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An efficient, one-pot, solvent-free synthesis of 1-amidoalkyl-2-naphthols using uranylacetateas new recyclable catalysts

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ABSTRACT

Solvent free and one-potcondensation of an aldehyde, acetamide or benzamide, and 2-naphthol was performed to obtain the corresponding 1-amidoalkyl-2-naphthol. The 1-amidoalkyl-2-naphtholswereproducedby using $UO_2(CH_3COO)_2{\cdot}2H_2O$as a catalyst at 90 $^{\circ}C$ in high yield. This method has several advantages, for example excellent products yields, short times reaction, easy work up and solvent free condition. Also, the catalyst was recyclable for five consecutive runs.

Keywords: 1-amidoalkyl-2-naphthols, Uranylacetate, Multicomponent reaction, Solvent free reaction

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INTRODUCTION

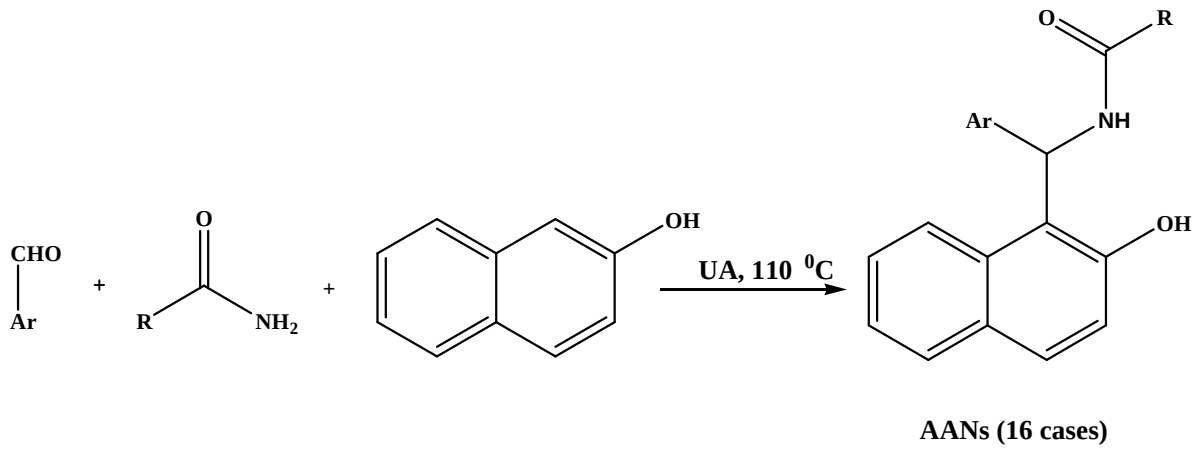
Amidoalkylnaphthols (**AAN**s) are synthetic building blocks for the synthesis of many important derivatives such as aminoalkylnapthols, oxazine derivatives and the other compounds bearing1,3-amino oxygenated groups.1,3-Amino oxygenated containing compounds have great deals biological activity (in example: cardiovascular activity [1], bradycardic effects [2], antibiotic [3], antirheumatic [4], anticonvulsant [5], antitumor [6], antianginal [7], antipsychotic [8], antimalarial [9],antibacterial [10] analgesic [11] and antihypertensive [12]) and chemical applications (such as asymmetric synthesis [13] and also as a catalyst [14]).

AAN scan be prepared by the coupling of an aldehyde, the 2-naphthol and an amide. Different methods for the synthesis of **AAN**s based on the use Lewis and Bronsted acids such as H_2NSO_3H [15], $Fe(HSO_4)_3$ [16], Al_2O_3/H_2SO_4 [17], $NaHSO_4.H_2O$ [18], $InCl_3$ [19], I_2 [20], $ZrOCl_2$ [21], SiO_2/H_2SO_4 [22], $Al(H_2PO_4)_3$ [23], $FeCl_3.SiO_2$ [24], $Sr(OTf)_2$ [25], NBS [26], nano-$TiCl_4$-SiO_2 [27], MoO_3/SiO_2 [28]), and varieties of other conditions such as ultrasonic irradiation [29] and ionic liquid [30],have been reported in the literature.

But there are few references for uranyl acetate (**UA**; $UO_2(CH_3COO)_2{\cdot}2H_2O$**)** as catalyst in organic reactions [31, 32]. In continuation of my investigations on catalytic effect of **UA**[33], here in, I report an efficient synthesis of **AAN**s by **UA** in one pot and solvent free condition.

RESULTS AND DISCUSSION

In order to study the efficiency of new methods, the catalytic effect of **UA**,2-naphthol, benzaldehyde and acetamide was heated in the presence of **UA** to give $\mathbf{AAN_1}$ (Scheme 1), under solvent free condition. In order to optimize the reaction conditions, several methods were tested on the same model reaction by conducting the reaction. The reactions were performed in different temperatures, times and different amount of **UA**. The results from this study are presented in Table 1, whereby the better yields (88%) were obtained when the temperature was at 110 °C with 3 hours reaction time and 2.5mol% of **UA**.



Scheme 1 Synthesis of 1-amidoalkyl-2-naphthol (AAN)

Table2 Synthesis of AAN_1[a] under different conditions for optimization of reactions by UA as catalyst

Temp °C of React.	Catalyst (mol %)	Time (h)	ProductYield(%)
90	20	4	38
100	20	4	77
110	20	4	88
120	20	4	89
110	20	5	88
110	20	3	88
110	20	2	75
110	10	3	88
110	5	3	88
110	***2.5***	***3***	***88***
110	1	3	68

[a]*Benzaldehyede (1 mmol), 2-naphthol (1mmol), acetamide (1.5 mmol), Solvent Free*

Several activated and deactivated aromatic aldehydes underwent the reaction with acetamide or benzamide to give the corresponding **AAN**s in high yields. The results are shown in Table 2. The experimental procedure was very simple, convenient, and had the ability to tolerate a variety of functional groups such as methoxy, nitro and halides under the reaction conditions (Table 3).

Table3 Details 1-aminoalkyl-2-naphthol synthesis

Entry	Ar-	R-	Product	product Yield (%)	mp °C	
					Found	Lit.
1	Ph-	CH_3-	AAN_1	88	244-246	241-243 [25]
2	2-Cl-Ph-	CH_3-	AAN_2	92	206-208	206-207 [34]
3	4-Cl-Ph-	CH_3-	AAN_3	90	234-236	237-238 [34]
4	4-MeO-Ph-	CH_3-	AAN_4	86	187-188	184-185 [25]
5	2-NO_2-Ph-	CH_3-	AAN_5	91	217-219	217-220 [25]
6	3-NO2-Ph-	CH_3-	AAN_6	92	242-244	241-243 [16]
7	4-NO2-Ph-	CH_3-	AAN_7	92	238-240	237-238 [34]
9	2-Furl-	Ph-	AAN_9	89	219-221	218-220 [35]
10	Ph-	Ph-	AAN_{10}	91	234-237	234-236 [36]
11	2-Cl-Ph-	Ph-	AAN_{11}	93	266-268	265-267 [37]
12	4-Cl-Ph-	Ph-	AAN_{12}	90	187-189	187-186 [38]
13	4-MeO-Ph-	Ph-	AAN_{13}	88	207-209	206-208 [34]
14	2-NO_2-Ph-	Ph-	AAN_{14}	92	267-269	266-267 [34]
15	3-NO2-Ph-	Ph-	AAN_{15}	94	243-245	241-243 [39]
16	4-NO2-Ph-	Ph-	AAN_{16}	93	238-240	237-239 [37]
18	2-furyl-	Ph-	AAN_{18}	89	235-237	234-236 [30]

The structures of the productswere confirmed by ^1H NMR, ^{13}C NMR, IR spectroscopy and CHN/O analysis,

Interestingly, the catalyst can be recycled for four consecutive runs without significant loss of activity (Table 3).For this purpose, after completion of the reaction, the reaction mixture was cooled to room temperature and then water was added. The precipitated solid was isolated by filtration; the catalyst was recovered from the filtrate by evaporation of the water, and reused for the similar reaction.

Table 3Recycled of UA in the synthesis of AAN_1 reactions

Catalyst type	Runs				
	1	2	3	4	5
Product yield (%)	89	88	82	76	45

Experimental

All reactions were carried out in an efficient hood. The starting materials were purchased from Merck and Fluka chemical companies. Melting points were determined with a BransteadElectrothermal model 9200 apparatus and are uncorrected. IR spectra were recorded on a Perkin Elmer RX1 Fourier transform infrared spectrometer. The ^{1}H and ^{13}C NMR spectra were recorded in DMSO-d_6 on BrukerAvance 300 MHz spectrometers. Elemental analyses were carried out by a Perkin Elmer 2400 series II CHN/O analyzer.

Synthesis of N-((2-Hydroxynaphthalen-1-yl)(phenyl)methyl)acetamide (AAN_1) as general procedure:A mixture of benzaldehyde (2mmol, 0.21 mL), 2-naphthol (2mmol, 0.29 gr), acetamide (2.5 mmol, 0.13 mL) and**UA**(2.5mol %, 0.022 gr) was heated on oil-bath with stirring at 110 °C for 3 hour (Scheme1, Tables 1 and 2). After cooling, the reaction mixture was poured in ice water and the precipitated solid was collected by filtration, washed with distilled water and dried. The crude product was recrystallized from hot ethanol to give the corresponding pure product (**AAN_1**). mp 244-246˚C ; IR (KBr): 3385, 3243, 3058, 1630, 1575, 1509, 1331, 1096, 800, 732cm^{-1}; ^{1}HNMR(300 MHz, DMSO-d_6): δ 1.96 (s, 3H), 7.13 - 7.38 (m, 9H),7.78 - 7.89 (m, 3H), 8.42 (d, *J* =8.8 Hz, 1H), 9.99 (s, 1H)ppm; ^{13}C NMR(75 MHz, DMSO-d_6): δ 23.7, 41.5, 119.0, 120.4,122.3, 124.1, 125.2, 125.9, 127.8, 128.3, 128.6, 128.9, 130.1, 134.5, 144.2, 152.8, 169.7ppm; Anal. Calcd for $C_{19}H_{17}NO$: C, 78.33; H, 5.88 Found: C, 78.28; H, 5.79.

CONCLUSION

In conclusion, I have successfully developed a quick, convenient and efficient method for the synthesis of **AAN**s under solvent-free conditions. The environmental advantages include omitting organic solvent, generality and simplicity of procedure, shorter reaction time, simple workup, reusable catalyst condition, and pure products in excellent yields.

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