Tetrahedron Letters 53 (2012) 4687–4691

Contents lists available at SciVerse ScienceDirect

Tetrahedron Letters

journal homepage: www.elsevier.com/locate/tetlet

Nano crystalline ZnO catalyzed one pot multicomponent reaction for an easy access of fully decorated 4*H*-pyran scaffolds and its rearrangement to 2-pyridone nucleus in aqueous media

Pranabes Bhattacharyya, Koyel Pradhan, Sanjay Paul, Asish R. Das *

Department of Chemistry, University of Calcutta, Kolkata 700009, India

ARTICLE INFO

Article history:
Received 29 May 2012
Revised 14 June 2012
Accepted 16 June 2012
Available online 29 June 2012

Keywords:
Nano structured ZnO
4*H*-Pyrans
3,4-Dihydropyridin-2-one
Organic acid catalyzed rearrangement

ABSTRACT

A green and highly efficient protocol has been developed for the synthesis of 4*H*-pyran scaffolds installing a one-pot three-component coupling reaction of an aldehyde, malononitrile, and a 1,3-diketo compound using nano structured ZnO as the catalyst in aqueous alcoholic medium. A greener method to synthesize 3,4-dihydropyridin-2-one has also been developed by rearranging 4*H*-pyran derivatives in aqueous medium applying *p*-TSOH as the right catalyst source. A wide spectrum of functional groups was tolerated in both the developed synthetic protocols with good to excellent yield of the targeted molecules.

Polyfunctionalized 4*H*-pyran derivatives are important to the synthetic chemists due to their pronounced biological and pharmacological activities.[1] These compounds are used as anti-coagulants, anticancer agents, spasmolytics, anti-anaphylactics, etc.[2,3] In addition,4*H*-pyrans containing heterocyclic rings (compound **I**, Fig. 1) are increasingly used for their pharmacological activities.[4] Furthermore, a number of 2-amino-4*H*-pyran derivatives are useful as photoactive materials.[5] Generally, 3-substituted-6-amino-4-aryl-5-cyano-2-methyl-4*H*-pyrans are prepared from arylidenemalononitriles and activated methylene compounds in the presence of organic bases.[6] It is also necessary to mention that the synthesis of 2-pyridone nucleus is a challenge to the synthetic and medicinal chemists as its derivatives have important biological and pharmacological activities.[7–9] It has been shown that 3,4-dihydropyridin-2-one derivative possesses HIV-1 specific reverse transcriptase inhibition capability.[10] Compounds **II** and **III** (Fig. 1) have been found to show hypolipidemic and **5a**-reductase inhibitory activities, respectively. In view of the immense importance of 4*H*-pyran and 2-pyridone derivatives there is renewed interest in developing new methodologies for the synthesis of these compounds.

Quite a good number of methods have been already reported in the literature for the synthesis of 6-amino pyran derivatives. Reagents, such as TMG-[bmim][X],[11] tetrabutylammonium bromide,[12] rare earth perfluorooctanoates,[13] hexadecyltrimethylammonium bromide,[14] silica nanoparticles,[15] and nano metal oxide/ionic liquid combocatalyst[16] have been already employed to achieve the synthesis of 6-amino pyran derivatives. But, so far only a few methods are available for the synthesis of 2-pyridone derivatives.[17] Although these methods have their own merits, still these classical reactions have significant limitations like harsh reaction conditions, use of mineral acids,[17] low yields, tedious work-up procedure. Thus, it is desirable to design an efficient and convenient method to construct such heterocyclic molecules.

In continuation of our interest in developing methodologies for carbon–carbon bond formation using nano metal oxide as catalyst[18,19] herein we disclose an efficient and high yielding protocol for the synthesis of 4*H*-pyran derivatives starting from aldehyde,

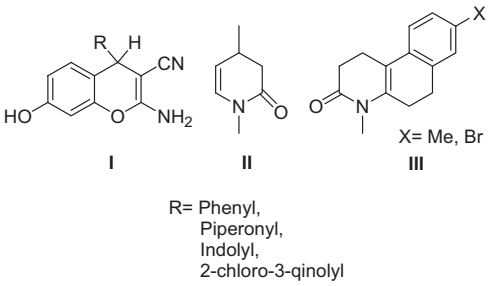
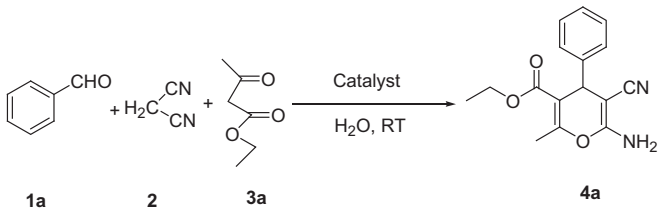


Figure 1. Biologically active 2-amino pyran and 3,4-dihydropyridin-2-one derivatives.

* Corresponding author. Tel.: +91 33 23501014/9433120265; fax: +91 3323519754.
E-mail addresses: ardas66@rediffmail.com, ardchem@caluniv.ac.in (A.R. Das).

0040-4039/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.tetlet.2012.06.086



Scheme 1. Synthesis of 6-amino pyran derivative via a three component coupling of benzaldehyde (**1a**), malononitrile (**2**), and ethylacetoacetate (**3a**).

Table 1
Optimization of reaction condition using different catalysts[a]

Entry	Catalyst	Time (h)	Yield[b] (%)
1	—	24	Trace
2	MgO (10 mol %)	6	32
3	CaO (10 mol %)	6	36
4	Nano Al_2O_3 (10 mol %)	6	42
5	Nano SiO_2 (10 mol %)	6	46
6	L-Proline (10 mol %)	6	58
7	Bu_4NBr (10 mol %)	6	52
8	Bulk ZnO (10 mol %)	3	68
9	Nano ZnO (10 mol %)	3	84

[a] Benzaldehyde (**1a**) (1 mmol), malononitrile (**2**) (1 mmol), and ethylacetoacetate (**3a**) (1 mmol) were stirred in 5 mL water at room temperature in the presence of 10 mol % of the catalyst.
[b] Isolated yield of the pure product.

malononitrile, and 1,3-diketone in aqueous alcoholic medium using nano crystalline ZnO as the catalyst. As 4*H*-pyran nucleus is very much sensitive to aqueous acid due to its cyclic enol-ether ligation, we also demonstrate here the acid-catalyzed rearrangement of 4*H*-pyran leading to the very easy access of 2-pyridone derivative.

Initially, we focused on systematic evaluation of different catalysts for the model reaction of benzaldehyde (**1a**), malononitrile (**2**), and ethylacetoacetate (**3a**) in water at room temperature (Scheme 1). We have applied a wide range of catalysts including MgO, CaO, Nano Al_2O_3, Nano SiO_2, L-proline, Bu_4NBr, Bulk ZnO, and Nano ZnO to improve the yield for the specific synthesis of 2-amino pyran derivatives. As shown in Table 1, the reaction did not take place without any catalyst (Table 1, entry 1). As mentioned in Table 1, most interesting result was obtained with ZnO as the catalyst and the yield of the desired product was maximized when nano crystalline ZnO was used replacing bulk ZnO (Table 1, entries 8 and 9).

We then tried to screen the reaction in various organic solvents in order to optimize the reaction conditions using nano ZnO as catalyst (Table 2). The results revealed that solvents show great effect on the catalytic activity of ZnO. The highest yield was obtained with solvent system water/ethanol (1:1) (Table 2, entry 9).

As nano ZnO had emerged as the most suitable catalyst for the reaction in 1:1 ethanol/water media, we then tried to optimize the catalyst load for the cyclocondensation reaction leading to the rapid formation of 4*H*-pyran nucleus. Our optimization studies revealed that the yield increased smoothly with catalyst load up to 10 mol % and then remained unaltered up to 25 mol % after that there was a sharp drop in the yield (Fig 2). This drop may be attributed to the coagulation of ZnO nano particles which decreased the effective surface area of the catalyst.

We next concentrated on the scope of this reaction with a variety of aldehydes and a series of 1,3-diketo compounds (Scheme 2)

Table 2
Solvent screening for the model reaction[a]

Entry	Solvent	Yield[b] (%)
1	Et_2O	38
2	$CHCl_3$	47
3	CH_2Cl_2	43
4	Toluene	56
5	Acetone	64
6	DMSO	68
7	DMF	72
8	H_2O	84
9	H_2O + ethanol (1:1)	96
10	Ethanol	92

[a] Benzaldehyde (**1a**) (1 mmol), malononitrile (**2**) (1 mmol), and ethylacetoacetate (**3a**) (1 mmol) were stirred in 5 mL solvent in the presence of 10 mol % nano ZnO at room temperature for 3 h.
[b] Isolated yield of the pure product.

to check the viability of this protocol in obtaining a library of substituted 4*H*-pyrans (Table 3).

The optimized methodology[21] tolerated a wide spectrum of aldehydes and dicarbonyl compounds with good to excellent yield of the targeted molecules. The aromatic aldehydes with electron withdrawing groups reacted faster with slightly improved yields than their electron donating counter parts. The method is also applicable to aliphatic aldehydes (Table 3, entry 9) and heterocyclic aldehydes (Table 3, entries 8, 18, 26, and 27). The catalyst can be recycled five times without significant loss of the activity. The reusability of the catalyst was checked for the synthesis of 6-amino-5-cyano-2-methyl-4-phenyl-4*H*-pyran-3-carboxylic acid ethyl ester (Table 3, entry 1). The study revealed that even after five cycles, the catalyst was able to carry out the reaction offering almost same catalytic activity.

As we observed the excellent catalytic activity of nano ZnO (rod like morphology)(characterized by SEM, X-ray diffraction study) (particle sizes are found to lie between 10 and 11 nm, Supplementary data) in this synthesis, we could propose a plausible mechanistic insight of the reaction which involves, consequent Knoevenagel condensation, Michael addition and finally intra molecular ring closure leading to the formation of 4*H*-pyran derivative catalyzed by nano ZnO as presented in Scheme 3. In the first step, the Knoevenagel condensation between aldehyde and malononitrile was catalyzed by amphoteric nano ZnO which during the removal of acidic proton from active methylene group of malononitrile unit acted like a base and during dehydration, it showed its acidic behavior. During ring closure, the catalyst played the key role where ZnO acted as a mild acid and not only minimized the 1,2 dipolar repulsion between the geminal nitrile groups but also activated one of the nitrile groups by polarization (through coordi-

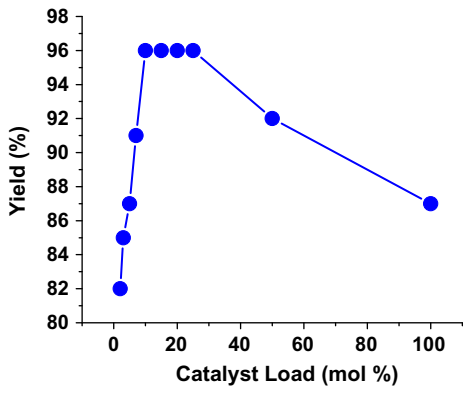
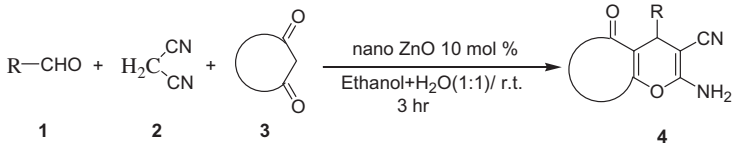


Figure 2. Catalyst load optimization study.

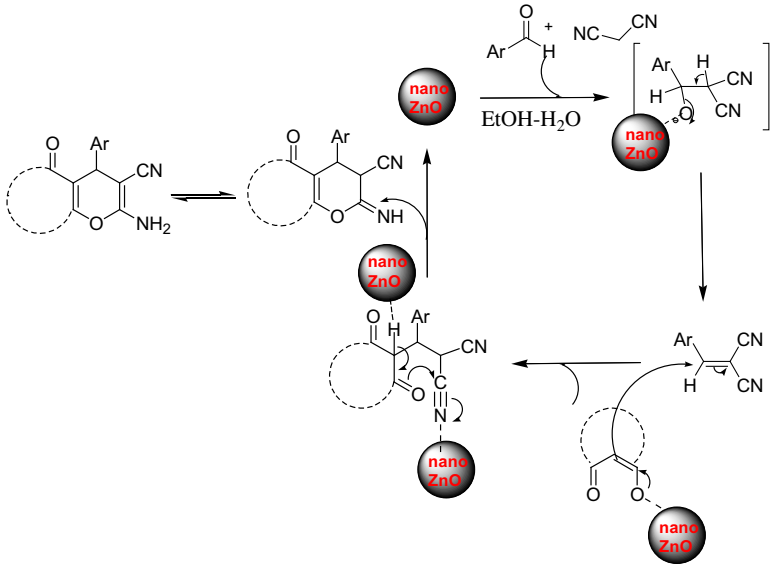


Scheme 2. Synthesis of 4*H*-pyran.

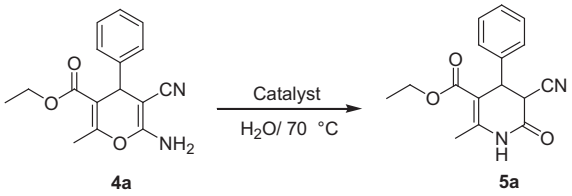
Table 3
Substrate scope[a]

Entry	Product No.	R	1,3-Diketone	Time (h)	Yield[b,c] (%)
1	**4a**	Ph	Ethylacetoacetate	3.0	96,93[d]
2	**4b**	4-NO_2–C_6H_4–	Ethylacetoacetate	2.5	98
3	**4c**	3-NO_2–C_6H_4–	Ethylacetoacetate	3.0	97
4	**4d**	4-F–C_6H_4–	Ethylacetoacetate	2.5	97
5	**4e**	4-Cl–C_6H_4–	Ethylacetoacetate	2.5	96
6	**4f**	4-CH_3–C_6H_4–	Ethylacetoacetate	3.5	92
7	**4g**	4-OCH_3–C_6H_4–	Ethylacetoacetate	4.0	88
8	**4h**	2-Fur	Ethylacetoacetate	2.0	93
9	**4i**	*n*-Propyl	Ethylacetoacetate	3.5	83
10	**4j**	4-OH–C_6H_4–	Ethylacetoacetate	4.0	86
11	**4k**	Ph-	Acetylacetone	3.5	92
12	**4l**	4-NO_2–C_6H_4–	Acetylacetone	2.5	95
13	**4m**	3-NO_2–C_6H_4–	Acetylacetone	3	94
14	**4n**	4-F–C_6H_4–	Acetylacetone	2.5	94
15	**4o**	4-Cl–C_6H_4–	Acetylacetone	3	93
16	**4p**	4-OCH_3–C_6H_4–	Acetylacetone	4.0	88
17	**4q**	4-CH_3–C_6H_4–	Acetylacetone	3.5	90
18	**4r**	2-Fur	Acetylacetone	3.5	89
19	**4s**	Ph–	Dimedone	3.5	86
20	**4t**	4-NO_2–C_6H_4–	Dimedone	3.0	91
21	**4u**	3-NO_2–C_6H_4–	Dimedone	3.5	88
22	**4v**	4-F–C_6H_4–	Dimedone	3.0	90
23	**4w**	4-OCH_3–C_6H_4–	Dimedone	4.0	81
24	**4x**	4-CH_3–C_6H_4–	Dimedone	3.5	83
25	**4y**	4-N$(CH_3)_2$–C_6H_4	Dimedone	4.0	79
26	**4z**	2-Fur	Dimedone	3.0	84
27	**4α**[e]	4-Pyr	Dimedone	4.0	81

[a] Aldehyde (1 mmol), malononitrile (1 mmol), and 1,3-diketone (1 mmol) were stirred in 5 ml 1:1 mixture of ethanol and H_2O in the presence of 10 mol % nano ZnO at room temperature.
[b] Isolated yield of the pure product.
[c] The products were characterized by ^{1}H NMR, ^{13}C NMR, IR, HRMS, and elemental analysis.
[d] Yield obtained after five catalytic cycles.
[e] The structure of the compound (**4α**) was confirmed by X-ray analysis of single crystal (Supplementary data).



Scheme 3. Proposed mechanism of formation of 4*H*-pyrans catalyzed by nano crystalline ZnO.



Scheme 4. Optimization of reaction condition for rearrangement of 4*H*-pyran into 3,4-dihydropyridin-2-one.

Table 4
Catalyst screening and catalyst load optimization[a]

Entry	Catalyst	Catalyst load (mol %)	Yield[b] (%)
1	CH_3COOH	10	—
2	$(COOH)_2$	10	28
3	Citric acid	10	21
4	Tartaric acid	10	24
5	Benzoic acid	10	11
6	Lactic acid	10	17
7	Al_2O_3	10	—
8	$ZnCl_2$	10	—
9	SiO_2	10	—
10	Alum	10	—
11	**p-CH_3–C_6H_4–SO_3H**	**10**	**87**
12	p-CH_3–C_6H_4–SO_3H	15	87

[a] 6-Amino-5-cyano-2-methyl-4-phenyl-4*H*-pyran-3-carboxylic acid ethyl ester (**4a**) (1 mmol), was heated at 70 °C in 5 ml water for 3 h in the presence of specific amount of catalyst.
[b] Isolated yield of **5a**.

Table 5
Solvent screening for the synthesis of 3,4-dihydropyridin-2-one[a]

Entry	Solvent	Time (h)	Yield[b] (%)
1	$CHCl_3$	12	—
2	CCl_4	12	—
3	THF	12	—
4	CH_3CN	12	—
5	Ethanol	6	48
6	DMF	6	52
7	Toluene	6	58
8	Dioxane	6	54
9	**H_2O**	**3**	**87**

[a] 6-Amino-5-cyano-2-methyl-4-phenyl-4*H*-pyran-3-carboxylic acid ethyl ester (**4a**) (1 mmol), was heated at 70 °C in 5 mL solvent for stipulated time in the presence of 10 mol % TsOH.
[b] Isolated yield of the pure product.

nation) and hence facilitated the intramolecular nucleophilic attack by the enolic OH group leading to the easy formation of the final product (4*H*-pyran) and the catalyst entered into the catalytic cycle.

To take an advantage of the elaborated green protocol a little amount of ethanol was added to the reaction mixture and the filtrate on concentrating gave the pure product. This procedure excluded the use of expensive silica gel chromatography, and application of small amount of ethanol instead of chromatographic eluent seemed to be advantageous from the green chemistry perception.

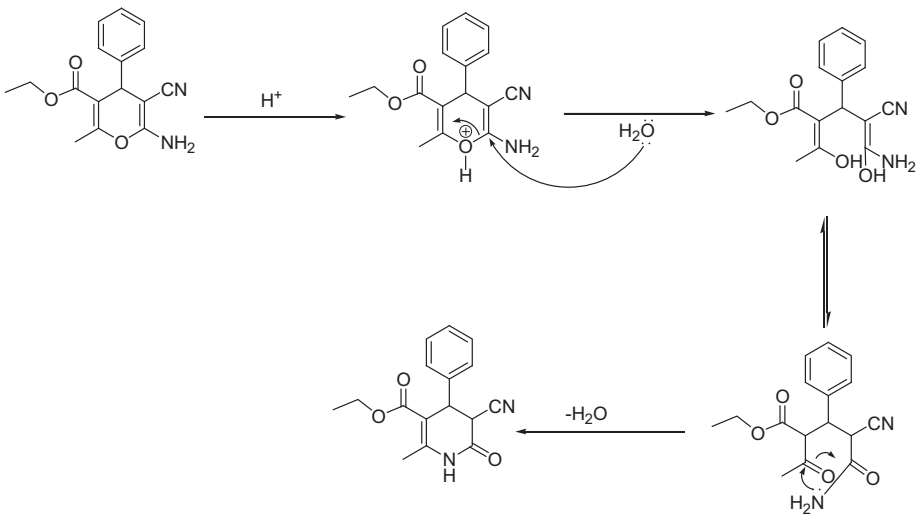
After the successful synthesis of 4*H*-pyran, we then focused on the rearrangement of 4*H*-pyran nucleus. We envisaged sequential ring opening followed by ring closure(Scheme 5) involving hydration and dehydration of the 4*H*-pyran nucleus leading to 3,4-dihydropyridone derivatives in aqueous medium. To find a suitable catalyst for this rearrangement, we initially screened the water soluble organic acids as well as Lewis acid catalysts. When the rearrangement was carried out with different organic acids and Lewis acids it did not give the expected rearranged product. However, treatment with *p*-TsOH transformed the 4*H*-pyran nucleus into 2-pyridone scaffold (with 10% loading) (Table 4, entry 11, bold). Subsequent increase in the catalyst load did not improve the product yield of the rearrangement reaction to any extent Table 5.

As shown in Table 5, the reaction was performed in $CHCl_3$, CCl_4, THF, CH_3CN, methanol, DMF, toluene, dioxane, and water (entries 1–9) to judge the right catalytic activity of *p*-TSOH and the best suited medium of transformation. Interestingly, we found that the reaction using water as the solvent (Table 5, entry 9, bold) resulted in higher yields than any other solvents.

We then investigated the scope of this synthetic methodology[22] (Table 6) toward the exploration of a new series of 3,4-dihydropyridin-2-one libraries by the rearrangement (Scheme 4) of substituted 4*H*-pyrans.

From Table 6, it is evident that the reaction proceeded smoothly for both electron rich and electron deficient aryl aldehydes with varied 1,3-diketone moieties. The methodology is so mild that it was also conveniently applied for the synthesis of 4-heteroaryl pyridin-2-one derivatives (Table 6, entries 8 and 22) with reasonably good yield.

We have demonstrated the use of nano structured-ZnO as a catalyst for the synthesis of substituted 4*H*-pyrans. We are also successful in developing a method to obtain the 3,4-dihydropyridin-2-



Scheme 5. Proposed mechanism of rearrangement of 4*H*-pyrans to 3,4-dihydropyridin-2-one.

Table 6
Substrate scope for synthesis of 3,4-dihydro pyridin-2-one

Entry	Product	Ar	1,3-Diketo compound	Time (h)[c]	Yield[a,b] (%)
1	**5a**	Ph	Ethylacetoacetate	3	87
2	**5b**	4-NO_2–C_6H_4–	Ethylacetoacetate	2	91
3	**5c**	3-NO_2–C_6H_4–	Ethylacetoacetate	2.5	90
4	**5d**	4-F–C_6H_4–	Ethylacetoacetate	2.5	88
5	**5e**	4-Cl–C_6H_4–	Ethylacetoacetate	3	87
6	**5f**	4-CH_3–C_6H_4–	Ethylacetoacetate	3.5	84
7	**5g**	4-OCH_3–C_6H_4–	Ethylacetoacetate	4	82
8	**5h**	2-Fur	Ethylacetoacetate	3	85
9	**5k**	Ph–	Acetylacetone	3	84
10	**5l**	4-NO_2–C_6H_4–	Acetylacetone	2.5	87
11	**5m**	3-NO_2–C_6H_4–	Acetylacetone	2.5	88
12	**5n**	4-F–C_6H_4–	Acetylacetone	2.5	86
13	**5o**	4-Cl–C_6H_4–	Acetylacetone	3	85
14	**5q**	4-CH_3–C_6H_4–	Acetylacetone	3.5	82
15	**5s**	Ph–	Dimedone	6	82
16	**5t**	4-NO_2–C_6H_4–	Dimedone	5	86
17	**5u**[d]	3-NO_2–C_6H_4–	Dimedone	5.5	85
18	**5v**	4-F–C_6H_4–	Dimedone	5	85
19	**5w**	4-OCH_3–C_6H_4-	Dimedone	7	78
20	**5x**	4-CH_3–C_6H_4–	Dimedone	7	80
21	**5y**	4-$N(CH_3)_2$–C_6H_4	Dimedone	7.5	76
22	**5z**	2-Fur	Dimedone	7	79

[a] Isolated yield of the pure product.
[b] The products were characterized by ^{1}H NMR, ^{13}C NMR, IR, HRMS, and elemental analysis.
[c] 1 mmol of 4*H*-pyran was heated at 70 °C in 5 ml water for stipulated time in the presence of 10 mol % TsOH.
[d] The structure of the compound (**5u**)was confirmed by X-ray analysis of single crystal (Supplementary data).

one derivatives applying the hydrolytic rearrangement reaction on 4*H*-pyran derivatives.

Acknowledgements

We acknowledge the financial support from the University of Calcutta. P.B. thanks CSIR, New Delhi, India for his fellowship (Grant No. 09/028(0768)/2010). K.P. and S.P. thank UGC for their respective fellowships. Crystallography was performed at the DST-FIST, India-funded Single Crystal Diffractometer Facility at the Department of Chemistry, University of Calcutta.

Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.tetlet.2012.06.086.

References and notes

1. *Comprehensive Heterocyclic Chemistry II*; Green, G. R., Evans, J. M., Vong, A. K., Katritzky, A. R., Rees, C. W., Scriven, E. F. V., Eds.; Pergamon Press: Oxford, 1995; Vol. 5, p 469.
2. Foye, W. O. *Prinicipi di Chemico Farmaceutica*; Piccin: Padova, Italy, 1991. 416.
3. Bonsignore, L.; Loy, G.; Secci, D.; Calignano, A. *Eur. J. Med. Chem.* **1993**, *28*, 517.
4. Martin, N.; Pascual, C.; Seoane, C.; Soto, J. L. *Heterocycles* **1987**, *26*, 2811.
5. Armesto, D.; Horspool, W. M.; Martin, N.; Ramos, A.; Seaone, C. *J. Org. Chem.* **1989**, *54*, 3069.
6. Quintela, J. M.; Peinador, C.; Moreira, M. J. *Tetrahedron* **1995**, *51*, 5901.
7. Li, Q.; Mitscher, L. A.; Shen, L. L. *Med. Res. Rev.* **2000**, *20*, 231.
8. Cox, R. J.; Ohagan, D. *J. Chem. Soc., Perkin Trans. 1* **1991**, 2537.
9. (a) Nagarajan, M.; Xiao, X. S.; Antony, S.; Kohlhagen, G.; Pommier, Y.; Cushman, M. *J. Med. Chem.* **2003**, *46*, 5712; (b) Hasvold, L. A.; Wang, W. B.; Gwaltney, S. L.; Rockway, T. W.; Nelson, L. T. J.; Mantei, R. A.; Fakhoury, S. A.; Sullivan, G. M.; Li, Q.; Lin, N. H.; Wang, L.; Zhang, H. Y.; Cohen, J.; Gu, W. J.; Marsh, K.; Bauch, J.; Rosenberg, S.; Sham, F. L. *Bioorg. Med. Chem. Lett.* **2003**, *13*, 4001.
10. (a) Dolle, V.; Fan, E.; Nguyen, C. H.; Aubertin, A. M.; Kirn, A.; Andreola, M. L.; Jamieson, G.; Tarragolitvak, L.; Bisagni, E. *Eur. J. Med. Chem.* **1995**, *38*, 4679; (b) Wai, J. S.; Williams, T. M.; Bamberger, D. L.; Fisher, T. E.; Hoffman, J. M.; Hudcosky, R. J.; Mactough, S. C.; Rooney, C. S.; Saari, W. S.; Thomas, C. M.; Goldman, M. E.; Obrien, J. A.; Emini, E. A.; Nunberg, J. H.; Quintero, J. C.; Schleif, W. A.; Anderson, P. S. *J. Med. Chem.* **1993**, *36*, 249.
11. Peng, Y.; Song, G. *Catal. Commun.* **2007**, *8*, 111.
12. Fotouhi, L.; Heravi, M. M.; Fatehi, A.; Bakhtiari, K. *Tetrahedron Lett.* **2007**, *48*, 5379.
13. Wang, L. M.; Shao, J. H.; Tian, H.; Wang, Y. H.; Liu, B. *J. Fluorine Chem.* **2006**, *127*, 97.
14. Babu, N.; Pasha, S. N.; Venkateswara Rao, K. T.; Sai Prasad, P. S.; Lingaiah, N. *Tetrahedron Lett.* **2008**, *49*, 2730.
15. Banerjee, S.; Horn, A.; Khatri, H.; Sereda, G. *Tetrahedron Lett.* **1878**, *2011*, 52.
16. Valizadeh, H.; Azimi, A. A. *J. Iran. Chem. Soc.* **2011**, *8*, 123.
17. Seoane, C.; Soto, J. L.; Zamorano, P.; Quinteiro, M. *J. Heterocycl. Chem.* **1981**, *18*, 309.
18. Paul, S.; Bhattacharyya, P.; Das, A. R. *Tetrahedron Lett.* **2011**, *52*, 4636.
19. Ghosh, P. P.; Das, A. R. *Tetrahedron Lett.* **2012**, *53*, 3140.
21. *General procedure of preparation of 4H-pyran*: A mixture of an aldehyde (1 mmol), malononitrile (1 mmol), 1,3-diketo compound (1 mmol) and nano ZnO (10 mol %) was stirred at room temperature in 5 ml ethanol–water mixture (1:1). The progress of the reaction was monitored by TLC (EtOAc/hexane = 3:7). After completion of the reaction 10 ml ethanol was added to the reaction mixture and filtered to remove ZnO. Upon concentrating the reaction mixture the desired product crystallized out. As the purity of the product was high no further recrystallization was required.
22. *General procedure of preparation of 3, 4-dihydropyridin-2-one via rearrangement*: A mixture of 4*H*-pyran (1 mmol) and *p*-toluelesulphonic acid (10 mol %) was heated at 70 °C in 5 ml H_2O. The progress of the reaction was monitored by TLC (EtOAc/hexane = 3:7). After completion of the reaction the reaction mixture was cooled to room temperature and the precipitated crude product was filtered. On recrystalizing from ethanol pure product (2-pyridone) was obtained.