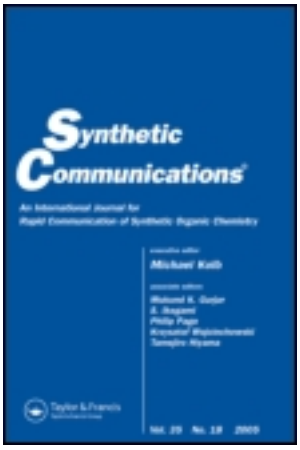

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P_2O_5-Hexamethyldisiloxane (HMDS): An Efficient System to Induce the Three-Component Reaction of Enolic Systems, Aromatic Aldehydes, and Acetonitrile

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P_2O_5-Hexamethyldisiloxane (HMDS): An Efficient System to Induce the Three-Component Reaction of Enolic Systems, Aromatic Aldehydes, and Acetonitrile

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Abstract: Three-component reaction of an enolizable compound, such as acetophenone, methyl acetoacetate, 4-hydroxycoumarin, 2-naphthol, or 3-hydroxy-2-naphthoic acid; an aromatic aldehyde, and acetonitrile induced by phosphorus pentoxide and hexamethyldisiloxane leads to 2-acetylamino ketones, methyl 3-(acetylamino aryl methyl)-3-oxobutanoates, 3-(acetylamino aryl methyl)-4-hydroxycoumarins, 1-(acetylamino aryl methyl)-2-naphthols, or 4-(acetylamino aryl methyl)-3-hydroxy-2-naphthoic acids in excellent yields.

Keywords: Aryl aldehydes, hexamethyldisiloxane, 4-hydroxycoumarin, 2-naphthol, phosphorus pentoxide, three-component reaction

Multicomponent reactions (MCRs) have received great attention from chemists because they can be widely employed for the rapid assembly of arrays with high molecular diversity.[1–5] The three-component reaction of enolic systems such as aryl methyl ketones or β-dicarbonyl compounds, aryl aldehydes, and acetonitrile has been recently used for the synthesis of β-acetylamino ketones or esters. This reaction is usually carried out in the presence of excess amounts of acetyl chloride as the oxygen

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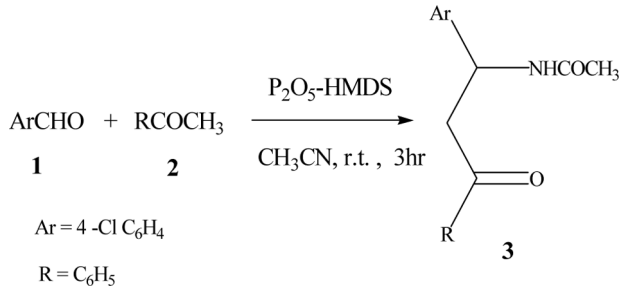
activator and catalyzed by a Brønsted or Lewis acid. A number of catalysts such as BiOCl,[6] $ZrOCl_2 \cdot 8H_2O$,[7] $CoCl_2$,[8] montmorillonite K-10 clay,[9] H_2SO_4/SiO_2,[10] heteropoly acids,[11] silica sulfuric acid,[12] $NaHSO_4$-SiO_2,[13] and $CsCl_3 \cdot 7H_2O$[14] have been employed to affect this transformation. The reaction of substituted acetophenones or β-ketoesters, aryl aldehydes, and acetonitrile has also been carried out in the presence of an excess amount of trimethylchlorosilane catalyzed by heteropoly acids.[15] Very recently, we reported that a similar reaction can be carried out with other enolic systems such as 4-hydroxycoumarin[16] or 2-naphthol,[17] aromatic aldehydes, and acetonitrile in the presence of chlorosulfonic acid.

The reaction between phosphorus pentoxide and hexamethyldisiloxane (HMDS) in organic solvents has been reported to produce a mixture of silylated three- and tetraphosphates, known as polyphosphoric acid trimethylsilylester (PPSE).[18] PPSE is a powerfull oxygen-activating and dehydrating agent that has been used to promote different organic processes such as dehydration of amides to nitriles,[19] pinacol rearrangements,[20] synthesis of hetrocycles,[21] aldol-like condensations,[22] and Friedel–Crafts[23] reactions. The employment of PPSE to promote the organic transformations has been recently reviewed.[24]

In continuation of our work[16,17] on the three-component reaction of enolic systems, aldehydes, and acetonitrile, here we report the results of our investigation on the reaction of acetophenone, methyl acetoacetate, 4-hydroxycoumarin, 2-naphthol, or 3-hydroxy-2-naphthoic acid; aromatic aldehydes, and acetonitrile induced by a mixture of phosphorus pentoxide and HMDS.

When a mixture of acetophenone and 4-chlorobenzaldehyde was stirred in acetonitrile in the presence of phosphorus pentoxide and HMDS, a smooth reaction took place and was completed within 3 h (monitored by thin-layer chromatography, TLC) (Scheme 1). After pouring the reaction mixture into ice water, 3-acetylamino-3-(4-chlorophenyl) propiophenone



Scheme 1. Three-component reaction of aromatic aldehydes, acetophenones, and acetonitrile promoted by P_2O_5/HMDS system.

Table 1. Optimization of the amounts of P_2O_5 and HMDS in the reaction of acetophenone, 4-chlorobenzaldehyde, and acetonitrile

Entry	P_2O_5 (eq)	HMDS (eq)	Yield (%)
1	—	1	—
2	1	—	—
3	1	1	95
4	0.5	0.5	68
5	2	2	95

was obtained as a pale-yellow solid, which was pure on the basis of the NMR data. As shown in Table 1, the best result was obtained by using 1 equivalent of phosphorus pentoxide and HMDS. No reaction was observed in the absence of phosphorus pentoxide or HMDS. Increasing the amount of phosphorus pentoxide or HMDS over 1 equivalent did not improve the yield considerably. To investigate the scope of the reaction, different acetophenones and aldehydes mixed together in acetonitrile in the presence of phosphorus pentoxide and HMDS. The results are shown in Table 2. As

Table 2. Synthesis of β-acetylamino ketones, and esters by the reaction of acetophenones, aromatic aldehydes or methyl acetoacetate, and acetonitrile in the presence of P_2O_5-HMDS system

Entry	Ar	R	Yield (%)[a]	Reference
1	C_6H_5	C_6H_5	95	15
2	3-$NO_2C_6H_4$	C_6H_5	97	15
3	4-$NO_2C_6H_4$	C_6H_5	88	13
4	4-ClC_6H_4	C_6H_5	95	15
5	2-ClC_6H_4	C_6H_5	92	13
6	4-$CH_3OC_6H_4$	C_6H_5	89	15
7	C_6H_5	4-$NO_2C_6H_4$	94	15
8	4-$CH_3C_6H_4$	4-$NO_2C_6H_4$	89	15
9	4-ClC_6H_4	4-$NO_2C_6H_4$	93	15
10	C_6H_5	4-MeC_6H_4	82	12
11	4-$NO_2C_6H_4$	4-MeC_6H_4	87	12
12	4-FC_6H_4	4-MeC_6H_4	85	12
13	C_6H_5	4-ClC_6H_4	82	12
14	4-ClC_6H_4	4-ClC_6H_4	89	15
15	4-FC_6H_4	4-ClC_6H_4	89	12
16	C_6H_5	$CH_3O_2CCH_2$	76	15
17	4-ClC_6H_4	$CH_3O_2CCH_2$	84	15

[a]Isolated yields.

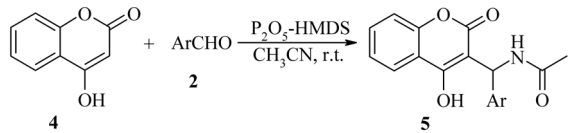
could be seen, the reaction is compatible with different aldehydes and acetophenones bearing electron-withdrowing and electron-releasing groups. The reaction was also carried out effectively with methyl acetoacetate instead of acetophenone (Table 2, entries 16 and 17).

The products reported in Table 2 are all known compounds, and their structures were deduced by comparison of melting points and spectral data with authentic samples. To develop the explained method, we also conducted the reaction with other enolic systems such as 4-hydroxycoumarin, 2-naphthol, or 3-hydroxy-2-naphthoic acid; aryl aldehydes; and acetonitrile in the presence of phosphorus pentoxide and HMDS. Thus, the reaction of 4-hydroxycoumarin, aromatic aldehydes, and acetonitrile in the presence of 1 equivalent of phosphorus pentoxide and HMDS afforded the corresponding 3-(acetylamino aryl methyl)-4-hydroxycoumarins in good yields (Scheme 2).

Similar products were obtained when 2-naphthol or 3-hydroxy-2-naphthoic acid was used instead of 4-hydroxycoumarin. The results obtained for these reactions are summarized in Table 3. As shown, good yields were obtained using aldehydes bearing electron-withdrawing and electron-releasing groups.

A reasonable mechanism for the formation of compounds **7** is presented in Scheme 3. Acetonitrile attacks the condensation product of naphthol derivative and aldehyde in the presence of PPSE to afford the diionic intermadiate **9**, which then hydrolyzes to product **7**.

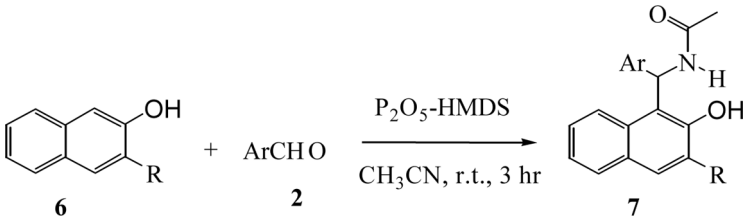
The ^{1}H NMR spectrum of compound **7a** exhibited a sharp line at $\delta = 1.9$ ppm for the protons of the methyl group. The methine and NH protons couple each other, and two doublets were observed for them at 7.1 and 8.7 ppm, respectively. When the ^{1}H NMR spectrum was recorded after addition of some D_2O to the d_6-DMSO solution of **7a**, the doublet



5	a	b	c	d	e	f	g	h	i
Ar	Ph	4-ClPh	4-BrPh	2-ClPh	3-NO_2Ph	4-NO_2Ph	3-CH_3OPh	2-CH_3OPh	2-HOPh
Yield (%)	95	95	90	90	95	95	90	97	90

Scheme 2. Three-component reaction of aromatic aldehydes, 4-hydroxycoumarin, and acetonitrile promoted by P_2O_5/HMDS system. Yields refer to the pure isolated products. All known products have been reported previously in the literature and were characterized by comparison of melting points and spectral data witha authentic samples.[16]

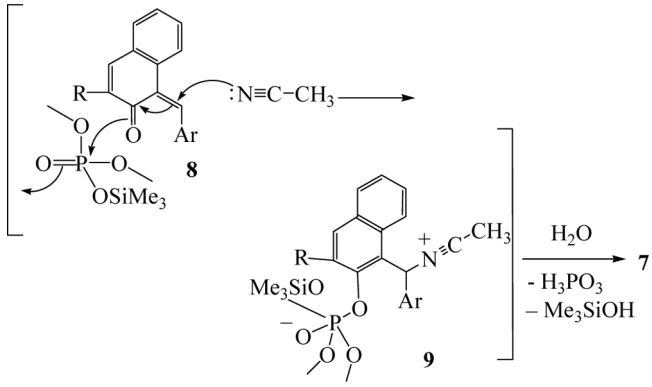
Table 3. Reaction between 2-naphthol or 3-hydroxy-2-naphthoic acid and aromatic aldehydes in acetonitrile in the presence of P_2O_5 and HMDS



7	R	Ar	Yield (%)[a]
a	CO_2H	4-ClC_6H_4	93
b	CO_2H	3-ClC_6H_4	95
c	CO_2H	2-ClC_6H_4	97
d	CO_2H	4-$CH_3C_6H_4$	90
e	CO_2H	4-$NO_2C_6H_4$	90
f	CO_2H	3-$NO_2C_6H_4$	95
g	CO_2H	4-BrC_6H_4	95
h	CO_2H	3-$CH_3OC_6H_4$	90
i	CO_2H	2-$CH_3OC_6H_4$	96
j	H	Ph	93
k	H	4-ClC_6H_4	95
l	H	2-$CH_3C_6H_4$	95
m	H	2-ClC_6H_4	95
n	H	3-$NO_2C_6H_4$	94
o	H	4-FC_6H_4	91
p	H	4-$CH_3C_6H_4$	93
q	H	4-BrC_6H_4	95
r	H	3-$CH_3OC_6H_4$	95
s	H	2-$CH_3OC_6H_4$	92

[a]Yields refer to the pure isolated products. Compounds **7a–i** were new, and their structures were deducted by elemental and spectral analysis. Compounds **7j–s** were known compounds, and their structures were deduced by comparing their melting points and spectral data with authentic samples.[17]

related to the NH proton disappeared and the doublet related to the methine proton converted to a singlet. The protons of two hydroxy groups resonated in the range of 12.00–13.00 ppm as a very broad signal, which in some cases was so broadened that was not observed. The ^{13}C NMR spectrum of compound **7a** showed 18 distinct signals, consistent with the proposed structure.



Scheme 3. Suggested mechanism for formation of compounds **7**.

In summary, here we report a simple and efficient one-pot synthesis of 2-acetylamino ketones, methyl 3-(acetylamino aryl methyl)-3-oxobutanoates, 3-(acetylamino aryl methyl)-4-hydroxycoumarins, 1-(acetylamino aryl methyl)-2-naphthols, or 4-(acetylamino aryl methyl)-3-hydroxy-2-naphthoic acids by three-component reaction of an enolizable compound, such as acetophenone, methyl acetoacetate, 4-hydroxycoumarin, 2-naphthol, or 3-hydroxy-2-naphthoic acid; an aromatic aldehyde; and acetonitrile induced by phosphorus pentoxide and hexamethyldisiloxane. The advantages of this method are simple, available starting materials, short reaction times, easy and clean workup, and excellent yields.

EXPERIMENTAL

Melting points were determined with an electrothermal 9100 apparatus. Elemental analyses were performed using a Heraeus CHN-O-Rapid analyzer. Mass spectra were recorded on a Finnigan-MAT 8430 mass spectrometer operating at an ionization potential of 70 eV. IR spectra were recorded on a Shimadzu IR-470 spectrometer. ^{1}H and ^{13}C NMR spectra were recorded on Bruker DRX-500 Avance spectrometer at solution in d_6-DMSO using TMS as internal standard. The chemicals used in this work were purchased from Fluka (Buchs, Switzerland) and were used without further purification.

General Procedure for the Preparation of Compounds 3, 5, and 7

To a magnetically stirred solution of acetophenone (3 mmol) and aldehyde (3 mmol) in 15 mL acetonitrile, phosphorus pentoxide (3 mmol)

and HMDS (3 mmol) were added at room temperature. The reaction mixture was then stirred for 3 h at room temperature. The reaction mixture was poured into 50 mL of ice water. The solid product was filtered, washed with ice water, and recrystallized from ethyl acetate/n-hexane to give the pure product.

Data

4-[Acetylamino(4-chlorophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7a**)

Yellow powder, mp 262–264 °C, IR (KBr) (ν_{max} cm^{-1}): 3350, 3125–2605, 1725, 1669. Anal. calcd. for $C_{20}H_{16}ClNO_4$: C, 64.96; H, 4.36; N, 3.79%. Found: C, 65.00; H, 4.28; N, 3.85. MS (m/z, %): 369 (11). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.98 (3 H, s, C*H*$_3$), 7.11 (1 H, d, $^3J_{HH} = 8$ H$_Z$, NCH), 7.25–8.62 (9 H, m, aromatic), 8.76 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.74 (2 H, broad s, 2 OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.97 (*C* H$_3$), 47.48 (*C* H), 120.58, 124.08, 128.32, 128.50, 129.17, 129.35, 130.99, 131.29, 133.50 and 153.06 (naphthol moiety), 114.82, 127.41, 135.27 and 141.52 (phenyl moiety), 169.98, 172.71 (2C=O).

4-[Acetylamino(3-chlorophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7b**)

Yellow powder, mp 258–260 °C, 95%; IR (KBr) (ν_{max} cm^{-1}): 3385, 3155–2615, 1708, 1668. Anal. calcd. for $C_{20}H_{16}ClNO_4$: C, 64.96; H, 4.36; N, 3.79%. Found: C, 65.00; H, 4.28; N, 3.85. MS (m/z, %): 369 (8). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.99 (3 H, s, C*H*$_3$), 7.01 (1 H, d, $^3J_{HH} = 8$ H$_Z$, NCH), 7.27–8.63 (9 H, m, aromatic), 8.71 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.73 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.95 (*C*H$_3$), 47.60 (*C* H), 120.41, 123.51, 124.13, 125.23, 126.11, 126.78, 130.10, 130.49, 133.59 and 154.92 (naphthol moiety), 114.87, 127.39, 131.01, 133.42, 135.26 and 145.16 (phenyl moiety), 170.07, 172.70 (2C=O).

4-[Acetylamino(2-chlorophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7c**)

Yellow powder, mp 246–248 °C, 95%; IR (KBr) (ν_{max} cm^{-1}): 3390, 3130–2580, 1729, 1667. Anal. calcd. for $C_{20}H_{16}ClNO_4$: C, 64.96; H, 4.36; N, 3.79%. Found: C, 65.00; H, 4.28; N, 3.85. MS (m/z, %): 369 (10). ^{1}H

NMR (500 MHz, d_6-DMSO): δ 1.94 (3 H, s, C*H*$_3$), 7.15 (1 H, d, $^3J_{HH} = 8$ Hz, NCH), 7.23–8.62 (9 H, m, aromatic), 8.74 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.63 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.76 (*C*H$_3$), 47.76 (*C*H), 119.34, 123.35, 123.93, 126.95, 128.84, 129.75, 129.86, 130.21, 132.66 and 155.38 (naphthol moiety), 114.65, 127.25, 131.01, 133.55, 135.81 and 139.57 (phenyl moiety), 169.29, 172.84 (2C=O).

4-[Acetylamino(4-methylphenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7d**)

White powder, mp 228–230 °C, IR (KBr) (ν_{max} cm^{-1}): 3350, 3182–2630, 1721, 1669. Anal. calcd. for $C_{21}H_{19}NO_4$: C, 72.19; H, 5.48; N, 4.01%. Found: C, 72.34; H, 5.40; N, 4.05. MS (m/z, %): 349 (10). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.97 (3 H, s, CH$_3$), 2.22 (3 H, s, C*H*$_3$), 6.95 (1H, d, $^3J_{HH} = 8$ Hz, NCH), 7.21–8.61 (9 H, m, aromatic), 8.75 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.73 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 20.99 and 23.03 (2 *C* H$_3$), 47.71 (*C* H), 121.24, 123.96, 126.41, 127.12, 129.13, 129.78, 130.90, 133.21, 135.41 and 154.80 (naphthol moiety), 114.75, 127.39, 135.73 and 139.32 (phenyl moiety), 169.75, 172.78 (2C=O).

4-[Acetylamino(4-nitrophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7e**)

Yellow powder, mp 253–255 °C, 90%; IR (KBr) (ν_{max} cm^{-1}): 3380, 3134–2580, 1729, 1671. Anal. calcd. for $C_{20}H_{16}N_2O_6$: C, 63.16; H, 4.24; N, 7.36%. Found: C, 63.20; H, 4.30; N, 7.40. MS (m/z, %): 380 (8). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.96 (3 H, s, C*H*$_3$), 7.12 (1 H, d, $^3J_{HH} = 8$ Hz, NCH), 7.25–8.65 (9 H, m, aromatic), 8.72 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.76 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.88 (*C*H$_3$), 47.96 (*C*H), 120.05, 123.38, 123.77, 124.20, 127.43, 130.25, 131.05, 133.83, 135.23 and 155.00 (naphthol moiety), 114.92, 127.56, 146.47 and 150.85 (phenyl moiety), 170.28, 172.66 (2C=O).

4-[Acetylamino(3-nitrophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7f**)

Yellow powder, mp 250–252 °C, 94%; IR (KBr) (ν_{max} cm^{-1}): 3385, 3136–2527, 1723, 1672. Anal. calcd. for $C_{20}H_{16}N_2O_6$: C, 63.16; H, 4.24; N,

7.36%. Found: C, 63.20; H, 4.30; N, 7.40. MS (m/z, %): 380 (7). ^{1}H NMR (500 MHz, d_6-DMSO): δ 2.01 (3 H, s, CH_3), 7.24–8.65 (10 H, m, aromatic and NCH), 8.77 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.74 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.92 (*C*H$_3$), 47.70 (*C*H), 119.10, 120.81, 121.92, 123.32, 124.25, 130.17, 130.33, 133.85, 135.20 and 155.38 (naphthol moiety), 114.91, 127.25, 131.07, 133.25, 145.08 and 148.22 (phenyl moiety), 170.28, 172.65 (2C=O).

4-[Acetylamino(4-bromophenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7g**)

Yellow powder, mp 235–237 °C, 90%; IR (KBr) (ν_{max} cm^{-1}): 3385, 3150–2590, 1740, 1663. Anal. calcd. for $C_{20}H_{16}BrNO_4$: C, 57.99; H, 3.89; N, 3.38%. Found: C, 58.06; H, 3.95; N, 3.40. MS (m/z, %): 414 (9). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.99 (3 H, s, CH_3), 7.01 (1 H, d, $^3J_{HH} = 8$ H$_Z$, NCH), 7.31–8.61 (9 H, m, aromatic), 8.72 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.70 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.97 (*C*H$_3$), 47.55 (*C*H), 119.77, 120.53, 124.27, 126.40, 128.70, 129.49, 129.70, 131.40, 132.99 and 156.50 (naphthol moiety), 115.67, 127.10, 137.68 and 141.97 (phenyl moiety), 170.03, 172.05 (2C=O).

4-[Acetylamino(3-methoxyphenyl)methyl]-3-hydroxy-2-naphthoic Acid (**7h**)

Yellow powder, mp 224–226 °C, IR (KBr) (ν_{max} cm^{-1}): 3385, 3118–2521, 1732, 1664. Anal. calcd. for $C_{21}H_{19}NO_5$: C, 69.03; H, 5.24; N, 3.83%. Found: C, 69.10; H, 5.30; N, 3.80. MS (m/z, %): 365 (11). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.98 (3 H, s, CH_3), 3.66 (3 H, s, OCH_3), 6.70 (1 H, d, $^3J_{HH} = 8$ Hz, NCH), 7.11–8.61 (9 H, m, aromatic), 8.70 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.75 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.99 (*C*H$_3$), 47.81 (*C*H), 55.35 (OCH_3), 118.86, 119.75, 121.05, 124.00, 127.37, 129.69, 130.92, 133.29, 135.40, and 154.80 (naphthol moiety), 111.33, 112.95, 114.75, 129.88, 144.06 and 159.64 (phenyl moiety), 169.81, 172.75 (2C=O).

4-[Acetylamino(2-methoxyphenyl)methyl]-3-hydroxy-2-naphthoic acid (**7i**)

Yellow powder, mp 244–246 °C, IR (KBr) (ν_{max} cm^{-1}): 3395, 3130–2570, 1703, 1669. Anal. calcd. for $C_{21}H_{19}NO_5$: C, 69.03; H, 5.24; N, 3.83%.

Found: C, 69.10; H, 5.30; N, 3.80. MS (m/z, %): 365 (9). ^{1}H NMR (500 MHz, d_6-DMSO): δ 1.93 (3 H, s, CH_3), 3.58 (3 H, s, OCH_3), 7.12–8.61 (10 H, m, aromatic and NCH), 8.74 (1 H, d, $^3J_{HH} = 8$ Hz, N*H*), 11.64 (2 H, broad s, 2OH). ^{13}C NMR (125.8 MHz, d_6-DMSO): δ 22.76 (*C*H_3), 47.75 (*C*H), 53.75 (OCH_3), 119.33, 123.34, 123.93, 126.95, 128.84, 129.75, 129.87, 130.21, 132.65 and 155.37 (naphthol moiety), 114.64, 127.24, 131.01, 133.55, 135.80 and 139.57 (phenyl moiety), 169.27, 172.83 (2C=O).

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