335, 1256, 1211, 1097. Anal. Calcd. for $C_{27}H_{18}N_2O_7$ (482.45): C, 67.22; H, 3.76; N, 5.81%. Found: C, 67.36; H, 3.88; N, 5.73%.

General procedure for multicomponent microwave-assisted syntheses of benzo[*f*]pyrrolo[1,2-*a*[quinolines (9–12)

A mixture of benzo[*f*]quinoline **1** (0.27 g, 1.5 mmol), 2-chloro-(*N*-phenyl)acetamide **2e–g** (1.5 mmol) and activated alkyne **3** (1.75 mmol) in 18 mL 1,2-epoxybutane was placed into a sealed microwave reactor at 120 °C for 120 minutes. The reaction mixture was cooled to room temperature and was worked-up as shown above for the compounds **4–8**. The crude products were recrystallized from $CHCl_3$/Et_2O.

Ethyl 3-[N-(3-chlorophenyl)carbamoyl]benzo[f]pyrrolo[1,2-a]quinoline-1-carboxylate **(9).** Beige crystals (0.28 g, 42 %). ^{1}H NMR (600.1 MHz, DMSO) δ (ppm): 1.39 (t, 3H, *J* = 7.1 Hz, CH_3-Et), 4.38 (q, 2H, *J* = 7.1 Hz, OCH_2), 7.22 (dd, 1H, *J* = 8.0, 2.8 Hz, H-4 from chlorophenyl), 7.45 (t, 1H, *J* = 8.0 Hz, H-5 from chlorophenyl), 7.72 (t, 1H, *J* = 7.0 Hz, H-8), 7.74 (dd, 1H, *J* = 8.2, 1.1 Hz, H-6 from chlorophenyl), 7.81 (td, 1H, *J* = 8.3, 1.2 Hz, H-9), 7.89 (s, H-2), 8.04 (t, 1H, *J* = 1.9 Hz, H-2 from chlorophenyl), 8.09 (d, 1H, *J* = 7.7 Hz, H-7), 8.11 (d, 1H, *J* = 9.7 Hz, H-5), 8.17 (d, 1H, *J* = 9.4 Hz, H-6), 8.42 (d, 1H, *J* = 9.6 Hz, H-12), 8.82 (d, 1H, *J* = 9.6 Hz, H-11), 8.85 (d, 1H, *J* = 8.5 Hz, H-10), 11.05 (s, 1H, NH). ^{13}C NMR (150.9 MHz, DMSO) δ (ppm): 14.4 (CH_3-Et), 59.7 (OCH_2), 105.0 (C-1), 117.3 (CH-12), 118.4 (CH-6 from chlorophenyl), 118.5 (CH-5), 119.4 (CH-2 from chlorophenyl), 119.9 (C-11a), 122.2 (CH-2), 122.3 (CH-11), 123.4 (C-3 and CH-10), 123.5 (CH-4 from chlorophenyl), 126.8 (CH-8), 127.9 (CH-9), 128.6 (CH-7), 129.2 (C-10a), 129.5 (CH-6), 130.2 (C-6a), 130.5 (CH-5 from chlorophenyl), 130.9 (C-4), 133.1 (C-3 from chlorophenyl), 137.0 (C-12a), 140.6 (C-1 from chlorophenyl), 162.4 (CO), 163.4 (COO). IR (KBr, cm^{-1}): 3257, 1693, 1640, 1593, 1517, 1477, 1429, 1320, 1238, 1194, 1095. Anal. Calcd. for $C_{26}H_{19}ClN_2O_3$ (442.91): C, 70.51; H, 4.32; N, 6.32%. Found: C, 70.69; H, 4.41; N, 6.19%.

Ethyl 3-[N-(2,6-diethylphenyl)carbamoyl]benzo[f]pyrrolo[1,2-a]quinoline-1-carboxylate **(10).** Orange crystals (0.27 g, 39 %). ^{1}H NMR (600.1 MHz, DMSO) δ (ppm): 1.29 (t, 6H, *J* = 7.6 Hz, 2xCH_3-Et), 1.41 (t, 3H, *J* = 7.1 Hz, CH_3-EtO), 2.75 (q, 4H, *J* = 7.4 Hz, 2xCH_2-Et), 4.41 (q, 2H, *J* = 7.1 Hz, OCH_2), 7.24 (d, 2H, *J* = 7.6 Hz, H-3 from diethylphenyl), 7.30 (t, 1H, *J* = 7.7 Hz, H-4 from diethylphenyl), 7.71 (t, 1H, *J* = 7.0 Hz, H-8), 7.81 (td, 1H, *J* = 7.0, 1.2 Hz, H-9), 7.83 (s, H-2), 8.13 (d, 1H, *J* = 7.5 Hz, H-7), 8.16 (d, 1H, *J* = 9.4 Hz, H-6), 8.27 (d, 1H, *J* = 9.4 Hz, H-5), 8.44 (d, 1H, *J* = 9.6 Hz, H-12), 8.81 (d, 1H, *J* = 9.6 Hz, H-11), 8.85 (d, 1H, *J* = 8.5 Hz, H-10), 10.37 (s, 1H, NH). ^{13}C NMR (150.9 MHz, DMSO) δ (ppm): 14.4 (CH_3-EtO), 14.8 (2xCH_3-Et), 24.5 (2xCH_2-Et), 59.8 (OCH_2), 104.9 (C-1), 117.4 (CH-12), 118.2 (CH-5), 119.8 (C-11a), 121.0 (CH-2), 121.9 (CH-11), 123.4 (CH-10), 123.8 (C-3), 126.4 (CH-3 from diethylphenyl), 126.7 (CH-8), 127.6 (CH-4 from diethylphenyl), 127.9 (CH-9), 128.6 (CH-7), 128.9 (CH-6), 129.2 (C-10a), 130.1 (C-6a), 130.9 (C-4), 133.1 (C-2 from diethylphenyl), 136.7 (C-12a), 141.8 (C-1 from diethylphenyl), 161.4 (CO), 163.4 (COO). IR (KBr, cm^{-1}): 3217, 2967, 1693, 1639, 1546, 1510, 1495, 1432, 1296, 1234, 1092, 1071. Anal. Calcd. for $C_{29}H_{26}N_2O_3$ (450.54): C, 77.31; H, 5.82; N, 6.22%. Found: C, 77.18; H, 5.91; N, 6.34%.

Ethyl 3-[N-(4-iso-propylphenyl)carbamoyl]benzo[f]pyrrolo[1,2-a]quinoline-1-carboxylate **(11).** Beige crystals (0.26 g, 38 %). ^{1}H NMR (600.1 MHz, DMSO) δ (ppm): 1.24 (d, 6H, *J* = 6.9 Hz, 2xCH_3), 1.39 (t, 3H, *J* = 7.1 Hz, CH_3-EtO), 2.90 (heptet, 1H, *J* = 6.8 Hz, CH), 4.38 (q, 2H, *J* = 7.1 Hz, OCH_2), 7.28 (d, 2H, *J* = 8.3 Hz, H-3 from 4-iso-propylphenyl), 7.72 (t, 1H, *J* = 7.2 Hz, H-8), 7.75 (d, 2H, *J* = 8.3 Hz, H-2 from 4-iso-propylphenyl), 7.79 (s, H-2), 7.81 (t, 1H, *J* = 7.8 Hz, H-9), 8.10 (d, 1H, *J* = 7.9 Hz, H-7), 8.13 (d, 1H, *J* = 9.4 Hz, H-5), 8.17 (d, 1H, *J* = 9.4 Hz, H-6), 8.43 (d, 1H, *J* = 9.6 Hz, H-12), 8.80 (d, 1H, *J* = 9.6 Hz, H-11), 8.85 (d, 1H, *J* = 8.5 Hz, H-10), 10.86 (s, 1H, NH). ^{13}C NMR (150.9 MHz, DMSO) δ (ppm): 14.4 (CH_3-EtO), 23.9 (2xCH_3), 32.9 (CH), 59.7 (OCH_2), 104.8 (C-1), 117.3 (CH-12), 118.3 (CH-5), 119.8 (C-11a), 120.1 (CH-2 from 4-iso-propylphenyl), 121.2 (CH-2), 121.9 (CH-11), 123.4 (CH-10), 124.0 (C-3), 126.5 (CH-3 from 4-iso-propylphenyl), 126.7 (CH-8), 127.9 (CH-9), 128.6 (CH-7), 129.2 (C-10a), 129.4 (CH-6), 130.1 (C-6a), 130.9 (C-4), 136.6 (C-12a), 136.7 (C-1 from 4-iso-propylphenyl), 144.0 (C-4 from 4-iso-propylphenyl), 160.2 (CO), 163.4 (COO). IR (KBr, cm^{-1}): 3275, 2955, 1694, 1640, 1591, 1519, 1456, 1427,, 1309, 1235, 1193, 1093, 1070. Anal. Calcd. for $C_{30}H_{28}N_2O_3$ (464.57): C, 77.56; H, 6.08; N, 6.03%. Found: C, 77.70; H, 6.21; N, 5.89%.

Dimethyl 3-[N-(3-chlorophenyl)carbamoyl]benzo[f]pyrrolo[1,2-a]quinoline-1,2-dicarboxylate **(12).** Beige crystals (0.29 g, 40 %). ^{1}H NMR (600 MHz, DMSO) δ (ppm): 3.82, 3.90 (2s, 6H, OMe), 7.27 (d, 1H, *J* = 7.8 Hz, H-4 from chlorophenyl), 7.45 (t, 1H, *J* = 8.0 Hz, H-5 from chlorophenyl), 7.54 (d, 1H, *J* = 8.0 Hz, H-6 from chlorophenyl), 7.73 (t, 1H, *J* = 7.4 Hz, H-8), 7.84 (t, 1H, *J* = 7.5 Hz, H-9), 7.92 (s, 1H, H-2 from chlorophenyl), 8.02 (d, 1H, *J* = 9.3 Hz, H-5), 8.09 (d, 1H, *J* = 8.0 Hz, H-7), 8.21 (d, 1H, *J* = 9.4 Hz, H-6), 8.33 (d, 1H, *J* = 9.7 Hz, H-12), 8.80 (d, 1H, *J* = 9.7 Hz, H-11), 8.86 (d, 1H, *J* = 8.5 Hz, H-10), 11.21 (s, 1H, NH). ^{13}C NMR (150.9 MHz, DMSO) δ (ppm): 51.7, 52.5 (OMe), 103.4 (C-3), 115.9

(CH-5), 117.5 (CH-12), 118.3 (CH-6 from chlorophenyl), 119.2 (CH-2 from chlorophenyl), 120.2 (C-11a), 122.1 (CH-11), 123.4 (CH-10), 123.5 (C-1), 123.8 (C-2), 124.2 (CH-4 from chlorophenyl), 127.1 (CH-8), 128.3 (CH-9), 128.7 (CH-7), 129.3 (C-10a), 130.3 (C-6a), 130.5 (CH-6), 130.8 (CH-5 from chlorophenyl), 133.3 (C-3 from chlorophenyl), 134.1 (C-12a), 139.7 (C-1 from chlorophenyl), 160.0 (CO), 163.1 (COO), 164.5 (COO). IR (KBr, cm^{-1}): 3315, 2948, 1704, 1696, 1666, 1595, 1526, 1468, 1420, 1249, 1184, 1098. Anal. Calcd. for $C_{27}H_{19}ClN_2O_5$ (486.92): C, 66.60; H, 3.93; N, 7.28%. Found: C, 66.78; H, 4.01; N, 7.14%.

CONCLUSIONS

We developed the most efficient one-pot, three component microwave-assisted synthesis of benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives which provides a direct and easy access to a range of novel benzo[*f*]pyrrolo[1,2-*a*]quinoline derivatives otherwise difficult to obtain. This method has the advantages of considerable shorter reaction time, reduced solvent consumption, operational simplicity and minimal impact on the environment.

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