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L-Proline catalyzed synthesis of naphthopyranopyrimidines via multicomponent reaction

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ABSTRACT

A green and efficient one pot, three component protocol for synthesis of naphthopyranopyrimidines by cyclocondensation of -naphthol, aldehyde, and 6-amino-1,3-dimethyluracil using L-proline as a beneficial catalyst with high catalytic activity under solvent-free conditions at 100 °C is described. In this study, several types of aromatic aldehyde, containing electron-withdrawing groups as well as electron-donating groups, were rapidly converted to the corresponding naphthopyrano-pyrimidine derivatives in good to excellent yields. The present approach offers several advantages such as short reaction times, simple work-up, excellent yields, non-toxicity of the catalyst, and solvent-free conditions.To the best of our knowledge, this is the first report on the synthesis of naphthopyranopyrimidine derivatives using L-proline as a catalyst under solvent-free conditions. The catalysts can be recovered for the subsequent reactions and reused without any loss of efficiency.

Keywords: L-proline; 6-Amino-1,3-dimethyluracil; Solvent-free; -naphthol.

1. Introduction

The discovery of novel synthetic methodologies to facilitate the preparation of compound libraries is a pivotal focal point of research activity in the field of modern medicinal and combinatorial chemistry. Molecular complexity and variety in natural and biologically relevant systems encourage chemists to investigate new methods and reactions [1]. The multicomponent reactions (MCRs) are effective methods in heterocyclic scaffolds for the creation of different chemical libraries of drug-like advanced compounds in organic and medicinal chemistry [2-4]. Multicomponent reactions are extremely convergent, producing a remarkably high increase of molecular complexity in just one step. In the mainstream of current interest, multicomponent reactions (MCRs) have attracted considerable attention due to significant advantages such as simplicity of operation, reduction of isolation and purification steps, facile execution, high atom-economy and high selectivity and mini-mization of costs, time and waste production [5-8]. MCRs are particularly useful to provide expedient approaches to a wide range of compounds of biological and pharmaceutical interest. Hence, most of the scientific efforts have been focused on the develment of multicomponent processes to prepare diverse heterocyclic compound libraries [9,10]. One such MCR that belongs to the latter category is synthesis of pyrimidine derivatives.

Green chemistry is the usage of a set of principles that will help decrease the use and generation of hazardous substances during the construction and application of chemical products. It is an approach to the synthesis, processing and use of chemicals that reduces risks to humans and the environment. Also, it is an important area in the chemical sciences. Much innovative chemistry has been developed ovebr the past several years that are effective, efficient and more environmentally benign [11].

Pyrimidine entity is one of the most prominent structures found in nucleic acid chemistry. Vitamin B (thiamine) is well known example of naturally occurring pyrimidine that is encountered in our daily lives. There is continuous wide spread interest in pyranopyrimidines because of diverse biological activities such as antitumor [12], antimicrobial [13], antihypertensive [14], physiological [15], analgesic, and anticonvulsant [16]. Moreover, naphthopyrano-pyrimidine and its derivatives are important structural

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motifs in medicinal and pharmaceutical chemistry as they show antibacterial and antifungal activities [17-19]. Neuropeptide S receptor, previously known as GPR-154, is highly expressed in brain areas that have been implicated in modulation of arousal, stress and anxiety. Therefore, Neuropeptide S receptor represents a novel drug target for the treatment of sleep and anxiety disorders. Naphthopyranopyrimidines are selective antagonists of Neuropeptide S receptor [20].

Though there is great importance of naphthopyranopyrimidines that there are only five reports for the synthesis of naphthopyranopyrimidines. Previously, Nandi et al reported the synthesis of naphthopyrano-pyrimidines by multicomponent condensation of -napthol, aldehyde, and 6-amino-1,3-dimethyluracil using $InCl_3$ under solvent free conditions [21]. Bandgar et al introduced simple, efficient and environmentally benign synthesis of naphthopyrano-pyrimidines using silicotungstic acid ($H_4[SiW_{12}O_{40}]$) as a catalyst [22]. Lately, Khurana et al reported on the synthesis of naphthopyranopyrimidines by three component cyclocondensation of -naphthol, aldehyde, and cyclic 1,3-dicarbonyl compounds using alum ($KAl(SO_4)_2.12H_2O$) as an inexpensive catalyst under solvent-free conditions [23]. Kumar et al presented iodine catalyzed one-pot three-component synthesis of naphthopyranopyrimidines [24]. $InCl_3$ or P_2O_5 also has been found as an effective catalyst towards this transformation [25].

In recent decades, the direction of technology and science has been shifting toward the commercially available and reusable catalysts. Proline is the only natural amino acid with a secondary amine functionally, which raises the pka value and displays better nucleophilicity as compared to other amino acides. It is bifuctional, with a carboxylic acid and an amine portion. These two functional groups can both act as acid or base and can also facilitate chemical transformations in concert, similar to enzymatic catalysis. While enzymes typically use several different functinoal groups in their cataytic machinery, bifunctional asymetric catalysis has become a very successful strategy in the laboratatory. In addition, proline is a chiral bidentate ligand that can form catalytically active metal complexes. It is evident from the recent literature that amino acid L-proline has invoked enormous interest as a highly active and inexpensive catalyst to construct carbon-carbon and carbon-heteroatom bonds in various organic transformations [26]. L-Proline and its derivatives are often used as asymmetric catalysts in organic reactions such as a direct asymmetric aldol reactions [27], asymmetric -amination of ketones [28], asymmetric Diels–Alder reactions [29], and synthesis of quinolins and coumarins [30]. As part of our continuing efforts on the development of new strategies for the synthesis of heterocyclic compounds [31], we decided to investigate the synthesis of naphthopyranopyrimidine derivatives via three component reaction of -naphthol, aldehydes, and 6-amino-1,3-dimethyluracil under solvent-free conditions by L-proline at 100 ºC (Scheme 1).

2. Experimental

2.1. General

All chemicals were purchased from Merck or Fluka Chemical Companies. All products were characterized by physical data (mp), and spectral data (IR, ¹H NMR). Melting points were measured on an Electrothermal 9200 apparatus. IR spectra were obtained on a Shimadzu FTIR-8400S spectrometer. ¹H and ¹³C NMR spectra were determined on a BRUKER DRX-400 AVANCE spectrometer.

2.2. General procedure for the preparation of naphthopyranopyrimidines (4a)

To a mixture of -naphthol (1.0 mmol, 0.144 g), benzaldehyde (1.0 mmol, 0.106 g), and 6-amino-1,3-dimethyluracil (1.1 mmol, 0.170 g), L-proline (40 mol %) was added under solvent-free condition. The reaction mixture was heated at 100 °C for the stipulated period of time till the full consumption of the starting materials (monitored by TLC). After completion of the reaction, the reaction mixture was allowed to cool to room temperature. The solid obtained was washed with water (2×10 mL) and recrystallized from ethanol to afford the pure product (4a).
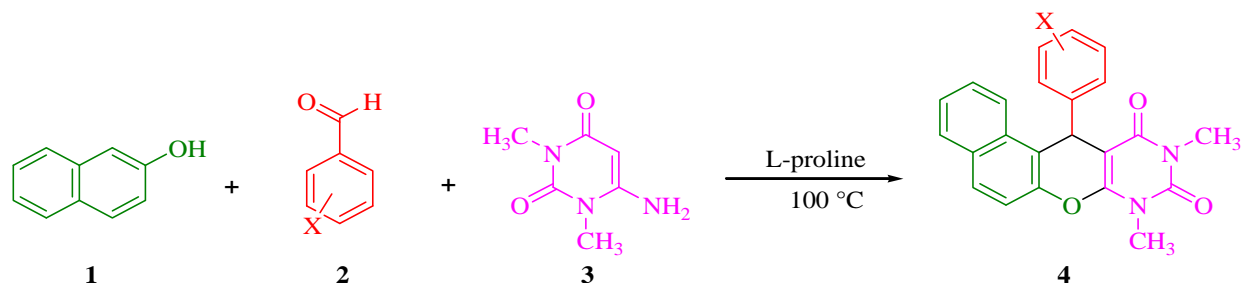
Scheme 1. Synthesis of naphthopyranopyrimidines in the presence of L-proline.

The same procedure was also used for the other products listed in Table 2.

Selected spectral data

8,10-Dimethyl-12-phenyl-8,12-dihydro-7-oxa-8,10-diazabenzo[a]anthracene-9,11-dione (4a):

m.p.= 225-227 °C. IR (KBr): $\bar{\nu}$ = 2932, 1703, 1657, 1475, 1227, 1179 cm^{-1}. ^{1}HNMR (400 MHz, CDCl$_3$): = 3.36 (3H, s), 3.58 (3H, s), 5.86 (s, 1H), 7.25-7.13 (3H, m), 7.39-7.42 (5H, m), 7.79 (2H, m), 7.93 (1H, d, *J* = 7.6 Hz) ppm. ^{13}CNMR (100 MHz, CDCl$_3$): = 27.8, 28.9, 35.7, 90.2, 116.8, 116.9, 124.3, 124.9, 127.1, 126.9, 127.9, 128.7, 128.2, 129.1, 131.2, 132.0, 144.1, 146.9, 150.2, 151.9, 161.7 ppm.

8,10-Dimethyl-12-p-tolyl-8,12-dihydro-7-oxa-8,10-diazabenzo[a]anthracene-9,11-dione (4k):

m.p.= 199-201 °C. IR (KBr): $\bar{\nu}$ = 2932, 1706, 1638, 1473, 1239, 1185 cm^{-1}. ^{1}HNMR (400 MHz, CDCl$_3$): = 7.98 (1H, d, *J* = 8.2 Hz), 7.84-7.80 (2H, m), 7.42–7.32 (3H, m), 7.31 (2H, d, *J* = 7.4 Hz), 7.02 (2H, d, *J*=7.4 Hz), 5.76 (1H, s), 3.65 (3H, s), 3.28 (3H, s), 2.26 (3H, s) ppm. ^{13}CNMR (100 MHz, CDCl$_3$): = 21.0, 27.9, 29.1, 35.2, 92.1, 115.8, 117.2, 124.3, 125.6, 127.1, 127.8, 128.2, 129.0, 129.4, 131.2, 131.3, 137.1, 141.0, 146.9, 150.2, 151.8, 162.1 ppm.

8,10-Dimethyl-12-(2,4-dimethoxyphenyl)-8,12-dihydro-7-oxa-8,10-diazabenzo[a]anthracene-9,11-dione (4m):

m.p. 280-282 °C. IR (KBr): $\bar{\nu}$ = 3245, 3152, 2963, 1689, 1654, 1656, 1343, 1245, 1172, 1054 cm^{-1}. ^{1}HNMR (400 MHz, CDCl$_3$): = 3.32 (3H, s), 3.58 (3H, s), 3.74 (3H, s, OCH$_3$), 3.80 (3H, s, OCH$_3$), 5.75 (1H, s), 7.08 (1H, d, *J* = 7.6 Hz), 7.30-7.36 (3H, m), 7.41-7.53 (2H, m), 7.79-7.87 (2H, m), 8.11 (1H, d, *J*= 8.0 Hz) ppm. ^{13}CNMR (100 MHz, CDCl$_3$): = 27.9, 28.9, 33.6, 42.1, 56.2, 89.3, 116.2, 123.7, 125.3, 127.4, 127.8, 129.1, 128.9, 129.8, 130.8, 131.4, 132.1, 132.5, 132.9, 139.3, 146.5, 151.0, 152.3, 160.8 ppm.

8,10-Dimethyl-12-(3-Bromophenyl)-8,12-dihydro-7-oxa-8,10-diazabenzo[a]anthracene-9,11-dione (4n):

m.p.= 232-234 °C. IR (KBr): $\bar{\nu}$ = 3052, 2947, 1703, 1657, 1580, 1475 cm^{-1}. ^{1}HNMR (400 MHz, CDCl$_3$): = 3.21 (3H, s), 3.52 (3H, s), 5.73 (3H, s), 8.13 (2H, m), 7.86 (2H, d, *J* = 8.0 Hz), 7.68 (1H, d, *J* = 8.0 Hz), 7.54 (2H, m), 7.43 (1H, s), 7.29 (2H, m), 7.21 (1H, d, *J*= 8.2 Hz) ppm. ^{13}CNMR (100 MHz, CDCl$_3$): = 27.5, 28.7, 29.1, 81.3, 115.4, 118.2, 122.2, 122.8, 125.9, 126.4, 126.5, 127.6, 128.0, 128.7, 128.8, 130.4, 133.4, 134.3, 144.4, 153.5, 153.7, 159.5, 161.4 ppm.

3. Results and Discussion

In continuation of our ongoing program in developing methods for the synthesis of heterocyclic compounds and identifying new catalysts [31], we herein report a novel and efficient protocol for synthesis of naphthopyranopyrimidines via three component reaction of -naphthol, aldehydes, and 6-amino-1,3-dimethyluracil by L-proline under solvent-free conditions (Scheme 1). Initially, the three component reaction involving -naphthol, benzaldehyde, and 6-amino-1,3-dimethyluracil in the absence of catalyst was performed for a period of 15-24 h at room temperature and at 100 °C. The desired naphthopyranopyrimidine **4a** was not obtained (Table 1, Entries 1, 2). This reaction was performed using various amounts of L-proline. Initially, 20 mol % L-proline was used to perform the reaction. But it requires slightly long reaction time. Therefore, the loading of the catalyst was gradually increased from 20 mol % to 50 mol %. It was found that 40 mol % of L-proline is optimal to carry out the reactions in a short duration (20 min). The use of excess of catalyst did not alter either reaction time or yield of the product (Table 1, Entry 9). When L-proline catalyzed preparation of **4a** was performed in various solvents on the same substrates, the product formation was observed in low yields (Table 1, Entries 11-14). Then finally in an effort to improve the yield further, the reaction was conducted without solvent in the presence of L-proline as a catalyst. Thus, the use of 40 mol % L-proline at 100 °C is ideal to achieve the desired product **4a** in good yields 96% (Table 1, Entry 8).

Under the optimized reaction conditions, the generality of the reaction was fully investigated with different aldehydes, -naphthol and 6-amino-1,3-dimethyluracil to produce naphthopyranopyrimidine derivatives. The results are summarized in Table 2. All these reactions showed rapid formation of naphthopyranopyrimidine derivatives with high efficiency. *Ortho* substituted aldehydes furnished desired product with longer reaction time and low yields compared to their *meta* and *para* counterparts. This might be due to steric hindrance. We also studied the reactions of different aliphatic aldehydes such as propanal, and butanal. Unfortunately, we failed to get the desired products. The aldol condensation of aliphatic aldehyde reaction takes place which prevents further reaction. The results represented that the reactions were performed within 20-40 min of heating, and the favorable products were provided in good yields (Table 2).

The structures of compounds **4a-n** were confirmed by IR and ^{1}H NMR spectroscopy. The IR spectrum of compound **4m**, for example, show absorption bands at 3245, 3152, 2963, 1689, 1654, 1656, 1343, 1245, 1172, and 1054 cm^{-1} indicating the presence of C-H, C=O, C=C, C-N, and C-O groups in this molecule. Aromatic protons of this compound were seen at 7.08-8.11 in its ^{1}H NMR spectrum resonating with proper integrals and splittings. Aliphatic region of this spectrum exhibits four singlet peaks at 3.32, 3.58, 3.74, and

Table 1. Optimization of the reaction conditions.[a]

Entry	Catalyst (g)	Conditions	Time (min)	Yield (%)	Ref.
1	-	Solvent-free/ 25 °C	24 h	-	-
2	-	Solvent-free/ 100 °C	15 h	-	-
3	$H_4[SiW_{12}O_{40}])$ (5)	Solvent-free/ 100 °C	20 min	90	[22]
4	$InCl_3$ (35)	Solvent-free/ 120 °C	20 min	80	[21]
5	L-proline (20)	Solvent-free/ 25 °C	12 h	Trace	-
6	L-proline (30)	Solvent-free/ 25 °C	10 h	Trace	-
7	L-proline (30)	Solvent-free/ 100 °C	120 min	78	-
8	L-proline (40)	Solvent-free/ 100 °C	20 min	96	-
9	L-proline (50)	Solvent-free/ 100 °C	25 min	94	-
10	L-proline (40)	Solvent-free/ 110 °C	20 min	95	-
11	L-proline (40)	CH_3CN/ reflux	70 min	57	-
12	L-proline (40)	CH_2Cl_2/ reflux	100 min	64	-
13	L-proline (40)	EtOH/ reflux	50 min	76	-
14	L-proline (40)	H_2O/ reflux	45 min	80	-

[a]A mixture of -naphthol **1** (1.0 mmol), benzaldehyde **2a** (1.0 mmol), and 6-amino-1,3-dimethyluracil (1.1 mmol) **3**.
[b]Isolated yield.

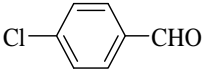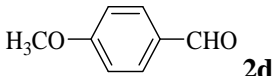**Table 2**. Synthesis of naphthopyranopyrimidines **4a-n.**
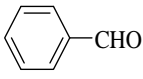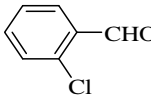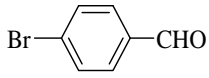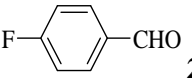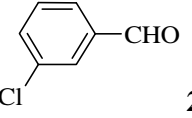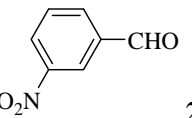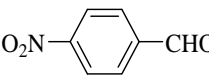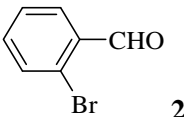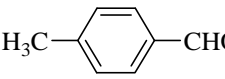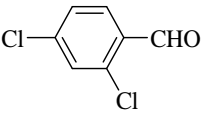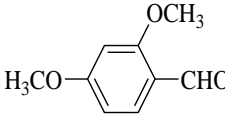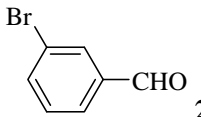
Entry	Aldehyde	Product[a]	Time (min)	Yield (%)[b]	m.p. (°C)		Ref.
					Found	Reported	
1	**2a**	**4a**	20	96	225-227	226-228	[25]
2	**2b**	**4b**	40	88	269-271	270-272	[25]
3	**2c**	**4c**	30	95	272-274	274-276	[24]
4	**2d**	**4d**	20	95	255-257	257-259	[22]

Table 2. *(Continued).*

5	Br—⬡—CHO **2e**	**4e**	25	93	248-250	250-252	[22]
6	F—⬡—CHO **2f**	**4f**	25	94	293-295	294-296	[23]
7	⬡—CHO (Cl) **2g**	**4g**	20	93	249-251	250-252	[22]
8	⬡—CHO (O₂N) **2h**	**4h**	20	95	309-311	310-312	[22]
9	O₂N—⬡—CHO **2i**	**4i**	25	93	287-289	288-290	[25]
10	⬡—CHO (Br) **2j**	**4j**	30	89	216-218	217-219	[24]
11	H₃C—⬡—CHO **2k**	**4k**	35	92	199-201	200-202	[24]
12	Cl—⬡—CHO (Cl) **2l**	**4l**	35	91	219-221	222-224	[25]
13	H₃CO—⬡(OCH₃)—CHO **2m**	**4m**	40	89	280-282	-	This work
14	(Br) ⬡—CHO **2n**	**4n**	35	92	232-234	-	This work

[a]All products were characterized by ^{1}H NMR and IR spectral data and comparison of their melting points with those of authentic samples.
[b]Isolated yield.

3.80 arising from protons of the methyl and methoxy groups along with the characteristic sharp signal of the methine proton at 5.75. The ^{13}CNMR spectrum of **4m** displays 24 distinct lines with appropriate chemical shifts corresponding to the structure of this compound. Workup procedure is so simple for the naphthopyranopyrimidines syntheses and includes addition of water at the end of reaction, filtration, and finally recrystallization of the products from ethanol. The L-proline was removed simply by dissolution in water added to the reaction mixture and then recovered by evaporation of water at 80 °C under reduced pressure. The recovered catalyst can be reused at least four additional times in subsequent reactions without significant loss in product yield (Fig. 1). Plausible mechanism for the formation of naphthopyranopyrimi-
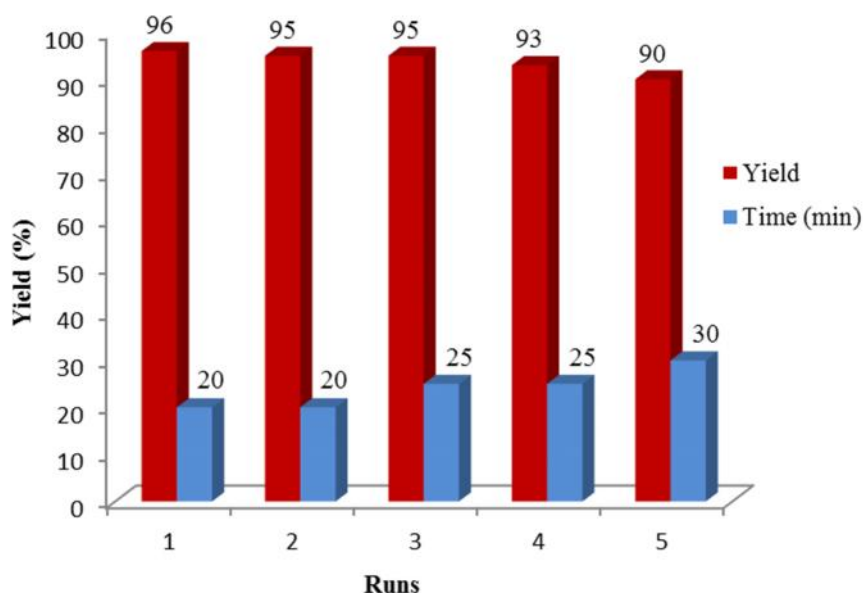
Fig. 1. Reusability of the catalyst in product **4a**.

dines is outlined in Scheme 2, which proceeds via *ortho* quinine methide intermediate **7** formation from -naphthol and aldehyde in the presence of L-proline as a catalyst. The 6-amino-1,3-dimethyluracil **3** attacks to adduct **7** in a Michael-type fashion to produce an open chain intermediate **8**. Intermediate **8** undergoes intramolecular cyclization by the reaction of nucleophilic amino function to carbonyl group. Cyclization of the intermediate **8** and subsequent loss of ammonia leads to the naphthopyranopyrimidine **4**.

4. Conclusions

In conclusion, we have developed an efficient, solvent-free and environmentally benign process for the synthesis of naphthopyranopyrimidines by one-pot, three-component reaction of -naphthol, aldehyde, and 6-amino-1,3-dimethyluracil catalyzed by L-proline as a readily available, economical, and environmentally safe catalyst. Simple experimental procedure, excellent yields of the products, recyclability of the catalyst with no loss in its activity, use of nontoxic, user friendly process and easy work up procedure are the major advantages of the present method.

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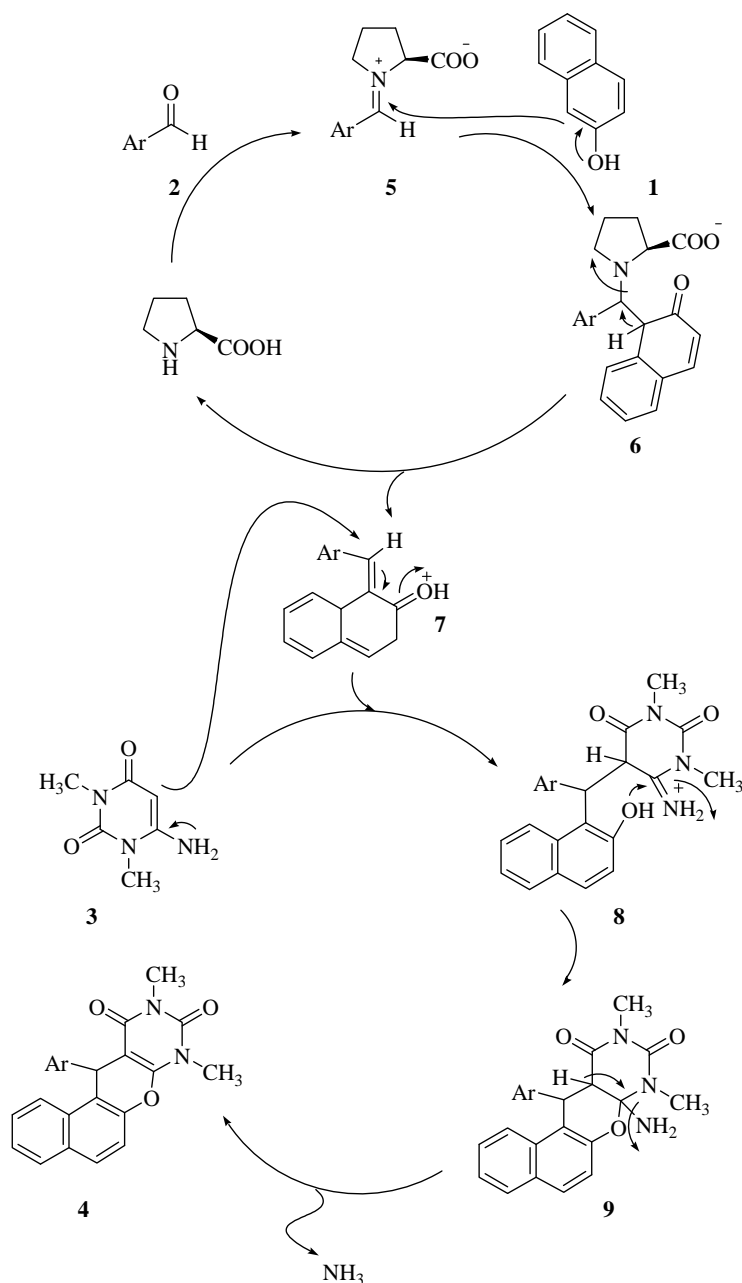
Scheme 2. A plausible mechanism for the synthesis of naphthopyranopyrimidine **4a-n** in the presence of L-proline.

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