

Strontium(II) triflate catalysed condensation of β -naphthol, aldehyde and urea or amides: a facile synthesis of amidoalkyl naphthols

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A novel and efficient synthesis of amidoalkyl naphthols has been developed by one-pot condensation of β -naphthol, aromatic aldehydes and urea or amides in the presence of strontium(II) triflate as a catalyst in high yields.

Key words: strontium(II) triflate, one-pot reaction, condensation, amidoalkyl naphthol, β -naphthol

Naphthalene derivatives have attracted considerable interest because of their pharmaceutical and agricultural activities.^{1–10} For example, naphazoline and pronethalol have been reported to act as cardiovascular agents.⁶ 1-naphthaleneacetic acid and 2-naphthoxyacetic acid have been reported to act as plant growth regulators.^{7,8} Whilst, 2-naphthol and its derivatives have also been reported as bactericides⁹ and antioxidants.¹⁰ Furthermore, many chiral amidoalkyl phenols have been reported as excellent chelating agents for metal-catalysed asymmetric synthesis.^{11–15} Particularly, chiral Mannich bases of β -naphthol are commonly used as metal-mediated and ligand-accelerated catalysts in enantioselective carbon–carbon bond formation.^{16–21}

Various methods have been reported for the aminoalkylation of β -naphthol, with aromatic aldehydes and heterocyclic amines,^{22,23} pyrrolidine,²⁴ iminium salts^{25,26} and methyl carbamate.²⁷ However, to the best of our knowledge, there have been only a few reports of the synthesis of amidoalkyl naphthols in the presence of urea or amides.^{28,29} Recently, Khodaei disclosed that the condensation of β -naphthol, aromatic aldehydes and urea or amides in the presence of *p*-toluenesulfonic acid, afforded amidoalkyl naphthols.²⁸ Patil and co-workers used indion-130 for the synthesis of these compounds.²⁹ However, these methodologies have some limitations such as prolonged reaction times, special apparatus, and the use of environmentally unfriendly catalysts and harsh reaction conditions. The challenge for a sustainable environment calls for clean procedures that can avoid using harmful catalysts. Therefore, the need for a new, simple, green, one-pot methods for the synthesis of amidoalkyl naphthols is highly desirable.

Multicomponent reactions (MCRs) have attracted considerable attention due to the fact that the products are formed in a single step without the need to isolate any intermediate. Diversity can be achieved simply by varying the reacting components and also complex molecules can be built up.^{30–33} Thus, development and discovery of new MCRs is of interest.

In recent years, rare earth metal triflate salts have been widely used in organic synthesis due to their low toxicity, low cost, high stability, ease of handling, water tolerance and ease of recovery from water.³⁴ Recently, we have successfully

Table 1 Catalyst effect on the condensation reaction of 4-chlorobenzaldehyde, β -naphthol and urea

| Entry | Catalyst | Catalyst/mol% | Time/h | Yield/% ^b |
|----------------|----------------------|---------------|--------|-----------------------|
| 1 | Y(OTf) ₃ | 10 | 12 | – |
| 2 | Eu(OTf) ₃ | 10 | 12 | – |
| 3 | Mg(OTf) ₂ | 10 | 12 | – |
| 4 | Yb(OTf) ₃ | 10 | 12 | <10 |
| 5 | Zn(OTf) ₂ | 10 | 12 | <10 |
| 6 | Cu(OTf) ₂ | 10 | 12 | 15 |
| 7 | Bi(OTf) ₃ | 10 | 12 | 30 |
| 8 | Sc(OTf) ₃ | 10 | 12 | 35 |
| 9 ^a | Sr(OTf) ₂ | 10 | 12 | 85, 87, 85, 85, 84 |
| 10 | Sr(OTf) ₂ | 1 | 12 | 28 |
| 11 | Sr(OTf) ₂ | 5 | 12 | 70 |
| 12 | Sr(OTf) ₂ | 15 | 12 | 84 |

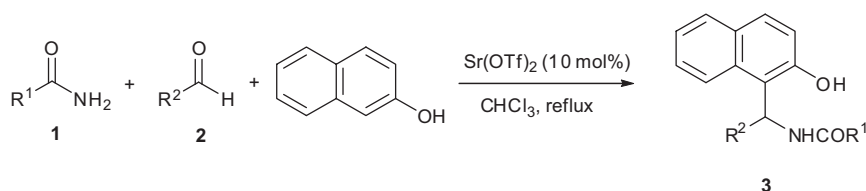
^aCatalyst was reused five times.

^bIsolated yield.

applied metal triflates to several reactions.^{35–37} However, up to now, Sr(OTf)₂ has seldom been studied as a catalyst. We have also reported a simple, efficient and practical procedure for the MCRs such as one-pot synthesis of dihydropyrimidones catalysed by strontium(II) triflate.³⁸

In continuation of our previous work on metal triflate catalysed MCRs, we report a simple, efficient and practical procedures for the synthesis of amidoalkyl naphthols using Sr(OTf)₂³⁹ as the catalyst in chloroform (Scheme 1).

Initially, we investigated the amidoalkylation of β -naphthol, 4-chlorobenzaldehyde and urea using metal triflates as catalyst at reflux temperature in chloroform and the results are listed in Table 1. It was found that all metal triflates showed catalytic effects except Y(OTf)₃, Eu(OTf)₃ and Mg(OTf)₂ (Table 1, entries 1–3). Most excitingly, when Sr(OTf)₂ was used, the reaction proceeded smoothly and gave the product in 85% yield (Table 1, entry 9). Moreover, we found that the yields were obviously affected by different amounts of Sr(OTf)₂. When using 1%, 5%, 10% and 15% of Sr(OTf)₂, the yields were 28%, 70%, 85% and 84% respectively (Table 1, entries 9–12). Therefore, 10% of Sr(OTf)₂ was sufficient and excess amount of catalyst did not increase the yields significantly (Table 1, entries 9–12). The activity of the recycled Sr(OTf)₂ was also examined. In this reaction, Sr(OTf)₂ could be reused



Scheme 1

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Table 2 Solvent effect on the reaction of 4-chlorobenzaldehyde, β -naphthol and urea catalysed by $\text{Sr}(\text{OTf})_2$

| Entry | Solvent | Time/h | Yield/% ^a |
|-------|-------------------------------------|--------|---|
| 1 | CHCl_3 | 12 | 85 |
| 2 | $\text{ClCH}_2\text{CH}_2\text{Cl}$ | 12 | 77 ^b , 78 ^c |
| 3 | MeCN | 12 | <30 |
| 4 | CH_3NO_2 | 12 | 78 ^d , 79 ^e , 65 ^f |
| 5 | EtOH | 12 | <30 |
| 6 | THF | 12 | <10 |
| 7 | H_2O | 12 | – |
| 8 | $[\text{bpy}]\text{BF}_4$ | 12 | 46 |
| 9 | None | 12 | 75 ^g |

^aIsolated yield.^bReaction conditions: 2 ml $\text{ClCH}_2\text{CH}_2\text{Cl}$ at 60°C; ^cReaction conditions: 2 ml $\text{ClCH}_2\text{CH}_2\text{Cl}$ at 80°C; ^dReaction conditions: 2 ml CH_3NO_2 at 60°C; ^eReaction conditions: 2 ml CH_3NO_2 at 80°C; ^fReaction conditions: 2 ml CH_3NO_2 at 100°C; ^gReaction conditions: solvent-free at 120°C.

five times without any loss of activity (Table 1, entry 9). In addition, we studied the reaction with these catalysts in chloroform at room temperature. It was found that the reaction also gave the moderate yields but required much longer reaction time for five days. When no catalyst was used, no products were detected. Showing that $\text{Sr}(\text{OTf})_2$ played an important role in this reaction. Thus our study showed the best results were obtained when 10 mol% $\text{Sr}(\text{OTf})_2$ was used in chloroform under reflux (Table 1, entry 9).

Then, we examined the above reaction in different solvents at 60°C (Table 2). It was found that this condensation reaction was affected by various solvents. The results indicated that acetonitrile, ethanol, tetrahydrofuran and $[\text{bpy}]\text{BF}_4$ are unsuitable for the reaction. When H_2O was used as solvent, no products were detected. Solvents such as 1,2-dichloroethane and nitromethane, proved to be effective. Different yields were obtained at different temperatures (Table 2, entries 2 and 4).

The reaction could be carried out under solvent-free condition at 120°C in 75% yield (Table 2, entry 9). Most excitingly, when chloroform was used as solvent at reflux temperature, the yield increased to 85% (Table 2, entry 1) which was better than any other solvents.

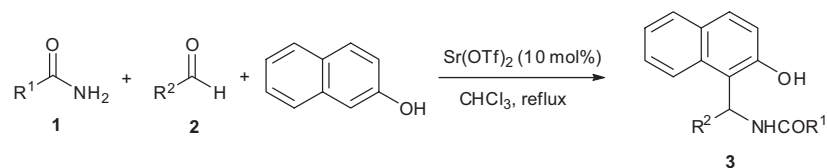
In order to study the generality of this procedure, a series of aldehydes and urea or amides were used. The results are shown in Table 3. The three component condensation reaction proceeded smoothly in chloroform at reflux temperature and were complete in 12 h. The reactions of aldehydes with β -naphthol and urea or different amides provided the corresponding amidoalkyl naphthols in good to excellent yields (Table 3).

In all case, aromatic aldehydes with substituents bearing either electron-donating or electron-withdrawing groups reacted successfully and gave the products in good yields. However, the substituents on the aromatic ring have an influence on the reaction. Aromatic rings bearing electron-withdrawing groups required a shorter time and gave higher yields (Table 3).

The condensation reactions of aromatic aldehydes, β -naphthol with urea gave the corresponding products in good yields. As compared to the urea, we studied that the reactions of aromatic aldehydes with β -naphthol and different amides, including acetamide, benzamide, propenionamide and thiophene-2-carboxamide under similar conditions. All the amides afforded the corresponding amidoalkyl naphthols in higher yields than urea. Furthermore, amidoalkyl naphthols were the sole products and no by-product was observed (Table 3).

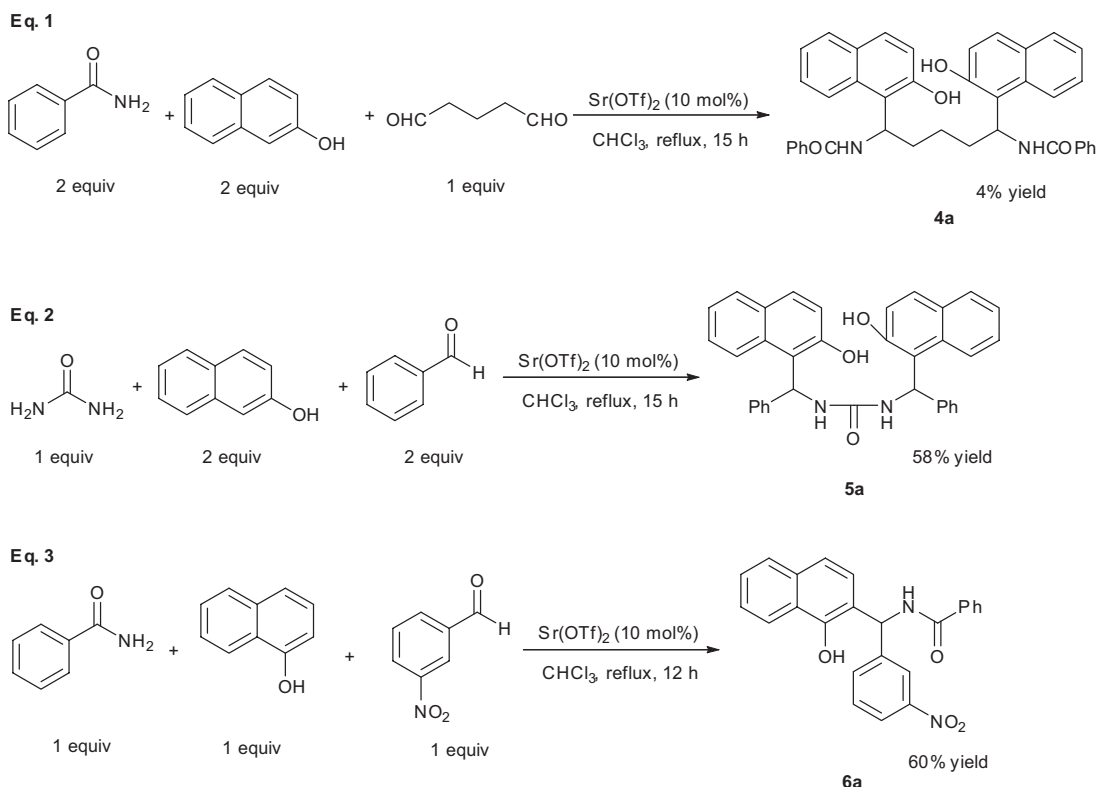
On the basis of the above results, this process was then extended to heterocyclic aldehyde and aliphatic aldehyde. The corresponding products (**3v**, **3w**) were obtained in 80% and 91% yields, respectively (Table 3, entries 22 and 23).

Interestingly, when glutaraldehyde was used as the aldehyde component and reacted with 2 equivalents of β -naphthol

Table 3 Synthesis of amidoalkyl naphthols in the presence of $\text{Sr}(\text{OTf})_2$ 

| Entry | R ¹ | R ² | Product | Time/h | Yield/% ^a |
|-------|-------------------------|---|-----------|--------|----------------------|
| 1 | NH_2 | <i>p</i> -(OCH_3) C_6H_4 | 3a | 15 | 80 |
| 2 | NH_2 | <i>p</i> -(CH_3) C_6H_4 | 3b | 15 | 82 |
| 3 | NH_2 | C_6H_5 | 3c | 12 | 84 |
| 4 | NH_2 | <i>p</i> -(Cl) C_6H_4 | 3d | 12 | 85 |
| 5 | NH_2 | <i>m</i> -(NO_2) C_6H_4 | 3e | 10 | 89 |
| 6 | C_6H_5 | <i>p</i> -(OCH_3) C_6H_4 | 3f | 10 | 89 |
| 7 | C_6H_5 | <i>p</i> -(CH_3) C_6H_4 | 3g | 10 | 90 |
| 8 | C_6H_5 | C_6H_5 | 3h | 9 | 93 |
| 9 | C_6H_5 | <i>p</i> -(Cl) C_6H_4 | 3i | 9 | 93 |
| 10 | C_6H_5 | <i>m</i> -(NO_2) C_6H_4 | 3j | 8 | 95 |
| 11 | 2-thiophene | <i>p</i> -(OCH_3) C_6H_4 | 3k | 10 | 90 |
| 12 | 2-thiophene | <i>p</i> -(CH_3) C_6H_4 | 3l | 10 | 90 |
| 13 | 2-thiophene | C_6H_5 | 3m | 9 | 92 |
| 14 | 2-thiophene | <i>p</i> -(Cl) C_6H_4 | 3n | 9 | 93 |
| 15 | 2-thiophene | <i>m</i> -(NO_2) C_6H_4 | 3o | 8 | 96 |
| 16 | CH_3 | <i>p</i> -(OCH_3) C_6H_4 | 3p | 12 | 89 |
| 17 | CH_3 | C_6H_5 | 3q | 10 | 90 |
| 18 | CH_3 | <i>m</i> -(NO_2) C_6H_4 | 3r | 8 | 93 |
| 19 | $\text{CH}_2=\text{CH}$ | <i>p</i> -(OCH_3) C_6H_4 | 3s | 12 | 90 |
| 20 | $\text{CH}_2=\text{CH}$ | C_6H_5 | 3t | 10 | 90 |
| 21 | $\text{CH}_2=\text{CH}$ | <i>m</i> -(NO_2) C_6H_4 | 3u | 8 | 94 |
| 22 | C_6H_5 | 2-furyl | 3v | 15 | 80 |
| 23 | C_6H_5 | CH_3CH_2 | 3w | 10 | 91 |

^aIsolated yield.



Scheme 2

and 2 equivalents of benzamide it gave the corresponding diamidoalkylation product in only 4% yield (Scheme 2, Eqn (1)). However, the reaction of urea was studied in a diamidoalkylation reaction. The yield of the diamidoalkylation product was 58% (Scheme 2, Eqn (2)), when the molar ratio of aldehyde, β -naphthol and urea was 2:2:1. In addition, using α -naphthol instead of β -naphthol, the desired product was also obtained in moderate yield (Scheme 2, Eqn (3)).

A tentative mechanism for the formation of amidoalkyl naphthols is proposed in Scheme 3. The reaction may proceed via the acylimine intermediate. Referring to the literature,⁴⁰⁻⁴³ we supposed that an intermediate is formed by the reaction of the aldehyde and urea by $\text{Sr}(\text{OTf})_2$. Subsequent addition of the β -naphthol afforded the corresponding amidoalkyl naphthols (Scheme 3).

In summary, we have developed a novel catalytic approach to the synthesis of amidoalkyl naphthols using strontium(II) triflate. The method has several advantages including its high yield, environmental suitability, simple work-up procedure and the easy isolation of its products. Studies on the extension of this protocol are ongoing in our laboratory.

Experimental

Chemicals and solvents were either purchased or purified by standard techniques. Melting points were determined on a Büchi B-540 melting apparatus and uncorrected. The NMR spectra were measured with a Bruker Advance III 500 or Varian Mercury Plus-400 instrument using DMSO as the solvent with TMS as internal standard. IR spectra were recorded using KBr pellets on a Nicolet Aviatar-370 instrument.

Mass spectra were measured with Thermo Finnigan LCQ-Advantage. Elemental analysis was determined on a Carlo-Erba 1108 instrument.

General procedure for the synthesis of amidoalkyl naphthols 3a–3w

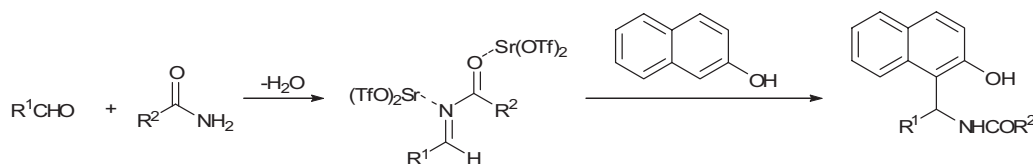
To a mixture of aldehyde (1 mmol), β -naphthol (1 mmol) and urea or amide (1.1 mmol) was added $\text{Sr}(\text{OTf})_2$ (0.1 mmol, 10 mol%) in chloroform (2 ml). The mixture was stirred at 60°C for the time as shown in Table 3. The progress of the reaction was monitored by TLC. After completion of the reaction, the mixture was filtered and the precipitate washed with H_2O . The crude products were purified by recrystallisation from ethyl acetate and the pure products were obtained in 80–96% yields.

General procedure for the synthesis of *N,N'*-(1,5-bis(2-hydroxynaphthalen-1-yl)pentane-1,5-diy)dibenzamide 4a

To a mixture of glutaraldehyde (1 mmol), β -naphthol (2 mmol) and benzamide (2 mmol) was added $\text{Sr}(\text{OTf})_2$ (0.1 mmol, 10 mol%) in chloroform (2 ml). The mixture was stirred at 60°C for 15 h. The progress of the reaction was monitored by TLC. After completion of the reaction, the mixture was filtered and the precipitate washed with H_2O . The crude product was purified by recrystallisation from ethyl acetate and the pure product was obtained in 4% yields.

General procedure for the synthesis of 1,3-bis((2-hydroxynaphthalen-1-yl)(phenyl)methyl)urea 5a

To a mixture of benzaldehyde (2 mmol), β -naphthol (2 mmol) and urea (1 mmol) was added $\text{Sr}(\text{OTf})_2$ (0.1 mmol, 10 mol%) in chloroform (2 ml). The mixture was stirred at 60°C for 15 h. The progress of the reaction was monitored by TLC. After completion of the reaction, the mixture was filtered and the precipitate washed with H_2O . The crude product was purified by recrystallisation from ethyl acetate and the pure product was obtained in 58% yields.



Scheme 3

General procedure for the synthesis of N-((1-hydroxynaphthalen-2-yl)(3-nitrophenyl)methyl)benzamide 6a

To a mixture of 3-nitrobenzaldehyde (1 mmol), α -naphthol (1 mmol) and benzamide (1.1 mmol) was added $\text{Sr}(\text{OTf})_2$ (0.1 mmol, 10 mol%) in chloroform (2 ml). The mixture was stirred at 60°C for 15 h. The progress of the reaction was monitored by TLC. After completion of the reaction, water was added and the product was extracted with ethyl acetate (3 \times 10 ml). The organic layer was dried (MgSO_4) and evaporated, and the crude product was purified by flash chromatography (ethyl acetate/petroleum ether, 1:12 to 1:4) to provide the corresponding product. The pure product was obtained in 60% yield.

Spectroscopic data for the products

1-((2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methyl)urea (3a): Yellow solid m.p. 99–100°C in 80% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.95 (1H, s, OH), 7.82–7.73 (2H, m, ArH), 7.40 (1H, s, NH), 7.29–7.19 (2H, m, ArH), 7.09–7.06 (2H, m, ArH), 6.89 (2H, s), 6.80 (2H, d, J = 8.4 Hz, ArH), 5.82 (2H, s, NH_2), 3.68 (3H, s, OCH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.6, 157.5, 152.9, 136.1, 132.3, 128.8 (CH \times 2), 128.6 (CH \times 3), 127.0, 126.4, 122.4, 120.3, 118.6, 113.3 (CH \times 2), 55.0, 47.8. IR ν_{max} (KBr): 3472, 3367, 1662, 1510, 1250, 1177, 1033, 815, 746, 595 cm^{-1} . MS (ESI) m/z 321 ($[\text{M}-1]^+$, 100). Found: C, 70.60; H, 5.79; N, 8.53. Calc. for $\text{C}_{19}\text{H}_{18}\text{N}_2\text{O}_3$: C, 70.79; H, 5.63; N, 8.69%.

1-((2-hydroxynaphthalen-1-yl)(p-tolyl)methyl)urea (3b): Yellow solid m.p. 117–118°C in 82% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.92 (1H, s, OH), 7.81 (1H, d, J = 8.0 Hz, ArH), 7.74 (1H, d, J = 8.8 Hz, ArH), 7.39 (1H, s, NH), 7.26 (1H, t, J = 7.6 Hz, ArH), 7.20 (1H, d, J = 9.2 Hz, ArH), 7.06–7.01 (4H, m, ArH), 6.89 (2H, s), 5.81 (2H, s, NH_2), 2.22 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.6, 152.9, 141.3, 134.7, 132.3, 128.9 (CH \times 2), 128.6, 128.5 (CH \times 4), 125.8, 122.4 (CH \times 2), 120.4, 118.6, 47.8, 20.6. IR ν_{max} (KBr): 3468, 3386, 1650, 1514, 1439, 1356, 1268, 813, 746 cm^{-1} . MS (ESI) m/z 306 (M^+ , 5), 231 (48), 144 (98). Found: C, 74.33; H, 6.05; N, 9.06. Calc. for $\text{C}_{19}\text{H}_{18}\text{N}_2\text{O}_2$: C, 74.49; H, 5.92; N, 9.14%.

1-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)urea (3c): White solid m.p. 174–175°C in 84% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.97 (1H, s, OH), 7.83–7.75 (3H, m, ArH), 7.40 (1H, s, NH), 7.29–7.11 (7H, m, ArH), 6.94 (2H, s), 5.86 (2H, s, NH_2). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.5, 152.8, 144.3, 128.9, 128.6 (CH \times 2), 127.8, 126.4 (CH \times 4), 125.8 (CH \times 2), 122.4 (CH \times 2), 120.2, 118.5, 48.1. IR ν_{max} (KBr): 3464, 3346, 3217, 1664, 1535, 1438, 1356, 1270, 813, 745, 699 cm^{-1} . MS (ESI) m/z 291 ($[\text{M}-1]^+$, 60), 248 (98), 215 (80), 143 (33). Found: C, 73.86; H, 5.66; N, 9.50. Calc. for $\text{C}_{18}\text{H}_{16}\text{N}_2\text{O}_2$: C, 73.95; H, 5.52; N, 9.58%.

1-((4-chlorophenyl)(2-hydroxynaphthalen-1-yl)methyl)urea (3d): White solid m.p. 115–116°C in 85% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.00 (1H, s, OH), 7.83–7.76 (3H, m, ArH), 7.43 (1H, s, NH), 7.31–7.14 (6H, m, ArH), 6.90 (2H, s), 5.86 (2H, s, NH_2). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.6, 152.9, 148.9, 143.5, 132.1 (CH \times 2), 131.6, 130.4 (CH \times 4), 129.2, 128.7, 127.8, 127.7, 122.5, 118.5, 47.7. IR ν_{max} (KBr): 3483, 3380, 3180, 1654, 1600, 1518, 1439, 1357, 1268, 815, 752 cm^{-1} . MS (ESI) m/z 326 (M^+ , 5), 265 (49), 231 (98), 144 (95). Found: C, 66.06; H, 4.78; N, 8.49. Calc. for $\text{C}_{18}\text{H}_{15}\text{ClN}_2\text{O}_2$: C, 66.16; H, 4.63; N, 8.57%.

1-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)urea (3e): Yellow solid m.p. 179–180°C in 89% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.13 (1H, s, OH), 8.09–7.99 (2H, m), 7.87–7.46 (7H, m), 7.33–7.21 (2H, m), 7.12 (1H, d, J = 8.4 Hz), 7.06–6.99 (1H, m), 5.96 (1H, s). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.5, 153.0, 147.7, 147.1, 132.6, 132.0, 129.6, 129.5, 128.7, 128.3, 126.9, 122.6, 121.0 (CH \times 2), 120.2, 119.1, 118.4, 48.4. IR ν_{max} (KBr): 3479, 3396, 1651, 1528, 1348, 1268, 813, 748 cm^{-1} . MS (ESI) m/z 336 ($[\text{M}-1]^+$, 100), 293 (40). Found: C, 64.01; H, 4.59; N, 12.37. Calc. for $\text{C}_{18}\text{H}_{15}\text{N}_3\text{O}_4$: C, 64.09; H, 4.48; N, 12.46%.

N-((2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methyl)benzamide (3f): White solid m.p. 208–209°C in 89% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.36 (1H, s, OH), 9.05 (1H, d, J = 8.4 Hz, NH), 8.09 (1H, d, J = 8.4 Hz, ArH), 7.87–7.79 (4H, m, ArH), 7.55–7.48 (4H, m, ArH), 7.33–7.23 (5H, m, ArH), 6.85 (2H, d, J = 8.4 Hz, ArH), 3.69 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 153.0, 134.4, 133.9, 132.2, 131.4, 129.2 (CH \times 4), 128.5 (CH \times 3), 127.7 (CH \times 2), 127.0 (CH \times 2), 126.7 (CH \times 2), 122.6 (CH \times 2), 118.7, 113.6 (CH \times 2), 55.0, 48.9. IR ν_{max} (KBr): 3422, 3065, 1630, 1511, 1437, 1345, 1262, 1171, 1029, 826, 712, 589 cm^{-1} . MS (ESI) m/z 382 ($[\text{M}-1]^+$, 100), 143 (20). Found: C, 78.22; H, 5.65; N, 3.53. Calc. for $\text{C}_{25}\text{H}_{21}\text{NO}_3$: C, 78.31; H, 5.52; N, 3.65%.

N-((2-hydroxynaphthalen-1-yl)(p-tolyl)methyl)benzamide (3g): White solid m.p. 216–217°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.35 (1H, s, OH), 9.03 (1H, d, J = 8.4 Hz, NH), 8.10 (1H, d, J = 8.8 Hz, ArH), 7.89–7.79 (4H, m, ArH), 7.55–7.45 (4H, m, ArH), 7.33–7.26 (3H, m, ArH), 7.20 (2H, d, J = 8.0 Hz, ArH), 7.09 (2H, d, J = 8.0 Hz, ArH), 2.24 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 165.6, 153.1, 139.0, 135.6, 134.4, 132.3 (CH \times 2), 131.3, 129.2 (CH \times 4), 128.7 (CH \times 2), 128.5 (CH \times 2), 127.0, 126.7 (CH \times 2), 126.4, 122.6, 118.7, 118.4, 49.1, 20.5. IR ν_{max} (KBr): 3417, 1631, 1530, 1438, 1345, 817, 712, 586 cm^{-1} . MS (ESI) m/z 368 ($[\text{M}-1]^+$, 100), 143 (23). Found: C, 81.65; H, 5.85; N, 3.79. Calc. for $\text{C}_{25}\text{H}_{21}\text{NO}_2$: C, 81.72; H, 5.76; N, 3.81%.

N-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)benzamide (3h): White solid m.p. 242–243°C in 93% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.34 (1H, s, OH), 9.03 (1H, d, J = 8.4 Hz, NH), 8.09 (1H, d, J = 8.8 Hz, ArH), 7.88–7.79 (4H, m, ArH), 7.57–7.45 (4H, m, ArH), 7.33–7.19 (8H, m, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 165.7, 153.1, 142.0, 134.3, 132.3, 131.4, 129.3 (CH \times 2), 128.6 (CH \times 2), 128.5 (CH \times 2), 128.3, 128.1, 127.1 (CH \times 2), 126.7, 126.5, 126.4, 122.6 (CH \times 2), 118.7, 118.3, 49.2. IR ν_{max} (KBr): 3419, 3061, 1631, 1435, 1348, 1272, 823, 697, 584 cm^{-1} . MS (ESI) m/z 353 (M^+ , 5), 231 (98). Found: C, 81.48; H, 5.55; N, 3.93. Calc. for $\text{C}_{24}\text{H}_{19}\text{NO}_2$: C, 81.56; H, 5.42; N, 3.96%.

N-((4-chlorophenyl)(2-hydroxynaphthalen-1-yl)methyl)benzamide (3i): White solid m.p. 187–188°C in 93% yield. ^1H NMR (500 MHz, $\text{DMSO}-d_6$): δ 10.36 (1H, s, OH), 9.02 (1H, d, J = 8.5 Hz, NH), 8.06 (1H, d, J = 8.5 Hz, ArH), 7.88–7.80 (4H, m, ArH), 7.57–7.45 (4H, m, ArH), 7.35–7.23 (7H, m, ArH). ^{13}C NMR (125 MHz, $\text{DMSO}-d_6$): 166.4, 153.7, 141.6, 134.7, 132.7, 132.0 (CH \times 2), 131.6 (CH \times 2), 130.1 (CH \times 2), 129.1 (CH \times 2), 128.9, 128.8, 128.6 (CH \times 2), 127.7, 127.3, 123.2 (CH \times 2), 119.1, 118.4, 49.2. IR ν_{max} (KBr): 3420, 3182, 1630, 1514, 1401, 1340, 1267, 1093, 812, 725 cm^{-1} . MS (ESI) m/z 386 ($[\text{M}-1]^+$, 100), 143 (20). Found: C, 74.21; H, 4.79; N, 3.53. Calc. for $\text{C}_{24}\text{H}_{18}\text{ClNO}_2$: C, 74.32; H, 4.68; N, 3.61%.

N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)benzamide (3j): White solid m.p. 241–242°C in 95% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.45 (1H, s, OH), 9.17 (1H, d, J = 8.0 Hz, NH), 8.14–8.10 (3H, m, ArH), 7.93–7.85 (4H, m, ArH), 7.74 (1H, d, J = 8.0 Hz, ArH), 7.62–7.55 (2H, m, ArH), 7.50 (3H, t, J = 7.6 Hz, ArH), 7.42 (1H, d, J = 8.0 Hz, ArH), 7.34 (1H, t, J = 7.2 Hz, ArH), 7.28 (1H, d, J = 8.4 Hz, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 166.2, 153.4, 147.8, 144.5, 134.0, 133.2, 132.2 (CH \times 2), 131.5, 129.9, 129.7, 128.7 (CH \times 2), 128.4, 127.3, 127.0, 122.8, 122.5, 121.6 (CH \times 2), 120.9, 118.6, 117.3, 48.9. IR ν_{max} (KBr): 3377, 3266, 1633, 1530, 1347, 820, 733, 655 cm^{-1} . MS (ESI) m/z 397 ($[\text{M}-1]^+$, 100), 143 (15). Found: C, 72.26; H, 4.65; N, 6.96. Calc. for $\text{C}_{24}\text{H}_{18}\text{N}_2\text{O}_4$: C, 72.35; H, 4.55; N, 7.03%.

N-((2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methyl)thiophene-2-carboxamide (3k): Yellow solid m.p. 119–120°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.24 (1H, s, OH), 8.94 (1H, d, J = 8.4 Hz, NH), 8.03 (1H, d, J = 8.8 Hz, ArH), 7.86–7.76 (4H, m, ArH), 7.43 (1H, t, J = 8.0 Hz, ArH), 7.31–7.13 (6H, m, ArH), 6.84 (2H, d, J = 8.4 Hz, ArH), 3.69 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 160.5, 158.1, 153.2, 139.4 (CH \times 2), 133.6, 132.3, 131.0, 129.3 (CH \times 2), 128.6, 128.4, 128.0, 127.9, 126.6, 122.9, 122.6, 118.7, 118.2, 113.6 (CH \times 2), 55.1, 48.9. IR ν_{max} (KBr): 3406, 1624, 1536, 1511, 1249, 1177, 816, 745 cm^{-1} . MS (ESI) m/z 388 ($[\text{M}-1]^+$, 100). Found: C, 70.81; H, 5.03; N, 3.53; S, 8.14. Calc. for $\text{C}_{23}\text{H}_{19}\text{NO}_3\text{S}$: C, 70.93; H, 4.92; N, 3.60; S, 8.23%.

N-((2-hydroxynaphthalen-1-yl)(p-tolyl)methyl)thiophene-2-carboxamide (3l): White solid m.p. 206–207°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.27 (1H, s, OH), 8.97 (1H, d, J = 8.8 Hz, NH), 8.04 (1H, d, J = 8.8 Hz, ArH), 7.90–7.76 (4H, m, ArH), 7.44 (1H, t, J = 8.0 Hz, ArH), 7.31–7.14 (6H, m, ArH), 7.09 (2H, d, J = 8.0 Hz, ArH), 2.24 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 160.5, 153.2, 139.3, 138.7 (CH \times 2), 135.6, 132.3, 131.0 (CH \times 2), 129.3 (CH \times 2), 128.7, 128.5, 128.4 (CH \times 2), 127.9, 126.5, 122.5 (CH \times 2), 118.6, 118.2, 49.0, 20.5. IR ν_{max} (KBr): 3387, 3186, 1628, 1536, 814, 722 cm^{-1} . MS (ESI) m/z 372 ($[\text{M}-1]^+$, 30), 143 (5). Found: C, 73.85; H, 5.26; N, 3.69; S, 8.70. Calc. for $\text{C}_{23}\text{H}_{19}\text{NO}_2\text{S}$: C, 73.97; H, 5.13; N, 3.75; S, 8.59%.

N-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)thiophene-2-carboxamide (3m): White solid m.p. 220–221°C in 92% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.24 (1H, s, OH), 8.96 (1H, d, J = 8.4 Hz, NH), 8.04 (1H, d, J = 8.4 Hz, ArH), 7.90–7.77 (4H, m, ArH), 7.44 (1H, t, J = 7.6 Hz, ArH), 7.32–7.14 (9H, m, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 160.7, 153.3, 141.7, 139.3 (CH \times 2), 132.4, 131.1, 129.4 (CH \times 2), 128.6, 128.4 (CH \times 2), 128.2 (CH \times 2), 128.0, 126.6, 122.9, 122.6 (CH \times 2), 118.6, 118.1, 49.2. IR ν_{max}

(KBr): 414, 1620, 1536, 1357, 1057, 820, 719 cm^{-1} . MS (ESI) m/z 358 ($[\text{M}-1]^+$, 25), 143 (3). Found: C, 73.35; H, 4.90; N, 3.83; S, 8.90. Calc. for $\text{C}_{22}\text{H}_{17}\text{NO}_2\text{S}$: C, 73.51; H, 4.77; N, 3.90; S, 8.92%.

N-((4-chlorophenyl)(2-hydroxynaphthalen-1-yl)methyl)thiophene-2-carboxamide (**3n**): White solid m.p. 130–131°C in 93% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.27 (1H, s, OH), 8.96 (1H, d, $J = 8.0$ Hz, NH), 8.01 (1H, d, $J = 8.4$ Hz, ArH), 7.91–7.78 (4H, m, ArH), 7.44 (1H, t, $J = 7.6$ Hz, ArH), 7.36–7.22 (7H, m, ArH), 7.15 (1H, t, $J = 4.4$ Hz, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 160.8, 153.4, 140.9, 139.2 (CH \times 2), 132.3, 131.2, 131.2, 129.7, 128.8, 128.7 (CH \times 2), 128.4, 128.1, 128.0, 126.8, 122.8, 122.7 (CH \times 2), 118.6, 117.7, 48.8. IR ν_{max} (KBr): 3417, 1627, 1536, 1503, 1356, 1276, 815, 719 cm^{-1} . MS (ESI) m/z 392 ($[\text{M}-1]^+$, 25), 149 (100). Found: C, 66.96; H, 4.15; N, 3.53; S, 8.20. Calc. for $\text{C}_{22}\text{H}_{16}\text{ClNO}_2\text{S}$: C, 67.08; H, 4.09; N, 3.56; S, 8.14%.

N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)thiophene-2-carboxamide (**3o**): Yellow solid m.p. 196–197°C in 96% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.37 (1H, s, OH), 9.12 (1H, d, $J = 8.4$ Hz, NH), 8.12–7.97 (4H, m, ArH), 7.88–7.80 (3H, m, ArH), 7.71 (1H, d, $J = 8.0$ Hz, ArH), 7.60 (1H, t, $J = 8.0$ Hz, ArH), 7.48 (1H, t, $J = 8.0$ Hz, ArH), 7.37–7.26 (3H, m, ArH), 7.19–7.17 (1H, m, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 161.0, 153.5, 147.8, 144.4, 138.9 (CH \times 2), 133.3, 131.4, 130.0, 129.7, 129.0, 128.7, 128.4, 128.0, 126.9, 122.7, 122.6, 121.6, 120.9, 118.6, 117.0, 48.9. IR ν_{max} (KBr): 3384, 3239, 1626, 1530, 1346, 814, 716 cm^{-1} . MS (ESI) m/z 403 ($[\text{M}-1]^+$, 100), 143 (18). Found: C, 65.14; H, 4.06; N, 6.93; S, 8.03. Calc. for $\text{C}_{22}\text{H}_{16}\text{N}_2\text{O}_4\text{S}$: C, 65.33; H, 3.99; N, 6.93; S, 7.93%.

N-((2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methyl)acetamide (**3p**): Yellow solid m.p. 171–172°C in 89% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.95 (1H, s, OH), 8.40 (1H, d, $J = 8.8$ Hz, NH), 7.86–7.74 (3H, m, ArH), 7.36 (1H, t, $J = 7.6$ Hz, ArH), 7.27–7.19 (2H, m, ArH), 7.08–7.04 (3H, m, ArH), 6.82–6.79 (2H, m, ArH), 3.68 (3H, s, OCH_3), 1.96 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 169.1, 157.7, 153.0, 134.4, 132.3, 129.1 (CH \times 2), 128.5 (CH \times 2), 127.2 (CH \times 2), 126.2, 122.3, 119.0, 118.5, 113.4 (CH \times 2), 55.0, 47.4, 22.7. IR ν_{max} (KBr): 3396, 3056, 1628, 1513, 1438, 1255, 1177, 813, 745 cm^{-1} . MS (ESI) m/z 320 ($[\text{M}-1]^+$, 100), 143 (8). Found: C, 74.66; H, 6.12; N, 4.30. Calc. for $\text{C}_{20}\text{H}_{19}\text{NO}_3$: C, 74.75; H, 5.96; N, 4.36%.

N-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)acetamide (**3q**): White solid m.p. 230–231°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.98 (1H, s, OH), 8.44 (1H, d, $J = 8.4$ Hz, NH), 7.84–7.75 (3H, m, ArH), 7.36 (1H, t, $J = 7.6$ Hz, ArH), 7.27–7.20 (4H, m, ArH), 7.17–7.12 (4H, m, ArH), 1.98 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 169.2, 153.1, 142.6, 132.3, 129.2, 128.5 (CH \times 2), 128.4, 128.0 (CH \times 2), 126.3, 126.0, 123.3, 122.4 (CH \times 2), 118.9, 118.5, 47.8, 22.6. IR ν_{max} (KBr): 3401, 3252, 1641, 1515, 1338, 1278, 808, 743, 697 cm^{-1} . MS (ESI) m/z 290 ($[\text{M}-1]^+$, 60), 149 (15). Found: C, 78.21; H, 6.01; N, 4.78. Calc. for $\text{C}_{19}\text{H}_{17}\text{NO}_2$: C, 78.33; H, 5.88; N, 4.81%.

N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)acetamide (**3r**): Yellow solid m.p. 255–256°C in 93% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.13 (1H, s, OH), 8.63 (1H, d, $J = 8.0$ Hz, NH), 8.06–8.01 (2H, m, ArH), 7.88–7.80 (3H, m, ArH), 7.59–7.53 (2H, m, ArH), 7.42 (1H, t, $J = 7.6$ Hz, ArH), 7.30 (1H, t, $J = 7.6$ Hz, ArH), 7.23–7.17 (2H, m, ArH), 2.02 (3H, s, CH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 169.7, 153.3, 147.7, 145.4, 132.8, 132.1, 129.9, 129.5, 128.7, 128.4, 126.8, 122.8, 122.6, 121.2, 120.4, 118.4, 117.8, 47.6, 22.5. IR ν_{max} (KBr): 3375, 3222, 1649, 1525, 1350, 806, 734 cm^{-1} . MS (ESI) m/z 335 ($[\text{M}-1]^+$, 70), 143 (5). Found: C, 67.76; H, 4.87; N, 8.30. Calc. for $\text{C}_{19}\text{H}_{16}\text{N}_2\text{O}_4$: C, 67.85; H, 4.79; N, 8.33%.

N-((2-hydroxynaphthalen-1-yl)(4-methoxyphenyl)methyl)acrylamide (**3s**): White solid m.p. 222–223°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.00 (1H, s, OH), 8.68 (1H, d, $J = 8.4$ Hz, NH), 7.88–7.75 (3H, m, ArH), 7.39–7.06 (6H, m, ArH), 6.84–6.80 (2H, m, ArH), 6.57 (1H, dd, $J_1 = 16.8$ Hz, $J_2 = 10.4$ Hz, CH), 6.11 (1H, dd, $J_1 = 17.0$ Hz, $J_2 = 2.0$ Hz, CH₂), 5.60 (1H, dd, $J_1 = 10.2$ Hz, $J_2 = 2.4$ Hz, CH₂), 3.68 (3H, s, OCH_3). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 164.4, 157.8, 153.2, 134.0, 132.3, 131.9, 129.2 (CH \times 2), 128.6, 128.5, 127.4, 126.3, 125.6, 123.3, 122.4, 118.6, 118.5, 113.5 (CH \times 2), 55.0, 47.7. IR ν_{max} (KBr): 3398, 3071, 1657, 1512, 1439, 1337, 1247, 1174, 818, 654 cm^{-1} . MS (ESI) m/z 332 ($[\text{M}-1]^+$, 15), 293 (50), 195 (100), 143 (3). Found: C, 75.58; H, 5.83; N, 4.15. Calc. for $\text{C}_{21}\text{H}_{19}\text{NO}_3$: C, 75.66; H, 5.74; N, 4.20%.

N-((2-hydroxynaphthalen-1-yl)(phenyl)methyl)acrylamide (**3t**): White solid m.p. 255–256°C in 90% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.02 (1H, s, OH), 8.71 (1H, d, $J = 8.4$ Hz, NH), 7.87–7.76 (3H, m, ArH), 7.37 (1H, t, $J = 7.6$ Hz, ArH), 7.28–7.15 (8H, m, ArH), 6.60 (1H, dd, $J_1 = 17.2$ Hz, $J_2 = 10.0$ Hz, CH), 6.13 (1H, dd,

$J_1 = 17.2$ Hz, $J_2 = 2.0$ Hz, CH₂), 5.61 (1H, dd, $J_1 = 10.4$ Hz, $J_2 = 2.0$ Hz, CH₂). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 164.5, 153.2, 142.2, 132.3, 131.8, 129.3 (CH \times 2), 128.5, 128.4, 128.0, 126.3, 126.2, 126.1, 125.6, 123.2, 122.4 (CH \times 2), 118.5, 118.4, 48.1. IR ν_{max} (KBr): 3398, 3067, 1655, 1515, 1438, 1339, 1270, 1069, 744, 650 cm^{-1} . MS (ESI) m/z 302 ($[\text{M}-1]^+$, 70), 195 (100), 143 (10). Found: C, 79.10; H, 5.73; N, 4.59. Calc. for $\text{C}_{20}\text{H}_{17}\text{NO}_2$: C, 79.19; H, 5.65; N, 4.62%.

N-((2-hydroxynaphthalen-1-yl)(3-nitrophenyl)methyl)acrylamide (**3u**): Yellow solid m.p. 247–248°C in 94% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.20 (1H, s, OH), 8.93 (1H, d, $J = 8.0$ Hz, NH), 8.09–7.83 (5H, m, ArH), 7.58–7.55 (2H, m, ArH), 7.44 (1H, t, $J = 7.2$ Hz, ArH), 7.33–7.25 (3H, m, ArH), 6.67 (1H, dd, $J_1 = 17.0$ Hz, $J_2 = 10.0$ Hz, CH), 6.21 (1H, dd, $J_1 = 17.0$ Hz, $J_2 = 1.2$ Hz, CH₂), 5.68 (1H, dd, $J_1 = 10.4$ Hz, $J_2 = 1.2$ Hz, CH₂). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 164.9, 153.5, 147.7, 145.0, 132.9, 132.2, 131.4, 130.0, 129.7, 128.7, 128.4, 126.9, 126.3, 122.7 (CH \times 2), 121.4, 120.5, 118.5, 117.5, 47.8. IR ν_{max} (KBr): 3381, 3161, 1656, 1619, 1521, 1347, 1274, 1221, 981, 808 cm^{-1} . MS (ESI) m/z 347 ($[\text{M}-1]^+$, 45), 143 (5). Found: C, 68.88; H, 4.73; N, 7.98. Calc. for $\text{C}_{20}\text{H}_{16}\text{N}_2\text{O}_4$: C, 68.96; H, 4.63; N, 8.04%.

N-((furan-2-yl)(2-hydroxynaphthalen-1-yl)methyl)benzamide (**3v**): White solid m.p. 222–223°C in 80% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.27 (1H, s, OH), 9.10 (1H, d, $J = 8.0$ Hz, NH), 8.18 (1H, d, $J = 8.4$ Hz, ArH), 7.86–7.77 (4H, m, ArH), 7.55–7.44 (5H, m, ArH), 7.30 (2H, t, $J = 7.6$ Hz, ArH), 7.22 (1H, d, $J = 8.8$ Hz, ArH), 6.37–6.35 (1H, m, ArH), 6.12 (1H, d, $J = 3.2$ Hz, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 165.7, 154.1, 153.4, 142.1, 134.2, 132.3, 131.5, 129.5 (CH \times 2), 128.6 (CH \times 2), 128.4, 127.3, 126.5, 122.9, 122.6 (CH \times 2), 118.5, 116.4, 110.5, 106.8, 44.6. IR ν_{max} (KBr): 3410, 3147, 1632, 1401, 1345, 819, 746, 598 cm^{-1} . MS (ESI) m/z 343 (M^+ , 15), 222 (100), 181 (60), 105 (55). Found: C, 76.86; H, 5.05; N, 4.01. Calc. for $\text{C}_{22}\text{H}_{17}\text{NO}_3$: C, 76.95; H, 4.99; N, 4.08%.

N-((1-(2-hydroxynaphthalen-1-yl)propyl)benzamide (**3w**): White solid m.p. 244–245°C in 91% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.10 (1H, s, OH), 8.62 (1H, d, $J = 8.0$ Hz, NH), 8.23 (1H, d, $J = 8.8$ Hz, ArH), 7.82–7.70 (4H, m, ArH), 7.54–7.44 (4H, m, ArH), 7.30 (1H, t, $J = 7.2$ Hz, ArH), 7.20 (1H, d, $J = 8.8$ Hz, ArH), 5.92 (1H, q, $J = 7.6$ Hz, ArH), 2.18–1.95 (2H, m, CH₂), 0.93 (3H, t, CH₃). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 165.4, 152.9, 134.7, 132.2, 131.0, 128.4 (CH \times 2), 128.3, 128.2 (CH \times 2), 126.9, 126.2, 122.5 (CH \times 2), 122.3, 119.5, 118.6, 48.5, 26.9, 11.3. IR ν_{max} (KBr): 3406, 3180, 1635, 1533, 1344, 817, 708 cm^{-1} . MS (ESI) m/z 305 (M^+ , 25), 276 (86), 169 (100), 105 (90). Found: C, 78.58; H, 6.35; N, 4.51. Calc. for $\text{C}_{20}\text{H}_{19}\text{NO}_2$: C, 78.66; H, 6.27; N, 4.59%.

N,N'-(1,5-bis(2-hydroxynaphthalen-1-yl)pentane-1,5-diyl)dibenzamide (**4a**): White solid m.p. 251–252°C in 4% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 10.15 (2H, s, OH), 8.52 (2H, s), 8.18 (2H, s), 7.80–7.68 (8H, m, ArH), 7.53–7.41 (8H, m, ArH), 7.29 (2H, t, $J = 7.6$ Hz, ArH), 7.18 (2H, d, $J = 8.8$ Hz, ArH), 5.99 (2H, q, $J = 8.0$ Hz), 2.27–2.25 (2H, m, CH₂), 2.03–1.96 (2H, m, CH₂), 1.64 (1H, s, CH₂), 1.43 (1H, s, CH₂). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 165.5 (CH \times 2), 152.9 (CH \times 2), 134.8 (CH \times 2), 132.1 (CH \times 2), 131.1 (CH \times 2), 128.4 (CH \times 6), 126.9 (CH \times 6), 126.4 (CH \times 4), 122.4 (CH \times 4), 119.9 (CH \times 2), 118.7 (CH \times 2), 46.8 (CH \times 2), 33.4 (CH \times 2), 23.6. IR ν_{max} (KBr): 3414, 3279, 1647, 1509, 1400, 1140, 694, 617 cm^{-1} . MS (ESI) m/z 595 ($[\text{M} + 1]^+$, 100). Found: C, 78.65; H, 5.91; N, 4.68. Calc. for $\text{C}_{39}\text{H}_{34}\text{N}_2\text{O}_4$: C, 78.77; H, 5.76; N, 4.71%.

1,3-bis(2-hydroxynaphthalen-1-yl)(phenyl)methylurea (**5a**): White solid m.p. 168–169°C in 58% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.85 (2H, s, OH), 7.77–7.69 (7H, m, ArH), 7.44–7.05 (19H, m, ArH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 158.0, 152.8 (CH \times 2), 144.6 (CH \times 2), 132.2 (CH \times 2), 128.9 (CH \times 4), 128.5 (CH \times 6), 127.9 (CH \times 4), 126.2 (CH \times 4), 125.8 (CH \times 2), 122.2 (CH \times 2), 120.2 (CH \times 2), 118.4 (CH \times 2), 48.0 (CH \times 2). IR ν_{max} (KBr): 3415, 3395, 1631, 1531, 1273, 1068, 814, 745, 698 cm^{-1} . MS (ESI) m/z 525 ($[\text{M} + 1]^+$, 100). Found: C, 80.01; H, 5.51; N, 5.28. Calc. for $\text{C}_{35}\text{H}_{28}\text{N}_2\text{O}_4$: C, 80.13; H, 5.38; N, 5.34%.

N-((1-hydroxynaphthalen-2-yl)(3-nitrophenyl)methyl)benzamide (**6a**): Sorrel solid m.p. 83–84°C in 60% yield. ^1H NMR (400 MHz, $\text{DMSO}-d_6$): δ 9.79 (1H, s, OH), 9.38 (1H, d, $J = 9.2$ Hz, NH), 8.27–8.25 (1H, m, ArH), 8.17–8.12 (2H, m, ArH), 7.96–7.81 (4H, m, ArH), 7.67–7.47 (8H, m, ArH), 7.15 (1H, d, $J = 8.4$ Hz, CH). ^{13}C NMR (100 MHz, $\text{DMSO}-d_6$): 166.4, 149.5, 147.8, 144.9, 134.3 (CH \times 2), 133.7, 131.6, 129.9, 128.4 (CH \times 2), 127.7, 126.2 (CH \times 2), 125.8 (CH \times 2), 125.3, 122.6 (CH \times 2), 122.2, 121.9, 121.7, 119.9, 50.6. IR ν_{max} (KBr): 3421, 3126, 1664, 1530, 1400, 1349 cm^{-1} . MS (ESI) m/z 398 (M^+ , 5), 230 (85), 105 (100). Found: C, 72.23; H, 4.62; N, 6.99. Calc. for $\text{C}_{24}\text{H}_{18}\text{N}_2\text{O}_4$: C, 72.35; H, 4.55; N, 7.03%.

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