

Recent advances in one-pot multicomponent catalytic synthesis of amidoalkyl naphthols

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ABSTRACT

Amidoalkyl-2-naphthols as synthetic intermediates play an important role in medicinal chemistry due to their remarkable biological and many pharmacological properties. Owing to the versatile biological activities, industrial and synthetic applications of these compounds, introduction of an alternative methodology is urgent in synthetic organic chemistry. Low yield and harsh reaction conditions promoted the researchers for the development of novel green catalysts for the synthesis of amidoalkyl naphthols. Therefore in the present review, we deal with green catalytic synthesis of amidoalkyl naphthols. The main purpose of this review is to present a survey of the literature on various heterogeneous and homogenous catalysts used for the synthesis of amidoalkyl naphthols along with mechanism of reaction since synthesis of such compounds have not been previously reviewed.

Keywords: Amidoalkyl naphthols, Green chemistry, One-pot multicomponent, Heterogeneous, Homogenous catalysts.

1. Introduction

Green chemistry which is also known as benign or sustainable chemistry is the design and development of chemical processes and products by reducing or eliminating the use of substances which is hazardous to human health and the environment. From the early 1990s, green chemistry has been a subject of increasing interest and is developed as a new and rapid emerging field of chemistry. This new branch of science that includes modification of engineering practices and bioremediation, also promotes catalytic processes and eco-friendly reaction media, as well as the concept of atom economy leading to almost zero waste. By using various green chemistry techniques and methodologies of organic synthesis, health and safety have been extended to industrial large scale production from small scale laboratory.

The Pollution Prevention Act of 1990 set the stage for green chemistry. Its focus is the prevention of pollution at the source rather than the treatment of pollutants after they are formed. This goal became a formal objective of the Environmental Protection Agency (EPA) in 1991.

The most widely accepted definition of green chemistry is the one offered by chemists Paul Anastas and John Warner, who defined "Green chemistry as the design of chemical products and processes that reduce or eliminate the use and generation of hazardous substances" [1].

Green chemistry efficiently utilizes toxic or non-hazardous raw materials and solvents in the manufacture by energy efficient process and disposes the waste material in best possible form so that it will not be harmful to environment. Paul Anastas, then of the United States Environmental Protection Agency, and John C. Warner developed twelve principles of green chemistry, which help to explain what the definition means in practice [2]. The principles cover such concepts as:

1. The design of processes to maximize the amount of raw material that ends up in the product
2. The use of safe, environment-friendly raw materials including solvents
3. The design of energy efficient processes
4. The best form of waste disposal: not to create it in the first place

Green Chemistry with its twelve principles would like to see changes in the conventional ways that were used

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for decades to make synthetic organic chemical substances and the use of less toxic starting materials. Green chemistry would like to increase the efficiency of synthetic methods, to use less toxic solvents, reduce the stages of the synthetic routes and minimize waste as far as practically possible. As a consequence, green chemistry focuses on the following [3]:

1. Prevention of waste
2. Atom Economy
3. Less Hazardous Chemical Synthesis
4. Designing Safer Chemicals
5. Safer solvents and auxiliaries
6. Energy efficient
7. Renewable feedstocks
8. Reduce derivatives
9. Catalysis
10. Design for degradation
11. Real time analysis for pollution prevention
12. Safety

The twelve principles of green chemistry [3] can be applied to almost every part of chemistry that includes synthesis of molecules with a desired structure and property, catalysis of a process, less polluting reaction conditions etc.

1. Prevention: It is better to prevent waste formation than to treat or clean up waste after it has been created.
2. Less hazardous chemical synthesis: Wherever possible, synthetic methods should be designed to use and generate substances that possess little or no toxicity to human health and the environment.
3. Designing buffer chemicals: Chemicals products should be designed to affect their desired function while minimizing their toxicity.
4. Atom economy: Synthetic methods should be designed to minimize the incorporation of all materials used in the process into the final product.
5. Safer solvents and auxiliaries: The use of auxiliary substances (solvents, separation, agents, etc.) should be minimized whenever possible and should be made innocuous when used.
6. Design for energy efficiency: Energy requirements of chemical processes should be minimized. If practicable, synthetic methods should be conducted at ambient temperature and pressure.
7. Use renewable feedstocks: Raw material or feedstock should be renewable rather than depleting whenever technically and economically practicable.
8. Reduce derivatives: Unnecessary derivatization (use of blocking groups, protection/deprotection and temporary modification of physical/chemical processes) should be minimized or avoided if possible, because such steps require additional reagents and can generate waste.
9. Catalysis: The use of catalysts for organic transformations should be encouraged. The catalysts promoted organic synthesis creates opportunities for

increased selectivity, better yield, and feasibility of non-feasible reaction.

10. Design for degradation: The end chemical products should be designed so that it would be broken down into non-toxic degradation products and hence not persists in the environment.

11. Real-time analysis for pollution prevention: Analytical methodologies need to be further designed to monitor and control the in-process generation of hazardous substances before it is formed.

12. Inherently safer chemistry for accident prevention: The raw materials including solvents used in chemical process should be chosen to minimize the potential for chemical accidents including releases, explosions and fires.

A shift in emphasis on organic synthesis is apparent with the desire to develop environmentally benign methods and routes to a number of biologically active molecules using non-toxic reagents, solvents, and catalysts. There is an increasing interest in the use of microwave techniques under neat conditions by various multicomponent reactions. These types of green chemistry approaches are very attractive for the synthesis of many heterocyclic and non-heterocyclic medicinal compounds.

Microwave-assisted organic synthesis [4] (MAOS) has developed as a new “lead” in organic synthesis and it has emerged as an expanding field in synthesis research. The principles of green chemistry apply to most of the synthetic routes with microwave irradiation. Microwave-assisted eco-friendly organic synthesis has become a new trend with many applications in synthesizing organic chemicals. Organic reactions under the microwave irradiation have many advantages compared to the conventional reactions which need very high temperatures. Microwave assisted reactions are “cleaner”, last only very few minutes, have high yield and produce minimum waste. The technique offers a large number of advantages, which encouraged many chemists to switch from traditional heating method to microwave assisted chemistry.

Microwave include following advantages, over the conventional heating.

1. Simple and clean
2. Uniform heating occurs throughout the material
3. Process speed is increased
4. High efficiency of heating
5. Reduction in unwanted side reaction
6. Purity in final product,
7. Improve reproducibility
8. Environmental heat loss can be avoided
9. Reduce wastage of heating reaction vessel
10. Low operating cost
11. Highly economic

Avoiding organic solvents during the reactions in organic synthesis leads to a clean, efficient, and economical technology (green chemistry). Earlier,

chemists thought that compounds react only in the liquid state or if dissolved. This has made solvents common in chemical syntheses, however, many compounds used as solvents were found to be environmentally unfriendly. The problem associated with waste disposal of solvents has been overcome by performing reactions in dry media conditions i.e. without a solvent under microwave irradiation [4] (MWI). These expeditious and solvent-free approaches involve the exposure of neat reactants to MWI in conjunction with the use of supported reagents or catalysts [5,6].

One of the methods belonging to such a green protocol is multicomponent solvent free synthesis. One of these MCRs is the preparation of amidoalkyl naphthols. One-pot multicomponent reactions (MCR's), sometimes also called as zipper reactions, have appeared as an imperative means for construction of diverse and complex organic molecules [7,8]. MCR's have gained significant importance as a tool for the synthesis of a wide variety of useful compounds, including pharmaceuticals. In recent decades, MCR's have gained wide applicability in the field of modern synthetic organic chemistry due to formation of carbon-carbon and carbon-hetero atom bonds in one-pot which contains essentially all the carbon atoms of the starting materials [9-11]. MCR's condensations involve three or more compounds mixed at once in one-pot to perform a sequence of reactions in a single event, but consecutively to form new products, which contains the essential parts of all the starting materials

without isolation of intermediates and other side products [12,13]. They also have additional merits over two-component reactions in other aspects including the simplicity of a one-pot procedure, possible structural variations, building up complex molecules as, increase the efficiency of the reaction and decrease the number of laboratory operations along with quantities of solvent and chemicals used. They also reduce the reaction time considerably and facilitate the yield of products than the normal multiple step methods. There has been tremendous development in three or four component reaction specially the Bignelli [14-16], Passerini [17], Ugi [18,19] and Mannich [20,21] reactions, which have further led to renaissance of MCRs. The search and discovery for new MCRs on one hand and the full exploitation of already known multicomponent reactions on the other hand, is therefore of considerable current interest in synthetic medicinal chemistry.

Amidoalkyl naphthols are the vital synthetic building blocks [22-23] and used as precursors for the synthesis of many important derivatives (Fig. 1). 1-Amidoalkyl-2-naphthols can be easily converted into 1-aminoalkyl-2-naphthols derivatives by an amide hydrolysis reaction which exhibits depressor and bradycardia effects in humans [24,25]. Aminoalkyl naphthols derivatives, exhibit important cardiovascular activity. Aminoalkyl naphthols have attracted strong interest to their useful biological and pharmacological properties such as adrenoceptor blocking, antihypertensive, and Ca^{2+} channel blocking activities.

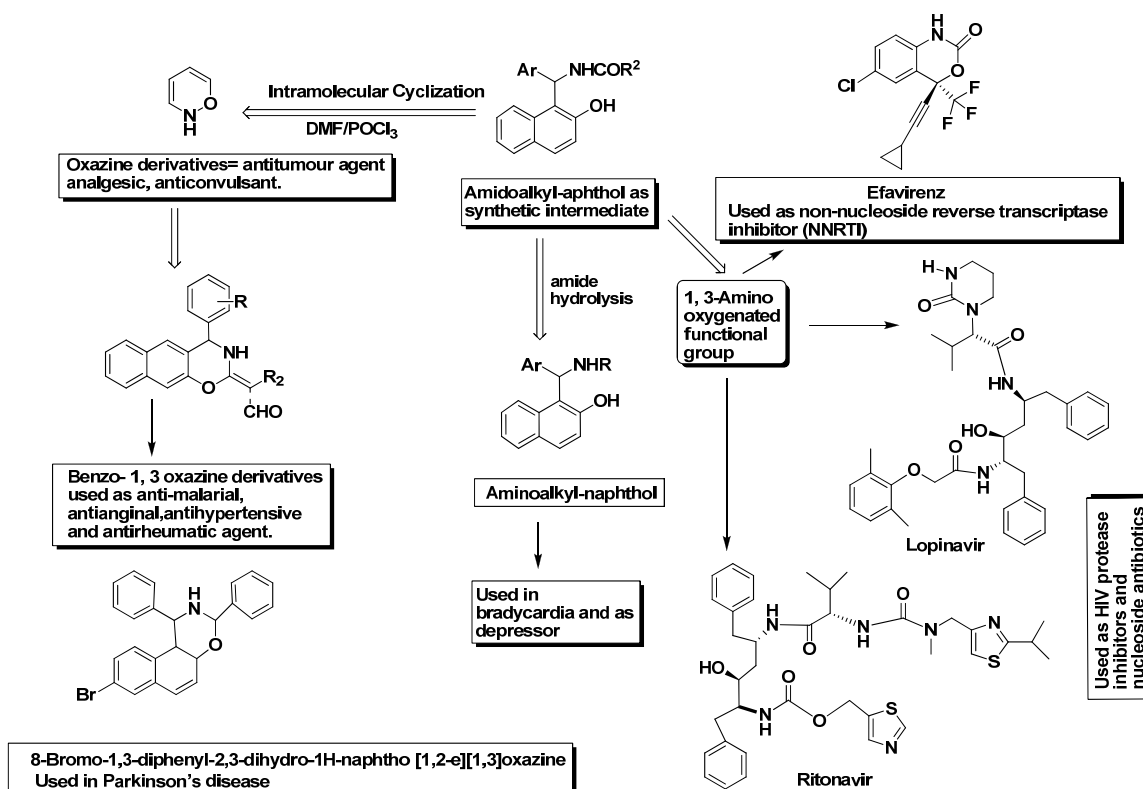
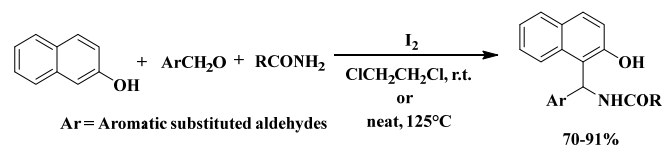


Fig. 1. The various pharmacological activities of amidoalkyl naphthols.

Moreover this 1-aminoalkyl alcohol-type ligand has been used for asymmetric synthesis and also as a catalyst [26,27]. 1-Amidoalkyl-2-naphthols can be also converted to 1,3-oxazine derivatives [28-32]. 1,3-Oxazines and have potentially numerous biological activities including antirheumatic [33], antibiotic [34], antitumor [35], antipsychotic [36], antimalarial [37], antianginal [38], analgesic [39], anticonvulsant [40], antihypertensive [41] and antibacterial [42] properties. The (quinolinyl- and isoquinolinyl-amino) alkyl-naphthols belong to the category of 1,3-amino-oxygenated functional group or 1,3-oxazine compounds which comprise varieties of biologically important natural products [43], bioactive molecules [44-47] and potent drugs such as nucleoside antibiotics and HIV protease inhibitors [48]. Two very important such compounds are ritonavir and lopinavir being HIV protease inhibitors of great significance. The most outstanding of these is Sustiva (Efavirenz), a non-nucleoside reverse transcriptase inhibitor that has been approved by the FDA in 1998 and is presently in clinical use for the treatment of AIDS [49]. In addition, naphthoxazine derivatives have exhibited therapeutic potential for the treatment of Parkinson's disease [50,51].

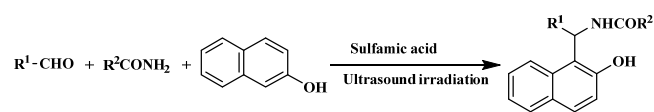
2. Literature Review

Das *et al.* [52] introduced the solvent-free method that has an operationally simple procedure. A mixture of 2-naphthol (1 mmol), aldehyde (1 mmol) and urea or amide (1.3 mmol) in dichloroethane (10 ml) and molecular iodine (5 mol%, catalyst) was stirred at room temperature to form corresponding amidoalkyl naphthols in high yields (Scheme 1).



Scheme 1.

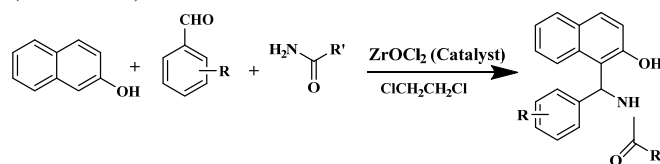
Patil *et al.* [53] reported a new method for the synthesis of 1-amidoalkyl-2-naphthols which were synthesized by three-component condensation of 2-naphthol with ureas/amides and aldehydes by ultrasound irradiation in the presence of sulfamic acid. The condensation reaction occurred under ambient conditions with excellent yields (Scheme 2).



Scheme 2.

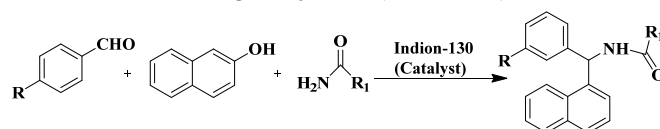
Nagawade and Shinde [54] synthesized 1-amidoalkyl-2-naphthols in very good yields in solvent-free conditions from the multicomponent condensation

reaction of β -naphthol, aromatic aldehydes and urea or amide in the presence of zirconyl (IV) chloride as catalyst. This method offers several advantages such as high conversions, shorter reaction times, cleaner reaction profiles and simple experimental procedures (Scheme 3).



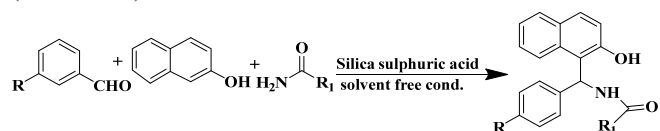
Scheme 3.

Patil *et al.* [55] introduced the cation-exchanged resins (indion-130) catalyzed synthesis of amidoalkyl naphthols by the reaction one-pot, three-component condensation of 2-naphthol, amides/ureas and aldehydes under solvent-free conditions in short reaction times and good yields (Scheme 4).



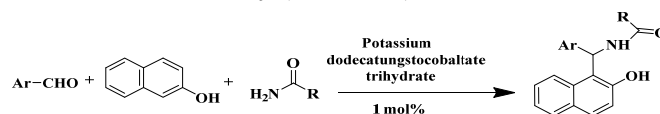
Scheme 4.

Srihari *et al.* [56] introduced one pot method for the synthesis of amidoalkyl naphthols by the condensation of aromatic or aliphatic aldehydes, α - or β -naphthols and amides or urea or thiourea in the presence of silica sulfuric acid as catalyst under the solvent-free conditions at room temperature in high yields (Scheme 5).



Scheme 5.

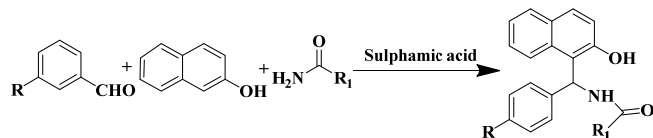
Nagarapu *et al.* [57] describes an efficient and direct procedure for the preparation of amidoalkyl naphthols from the condensation of aryl aldehydes, β -naphthol and urea or amides catalyzed by potassium dodecatungstocobaltate trihydrate (1 mol%) as heterogeneous catalyst. The reaction proceeds well in the presence of solvent as 1,2-dichloroethane at room temperature or under solvent-free conditions at elevated temperature. Moreover, the catalyst exhibited remarkable reusability (Scheme 6)



Scheme 6.

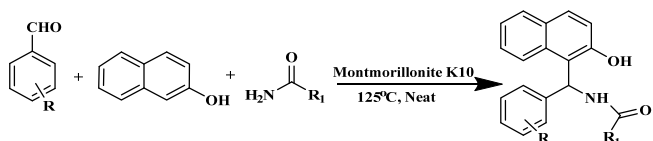
Nagawade and Shinde [58] reported a new procedure by using sulphamic acid (H_2NSO_3H) as an efficient catalyst for the synthesis of amidoalkyl naphthols

under solvent-free conditions. This method has advantages of high conversions, short reaction time and clean reaction profiles with simple experimental and work-up procedures (Scheme 7).



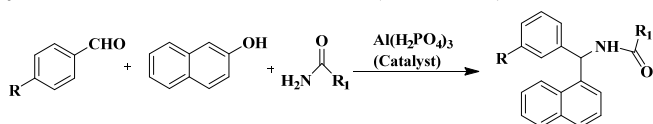
Scheme 7.

Kantevari *et al.* [59] introduced a simple and versatile method for the preparation of amidoalkyl naphthols *via* three-component condensation reaction β -naphthol, aromatic aldehyde, amides or urea in the presence of catalytic amount of montmorillonite K10 clay. This method has advantages of shorter reaction time (<2 h), simple workup, excellent yields, recovery and reusability of catalyst (Scheme 8).



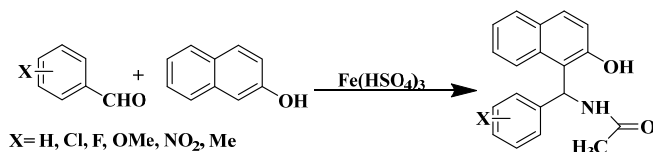
Scheme 8.

Shaterian *et al.* [60] introduced aluminium tris (dihydrogen phosphate) as a very stable and recovered catalyst for the preparation of amidoalkyl naphthols. The reaction proceeds efficiently under thermal conditions and microwave irradiation with excellent yields and short reaction times (Scheme 9).



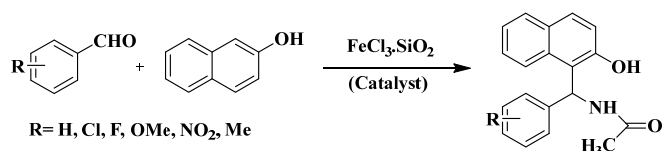
Scheme 9.

Shaterian *et al.* [61] describes efficiently the condensation reaction of 2-naphthol, aromatic aldehydes and acetamide in the presence of ferric hydrogensulfate as catalyst to afford the corresponding amidoalkyl naphthols under solvent, solvent-free and microwave conditions. Regarding the optimum quantity of the catalyst, it was found that 5 mol% of catalyst is necessary to promote the reaction in high to excellent yields (Scheme 10).



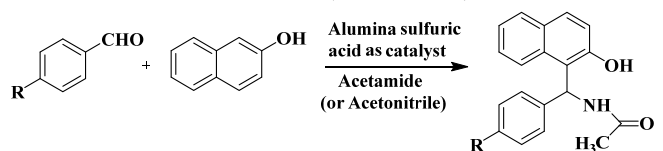
Scheme 10.

Shaterian and Yarahmadi [62] synthesized amidoalkyl naphthols using $\text{FeCl}_3 \cdot \text{SiO}_2$ as a heterogeneous catalyst under thermal solvent less conditions (Scheme 11).



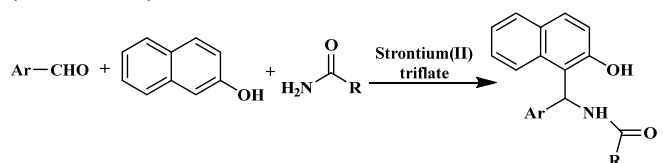
Scheme 11.

Shaterian *et al.* [63] reported an efficient and expeditious direct protocol for the synthesis of amidoalkyl naphthols employing alumina sulfuric acid as heterogeneous catalyst under solvent, solvent-free and microwave conditions (Scheme 12).



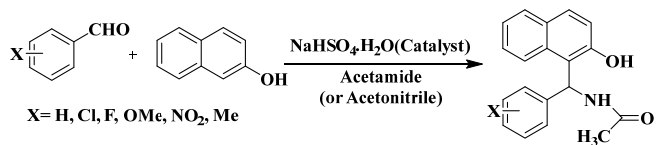
Scheme 12.

Li *et al.* [64] synthesized 1-amidoalkyl-2-naphthols 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 (Scheme 13).



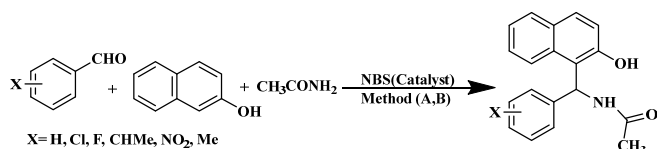
Scheme 13.

Shaterian and Yarahmadi [65] reported the reaction between aryl aldehydes, 2-naphthol and acetonitrile (reactant as well as solvent, 5.0 ml) or acetamide in the presence of $\text{NaHSO}_4 \cdot \text{H}_2\text{O}$ (45 mg) that gave rise to good to excellent yields of the 1-amidoalkyl-2-naphthol derivatives in short filtration. To show reusability of the catalyst, the recovered catalyst from the reaction of benzaldehyde, acetamide and 2-naphthol was used for the same reaction for five times, it showed the same activity such as fresh catalyst without loss of its activity. The reaction proceeds efficiently under thermal solvent-free and microwave green conditions (Scheme 14).



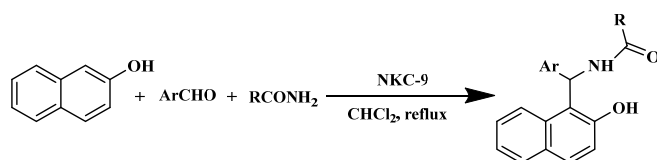
Scheme 14.

Shaterian *et al.* [66] introduced the *N*-bromo succinimide (NBS) catalyzed multi-component and one-pot condensation reaction of 2-naphthol, benzaldehydes, and acetamide for the synthesis of acetamidobenzyl-naphthols under thermal and microwave irradiation conditions (Scheme 15).



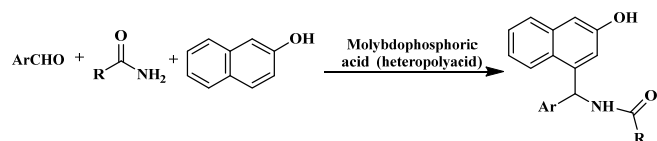
Scheme 15.

An *et al.* [67] reported the Synthesis of α -amidoalkyl- β -naphthols by three-component condensation reaction of 2-naphthol, aromatic aldehyde and urea/amide in the presence of polymer-supported sulphonic acid NKC-9 as catalyst. The method offers the advantages of catalyst reusability, good to high yields and environmental friendliness (Scheme 16).



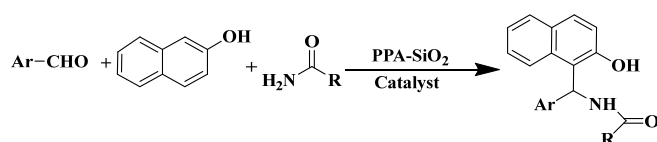
Scheme 16.

Jiang *et al.* [68] introduced molybdophosphoric acid (heteropolyacid) catalyzed condensation of 2-naphthol, aldehyde, urea or amide to afford amidoalkyl naphthols in high yields (Scheme 17).



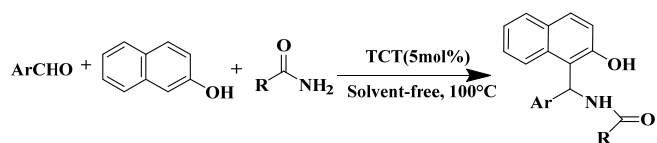
Scheme 17.

Shaterian *et al.* [69] introduced silica gel-supported polyphosphoric acid (PPA-SiO₂) catalyzed multicomponent condensation reaction of aryl aldehydes, 2-naphthol and urea or amides under solvent-free conditions afforded 1-amidoalkyl-2-naphthols in good to excellent yields (Scheme 18).



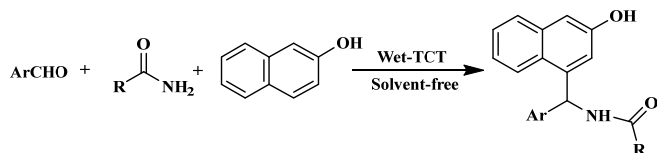
Scheme 18.

Zhang and Zhang [70] introduced 2, 4, 6-trichloro-1, 3, 5-triazine (TCT, Cyanuric Chloride) efficiently catalyzed the condensation of 2-naphthol, aromatic aldehydes and various amides to afford amidoalkyl naphthols in good to excellent yields. 2, 4, 6-trichloro-1, 3, 5-triazine (TCT) has received considerable attention due to its commercial availability, stable, non-volatile, inexpensive and an easy to handle reagent. Regarding the optimum quantity of the catalyst, it was found that 5 mol% of catalyst is necessary to promote the reaction with good to fairly high yields (Scheme 19).



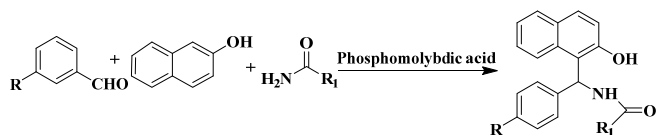
Scheme 19.

Mahdavinia and Bigdeli [71] synthesized an amidoalkyl naphthols by a one-pot condensation of aryl aldehydes, 2-naphthol and urea or amides, in the presence of wet-cyanuric chloride (wet-TCT) as a catalyst. The reaction offers several advantages such as excellent yields, simple procedure, solvent-free and eco-friendly reaction conditions (Scheme 20).



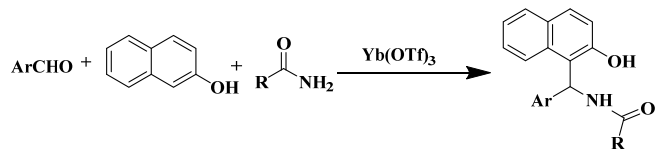
Scheme 20.

Gawand *et al.* [72] reported the one-pot synthesis of amidoalkyl naphthols using aromatic aldehydes, β -naphthol, and urea or amides in the presence of solid H₃Mo₁₂O₄₀P as an efficient catalyst. The catalyst was efficiently recovered from the reaction mixture and reused with negligible loss of catalyst activity (Scheme 21).



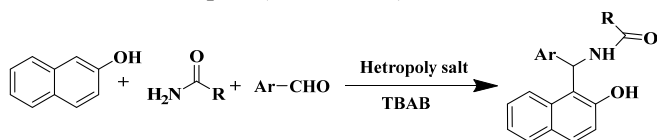
Scheme 21.

Kumar *et al.* [73] introduced ytterbium triflate catalyzed condensation of β -naphthol, aromatic aldehydes and amides in ionic liquids to afford amidoalkyl naphthols under eco-friendly and non-corrosive conditions in high yields (Scheme 22).



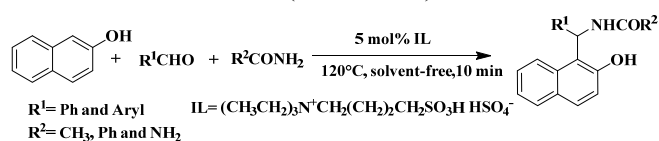
Scheme 22.

Khabazzadeh *et al.* [74] introduced a facile method for the synthesis of amidoalkyl naphthols by condensation of various aldehydes, 2-naphthol and urea or amides with Cu-exchanged heteropoly acids as efficient and reusable catalysts in a temperature controlled oil bath at 100°C for 90 min. The reaction was conducted in molten tetrabutylammonium bromide as a cost effective ionic liquid (Scheme 23).



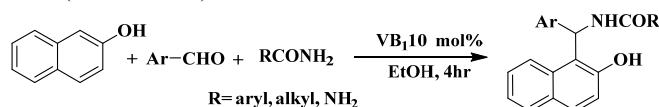
Scheme 23.

Hajipour *et al.* [75] introduced the condensation reaction of aldehydes with amides or urea and 2-naphthol in the presence of catalytic amount of 5 mol% Bronsted acidic ionic liquid ([TEBSA][H₂SO₄]) under thermal solvent-free conditions in excellent high yields and short reaction time (Scheme 24).



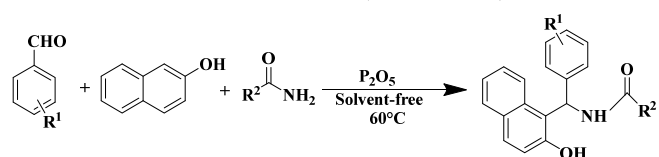
Scheme 24.

Lei *et al.* [76] introduced a new approach in which condensation of aryl aldehydes, 2-naphthol and various amides with ethanol in the presence of 10 mol% thiamine hydrochloride (VB₁) as a novel catalyst for 4 hr afforded 1-amidoalkyl-2-naphthol derivatives in 75-93 % yield. This method offers salient features of catalyst such as non-toxic, inexpensive and metal ion free (Scheme 25).



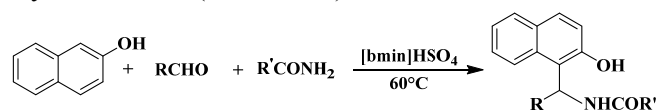
Scheme 25.

Nandi *et al.* [77] carried out the synthesis of amidoalkyl naphthols *via* multi-component one-pot reaction of β -naphthol, aromatic aldehydes and amides catalyzed by P₂O₅ at moderate temperature of 60°C under solvent-free conditions (Scheme 26).



Scheme 26.

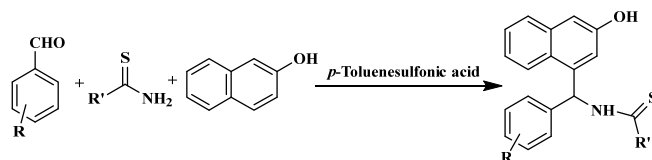
Sapkal *et al.* [78] introduced a facile method for the synthesis of amidoalkyl naphthols by the condensation of aromatic or heteroaromatic or aliphatic aldehydes, 2-naphthol and amides or urea in the presence of acidic ionic liquid 1-butyl-3-methylimidazolium hydrogen sulphate ([bmin]HSO₄) as a catalyst at 60°C with stirring. The reaction mixture was extracted with ethyl acetate (2 × 20 mL) to separate and leaving behind the ionic liquid catalyst. The organic layer washed by brine (2 × 10 mL) solution and dried over sodium sulphate. The organic layer was evaporated under reduced pressure and the solid product obtained was crystallized out (Scheme 27).



Scheme 27.

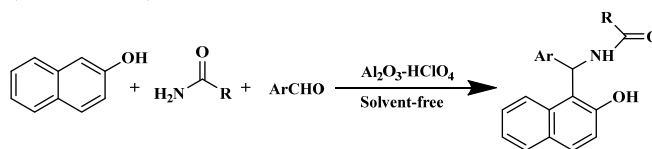
Zhang *et al.* [79] introduced *p*-toluenesulfonic acid catalyzed an efficient, inexpensive and mild method

for the synthesis of α -thiocarbamidoalkyl- β -naphthols by the condensation reaction of aromatic aldehydes, β -naphthol and *N*-phenylthiourea or thiourea at room temperature in short reaction times and high yields (Scheme 28).



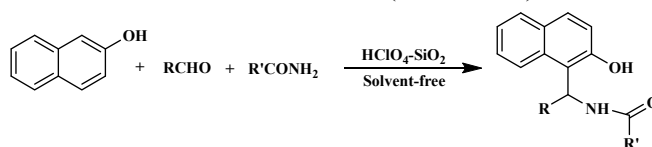
Scheme 28.

Shaterian *et al.* [80] introduced preparation of perchloric acid supported on alumina and its primary application as a solid supported heterogeneous catalyst for the synthesis of α -(α -amidobenzyl)- β -naphthols by a one-pot, three-component condensation of benzaldehydes, β -naphthol and acetamide or benzamide under thermal solvent-free conditions (Scheme 29).



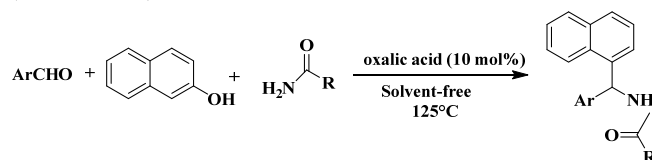
Scheme 29.

Shaterian *et al.* [81] introduced HClO₄-SiO₂ as a versatile heterogeneous catalyzed condensation reaction of a series of new and novel *N*-protected 1-aminoalkyl-2-naphthol derivatives under thermal solvent-free reaction conditions (Scheme 30).



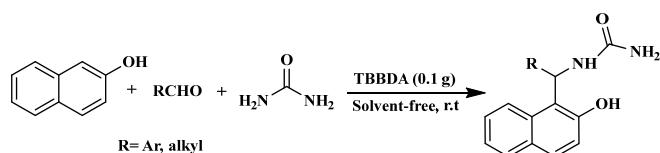
Scheme 30.

Ansari *et al.* [82] introduced the reactions which were carried out at 125°C and magnetically stirred for 7-24 min by taking a 1:1:1.1 mmol ratio mixture of aromatic aldehydes, 2-naphthol and urea/amide in the presence of catalytic amount of 10 mol% oxalic acid to give desired products under solvent-free conditions (Scheme 31).



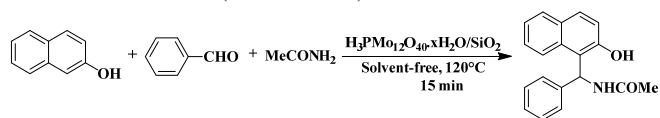
Scheme 31.

Vaghei and Malaekehpaour [83] introduced *N, N, N, N'*-tetrabromo benzene-1,3-disulfonamide [TBBDA] catalyzed an efficient synthesis of various amidoalkyl naphthols from β -naphthol, aromatic aldehydes and urea in good to high yields under solvent-free conditions (Scheme 32).



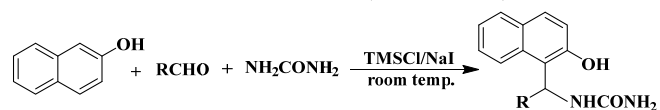
Scheme 32.

Zare *et al.* [84] describes an efficient, clean and green method for the synthesis of 1-amidoalkyl-2-naphthols by the condensation reaction of 2-naphthol, acetamide and aromatic aldehyde with Silica-Supported Molybdotophosphoric Acid ($\text{H}_3\text{PMo}_{12}\text{O}_{40} \cdot x\text{H}_2\text{O}/\text{SiO}_2$, 3.17 mol%) catalyst under solvent-free conditions refluxed with stirring in an oil-bath at 120 °C. The reaction mixture was cooled to room temperature and the product was separated by silica gel column chromatography using an ethyl acetate-hexane (1:2) mixture as eluent (Scheme 33).



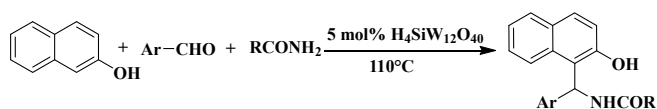
Scheme 33.

Sabitha *et al.* [85] introduced the new approach for the synthesis of amidoalkyl-naphthol derivatives by one-pot condensation of β -naphthol, aromatic aldehydes and urea in the presence of TMSCl/NaI catalyst at room temperature under solvent-free conditions. Ring closure of amidoalkyl-naphthol derivatives occurred at 140°C to afford 1,2-dihydro-1-arylnaphthol [1,2-e] [1,3]oxazin-3-one derivatives (Scheme 34).



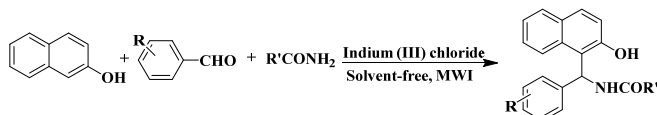
Scheme 34.

Supale and Gokavi [86] introduced the application on of silicotungstic acid as a recyclable catalyst (5 mol%) for the synthesis of amidoalkyl naphthols by the condensation reaction of aromatic aldehydes, 2-naphthol and amide/urea with stirring at 110°C in an oil bath. When the amount of catalyst was used less than 5 mol% yields of amidoalkyl naphthols derivatives were decreased with more time required and further increase in concentration of catalyst upto 10 mol% decreases the yield within same time period. After the completion of reaction, the catalyst was easily separated by addition of ethyl acetate and simple filtration. The filtrate was washed with aqueous $\text{Na}_2\text{S}_2\text{O}_3$ and the organic layer was concentrated and the product was separated. To show the recyclability of catalyst, the recovered catalyst from the reaction mixture was filtered off to get catalyst back. It was recycled for five times without loss in its activity. Heteropolyacids (HPAs) with keggin structure are widely used as solid acid catalysts due to their very strong Bronsted acidity and their structure properties (Scheme 35).



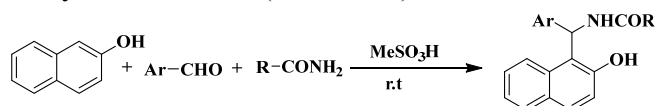
Scheme 35.

Chavan *et al.* [87] introduced indium (III) chloride catalyzed condensation reaction of β -naphthol, aromatic aldehydes and acetamide/urea to afford 1-amidoalkyl-2-naphthols derivatives under solvent-free and microwave irradiation conditions (Scheme 36).



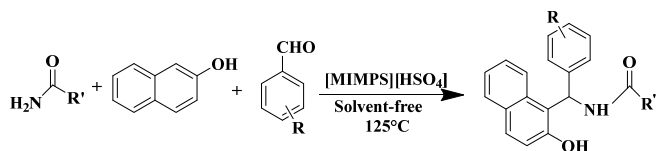
Scheme 36.

Rani *et al.* [88] reported the synthesis of 1-amidoalkyl-2-naphthols by the condensation reaction, of various aromatic aldehydes, 2-naphthols, and amide or urea in the presence of 2.5 mol% methane sulphonic acid (MSA) as a catalyst at room temperature in water under ambient conditions. The yields are high and reactions go to completion within 1-5 min. MSA, which is an inexpensive and safe, can efficiently catalyze this reaction (Scheme 37).



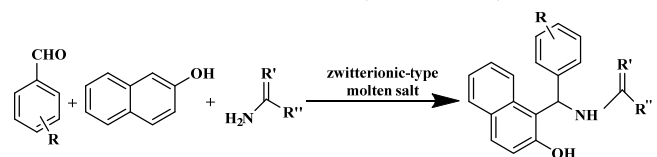
Scheme 37.

She *et al.* [89] reported the synthesis of 1-amidoalkyl-2-naphthols by the three-component Ritter type reaction of β -naphthol, aromatic aldehydes and amide/urea catalyzed by sulfonic ionic liquid, 1-methyl-3-propanesulfonic acid imidazolium hydrogen sulfate [MIMPS][HSO₄] in the ratio of 1:1:1.3:0.1 mmol at 125°C for 5-40 min in 85-97% yields (Scheme 38).



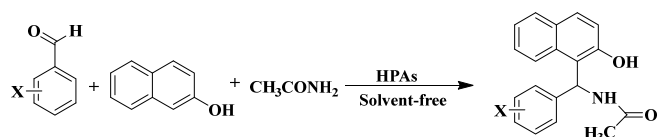
Scheme 38.

Dhiman *et al.* [90] carried out the synthesis of 2-amidoalkyl and 2-carbamatoalkyl naphthol derivatives by condensing the reaction mixture of 2-naphthol, aldehydes and amide or carbamates in the presence of zwitterionic-type molten salt as mild organocatalyst under solvent-free conditions (Scheme 39).



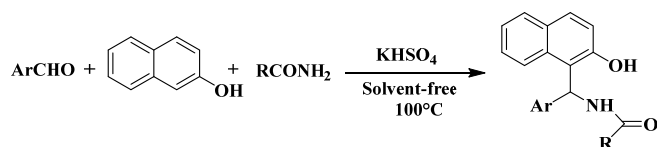
Scheme 39.

Bamoharram *et al.* [91] involves the simple method for the synthesis of amidoalkyl naphthols using catalytic amounts of different types of vanadium (V)-substituted polyoxomolybdates (HPAs) including $H_4[PMo_{11}VO_{40}]$, $H_5[PMo_{10}V_2O_{40}]$ and $H_6[PMo_9V_3O_{40}]$. In all cases, $H_6[PMo_9V_3O_{40}]$ gave the highest yield under solvent-free conditions. The presence of both bronsted acidity and vanadium in the structure of $H_6[PMo_9V_3O_{40}]$ heteropolyacid is responsible for the catalytic activity. The greater protons number may lower, the activation energy barrier and the greater vanadium atoms may provide many sites for the catalytic reaction. The catalyst can be easily recovered, regenerated and reused without loss of structure and activity (Scheme 40).



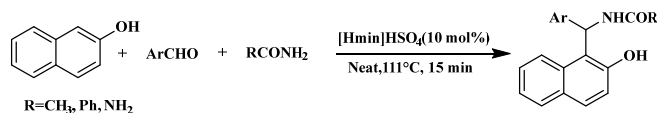
Scheme 40.

Cai *et al.* [92] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of β -naphthol, aromatic aldehydes, acetamide or urea in the presence of catalytic amount of potassium hydrogen sulfate at 100°C for the appropriate time in 83~96% yields (Scheme 41).



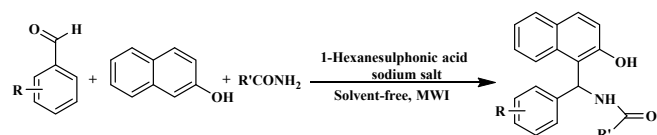
Scheme 41.

Khazdooz *et al.* [93] reported methylimidazolium hydrogensulfate as a bronsted ionic liquid ($[Hmin]HSO_4$) catalyzed synthesis of 1-amidoalkyl-2-naphthols under solvent-free conditions (Scheme 42).



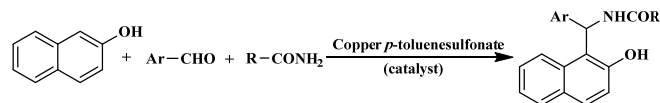
Scheme 42.

Kirti *et al.* [94] introduced an easy workup practical protocol for the synthesis of amidoalkyl naphthols in the presence of an inexpensive solid catalyst as 1-hexanesulphonic acid sodium salt under solvent-free and microwave-irradiation. This method has advantages of clean conversion, greater selectivity and economically attractive (Scheme 43).



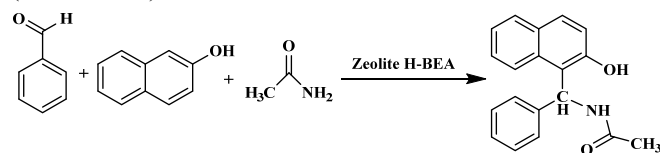
Scheme 43.

Wang and Liag [95] reported an efficient solvent-free synthesis of 1-amidoalkyl-2-naphthols by multi-component reaction of 2-naphthol, aromatic aldehydes and amide or urea in the presence of catalytic amount of copper *p*-toluenesulfonate (Scheme 44).



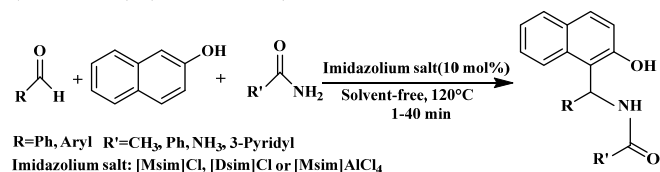
Scheme 44.

Mistry *et al.* [96] introduced a facile method for the synthesis of 1-amidoalkyl-2-naphthols by the condensation of 2-naphthol, aldehydes and acetamide in the ratio of 1:1:1.2 mmol with zeolite H-beta (H-BEA) in the presence of solvent-free conditions heated with stirring in a temperature controlled oil bath at 120°C. The reaction mixture was cooled to 25°C and solid residue was dissolved in EtOAc and the mixture stirred for 5 min. The catalyst was recovered and the product was recrystallized from aqueous EtOH (15%) (Scheme 45).



Scheme 45.

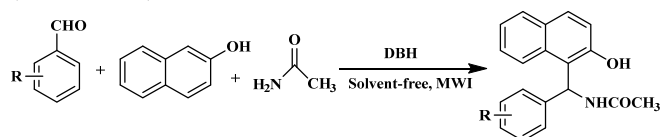
Zolfigol *et al.* [97] introduced the applications of novel sulfonic acid functionalized imidazolium salts including 3-methyl-1-sulfonic acid imidazolium chloride $\{[Msim]Cl\}$ (an ionic liquid), 1,3-disulfonic imidazolium chloride $\{[Dsim]Cl\}$ (an ionic liquid) and 3-methyl-1-sulfonic acid imidazolium tetrachloro aluminate $\{[Msim]AlCl_4\}$ (a solid) which efficiently catalyze one-pot multi-component condensation of 2-naphthol with aromatic aldehydes and amide derivatives to afford 1-amidoalkyl-2-naphthols. The reaction proceeds well under solvent-free conditions giving excellent yields (81-96%) in very short reaction times (1-40 min) (Scheme 46).



Scheme 46.

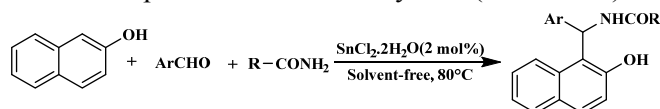
Habibzadeh and Ghasemnejad-Bosra [98] describes an efficient and operationally simple new method for the synthesis of 1-amidoalkyl-2-naphthols under microwave irradiation versus conventional thermal method for comparative purposes. The procedure consists of condensation of 2-naphthols, aromatic aldehydes and acetamide in the presence of catalytic amount of 1,3-dibromo-5,5-dimethylhydantoin (DBH).

This greener protocol proceeds efficiently in the absence of any organic solvent offers mild, clean, high yields and environmentally benign reaction conditions (Scheme 47).



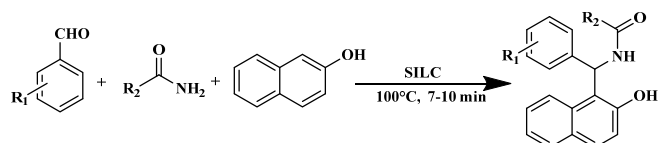
Scheme 47.

Wang *et al.* [99] introduced the reactions which were carried out at 80°C with one-pot three-component condensation of 2-naphthol, aldehydes and primary amides in the presence of 2 mol% SnCl₂·2H₂O to give the desired products in excellent yields (Scheme 48).



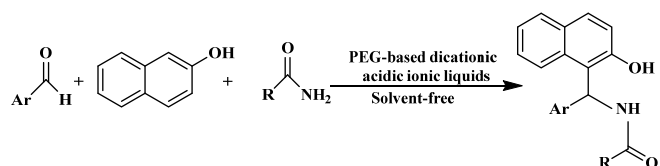
Scheme 48.

Kotadia and Soni [100] reported the condensation reactions of 1-amidoalkyl-2-naphthols from 2-naphthol, amides and aldehydes using silica gel supported-SO₃H functionalized benzimidazolium based ionic liquid as catalyst for the rapid synthesis of 1-amidoalkyl-2 naphthols which was found to be an efficient, mild, safer, high product quality with catalyst recycling (Scheme 49).



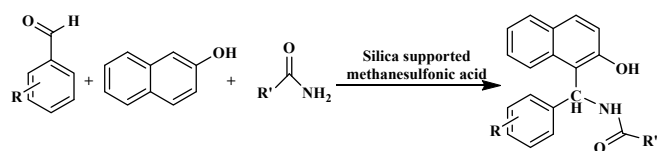
Scheme 49.

Luo and Zhang [101] introduced the synthesis of 1-amidoalkyl-2-naphthols catalyzed by PEG-based dicationic acidic ionic liquids under solvent-free conditions (Scheme 50).



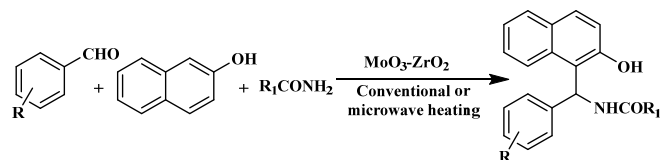
Scheme 50.

Wang *et al.* [102] introduced a simple and an efficient method for the synthesis of amidoalkyl naphthols in the presence of silica supported methanesulfonic acid as a reusable heterogeneous catalyst (Scheme 51).



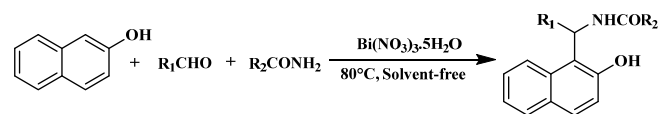
Scheme 51.

Samantaray *et al.* [103] introduced the novel application of MoO₃-ZrO₂ composite oxides for the synthesis of amidoalkyl naphthols. The series of xMoZr catalysts were prepared by coprecipitation and impregnation methods and characterized by XRD, Raman, UV-Vis, TEM and sorptometric techniques. The MoO₃ species are present predominantly in the form of isolated and cluster molybdate on zirconia surface for the coprecipitated samples, whereas the impregnated sample shows the presence of bulk type MoO₃. The MoO₃(20 mol%)-ZrO₂ material was used as efficient catalyst for synthesis of amidoalkyl naphthols under solvent free conditions using conventional as well as microwave heating. The results obtained clearly showed that the composite oxide catalyst was recyclable and highly efficient for the reaction giving good yield and purity of the products (Scheme 52).



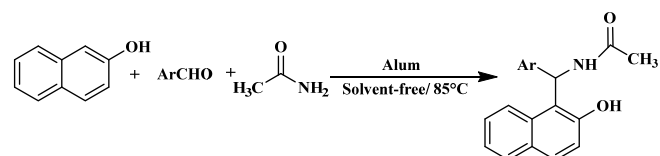
Scheme 52.

Wang *et al.* [104] reported bismuth (III) nitrate pentahydrate catalyzed the three-component condensation of β-naphthol, aldehydes and amines/urea under solvent-free conditions to afford the corresponding amidoalkyl naphthols in excellent yields (Scheme 53).



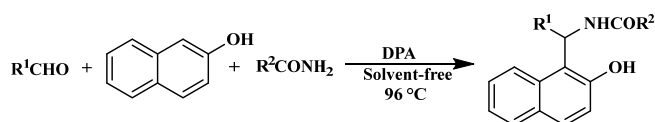
Scheme 53.

Oskooie *et al.* [105] introduced the simple practical procedure catalyzed by KAl(SO₄)₂·12H₂O for the synthesis of amidoalkyl naphthols in good yields and short reaction times (Scheme 54).



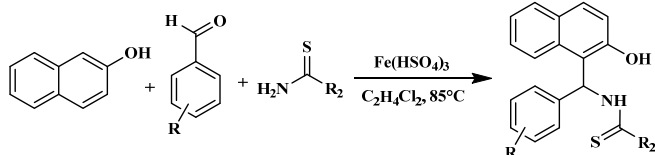
Scheme 54.

Zandi *et al.* [106] reported the green protocol for the synthesis of 1-amidoalkyl-2-naphthols employing a multi-component, one-pot condensation reaction between β-naphthol, aromatic or aliphatic aldehydes and benzamide or acetamide in the presence of dodecylphosphonic acid as catalyst under solvent-free conditions (Scheme 55).



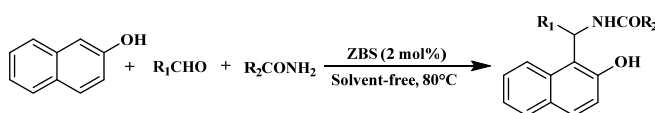
Scheme 55.

Eshghi *et al.* [107] reported the new approach for the synthesis of thioamidoalkyl naphthols which were synthesized by multi-component reaction of β -naphthol, aromatic aldehydes and thioamide derivatives in the presence of ferric hydrogensulphate [$\text{Fe}(\text{HSO}_4)_3$] as a suitable heterogeneous catalyst in good yields (Scheme 56).



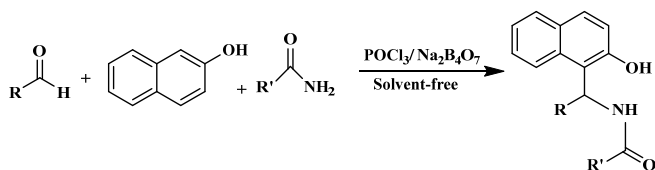
Scheme 56.

Wang and Song [108] introduced zinc benzenesulfonate (ZBS) catalyzed condensation reaction of amidoalkyl naphthols at 80°C in good yields under solvent-free conditions. After the reaction, the catalyst can be recycled (Scheme 57).



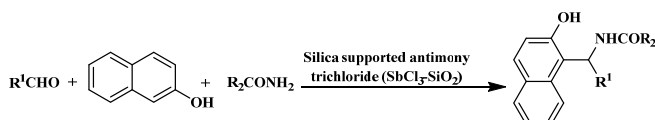
Scheme 57.

Hadi and Hasan [109] introduced convenient and efficient procedure for synthesis of 1-amidoalkyl-2-naphthols by reacting a mixture of β -naphthol, aldehydes and amide/urea in the presence of $\text{POCl}_3/\text{Na}_2\text{B}_4\text{O}_7$ as catalyst. This method offers several advantages such as low cost and easy availability of the catalyst, environmentally friendly procedure and easy work-up under solvent-free conditions (Scheme 58).



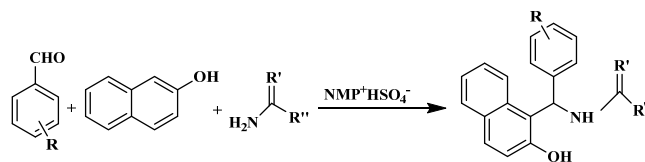
Scheme 58.

Moeinpour *et al.* [110] introduced a new approach for synthesis of 1-amidoalkyl-2-naphthols catalyzed by silica-supported antimony trichloride ($\text{SbCl}_3\text{-SiO}_2$) which was found to be an inexpensive, effective and environmentally benign and gives well to excellent yields (Scheme 59).



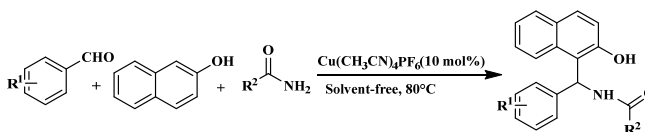
Scheme 59.

Deshmukh *et al.* [111] introduced a novel and green approach which has been described for the synthesis of 1-amidoalkyl-2-naphthols and 1-carbamatoalkyl-2-naphthols using *N*-methyl-2-pyrrolidone hydrogen sulfate ($\text{NMP}^+\text{HSO}_4^-$) as a reusable Brønsted acidic ionic liquid under solvent-free conditions. This method offers several advantages such as excellent yields, short reaction times, easy work-up procedure and reusable catalyst (Scheme 60).



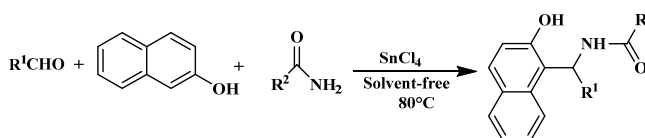
Scheme 60.

Soleimani and Zainali [112] introduced a new approach for synthesis of 1-amidoalkyl-2-naphthols which was carried out by condensation reaction of β -naphthol, aromatic aldehydes, and amides (acetamide, benzamide, and urea) in the presence of tetrakis (acetonitrile) copper(I) hexafluorophosphate [$\text{Cu}(\text{CH}_3\text{CN})_4\text{PF}_6$] under solvent-free conditions (Scheme 61).



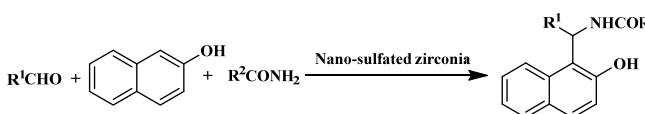
Scheme 61.

Wang *et al.* [113] introduced the reactions which were carried out at 80°C with one-pot three-component condensation of 2-naphthol, aldehydes and amides/urea in the presence of tin tetrachloride as catalyst to give the desired 1-amidoalkyl-2-naphthols in short reaction times (0.1-4h) and excellent yields (45-97%) (Scheme 62).



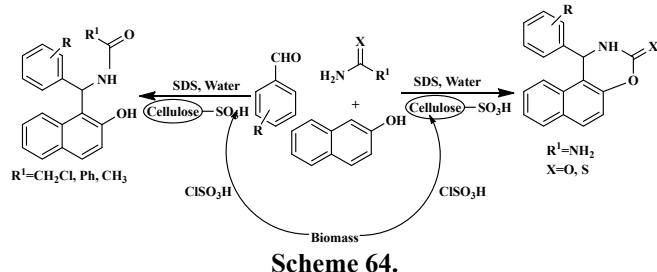
Scheme 62.

Zali and Shokrolahi [114] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using nano-sulfated zirconia as a catalyst under solvent-free conditions. The catalyst is recycled for five consecutive times without loss of activity (Scheme 63).

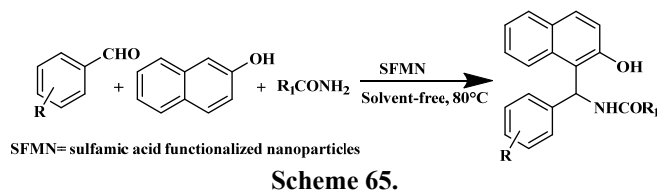


Scheme 63.

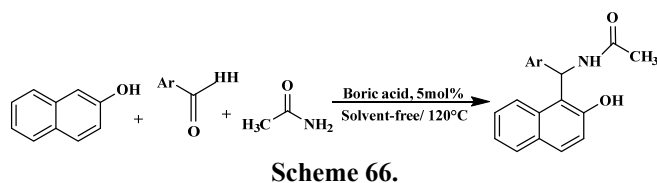
Kumar *et al.* [115] introduced micelle promoted natural carbohydrate scaffold catalyzed synthesis of 1,2-dihydro-1-aryl-3H-naphth[1,2-e][1,3]oxazin-3-one and amidoalkyl-naphthol derivatives have been developed *via* a multicomponent one pot reaction of 2-naphthol / 1-naphthol, aromatic / heteroaromatic aldehyde and urea / thiourea / amide in water. The advantages of this method are efficient catalysis, excellent cost effectiveness, simple work-up and recyclability of catalyst (Scheme 64).



Yarahmadi and Shaterian [116] synthesized sulfamic acid functionalized magnetic nanoparticles (SFMN) and have been used as efficient heterogeneous solid acid catalysts for the condensation of aromatic aldehydes with 2-naphthol and amides (or carbamates) *via* three-component reactions under solvent-free conditions at 80°C catalyzed an efficient synthesis of various amidoalkyl naphthols from β -naphthol, aromatic aldehydes and urea in good to high yields under solvent-free conditions (Scheme 65).

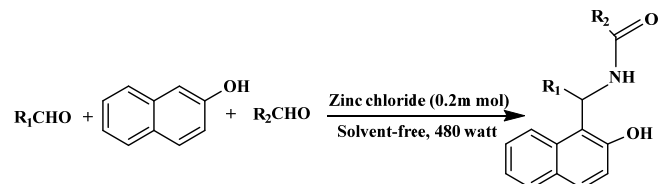


Shahrissa *et al.* [117] introduced a new approach for synthesis of 1-amidoalkyl-2-naphthols by multi-component reaction of 2-naphthol, aromatic aldehyde and acetamide catalyzed by boric acid as solvent-free conditions having temperature 120°C in oil bath. Boric acid has been used in organic synthesis because it is commercially available, environmentally benign, cheap, stable, and easy to handle (Scheme 66).

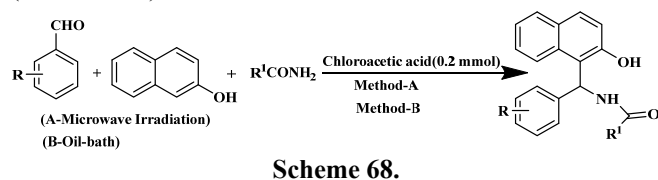


Malik *et al.* [118] reported the new approach for the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehyde, 2-naphthol and amido/urea using anhydrous zinc chloride as a heterogeneous catalyst under microwave irradiation.

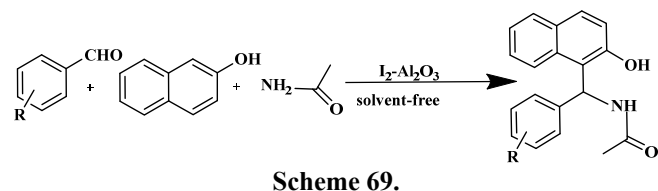
Anhydrous zinc chloride is highly efficient and recyclable (Scheme 67).



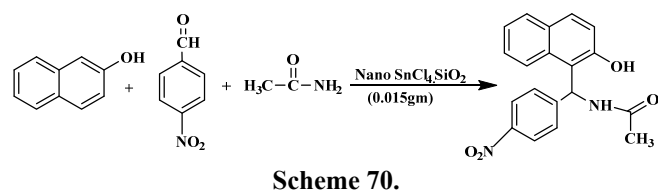
Duvedi and Singh [119] synthesized 1-amidoalkyl-2-naphthols by *via* the one-pot multi-components condensation of 2-naphthol, aromatic aldehydes and amides using chloroacetic acid as catalyst in the absence of solvent media under microwave (method A) and thermal techniques (method B). This greener protocol offer many advantages such as shorter reaction time, simple work-up and excellent yield (Scheme 68).



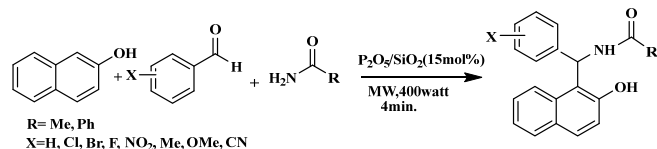
Ahad *et al.* [120] introduced a new approach for the preparation of 1-amidoalkyl-2-naphthols by *via* the one-pot multi-components condensation of 2-naphthol, aromatic aldehydes and amides using I₂-Al₂O₃ as catalyst under solvent-free conditions. I₂-Al₂O₃ is efficient catalyst with high catalytic activity and short reaction time (Scheme 69).



Mirjalili *et al.* [121] introduced a new approach for the preparation of 1-Amidoalkyl-2-naphthols by *via* the one-pot multicomponents condensation of 2-naphthol, aromatic aldehydes and amides using nano SnCl₄-SiO₂ as catalyst under solvent-free conditions. Nano silica supported tin chloride as an efficient acidic catalyst prepared *via* a reaction of nano-SiO₂ with SnCl₄ at room temperature. The structure of catalyst evaluated by XRD, TEM and SEM. The advantages of this protocol is short reaction time, high yield, scale up and easy work up (Scheme 70).

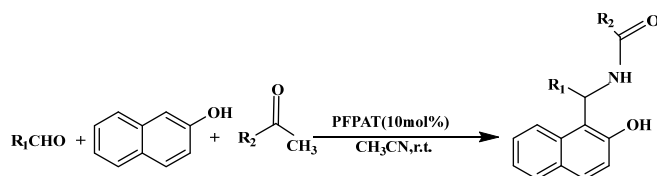


Zarei [122] introduced a novel and green approach which has been described for the synthesis of 1-amidoalkyl-2-naphthols from simple, one-pot, three component reaction between aldehydes, 2-naphthol and amides or ureas using phosphorus pentoxide as a catalyst under solvent free condition. This method offers several advantages such as high yields, simple and clean work-up, inexpensive material and short reaction time (Scheme 71).



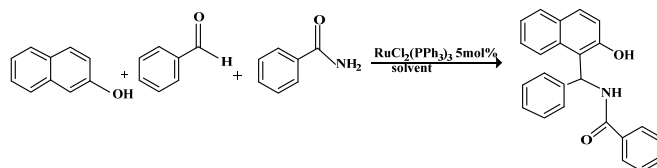
Scheme 71.

Khaksar *et al.* [123] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using pentafluorophenylammonium triflate (PFPAT) as a new organocatalyst under solvent-free conditions. PFPAT as an efficient, non-volatile, noncorrosive recyclable, air-stable, cost-effective, easy to handle and easily removed from the reaction mixtures (Scheme 72).



Scheme 72.

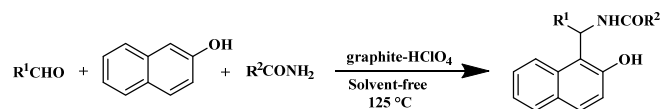
Zhu *et al.* [124] described the synthesis of 1-amidoalkyl-2-naphthols which was carried out by condensation reaction of β -naphthol, aromatic aldehydes, and amides (acetamide, benzamide, and urea) in the presence $\text{RuCl}_2(\text{PPh}_3)_3$ as a catalyst with solvent media. This catalyst tris (triphenylphosphine) ruthenium(II) dichloride is a viable alternative and may be a promising catalyst for the synthesis of 1-amidoalkyl-2-naphthols due to its easy availability, sustainability, non-toxicity, and environmentally friendly properties (Scheme 73).



Scheme 73.

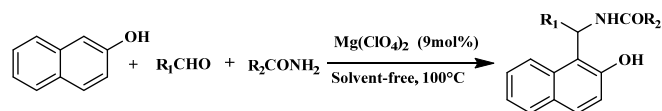
Lei *et al.* [125] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using graphite supported perchloric acid ($\text{HClO}_4\text{-C}$) as a new heterogeneous catalyst under solvent-free conditions. Silica-supported perchloric

acid ($\text{HClO}_4\text{-SiO}_2$) has been used as a recyclable heterogeneous catalyst and the graphite-supported catalysts show a certain catalytic activity in chemical reactions. Simple work-up, shorter reaction times and higher product yields, and the catalyst reactivity and can be recycled are the advantages of this protocol (Scheme 74).



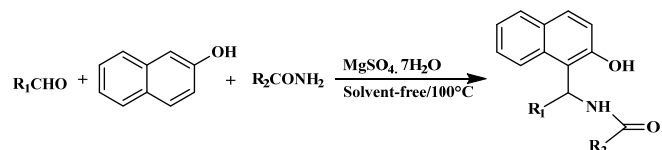
Scheme 74.

Amrollahi *et al.* [126] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using $\text{Mg}(\text{ClO}_4)_2$ as a catalyst under solvent-free conditions. Magnesium perchlorate ($\text{Mg}(\text{ClO}_4)_2$) is more effective electrophilic activation agent and can accelerate the overall reaction rate. $\text{Mg}(\text{ClO}_4)_2$ is an inexpensive solid acid catalyst which can be handled easily and removed from the reaction mixtures by simple filtration. This method offers several advantages such as environmentally benign reaction condition, easy purification and economic availability of the catalyst. (Scheme 75)



Scheme 75.

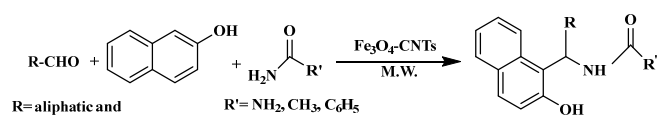
Ashalu and Rao [127] introduced the reactions which were carried out at 100°C with one-pot three-component condensation of 2-naphthol, aldehydes and amides/urea in the presence of eco-friendly anhydrous magnesium sulfate ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$) as catalyst to give the desired 1-amidoalkyl-2-naphthols in short reaction time (0.5-1h) and excellent yield (72-94%). Anhydrous magnesium sulfate acts as a Lewis acid catalyst for transformation in this synthesis (Scheme 76).



Scheme 76.

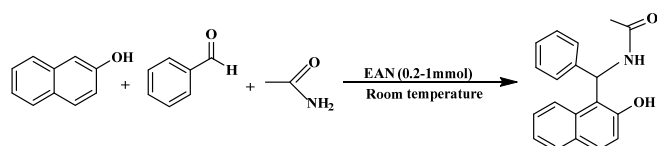
Safari and Zarnegar [128] describe a practical and effective method for the preparation of 1-amidoalkyl-2-naphthol *via* the one-pot three-component reaction of β -naphthol, aldehydes derivatives, amides or urea in the presence of magnetically $\text{Fe}_3\text{O}_4\text{-CNTs}$ as a heterogeneous catalyst under solvent-free conditions and microwave-irradiation. Carbon nanotubes (CNTs) consider as catalytic support of catalytic reaction intriguing nanoscale dimensions, uniform nano-structure, electric properties, high specific surface area,

thermal and chemical stability. The structure of catalyst evaluated by XRD, TEM, SEM. and EDS. The notable advantages of the present method are mild, solvent-free conditions, ecofriendly catalyst, ease of product isolation, and recyclability of the magnetic catalyst (Scheme 77).



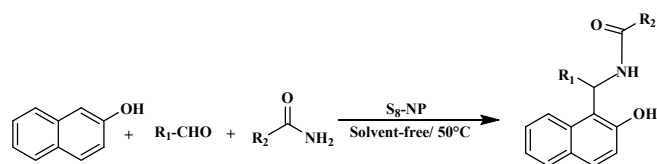
Scheme 77.

Mulla *et al.* [129] described a practical and effective method for the preparation of 1-amidoalkyl-2-naphthol *via* the one-pot three-component reaction of β -naphthol, aldehydes derivatives, amides or urea in the presence of ethylammonium nitrate (EAN) as ionic liquid catalyst under solvent-free conditions at room temperature. EAN recovered and recycled several times without loss of catalytic activity. Simple work-up, shorter reaction times and higher product yields, and the can be recycled are the advantages of this protocol (Scheme 78).



Scheme 78.

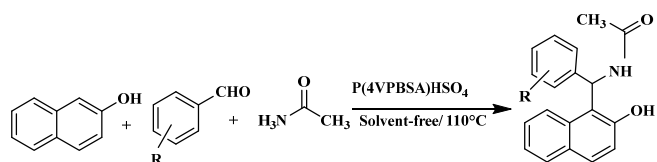
Das *et al.* [130] introduced a new approach for synthesis of 1-amidoalkyl-2-naphthols which was carried out by condensation reaction of β -naphthol, aromatic aldehydes, and amides (acetamide, benzamide, and urea) in the presence of piper-bettle-shaped nano-S as a catalyst under solvent-free media. Nano-S prepared by annealing process showed excellent catalytic activity. The structure of nano-S confirmed by the XRD, TEM, EDX and TGA. Potent, benign, highly active, cheaper and reusable are the advantages of nano-S used in the synthesis of amidoalkyl-2-naphthols (Scheme 79).



Scheme 79.

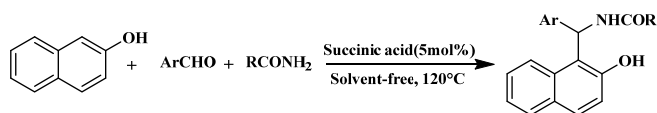
Kaisat *et al.* [131] used poly(vinylpyridinium butane sulfonic acid) hydrogen sulfate P(4VPBSA)HSO₄ as an efficient dual acidic catalyst in the synthesis of 1-amidoalkyl-2-naphthols which was carried out by condensation of β -naphthol, aromatic aldehyde and amides under solvent-free conditions. The catalyst was characterized by Fourier transform infrared spectroscopy, scanning electron microscopy, thermogravimetric analysis. This method offers several

advantages like high yield, recyclable catalyst, short reaction time, facile work-up and ability to perform the reaction under solvent-free conditions (Scheme 80).



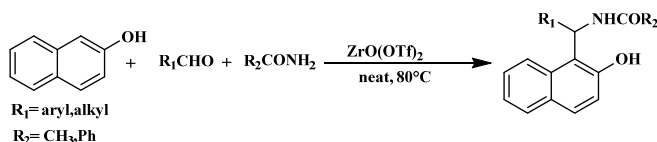
Scheme 80.

Hazeri *et al.* [132] described the green and efficient synthesis of 1-amidoalkyl-2-naphthols using succinic acid as a catalyst for the three-component reaction of 2-naphthol, aldehyde and amide/urea under solvent-free conditions. Succinic acid is readily available, low cost reagent; it can conveniently be handled, and removed from the reaction mixture. This protocol has the advantages of shorter reaction times, low cost, excellent yield, and environmentally friendly, economical, easy work-up and performs the reaction under solvent-free conditions (Scheme 81).



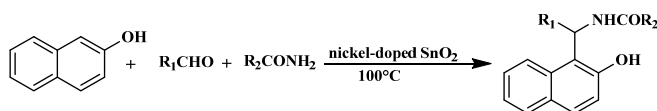
Scheme 81.

Hashemi and Sardarian [133] described the new approach for the preparation of 1-amidoalkyl-2-naphthols *via* one-pot three component condensation of amides, aldehydes, and β -naphthols in the presence of zirconyl triflate ZrO(OTf)₂ as a highly efficient, low toxic, stable and non-hygroscopic catalyst under solvent-free conditions. Low cost procedure, short reaction time, excellent yield and ability to perform the reaction under solvent-free conditions are the advantages of this protocol (Scheme 82).



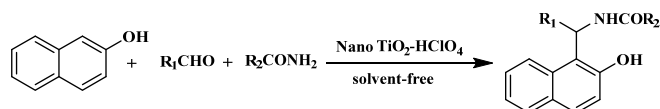
Scheme 82.

Dehbashi *et al.* [134] described the preparation of 1-amidoalkyl-2-naphthols *via* one-pot three component condensation of amides, aldehydes, and β -naphthols in the presence nickel doped tin oxide nanoparticle (Ni-doped SnO) as a catalyst. The size of nano nickel doped tin oxide conformed by the XRD, TEM, XRF and FT-IR. This catalyst offers the High yield, and can be recovered and recyclable several times without, loss of activity (Scheme 83).



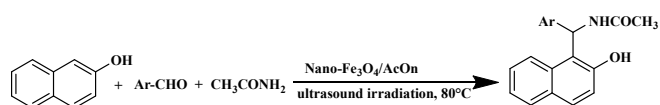
Scheme 83.

Shaterian and Mohammadian [135] described the new approach for the preparation of 1-amidoalkyl-2-naphthols *via* one-pot three component condensation of amides, aldehydes, and β -naphthols in the presence perchloric acid coated tin oxide nanoparticles as an acidic heterogenous catalyst in solvent-free conditions. The size of the catalyst conformed by the XRD, TEM, XRF and FT-IR. This protocol offers the high yield, and can be recovered and recyclable several times without, loss of activity (Scheme 84).



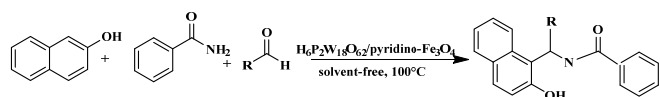
Scheme 84.

Mokhtary and Torabi [136] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using nano-magnetite Fe_3O_4 as an efficient catalyst under ultrasound irradiation. High yields, clean methodologies, easy workup procedure, recoverable easily are the advantages of this protocol (Scheme 85).



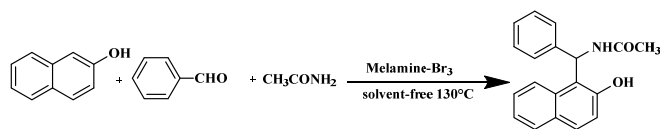
Scheme 85.

Tayebee *et al.* [137] introduced a new approach for synthesis of 1-amidoalkyl-2-naphthols which was carried out by condensation reaction of β -naphthol, aromatic aldehydes, and amides in the presence of magnetic inorganic-organic nano hybrid material $\text{H}_6\text{P}_2\text{W}_{18}\text{O}_{62}/\text{pyridino-Fe}_3\text{O}_4$ as a catalyst under solvent-free media. This protocol offers high yield, highly recyclable, efficient, eco-friendly catalyst for the solvent-free, one-pot, and multi-component synthesis of 1-amidoalkyl-2-naphthols (Scheme 86).



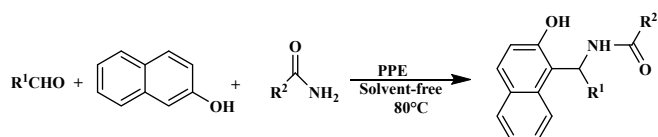
Scheme 86.

Choghamarani and Rashidmoghadam [138] introduced the one-pot three-component condensation of 2-naphthol, aromatic aldehydes and acetamide in the presence of tribromo-melamine as a homogenous and non-hygroscopic solid catalyst to give the desired 1-amidoalkyl-2-naphthols. There are several advantages to this protocol, including high yield, short reaction time, and high catalytic efficiency (Scheme 87).



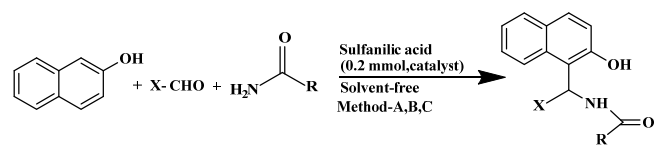
Scheme 87.

Moghanian and Ebrahimi [139] introduced a new approach for the preparation of 1-Amidoalkyl-2-naphthols by a *via* the one-pot multi-components condensation of 2-naphthol, aromatic aldehydes and amides or urea using polyphosphate ester (PPE) as catalyst under solvent-free conditions. The advantages of this protocol are short reaction time, high yield, scale up, easy work-up and elimination of solvents and toxic catalysts (Scheme 88).



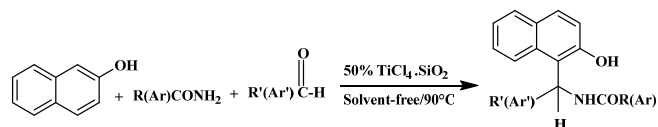
Scheme 88.

Singh *et al.* [140] introduced the one-pot three-component condensation of 2-naphthol, aromatic aldehydes and acetamide in the presence of *p*-aminobenzene sulfonic acid (sulfanilic acid) as a organoacid catalyst to give the desired 1-amidoalkyl-2-naphthols under thermal (oil bath and hot plate) and microwave techniques. There are several advantages to this procedure including high yield, short reaction time, high catalytic efficiency and ability to perform the reaction under solvent free conditions (Scheme 89).



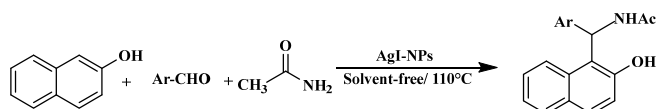
Scheme 89.

Zamani *et al.* [141] introduced Nano- $\text{TiCl}_4.\text{SiO}_2$ to be an extremely efficient catalyst for the preparation of 1-amidoalkyl-2-naphthols from various aldehyde and amides under mild conditions in solvent-free media. The structure and size of nano- $\text{TiCl}_4.\text{SiO}_2$ as catalyst conformed by the XRD, TEM, SEM, TGA and UV-IR. This protocol offers high yields, simple methodology and easy work-up (Scheme 90.)



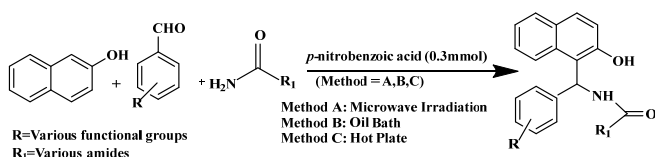
Scheme 90.

Ghomi *et al.* [142] introduced the one-pot three-component condensation of 2-naphthol, aromatic aldehydes and acetamide in the presence of silver iodide (AgI) nanoparticles as effectual heterogeneous catalyst to give the desired 1-amidoalkyl-2-naphthols under mild conditions. There are several advantages to this protocol including high yield, short reaction time, moderate temperature, eco-friendly reaction conditions, simple purification and good reusability of catalyst (Scheme 91).



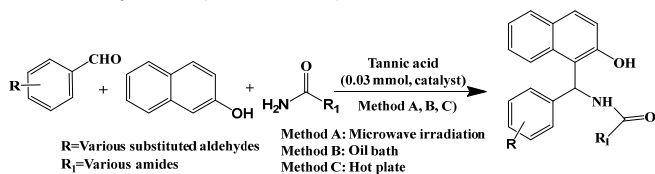
Scheme 91.

Li *et al.* [143] carried out the synthesis of 1-amidoalkyl-2-naphthols by condensing the reaction mixture of aromatic aldehydes, 2-naphthol and amide/urea using *p*-nitrobenzoic acid as catalyst by thermal and microwave methods. The *p*-nitrobenzoic acid is readily available, inexpensive reagents and can conveniently be handled and removed from the reaction mixture. There are several advantages to this procedure including high yield, high catalytic efficiency and ability to perform the reaction under solvent free conditions (Scheme 92)



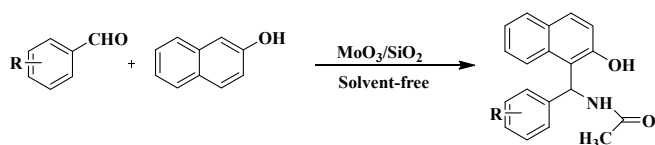
Scheme 92.

Singh and Duvedi [144] investigated the catalytic potential of tannic acid for an efficient synthesis of amidoalkyl naphthols. The β -naphthol were condensed with substituted aromatic aldehydes and various amides using catalytic amount of tannic acid (3.0 mol%) in absence of solvent under thermal (hot plate and oil bath) and microwave irradiation techniques. Tannic acid was found to be novel, cheap and biodegradable catalyst. This green protocol offered many advantages such as short reaction time, use of environment-friendly and cheap catalyst and good to excellent yields (Scheme 93).



Scheme 93.

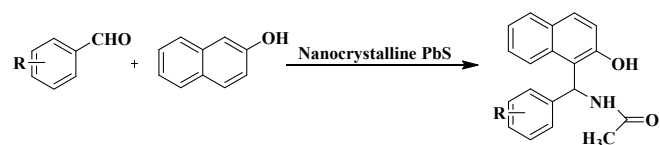
Moeinpour *et al.* [145] introduced the catalytic activity of MoO₃/SiO₂ as efficient catalyst for the synthesis of amidoalkyl-naphthols under solvent-free conditions with high yields upto 93 % and other advantages such as short reaction times, low cost of catalyst and mild reaction conditions (Scheme 94).



Scheme 94.

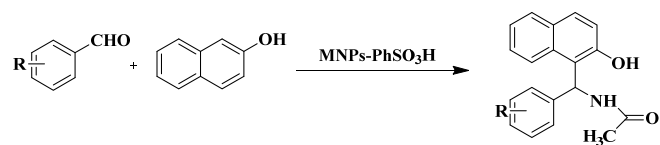
Borhade *et al.* [146] synthesized amido-alkyl naphthols under solvent-free conditions with yield of 85-95 %

using recyclable nanocrystalline lead sulphide (PBS) catalyst after characterizing it by XRD, TEM and adsorption/desorption analysis (BET) (Scheme 95).



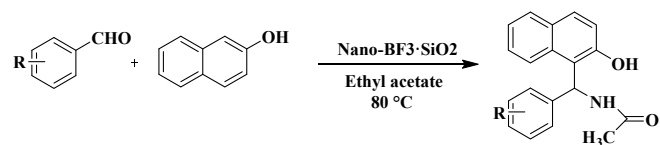
Scheme 95.

Moghaniai *et al.* [147] synthesized sulfanilic acid-functionalized silica-coated nano-Fe₃O₄ particles (MNPs-PhSO₃H) as a heterogeneous and reusable catalyst and studied its catalytic performance by synthesizing 1-amido and 1-aminoalkyl-2-naphthols *via* a one-pot three-component condensation reaction of aldehydes, 2-naphthol, and amides/urea/amines in good to excellent yields, under solvent-free classical heating conditions. The catalyst was easy to separate by applying external magnetic field and reused for several consecutive runs without significant loss of its catalytic efficiency. The catalyst was characterized by transmission electron microscopy (TEM), scanning electron microscope (SEM), powder X-ray diffraction (XRD) and sample magnetometry (VSM) and results showed that the synthesized magnetic nanocomposites are superparamagnetic with a size range of 15–30 nm (Scheme 96).



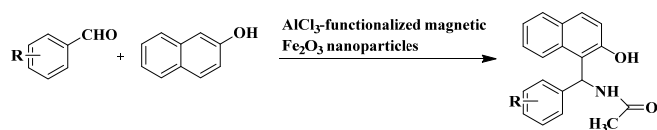
Scheme 96.

Mirjalili *et al.* [148] synthesized the 1-amidoalkyl-2-naphthols *via* one-pot multi-component reaction of 2-naphthol, aldehydes, and amides or urethane in the presence of nano-silica supported boron trifluoride (nano-BF₃·SiO₂) in ethyl acetate at 80°C. This method offered significant advantages, such as high conversions, easy handling, cleaner reaction profile and appropriate reaction times which make it a useful and attractive process for the rapid synthesis of substituted 1-amidoalkyl-2-naphthols (Scheme 97).



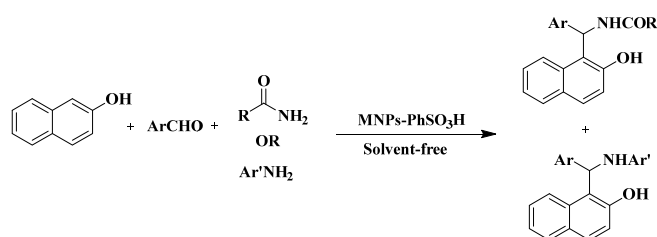
Scheme 97.

Ghader and Kassae [149] synthesized AlCl₃-functionalized magnetic Fe₂O₃ nanoparticles and used it for the cost effective and efficient synthesis of amidoalkyl-naphthols. The catalyst was easy to remove by an external magnet and reused. It was characterized by SEM, XRD, ICP, TGA and VSM (Scheme 98).



Scheme 98.

Kiyani *et al.* [150] described the catalytic activity of 2-hydroxy-5-sulfobenzoic acid as recyclable organocatalyst for an efficient synthesis of 1-amidoalkyl-naphthols (10 mol% at 100 °C) and 3,4-disubstituted isoxazol-5 (4*H*)-ones (15 mol% at room temperature) with the yields of 82-97 %. The reaction was simple and completed within 3–25 min (for amidoalkyl naphthols), and 70–120 min (for 3,4-disubstituted isoxazol-5(4*H*)-ones). This procedure offers the advantages of convenience, simple operational procedure, cost-effectiveness, no use of hazardous organic solvents, and the commercial availability of the catalyst (Scheme 99).



Scheme 99.

3. Mechanism of Reaction and Role of Catalyst

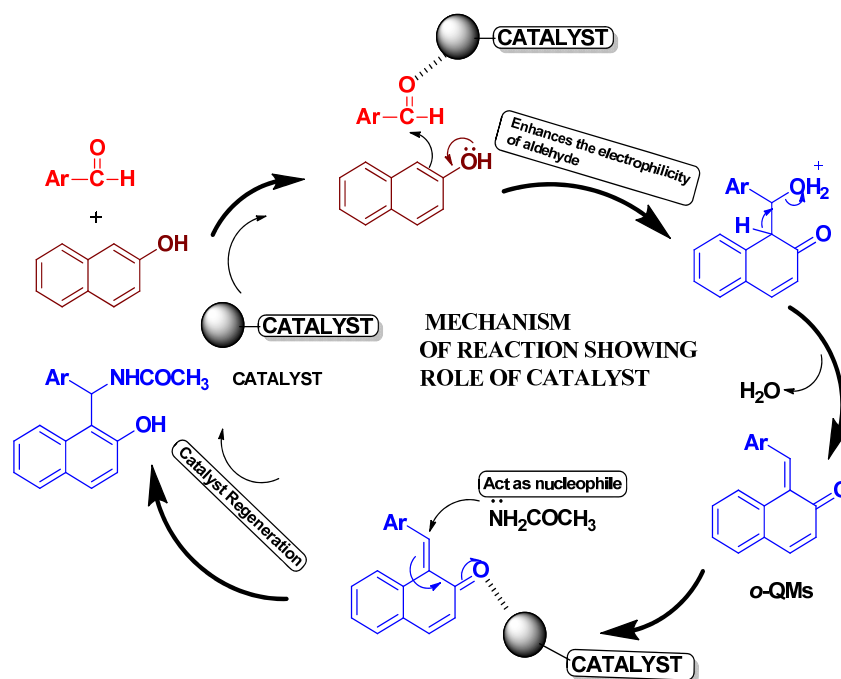
Mechanism for the synthesis of 1-amidoalkyl-2-naphthols is shown in Scheme 100. In general, the any

type of catalyst whether it is Lewis acid, ionic liquids or polymer supported catalysts, have been proposed to activate the aldehyde by coordinating to oxygen atom which ultimately enhances the electrophilicity of the aldehyde and leads to reduction in reaction time [61-63].

The condensation of 2-naphthol with the activated aldehyde give *ortho*-quinone methides (*o*-QMs) as a highly reactive and ephemeral intermediate. The same *o*-QMs, generated *in-situ*, have been reacted with amide under the influence of catalyst to form 1-amidoalkyl-2-naphthol derivatives. Thus, the 2-naphthol act as Michael acceptors and arylaldehydes as nucleophile resulting in a Michael adduct under the influence of catalyst. The electron withdrawing groups (EWD) substituted on benzaldehyde in *o*-QM intermediate increase the rate of 1, 4-nucleophilic addition reaction because of alkene LUMO is at lower energy in the neighboring withdrawing groups than electron donating groups (EDG). Hence the benzene ring with electron withdrawing groups gave better yield as compared to electron donating groups.

4. Conclusion

1-Amidoalkyl-2-naphthols are the vital synthetic building blocks and used as intermediates for the synthesis of many important derivatives such as 1,3-oxazine derivatives and 1-aminomethyl-2-naphthol derivatives which are widely used class of bioactive compounds due to their remarkable biological and many pharmacological properties.



Scheme 100. Mechanism of action showing the role of catalyst for the synthesis of 1-amidoalkyl-2-naphthols.

Because of their important therapeutic value, better search of reagent, catalysts and routes are continually being developed. In this review, we have focused on green catalytic approaches for an efficient synthesis amidoalkyl naphthols. Our knowledge of exploring novel catalysts should aid in future research to synthesize various other synthetic intermediates.

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