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New GO/Mn/Co Nanocatalysts: An effective catalyst for the preparation of 14-aryl-14*H*-dibenzo[a,j]xanthenes derivatives and amido alkyl naphthols under solvent-free conditions

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Abstract: In this study, nanocatalysts of GO/ZnO/Mn/Co for the reaction of xanthene and amido alkyl naphthol were studied. First, the optimal conditions for these two reactions were obtained and then the effect of zinc oxide nanoparticles on this reaction was studied. To do this, study the graphene oxide catalyst with the presence and absence of ZnO and then different conditions such as the amount of catalysis, the reaction temperature, the reaction time, and finally the appropriate solvent to perform both reactions optimally. Finally, the conditions of these reactions for the synthesized of derivatives under thermal and ultrasonic conditions are studied and the results show that this catalyst has a good ability to perform these two reactions because with a small amount of catalyst excellent yield is obtained with high efficiency and also recyclability several times for xanthene and amido alkyl naphthol derivatives. The products were characterized by FT-IR, NMR spectra, and melting point.

Keywords: Graphene oxide, Nano-catalyst, Xanthene, Amido alkyl naphthol, Ultrasound condition.

Introduction

Due to the importance of heterocyclic compounds, which have more medicinal properties, they have wide applications in the treatment of patients with various diseases [1-4]. This makes researchers more encouraged to synthesize these compounds in different forms. Among these compounds, xanthene and its derivatives are very important due to their wide range of applications, including anti-inflammatory activities, antibacterial activities, photodynamic therapy, antiviral effects, antifungal activities, and enzyme inhibition [5-7]. The amido alkyl naphthol derivatives are one of the useful methods for synthesized drug compounds whose derivatives, like 2-amino benzothiazole like 2′-aminobenzothiazolo methyl naphthols, have many pharmaceuticals active, and another application of these compounds is their anti-inflammatory, anti-epileptic, antimicrobial, antitumor, antidiabetic and anticonvulsant agents [8-9]. It also has a wide range of pharmaceutical applications such as antibiotic, antitumor, analgesic, antianginal, antipsychotic, antimalarial, antirheumatic, and antihypertensive features because amido alkyl naphthol derivatives are easily converted to auxin 1,3-oxazine [10-12]. Although several catalytic methods have been reported for the manufacture of amido alkyl naphthol and xanthene, most of them are associated with defects in synthetic chemistry such as harsh reaction conditions, low efficiency, increased reaction temperature, long time, and finally hard purification. Therefore, the introduction of methods that minimize these problems has always been considered by researchers [13-18]. The application of nano-catalysts has many advantages such as

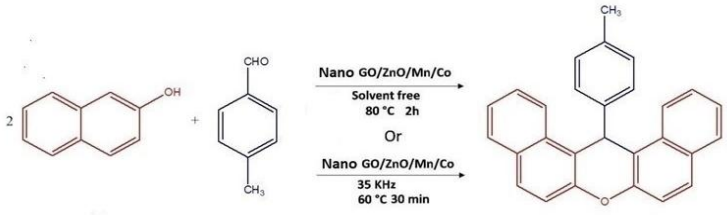
losing the amount of catalyst used, improving reaction conditions high yields of products, and environmental affairs. They are the best choice in the lab and industry because of their advantages in use. On the other hand, environmental pollution control is an important reason for using these catalysts. In recent years, graphene oxide (GO) nanosheets, which can be easily prepared from graphite powder using modified Hummer's method [19], have attracted great interest as promising low-cost ideal support for a number of nanoparticles, metals, and organic compounds due to their extremely high specific surface area, high thermal conductivity, high hydrophilic nature, good accessibility adsorption of organic dye high thermal and chemical stability [20-21]. In addition to their use as effective heterogeneous catalysts in various chemical transformations functionalized GO based materials have a wide range of applications in domains such as solar cells [22 23] Multicomponent reactions MCRs are convergent reactions in which three or more starting components combine to produce a product in which all or most atoms contribute to the newly produced product In MCR the product is constructed by a cascade of initial chemical reactions and is frequently used to synthesizes organic molecules [24] Natural hydroxyapatite supported ZnCl2 produced from bovine bone as a sustainable high efficiency heterogeneous biocatalyst for the synthesis of amido alkyl naphthol's[25] A novel magnetic phosphonium ionic liquid facilitated the synthesis of 2' aminobenzothiazolo methyl naphthols and amido alkyl naphthol derivatives providing new and promising insight into the rational design synthesis and applications of task specific multi rule magnetic ionic liquids for a variety of applications[26] . Recently, a new nano-catalyst based on graphene oxide sheets has been developed for the synthesis of benzimidazoles from aldehydes and phenylene diamine using nano Co/Mn supported on GO [27]. Now in this research, using graphene oxide and zinc oxide, methods for synthesis of Xanthene and Amido alkyl naphthol derivatives have been presented, which study different reaction conditions, including reaction temperature, reaction time, solvent conditions, and finally comparing thermal and ultrasonic conditions has been studied.

Results and discussion

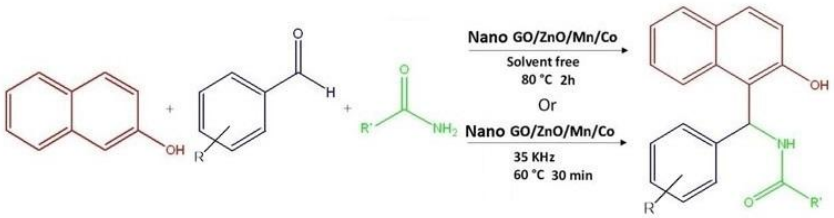
After the nanocatalyst was developed and characterized by various research, it has been reported in previous research. Reaction conditions such as temperature, reaction time, and suitable solvent for reaction with GO/ZnO/Mn/Co and GO/Mn/Co were studied for two well-known chemical reactions such as the synthesis of xanthene and amido alkyl naphthol derivatives.

Optimization of reaction condition

2-Naphthol (1 mmol) and 4-methyl benzaldehyde (2 mmol) as shown in scheme1 and 2-naphthol (1 mmol), 4-chlorobenzaldehyde (1 mmol), urea (1 mmol) was selected as a model for optimization as shown in scheme2. First of all, the reactions were carried out in different solvents and in the absence of them at different temperatures for 2 h in the presence of GO/Mn/Co and GO/ZnO/Mn/Co.



Scheme 1: Synthesis of xanthene derivatives under solvent-free and ultrasonic method.



Scheme 2: Synthesis of amido alkyl naphthol with urea under solvent-free and ultrasonic method.

As shown in Table **1** and Table **3**, the best condition is solvent-free at 80°C in the presence of GO/ZnO/Mn/Co. Furthermore, the different amounts of GO/ZnO/Mn/Co as catalyst were used (Table **1** and Table **3**, Entry f-h) and illustrated the suitable amount of catalyst is 0.1 g. When the reactions were carried out at different times from ambient temperature to 120 ℃ the yield of the product increased to 80 ℃ followed by it falling steadily (Table **1** and Table **3**, Entry b-e). It seems the best time with excellent yield is about 2h for xanthene derivatives and amido alkyl naphthol derivatives in solvent-free conditions. In addition, to investigate the catalyst design GO/ZnO/Mn/Co, 2-naphthol (1 mmol) and 4-methyl benzaldehyde (2 mmol) and 2-naphthol (1 mmol), 4-chlorobenzaldehyde (1 mmol), urea (1 mmol) was selected as a model for optimization under ultrasonic conditions, which are the best conditions for this reaction at a 35 KHz at 60 ℃ for 30 min (Table 1 and Table 3, entry i). Finally, the reaction of synthesis of different derivatives of xanthene Table 3 and also amido alkyl naphthol derivatives in the optimized conditions is reported in Table **4-9**.

The method using the present GO/ZnO/Mn/Co catalyst was also compared with other inorganic and organic catalysts reported in the literature for the synthesis of xanthene and amido alkyl naphthol derivatives (Table **10, 11**), clearly showing that the present nano cat GO/ZnO/Mn/Co was more efficient compared with reported procedures.

Catalyst recovery

One of the parameters that are very important in estimating the efficiency of the catalyst is the amount of recycling and reuse of them, so in order to study the ability of the GO/ZnO/Mn/Co catalyst, reaction 2-naphthol (1 mmol) and 4-methyl benzaldehyde (2 mmol) was used as a sample in optimal conditions. Recyclable change with very low reaction efficiency, which can be due to saturation of the active surface or contamination of the active points of the catalyst, and to recycle the catalyst, a solvent of water and ethanol at 110 °C has been used for two hours.; M.p. 262–264 °C (lit. 264–265 °C); 14-(4-Methoxyphenyl)-14H-dibenzo[a,j]xanthene. 202-204 ˚C; FT-IR (KBr, cm^{-1}): 3042 (aromatic CH), 1625 (C,C), 1594, 1512 (C,C), 1251 (C–O), 1236 (C–O), 811; ^{1}HNMR ($CDCl_3$, 300 MHz) δ (ppm): 3.62 (3H, s, OCH_3), 6.45 (1H, s, CH), 6.67–6.69 (2H, d, J = 7.9 Hz, Ar-H), 7.27–7.40 (4H, m), 7.42–7.44 (4H, m), 7.78–7.80 (2H, d, J = 8.0 Hz, Ar-H), 8.82–8.84 (2H, d, J = 8.0 Hz, Ar-H), 8.38–7.40 (2H, d, J = 7.9 Hz, Ar-H). ^{13}C NMR ($CDCl_3$, 50 MHz) δ (ppm): 37.1, 55.1, 113.8, 117.5, 118.0, 122.7, 124.7, 126.7, 128.7, 128.8, 129.2, 131.1, 131.4, 137.4, 148.6, and 157.8.

The reactions were performed with 2-naphthol (1 mmol), 4-chlorobenzaldehyde (1 mmol), urea (1 mmol), and catalyst (0.1 g), a Reflux condition, b–h Under solvent-free conditions. bAt room temperature. c At 50 °C.d At 80 °C. e At 120 °C. f At 80 °C/0.025 g catalyst. g At 80 °C/ 0.05 g catalyst. hAt 80 °C/0.1 g catalyst, the reaction was completed after 2 h, i At 80 °C/0.1 g GO /Mn/Co catalyst.

Table 1: Optimization of reaction in different conditions

Entry	Solvent,[a]	Time, h	Yield, %
1	CH_2Cl_2	2	7
2	$CHCl_3$	2	10
3	CH_3OH	2	15
4	CH_3CH_2OH	2	25
5	H_2O	2	Trace
6	THF	2	Trace
7	$(CH_3CH_2)_2O$	2	Trace
8	Toluene	2	Trace
9	None [b]	2	70
10	None [c]	2	80
11	None [d]	2	95
12	None [e]	2	95
13	None [f]	2	80
14	None [g]	2	85
15	None [h]	2	95
16	None [i]	2	70

Table 2. synthesis of 14-aryl-14H-dibenzo[a,j]xanthene using GO/Mn/Co Nanocatalyst

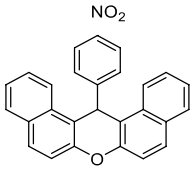
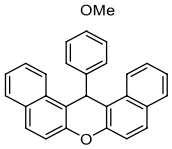
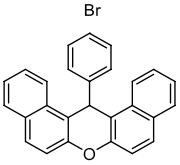
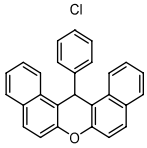
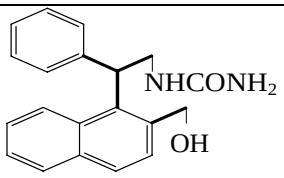
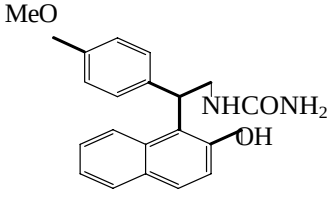
Entry	Aldehyde	Product	Time/min	Yield (%)
1	4-$NO_2C_6H_4$		15	98
2	4-$OMeC_6H_4$		15	96
3	4- BrC_6H_4		15	92
4	4- ClC_6H_4		15	90

Table 3: Optimization of reaction in different conditions

Entry	Solvent,[a]	Time, h	Yield, %
1	CH_2Cl_2	2	7
2	$CHCl_3$	2	10
3	CH_3OH	2	15
4	CH_3CH_2OH	2	25
5	H_2O	2	Trace
6	THF	2	Trace
7	$(CH_3CH_2)_2O$	2	Trace
8	Toluene	2	Trace
9	None [b]	2	70
10	None [c]	2	80
11	None [d]	2	95
12	None [e]	2	95
13	None [f]	2	80
14	None [g]	2	85
15	None [h]	2	95
16	None [i]	2	70

The reactions were performed with 2-naphthol (1 mmol), 4-chlorobenzaldehyde (1 mmol), urea (1 mmol), and catalyst (0.1 g), a Reflux condition, b–h Under solvent-free conditions. bAt room temperature. c At 50 °C.d At 80 °C. e At 120 °C. f At 80 °C/0.025 g catalyst. g At 80 °C/ 0.05 g catalyst. hAt 80 °C/0.1 g catalyst, the reaction was completed after 2 h, i At 80 °C/0.1 g GO /Mn/Co catalyst.

Table 4: Synthesis of [α-(2-Hydroxynaphthalen-1-yl) -(4-chlorobenzyl) methyl] urea using GO/Mn/Co Nano catalyst/ZnO under solvent-free conditions at 80-100 ˚C

Entry	Aldehyde	Product	Time/h	Yield (%)
1	C_6H_5		4	96
2	4-$OMeC_6H_4$		4	86

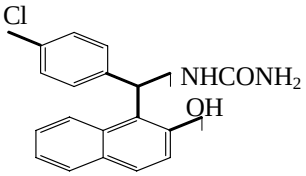
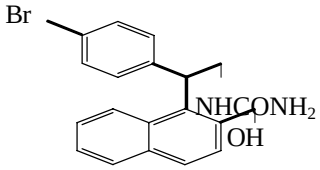
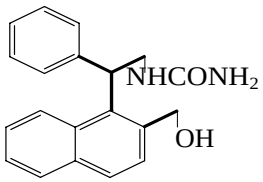
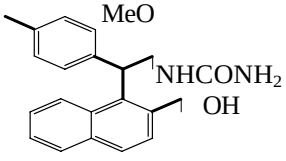
3	4-ClC_6H_4		4	88
4	4-BrC_6H_4		4	89

Table 5: Synthesis of [α-(2-Hydroxynaphthalen-1yl) (4chlorobenzyl) methyl] urea in the absence of GO/Mn/Co Nanocatalyst under solvent-free conditions at 80-100 ˚C

Entry	**Aldehyde**	**Product**	**Time/h**	**Yield (%)**
1	C_6H_5		2	96
2	4-$OMeC_6H_4$		4	86

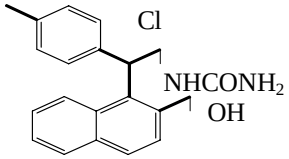
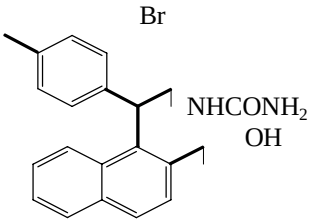
3	4-ClC_6H_4		4	88
4	4-BrC_6H_4		4	89

Table 6: Synthesis of [α-(2-Hydroxynaphthalen-1-yl) -(4-chlorobenzyl) methyl] urea using GO/Mn/Co Nanocatalyst with the ultrasonic method in water solvent at 60 ˚C

Entry	**Aldehyde**	**Product**	**Time (min)**	**Yield (%)**
1	C_6H_5		20	90

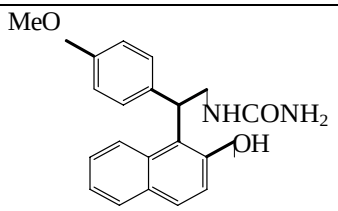
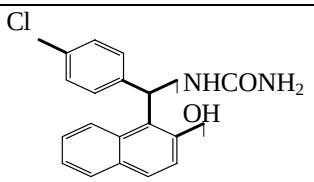
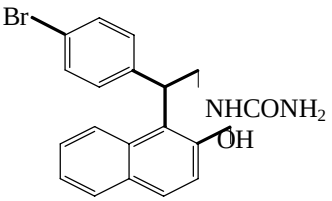
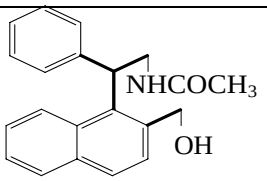
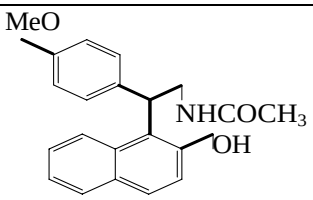
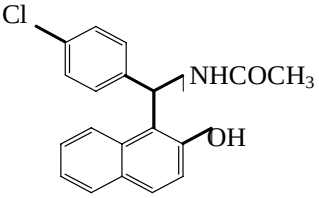
2	4-$OMeC_6H_4$		25	86
3	4-ClC_6H_4		18	88
4	4-BrC_6H_4		18	89

Table 7: Synthesis of [α-(2-Hydroxynaphthalen-1-yl) -(4-chlorobenzyl) methyl] acetamide using GO/Mn/Co Nano catalyst /ZnO under solvent-free conditions at 80-100 ˚C

Entry	**Aldehyde**	**Product**	**Time/h**	**Yield (%)**
1	C_6H_5		2	95
2	4-$OMeC_6H_4$		4	94
3	4-ClC_6H_4		4	94

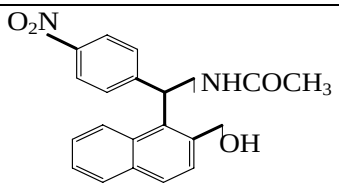
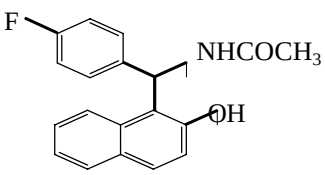
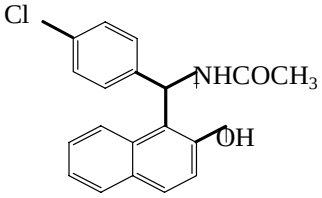
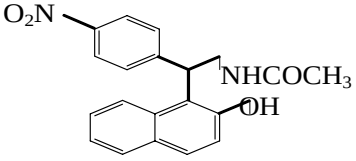
4	4-$NO_2C_6H_4$		4	94
5	4-FC_6H_4		4	88

Table 8: Synthesis of [α-(2-Hydroxynaphthalen-1-yl) -(4-chlorobenzyl) methyl] acetamide using GO/Mn/Co Nano catalyst /without ZnO under solvent-free conditions at 80-100 ˚C

Entry	**Aldehyde**	**Product**	**Time (min)**	**Yield (%)**
1	C_6H_5		2	95
2	4-$OMeC_6H_4$		4	94
3	4-ClC_6H_4		4	94
4	4-$NO_2C_6H_4$		4	94

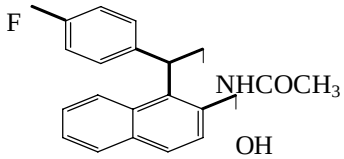
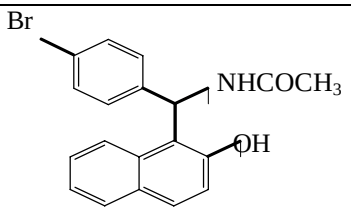
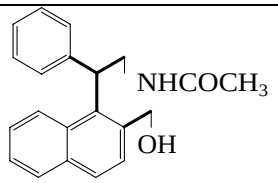
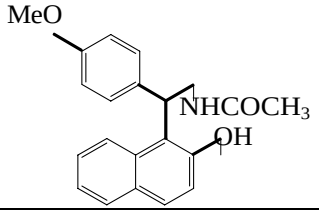
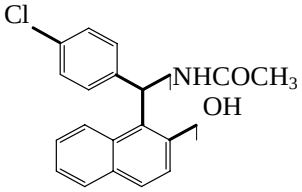
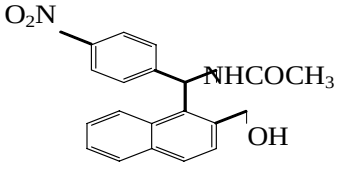
5	4-FC_6H_4		4	88
6	4-BrC_6H_4		4	90

Table 9: Synthesis of [α-(2-Hydroxynaphthalen-1-yl) -(4-chlorobenzyl) methyl] acetamide using GO/Mn/Co Nanocatalyst with the ultrasonic method in water solvent at 60 ˚C

Entry	**Aldehyde**	**Product**	**Time (min)**	**Yield (%)**
1	C_6H_5		20	95
2	4-$OMeC_6H_4$		22	94
3	4-ClC_6H_4		22	94
4	4-$NO_2C_6H_4$		22	94

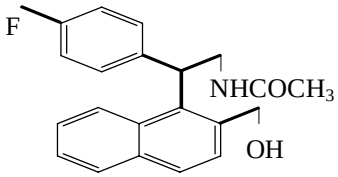
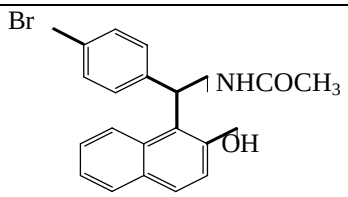
5	4-FC_6H_4		24	88
6	4-BrC_6H_4		22	90

Table 10. Comparison of synthesis of xanthene using nano cat GO/ZnO/Mn/Co with other catalysts reported in the literature

Entry	Catalyst(g)	Conditions	Time	Yield(%)	References
1	$ZrCl_4$@Arabic Gum (0.01)	Ethanol,	60 °C,1 h	50	[24]
2	$ZrCl_4$@Arabic Gum (0.01)	Solvent-free,	25 °C,20 min	92	[24]
3	$ZrCl_4$@Arabic Gum (0.02)	Solvent-free,	25 °C,15 min	92	[24]
4	Ag Nanoparticles	Solvent-free,	25 °C,5 min	95	[30]
5	$BF_3.SiO_2$ (0.10)	Solvent-free,	60 °C,15 min	94	[31]
6	SiO2-Cl Solvent- free, ultrasound		28–30 Ca	95	[28]
7	GO/ZnO/Mn/Co (0.10)	Solvent-free, 80 °C,2 h		95	Present work

Table 11 Comparison of synthesis of amido alkyl naphthol using nano cat GO/ZnO/Mn/Co with other catalysts reported in the literature

Entry	Catalyst(g)	Conditions	Yield (%)	References
1	Ce (SO4)2	CH3CN reflux, 36 h	66-78	[32]
2	Iodine	Solvent free, 125 °C, 5 h	71-84	[33]
3	Montmorillonite K10	Solvent free, 125 °C, 1.5 h	70-78	[34]
4	MgSO4	Solvent free, 100 °C, 1 h	72-90	[35]
5	ZnO NPs	Solvent free, (a) 130 °C, 30 min (b) 6 min	81-94	[36]
6	Imidazolium salt	Solvent free, 120 °C, 40 min	81-95	[37]
7	H4SiW12O40	Solvent free, 110 °C	78-94	[38]
7	Trityl chloride	Room temp., 1-4 h	83-94	[39]

8	p-TSA	Solvent free, 125 °C, 4-8 h	83-93	[40]
9	MNPs-PhSO3H	Solvent free, 120 °C, 40 min	81-94	[41]
10	PPA-SiO2	Solvent-free, 120°C, 7 min	86	[29]
11	MSNs-HPZ-	Solvent free, 120 °C, 60 min	75-94	[42]
12	Ferrite-Mo	Solvent-free, MW, 3 min	86-94	[25]
13	GO/ZnO/Mn/Co	Solvent-free, 80 °C,2 h	95	Pres work
				wor
				work

Experimental

Materials and Methods

All chemicals were purchased from Fluka or Merck Chemical Company. A Bruker Avance spectrometer (300 MHz) 1HNMR. was recorded in the range 400-4000 cm^{-1} wave number (Thermo (AVATAR). the surface morphology of nanoparticles was studied by scanning electron microscope (SEM) (TESCAN, MIRA III) that operated at an accelerating voltage of 15 KV. All melting points were checked by electrothermal 9300.

Preparation of GO nanosheets

Graphene oxide, made by the well-known Hummer method, briefly mix 5 grams of graphic acid with sodium nitrate and add 115 ml of concentrate to it and continue stirring at 5 ° C for 15 minutes and then Add 15 g of potassium permanganate to the mixture at the specified time and keep at a maximum temperature of 10 ° C for 12 hours. Then add 230 ml of distilled water to the mixture and continue stirring at 98 ° C for 15 minutes, and after sufficient time add 700 ml of water until the ambient temperature reaches 40 ° C and at this stage 50 ° C. 30 ml of 30% oxygenated water was added dropwise and finally the yellow precipitate was removed by centrifugation and washed several times using 5% hydrochloric acid.

Synthesis of GO/ZnO/Mn/Co [27]

Mix 1 gram of graphene oxide with 200 mg of ZnO and a 150 mL round bottom balloon, then added 0.5 mL of cobalt nitrate to 0.5 M and 3 ml of manganese nitrate 0.5 M and heat until 10 min so gently with 0.5 M of sodium carbonate solution the pH of the solution reaches 10. At 15 min the reaction is complete then reflux for 5 to 6 h. At the end, filter and rinse several times with distilled water and dry for 16 h at 50 °C.

Synthesis of xanthene derivatives using GO/ZnO/Mn/Co and GO/Mn/Co

0.288 g 2-naphthol (2 mmol) and 0.20 g 4-methyl benzaldehyde (4 mmol) and 0.1 g of GO/ZnO/Mn/Co or GO/Mn/Co were mixed and heated at 80 ˚C for 2 h. check the progress of the reaction by TLC. After completing the reaction, the mixture was dissolved in ethanol at room temperature and in the lowest amount of water, and by centrifugation the solvent was separated from the solid, then the solid was recrystallization with ethanol and cold water.

Some selected spectral data of the products

14-(4-Methoxyphenyl)-14H-dibenzo[a,j]xanthene: , GO/Mn/Co with ZnO

Table 2, entry 2; M.p. 202–204 °C (lit. 203–205 °C) [28]; FT-IR (KBr, cm-1): 3042 (aromatic CH), 1625 (C,C), 1594, 1512 (C,C), 1251 (C–O), 1236 (C–O), 811; 1HNMR (CDCl3, 300 MHz) δ (ppm): 3.62 (3H, s, OCH3), 6.45 (1H, s, CH), 6.67–6.69 (2H, d, J = 7.9 Hz, Ar-H), 7.27–7.40(4H, m), 7.42–7.44 (4H, m), 7.78–7.80 (2H, d, J = 8.0 Hz, Ar-H), 8.82–8.84(2H, d, J = 8.0 Hz, Ar-H), 8.38–7.40 (2H, d, J = 7.9 Hz, Ar-H). 13C NMR (CDCl3, 50 MHz) δ (ppm): 37.1, 55.1, 113.8, 117.5, 118.0, 122.7, 124.7,126.7, 128.7, 128.8, 129.2, 131.1, 131.4, 137.4, 148.6, 157.8.

Synthesis of xanthene derivatives by the ultrasonic method using GO/ZnO/Mn/Co

0.288 g 2-naphthol (2 mmol) and 0.120 g 4-methyl benzaldehyde (4 mmol) and 0.1 g of GO/ZnO/Mn/Co mixed with 80 mL of water and put it in a 120-watt ultrasonic device equal to 10% of the total power of the device and 35 KHz at 60

°C for 30 min (Check the progress of the reaction by TLC). The reaction was completed after 30 min. After the reaction was completed, the solid was recrystallized with ethanol and cold water.

14-(4-Methoxyphenyl)-14H-dibenzo[a,j]xanthene: GO/Mn/Co with ZnO, ultrasonic method

m.p. 202-204 ˚C; FT-IR(KBr, cm-1): 3042 (aromatic CH), 1625 (C,C), 1594, 1512 (C,C), 1251 (C–O), 1236 (C–O), 811; 1HNMR (CDCl3, 300 MHz) δ (ppm): 3.62 (3H, s, OCH3), 6.45 (1H, s, CH), 6.67–6.69 (2H, d, J = 7.9 Hz, Ar-H), 7.27–7.40(4H, m), 7.42–7.44 (4H, m), 7.78–7.80 (2H, d, J = 8.0 Hz, Ar-H), 8.82–8.84(2H, d, J = 8.0 Hz, Ar-H), 8.38–7.40 (2H, d, J = 7.9 Hz, Ar-H). 13C NMR (CDCl3, 50 MHz) δ (ppm): 37.1, 55.1, 113.8, 117.5, 118.0, 122.7, 124.7,126.7, 128.7, 128.8, 129.2, 131.1, 131.4, 137.4, 148.6, 157.8.

Synthesis of amido alkyl naphthol with urea using GO/ZnO/Mn/Co and GO/Mn/Co

0.144 g 2-naphthol (1 mmol), 0.140 g 4-chlorobenzaldehyde (1 mmol), 0.099 g urea (1 mmol), and 0.1 g GO/ZnO/Mn/Co or GO/Mn/Co mixed at 80 ° C for 2h. The progress of the reaction was monitored by TLC. The solid was recrystallized with ethanol and cold water.

[α- (2-Hydroxynaphthalen-1-yl) - (4-chlorobenzyl) methyl] urea, GO/Mn/Co with ZnO

m.p. 168– 170 ˚C, IR(KBr): 3456, 3360, 3200, 2240, 1632, 1580, 1513, 1430, 1370,1238, 816 cm-1; 1HNMR (DMSO-d6, 500 MHz) δ: 10.32 (s, 1H), 7.95-7.75 (m, 3H), 7.50-7.10 (m, 7H), 6.80 (s, 2H), 5.80 (s, 2H) ppm; 13CNMR (DMSO-d6, 125 MHz): 159.3, 153.6, 144.3, 132.8, 131.1, 130.0, 129.4,129.1, 128.9, 128.6, 128.4,127.4, 123.3, 120.4, 119.2, 48.4 ppm.

[α- (2-Hydroxynaphthalen-1-yl) - (4-chlorobenzyl) methyl] urea: GO/Mn/Co in the absence of ZnO

m.p. 168– 170 ˚C, IR(KBr): 3456, 3360, 3200, 2240, 1632, 1580, 1513, 1430, 1370,1238, 816 cm^{-1}; ^{1}HNMR (DMSO-d_6, 300 MHz) δ: 10.32 (s, 1H), 7.95-7.75 (m, 3H), 7.50-7.10 (m, 7H), 6.80 (s, 2H), 5.80 (s, 2H) ppm; ^{13}CNMR(DMSO-d_6, 125 MHz): 159.3, 153.6, 144.3, 132.8, 131.1, 130.0,129.4, 129.1, 128.9, 128.6, 128.4, 127.4, 123.3, 120.4, 119.2, 48.4ppm

Synthesis of amido alkyl naphthol derivatives by the ultrasonic method using GO/ZnO/Mn/Co

00.144 g 2-naphthol (1 mmol), 0.140 g 4-chlorobenzaldehyde (1 mmol), 0.099 g urea (1 mmol), and 0.1 g GO/ZnO/Mn/Co mixed with 80 mL of water and put it in a 120-watt ultrasonic device equal to 10% of the total power of the device and 35 KHz at 60 °C for 30 min (Check the progress of the reaction by TLC). The reaction was completed after 30 min. After the reaction was completed, the solid was recrystallized with ethanol and cold water

[α-(2Hydroxynaphthalen1yl) (4chlorobenzyl) methyl] urea:ultrasonicmethod

Table 6, entry 2; m.p. 168–170˚C, (lit. 203–205 °C) [29]; IR(KBr): 3456, 3360, 3200, 2240, 1632, 1580,1513,1430,1370, 1238, 816 cm^{-1}; ^{1}HNMR (DMSO-d_6, 300 MHz) δ: 10.32 (s,1H), 7.95-7.75 (m, 3H), 7.50-7.10 (m, 7H), 6.80 (s, 2H), 5.80 (s, 2H) ppm;^{13}CNMR (DMSO-d_6, 125 MHz): 159.3, 153.6, 144.3, 132.8, 131.1, 130.0,129.4, 129.1, 128.9, 128.6, 128.4, 127.4, 123.3, 120.4, 119.2, 48.4 ppm.

General procedure for the synthesis of 1-amido alkyl-2-naphthols

A mixture of aldehydes (1 mmol), 2-naphthol (1 mmol), acetamide (1.2 mmol), and nano-graphene oxide (0.007 g) was heated at 90 °C for an appropriate time under solvent-free conditions. The progress of the reaction was monitored by TLC (*n*-hexane: ethyl acetate 1:2). After completion of the reaction, the mixture was washed with ethyl acetate and the crude product was recrystallized by ethanol in order to obtain the pure 1-amidoalkyl-2-naphthol derivatives in 80-96% yields.

[α-(2-Hydroxynaphthalen-1-yl) (phenyl) methyl] acetamide

Table7; m.p. 238-241 °C, IR(KBr): 3436, 3350, 3190, 2220, 1630, 1570,1500,1420,1360, 1235, 812 cm-1-; ^{1}HNMR (DMSO-d6): δ 2.05 (s, 3H), 7.00–7.19 (m, 9H), 7.63–7.70(m, 3H), 8.12 (d, J = 7.6 Hz, 1H), 9.68 (s, 1H); 13C NMR (DMSOd6):δ 23.4, 41.2, 117.8, 121.2, 122.1, 124.0, 124.0, 125.6, 127.5,128.3, 128.4, 128.5, 128.7, 134.3, 144.2, 152.8, 169.8.

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