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# Green and catalyst-free synthesis of aminoanthraquinone derivatives in solvent-free conditions

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## Abstract

In this study, new derivatives of aminoanthraquinone have been synthesized via one-pot three-component condensation reaction of 1- and 2-amino anthraquinones, triethyl orthoformate and CH-acid compounds without using any solvent or catalyst in mild temperature (50 °C). This simple and efficient method yields the desired products in short reaction time (14–50 min) and good to excellent yields (85–96%). Moreover, chemical structures of synthesized products have been entirely confirmed by FT-IR, $^{1}$H and $^{13}$CNMR and mass spectroscopy and melting points.

**Keywords** Multicomponent reactions · Anthraquinone · Catalyst-free · Triethyl orthoformate

## Introduction

Environmental pollution is one of the most severe problems that affects human health worldwide by contaminating air, soil and water. Water is the most essential resource in the world, as the existence of life is not possible without it. Water quality is an essential factor for human beings, plants, animals, and all living organisms. Nowadays, the increase in using organic pollutants in various industries causes harmful effects to humans and ecological systems at a fast-growing rate. The wastewater from industries is directly released into the water, thus contaminating the entire fresh water resources. Hence, water pollution has attracted the attention of various researchers and scientists around the world and several solutions have been offered. One of the solution ways of this problem is to use a clean process or the laws of green chemistry [1–9].

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Green chemistry is one of the most important branches of chemistry which deals with sustainable and environmentally friendly materials to achieve better human life. This new emerging field has attracted vast interest these days due to the decrease and removal of hazardous materials and introduces sustainable and useful processes. It is also defined as the careful and accurate planning for molecular design and organic synthesis to eliminate adverse consequences and improve human health and the environment [10–12]. Green Chemistry has a framework of a cohesive set of twelve principles which the use of green conditions is the most important one [13–15]. Solvents and catalysts undoubtedly are the main part of green chemistry, because many of them are flammable, toxic and corrosive and the use of solvents and catalysts in organic reactions and after the reaction, their separation procedures are a huge challenge [16–19]. Certainly, the catalyst- and solvent-free conditions are the safe and useful conditions that are in accordance with principles of green chemistry [20–24]. Thus, it is of supreme importance to employ these conditions in organic synthesis [25–29].

Nowadays, multicomponent reactions (MCRs) have received considerable attention in modern organic chemistry because they combine three or more compounds in a single step to produce a high yield of product, without formation any intermediate or separation procedure [27, 30–34]. Recently, this strategy for drug discovery using the synthesis of biologically active compounds has attracted widespread attention in organic and pharmaceutical chemistry research [35]. It has been generally employed due to its high yields and reduced reaction time compared to conventional multistage reactions [36–39].

Anthraquinones derivatives are polycyclic compounds containing two carbonyl groups in the central ring based on the 9, 10-anthraquinonoid framework [40]. Natural and synthetic anthraquinones are widely used in industry and medicine [41–43]. The importance of studies on anthraquinones in the industry is due to their different applications in many fields, especially in the field of colors, and it has attracted the attention of the paint industry in the fields of food [44], industrial dyes [45], clothing [46] and cosmetics [47]. On the other hand, anthraquinones and their derivatives have received a great deal of attention for specific therapeutic uses such as antimicrobial [48], antibacterial [49], antiviral [50], antioxidant [51], anticancer [52], and antidiabetes activities [53]. For example, anthraquinones derivatives such as idarubicin, mitoxantrone and pixantrone are the members of this class of compounds that are approved by the Food and Drug Administration (USA), for clinical uses (Fig. 1) [42].

Due to the widespread applications of anthraquinone derivatives in industry and their unique biological activity and therapeutic effects. Our research group has been excited to study and synthesis of this fantastic class of organic compounds [54–56]. In our previous research, anthraquinone-scaffold based-enaminodiones (2, 2-diacylethenamines) compounds have been synthesized via one-pot three-component reaction of amino anthraquinones, triethyl orthoformate and 1, 3-dicarbonyl compounds at room temperature and the presence of acetic acid as an acidic catalyst [56]. In this study, a new and green method has been developed to synthesize anthraquinone derivatives via three-component reaction of 1- and

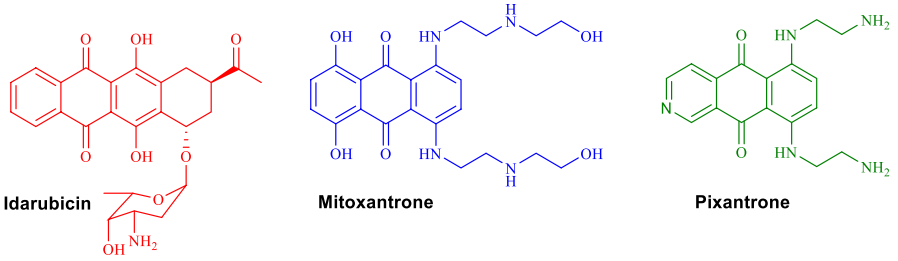


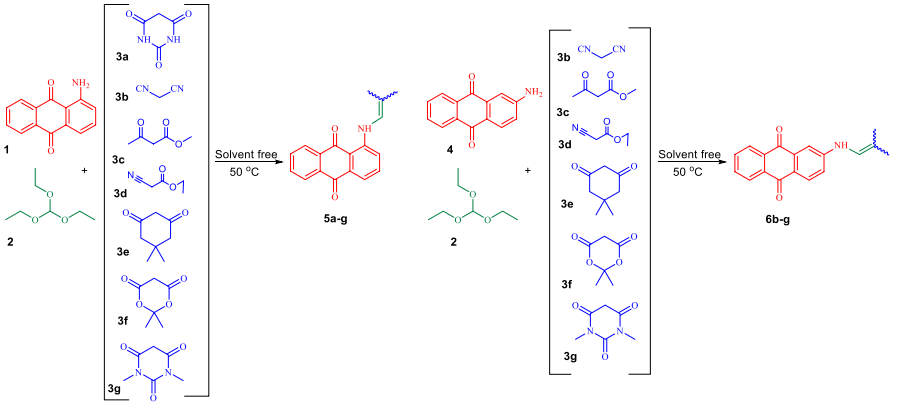

**Fig. 1** Clinically approved anthraquinone derivatives

2-amino anthraquinones, triethyl orthoformate and CH-acid compounds in catalyst- and solvent-free conditions (Scheme 1).

$^{1}$HNMR and $^{13}$CNMR spectroscopy, mass, IR spectra, and the melting point have been exploited to determine the chemical structures of new compounds.

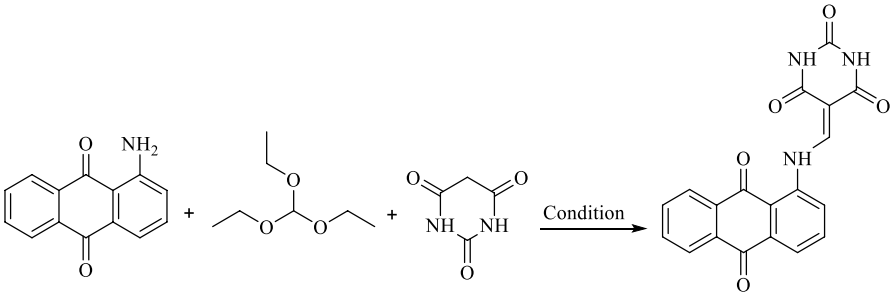
## Results and discussion

The reaction of 1-amino anthraquinone, triethyl orthoformate and barbituric acid has been selected as a model reaction (Table 1). Then, to find the best solvent, the effects of different polar and nonpolar solvents such as $H_2O$, ethanol, acetone, diethyl ether, methanol and solvent-free condition (entries 1–10) have been examined for the synthesis of anthraquinone derivative **5a**. Interestingly, the yield of the model reaction was not satisfying in the presence of different solvents, while, in solvent-free condition, only after 16 min, the yield of the reaction was 96% (entry 9). Thus, the best condition was selected solvent-free condition. After obtaining the optimized condition, the synthesis of anthraquinone derivative **5a** was investigated at different temperatures (entries 8–10). The best temperature was 50 °C, and with increasing the




**Scheme 1** Synthesis of anthraquinone derivatives in catalyst- and solvent-free conditions

**Table 1** Optimization of the reaction condition for the synthesis of **5a**[a]




| Entry | Solvent | Temperature (°C) | Time (min) | Yield (%)[b] |
|---|---|---|---|---|
| 1 | Lactic acid | RT | 180 | – |
| 2 | $H_2O$ | RT | 180 | – |
| 3 | Ethanol | RT | 180 | 63 |
| 4 | Methanol | RT | 180 | 60 |
| 5 | Acetone | RT | 180 | 42 |
| 6 | Diethyl ether | RT | 180 | 38 |
| 7 | Ethanol | Reflux | 180 | 77 |
| 8 | Solvent-free | RT | 180 | 61 |
| 9 | Solvent-free | 50 | 16 | 96 |
| 10 | Solvent-free | 100 | 20 | 96 |

[a]Mixture of 1-amino anthraquinone (1.0 mmol), triethyl orthoformate (1.0 mmol) and barbituric acid (1.0 mmol) in the presence of different solvents and temperatures

[b]Isolated Yield

temperature above this temperature, the yield of the reaction was unchanged (entry 10). Therefore, the highest amount of compound **5a** was obtained in solvent-free condition at 50 °C after only 16 min running the reaction (entry 9).

More importantly, the scope and generality of the method have been studied for the synthesis of a wide variety of amino anthraquinone derivatives through one-pot three-component condensation reaction of 1- and 2-amino anthraquinone, triethyl orthoformate and various CH acids like barbituric acid, malononitirile, dimedone, meldrum's acid, 1, 3-dimethyl barbituric acid, methyl acetoacetate and ethyl cyanoacetate (Table 2). It is worth mentioning that the barbituric acid had the most active CH acid and it reacted better with amino anthraquinone and triethyl orthoformate and the highest amount of yield (96%) was obtained (entry 1). Taking into account other CH-acid compounds, malononitrile was better compared to meldrum's acid with a 95% yield of product (entries 2 and 6). The lowest amount of product was achieved with the use of meldrum's acid as a CH-acid compound (85%) (entry 6). These observations showed that the most and least active CH-acid compounds were barbituric acid and meldrum's acid, respectively, for this method. The new products have been identified by FT-IR, $^{1}H$ and $^{13}C$NMR and mass spectroscopy.

**Table 2** Synthesis of amino anthraquinone derivatives in solvent-free conditions[a]

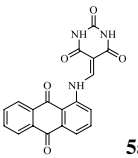
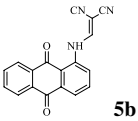
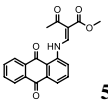
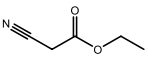
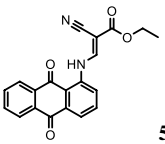
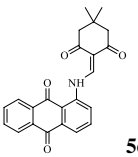
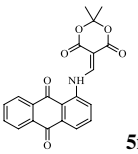
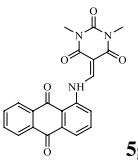
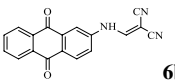
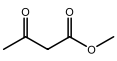
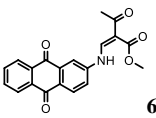
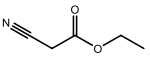
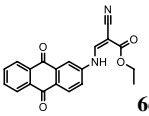
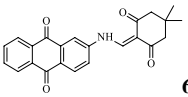
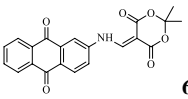
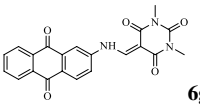
| Entry | CH acid | Product | Time (min) | Yield (%)[b] | Melting point (°C) Found/reported [ref] |
|---|---|---|---|---|---|
| 1 |  |  **5a** | 16 | 96 | > 360/this study |
| 2 |  |  **5b** | 23 | 95 | 320–322/this study |
| 3 |  |  **5c** | 25 | 90 | 250–252/this study |
| 4 |  |  **5d** | 24 | 93 | 266–268/this study |
| 5 |  |  **5e** | 50 | 90 | 258–260/257–259 [56] |
| 6 |  |  **5f** | 14 | 85 | 256–258/256–257 [56] |
| 7 |  |  **5g** | 14 | 94 | > 360/ > 330 [56] |
| 8 |  |  **6b** | 3293 | | 310–312/this study |
| 9 |  |  **6c** | 35 | 91 | 222–223/this study |

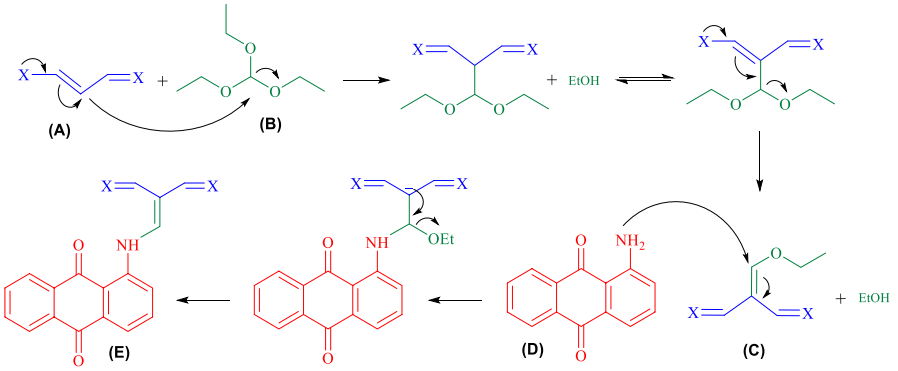
**Table 2** (continued)

| Entry | CH acid | Product | Time (min) | Yield (%)[b] | Melting point (°C) Found/reported [ref] |
|---|---|---|---|---|---|
| 10 |  |  **6d** | 33 | 93 | 263–267/this study |
| 11 |  |  **6e** | 44 | 88 | 259–262/258–260 [56] |
| 12 |  |  **6f** | 50 | 85 | 249–252/258–259 [56] |
| 13 |  |  **6g** | 14 | 90 | 338–340/ > 330 [56] |

[a]Mixture of amino anthraquinone (1.0 mmol), triethyl orthoformate (1.0 mmol) and CH-acid compounds (1.0 mmol) in solvent-free condition at 50 °C temperatures

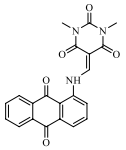
To better understanding product synthesis, the proposed mechanism was illustrated in Scheme 2. In the first step, the CH-acid compound (**A**) attacks the triethyl orthoformate (**B**) and then removes two molecules of EtOH, and the intermediate (**C**) was formed consequently. Then, from the rapid Knoevenagel condensation reaction of aminoanthraquinone (**D**) with the previous intermediate, the target anthraquinone-based enamino-dicarbonyl compound (**E**) was obtained.

The last experiment in this research was the comparison of our method with other previously published approaches. After literature survey, only two published articles observed for the synthesis of amino anthraquinone derivatives. As shown in Table 3, our method has some worthwhile advantages over previous approaches. For




**Scheme 2** Proposed mechanism for the synthesis of amino anthraquinone derivatives

**Table 3** The comparison of our method with other approaches for the synthesis of product **5 g**

| Entry | Product | Conditions | Temp. (°C) | Time (h) | Yield (%)[a] | Reference |
|---|---|---|---|---|---|---|
| 1 |  | Acetic acid | r.t | 24 | 93 | [56] |
| 2 | | Formic acid | reflux | 30 | 94 | [57] |
| 3 | | Solvent-free | 50 | 14[b] | 94 | This work |

[a]Isolated yields

[b]Minutes

example, we synthesized amino anthraquinone derivatives in milder reaction conditions without using any acidic or basic catalysts and also solvents. More importantly, the product **5 g** has been prepared after only 14 min. running the reaction, while in previous methods, the synthesis of product **5 g** was carried out in longer reaction times (24 and 30 h) (Table 3, entries 1 and 2).

## Experimental section

### Materials and instruments

Melting points were analyzed on an Electrothermal type 9100 melting point apparatus. The FT-IR spectra were performed on an Avatar 370 FT-IR Thermo-Nicolet spectrometer. Proton and Carbon NMR spectra were obtained with a Bruker DRX-300 Avance spectrometer at 300 MHz. All materials were purchased from Sigma-Aldrich and Merck companies.

### General procedure for the synthesis of aminoanthraquinone derivatives

A mixture of 1- or 2-amino anthraquinone (1.0 mmol), triethyl orthoformate (1.0 mmol) and CH-acid compounds (1.0 mmol) was stirred in an oil bath at 50 °C for the time mentioned in Table 2. After finalizing the reaction (monitored by TLC (n-hexane: ethyl acetate 1:1), the obtained solid products were collected to give desired products. The structures of the synthesized products described by their physical and spectral data ($^{1}$H NMR, $^{13}$C NMR, and FT-IR spectra and mass).

### 5-(((9,10-Dioxo-9,10-dihydroanthracen-1-yl)amino)methylene) pyrimidine-2,4,6(1H,3H,5H)-trione (5a)

Yield: 96%; in the form of yellow solid; mp > 360 °C; FT-IR ($cm^{-1}$) 3171, 3030, 2992, 2833, 1734, 1689, 1653, 1592, 1561, 1498, 1445, 1426, 1361, 1302, 1260, 1163, 1048, 1000, 859, 803, 781, 733, 705, 608, 524, 511, 492, 448, 408;

$^{1}$H NMR (300 MHz, DMSO-$d_6$): 7.92–7.97 (m, 3H, 3CH arom), 8.07 (d, $J$ = 6.9, 1H, CH arom), 8.19 (d, $J$ = 9, 1H, CH arom), 8.23–8.28 (m, 2H, 2CH arom), 8.75 (d, $J$ = 13.5, 1H, CHNH), 11.01–11.10 (2S, 2H, 2 CONHCO), 14.11 (d, J = 13.5, CHNH); $^{13}$C NMR (300 MHz, DMSO-$d_6$): 96.02, 119.94, 122.99, 124.19, 126.95, 127.41, 132.72, 134.17, 134.76, 135.08, 135.27, 136.13, 140.73, 150.34 (CHNH), 151.18 (HNCONH), 164.05 (CCONH), 165.32 (CCONH), 182.45, 185.28; Mass (MALDI-TOF) m/z calcd for C19H11N3O5 [M]þ 361.07 found: 361.2.

### 2-(((9,10-Dioxo-9,10-dihydroanthracen-1-yl)amino)methylene)malononitrile (5b)

Yield: 95%; in the form of warm red solid; mp 320–322 °C; FT-IR (cm$^{-1}$) 3067, 2229, 1670, 1650, 1614, 1584, 1572, 1494, 1400, 1364, 1341, 1261, 1232, 1162, 1076, 1001, 969, 887, 827, 793, 728, 700, 625, 610, 587, 555, 425; $^{1}$H NMR (300 MHz,DMSO-$d_6$): 7.95–8.05 (m, 4H, 4CH arom), 8.20–8.30 (m, 4H, 4CH arom), 9.19 (d, $J$ = 9.9, 1H, CHNH), 12.87 (d, $J$ = 9.6, 1H, CHNH); $^{13}$C NMR (300 MHz, DMSO-$d_6$) 57.82, 113,42, 115,26 (CN), 118.27 (CN), 122.31, 123.78, 127.13, 127.68, 132.81, 133.87, 134.50, 135.43, 135.59, 136.47, 140.87, 156.31 (CHNH), 182.26, 187.26; Mass (MALDI-TOF) m/z calcd for C18H9N3O2 [M]þ 299.07 found: 299.1.

### Methyl (E)-2-(((9,10-dioxo-9,10-dihydroanthracen-1-yl)amino)methylene)-3 oxobutanoate (5c)

Yield: 90%; in the form of dark orange solid; mp: 250–252 °C; FT-IR (cm$^{-1}$) 3154, 3088, 3013, 2956, 1703, 1664, 1642, 1574, 1558, 1475, 1434, 1397, 1339, 1307, 1250, 1195, 1164, 1139, 1085, 1062, 1003, 968, 881, 832, 806, 730, 706, 659, 618, 571, 545, 496, 415; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 2.22 (s, 3H, $CH_3$), 3.776 (s, 3H, $CH_3$), 7.93–8.02 (m, 4H, 4CH arom), 8.16–8.24 (m, 3H, 3CH arom),8.67 (d, $J$ = 12.9, 1H, CHNH), 14.34 (d, $J$ = 12.6, 1H,CHNH); $^{13}$C NMR (300 MHz, $CDCl_3$) 31.478, 51.69, 120.65, 123.74, 127.06, 128.02, 134.38, 134.71, 135.29, 141.83, 148.79(CHNH), 167.41(C$COOCH_3$), 199.28 ($H_3CCO$C); Mass (MALDI-TOF) m/z calcd for C20H15NO5 [M]þ 349.1 found: 349.

### Ethyl (E)-2-cyano-3-((9,10-dioxo-9,10-dihydroanthracen-1-yl)amino)acrylate (5d)

Yield: 93%; in the form of yellow solid; mp: 266–268 °C; FT-IR (cm$^{-1}$) 3076, 2980, 2905, 2218, 1707, 1673, 1613, 1573, 1381, 1339, 1268, 1230, 1184, 1165, 1112, 1028, 999, 971, 887, 829, 797, 728, 702, 656, 608, 537, 502, 423, 406; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 1.34 (t, 3H, $CH_3$), 4.33 (q, 2H, $CH_2$), 7.83–7.94 (m, 4H, 4CH arom), 8.11–8.23 (m, 3H, 3CH arom) 8.708 (d, $J$ = 8.1, 1H, CHNH), 13.48 (d, $J$ = 8.1,1H, CHNH); $^{13}$C NMR (300 MHz, DMSO-$d_6$): 9.67, 10.35, 14.29, 29.58, 30.28, 60.93, 122.94 (CN), 134.81, 135.82 (CHNH); Mass (MALDI-TOF) m/z calcd for C20H14N2O4 [M]þ 346.10 found: 346.

**1-(((4,4-Dimethyl-2,6-dioxocyclohexylidene) methyl) amino) anthracene-9,10-dione (5e)**

Yield: 90%; in the form of dark yellow solid; mp: 258–260 °C; FT-IR ($cm^{-1}$) 3086, 2952, 2924, 1656, 1610, 1548, 1455, 1410, 1378, 1328, 1231, 1157, 1085, 988, 925, 870, 832, 800, 732, 703,

652, 624; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 1.07 (s, 6H, $2CH_3$), 2.46 (s,2H, $CH_2$), 7.50–8.0 (m, 3H, 3CH arom), 8.11 (dd, *J1* = 7.95, *J2* = 0.9, 1H, CH arom), 8.20–8.31 (m, 3H, 3CH arom), 8.67 (d, *J* = 13.2, 1H, CHNH), 14.47 (d, *J* = 13.2, 1H, CHNH).

**5-(((9,10-Dioxo-9,10-dihydroanthracen-1-yl)amino) methylene)-2,2-dimethyl-1,3-dioxane-4,6-dione (5f)**

Yield: 85%; in the form of saffron yellow solid; mp: 256–258 °C; FT-IR ($cm^{-1}$) 3118, 3076, 2986, 1730, 1676,1649, 1597, 1564, 1493, 1461, 1416, 1373, 1341, 1251, 1194, 1139, 1078, 1002, 933, 857, 807, 785, 737, 710, 673, 643, 546, 504, 483, 428; $^{1}$H NMR (300 MHz, DMSO $d_6$): 1.74 (s, 6H, $2CH_3$), 7.97–8.01 (m, 3H, 3CH arom), 8.12 (d, *J* = 7.5 1H, CH arom), 8.25 (dd, *J1* = 20.4, *J2* = 8.4, 2H, 2CH arom), 8.35 (d, *J* = 8.4, 1H, CH arom), 8.91 (d, *J* = 13.8, 1H, CHNH), 13.84 (d, *J* = 14.1, 1H, CHNH); $^{13}$C NMR (300 MHz, DMSO): 27.12, 90.25, 104.90, 123.32, 124.63, 127.01, 127.57, 132.78, 134.16, 134.74, 135.25, 135.38, 136.17, 140.38, 152.72 (CHNH), 163.15 (CCOO), 163.57 (CCOO), 180.22, 182.47.

**5-(((9,10-Dioxo-9,10-dihydroanthracen-1-yl)amino) methylene)-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (5 g)**

Yield: 94%; in the form of light yellow solid; mp > 360 °C; FT-IR ($cm^{-1}$) 3116, 3081, 1722, 1668, 1636, 1595, 1568, 1503, 1464, 1431, 1357, 1307, 1268, 1240, 1177, 1085, 999, 984, 860, 830, 803, 780, 752, 732, 704, 655, 616, 545, 483, 415.

**2-(((9,10-Dioxo-9,10-dihydroanthracen-2-yl)amino)methylene)malononitrile (6b)**

Yield: 93%; in the form of dark goldenrod solid; mp 310–312 °C; FT-IR ($cm^{-1}$) 3293, 3203, 3114, 3061, 2225, 1669, 1655, 1640, 1572, 1486, 1282, 1101, 977, 929, 862, 709, 659, 639, 582, 551, 472, 434; $^{1}$H NMR (300 MHz,DMSO-$d_6$): 7.93–7.97 (m, 3H, 3CH arom), 8.17–8.24 (m, 4H, 4CH arom), 8.84 (s, 1H, CHNH), 11.57 (s, 1H, CHNH); $^{13}$C NMR (300 MHz, DMSO-$d_6$) 55.73, 113.96, 115.85 (CN), 116.22 (CN), 122.86, 127.19, 127.27, 129.29, 129.58, 133.51, 133.54, 134.85, 134.94, 135.17, 144.74, 156.08 (CHNH), 181.79, 182.56; Mass (MALDI-TOF) m/z calcd for C18H9N3O2 [M]þ 299.07 found: 299.1.

**Methyl (Z)-2-(((9,10-dioxo-9,10-dihydroanthracen-2-yl)amino) methylene)-3-oxobutanoate (6c)**

Yield: 91%; in the form of brown solid; mp: 222–223 °C; FT-IR ($cm^{-1}$) 3376, 3203, 2943, 1712, 1670, 1643, 1585, 1563, 1432, 1403, 1376, 1356, 1319, 1286, 1237,

1189, 1151, 1072, 978, 931, 844, 795, 713, 662, 641, 573, 552, 501, 430, 413; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 2.48 (s, 3H, $CH_3$), 3.79 (s, 3H, $CH_3$), 7.93–7.99 (m, 3H, 3CH arom), 8.13 (d, $J=1.8$ 1H, CH arom) 8.23–8.26 (m, 3H, 3CH arom) 8.56 (d, $J=30$, 1H, CHNH), 12.56 (d, $J=8.1$. 1H, CHNH); $^{13}$C NMR (300 MHz, $CDCl_3$): 31.46, 51.70, 104.79, 114.20, 122.65, 127.47, 130.01, 133.47, 133.66, 134.23, 134.65, 135.45, 144.17, 150.24 (CHNH), 166.80 (CCOOCH$_3$), 181.82, 182.74, 201.16 ($CH_3$COC); Mass (MALDI-TOF) m/z calcd for C20H15NO5 [M]þ 349.10 found: 349.

**Ethyl (Z)-2-cyano-3-((9,10-dioxo-9,10-dihydroanthracen-2-yl)amino)acrylate (6d)**

Yield: 93%; in the form of greenish yellow solid; mp: 263–267 °C; FT-IR ($cm^{-1}$) 3392, 3178, 3082, 2944, 2230, 1708, 1678, 1642, 1587, 1491, 1366, 1329, 1291, 1257, 1181, 1126, 1018, 977, 932, 909, 842, 760, 709, 660, 640, 608, 568, 545, 500; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 2.47 (t, 3H, $CH_3$), 3.78 (q, 2H, $CH_2$), 7.94–7.97 (m, 3H, 3CH arom), 8.11–8.26 (m, 4H, 4CH arom) 8.59 (s, 1H, CHNH), 12.56 (s, 1H, CHNH); $^{13}$C NMR (300 MHz, DMSO-$d_6$): 10.29, 14.29, 60.75, 77.79, 114.95 (CN), 122.05, 126.72, 128.57, 128.89, 132.92, 134.26, 134.62, 151.35 (CHNH), 164.04, 181.14, 182.03; Mass (MALDI-TOF) m/z calcd for C20H14N2O4 [M]þ 346.10 found: 346.

**2-(((4,4-Dimethyl-2,6-dioxocyclohexylidene)methyl)amino) anthracene-9,10-dione (6e)**

Yield: 88%; in the form of light brown solid; mp: 259–262 °C; FT-IR ($cm^{-1}$) 3050, 2933, 2866, 2314, 1661, 1574, 1447, 1407, 1325, 1267, 1211, 1130, 1015, 972, 924, 897, 845, 791, 748, 710, 663, 621; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 1.05 (s, 6H, $2CH_3$), 2.44 (s, 2H, $CH_2$), 7.96–8.0 (m, 2H, 2CH arom), 8.07 (dd, $J1=8.4$, $J2=2.4$ 1H, CH arom), 8.20 (d, $J=2.1$, 1H, CH arom), 8.23–8.27 (m, 3H, 3CH arom), 8.62 (d, $J=13.2$, 1H, CHNH),12.67 (d, $J=13.5$, 1H, CHNH).

**5-(((9,10-Dioxo-9,10-dihydroanthracen-2-yl)amino) methylene)-2,2-dimethyl-1,3-dioxane-4,6-dione (6f)**

Yield: 85%; in the form of cool brown solid; mp: 249–252 °C; FT-IR ($cm^{-1}$) 3182, 2997, 1733, 1672, 1624, 1573, 1504, 1463, 1380, 1263, 1198, 1136, 1097, 1025, 987, 926, 858, 824, 782, 715, 640.

**5-(((9,10-Dioxo-9,10-dihydroanthracen-2-yl)amino) methylene)-1,3-dimethylpyrimidine-2,4,6(1H,3H,5H)-trione (6 g)**

Yield: 90%; in the form of cool brown solid; mp: 338–340 °C; FT-IR($cm^{-1}$) 3108, 1716, 1656, 1585, 1498, 1469, 1425, 1297, 1205, 1176, 1151, 1094, 1052, 981, 933, 895, 802, 753, 710, 657; $^{1}$H NMR (300 MHz, DMSO-$d_6$): 3.24 (s, 3H, $CH_3$), 3.26 (s, 3H, $CH_3$), 7.96–7.99 (m, 2H, 2CH arom), 8.10–8.13 (dd, 1H, CH arom), 8.23–8.29 (m, 4H, 4CH arom), 8.76 (d, 1H, CHNH); 8.81 (d, 1H, CHNH).

## Conclusions

In summary, the new strategy has been introduced for the synthesis of different derivatives of aminoanthraquinone through a one-pot three-component condensation reaction of amino anthraquinone, triethyl orthoformate and CH-acid compounds. One of the most advantages of this method was the environmentally friendly conditions without using any solvent or catalyst and in agreement with the principles of green chemistry. Besides, this strategy was simple and rapid to give the desired products with good to excellent yields with easy workup procedure.

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