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Vilsmeier Reaction of 3-Aminopropenamides: One-Pot Synthesis of Pyrimidin-4(3*H*)-ones

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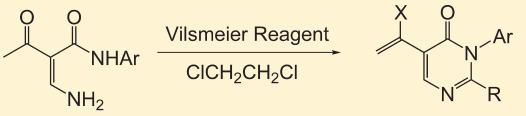
Supporting Information

ABSTRACT: A facile one-pot synthesis of pyrimidin-4(3*H*)-ones was developed via reactions of a series of readily available 3-aminopropenamides with varied Vilsmeier reagents, and a mechanism involving sequential halogenation, formylation, and intramolecular nucleophilic cyclization is proposed.

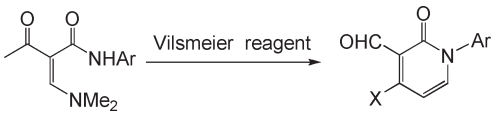


R=H,CH_3; X=Cl,Br; 11 examples; 54-85% yield

Pyrimidin-4(3*H*)-ones and their benzo-/hetro-fused analogues represent an important class of organic aza-heterocycles for their presence in numerous natural products and synthetic organic compounds along with diverse bio-, physio-, and pharmacological activities.[1−3] For example, some related 4-(phenylamino)-pyrido[*d*]pyrimidines have been reported as selective inhibitors of tyrosine phosphorylation by the epidermal growth-factor receptor (EGFR), and have become an important class of potential anticancer drugs.[4,5] The development of synthetic approaches for such aza-heterocyclic compounds has been the focus of intense research for decades and continues to be an active area of research today. So far, a variety of synthetic approaches are already available based on either the modification of the preconstructed heterocyclic ring or construction of the heterocyclic skeleton from appropriately substituted open chain precursors, such as β-aminoester, ethyl cyanoacetate, and diethyl malonate.[6,7] However, many of these methods suffer from drastic reaction conditions, long reaction times, and complex procedures.

On the other hand, Vilsmeier−Haack reaction associated with its mild reaction condition, commercial availability of the reagents, and improved understanding of the reaction mechanism has been widely used for the formylation of activated aromatic compounds and β-halovinyl aldehydes.[8] The versatile reactivity of carbonyl compounds with halomethylene iminium salts and a variety of cyclization reactions leading to heterocycles induced by Vilsmeier reagent has been well-documented.[9] In our recent work, we have demonstrated the utility of Vilsmeier reagent in the synthesis of functionalized heterocycles, such as substituted 2*H*-pyrans,[10] pyridines,[11] quinolines,[12] and pyridin-2(1*H*)-ones.[13] Most recently, we synthesized a series of 2-[(dimethylamino)methylene]-3-oxo-*N*-arylbutanamides from β-oxo amides, examined their reaction behavior under Vilsmeier conditions, and achieved a facile synthesis of substituted pyridin-2(1*H*)-ones bearing formyl at the 3-position (Scheme 1).[14]

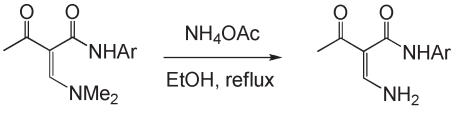
Scheme 1. Synthesis of Pyridin-2(1*H*)-ones



In connection with the previous work and following with our interest in the synthesis of highly valuable heterocycles from β-oxo amide derivatives,[15] we synthesized a series of 2(amino-methylene)-3-oxo-*N*-arylbutanamides **2**, and examined their reactivity toward different Vilsmeier reagents. By this research, we developed a facile one-pot synthesis of substituted pyrimidin-4(3*H*)-ones **3** via Vilsmeier reaction of the readily available 3-aminopropenamides **2**. Herein, we wish to report our experimental results and present a proposed mechanism for the cyclization.

According to our reported procedure, a series of 2-[(dimethylamino)methylene]-3-oxo-*N*-arylbutanamides **1** was synthesized from commercially available β-oxo amides and *N*,*N*-dimethylformamide dimethyl acetal (DMFDMA) in the presence of K_2CO_3 in *N*,*N*-dimethylformamide (DMF) at room temperature in good yields (up to 80%).[14] Then, 3-aminopropenamides **2** were prepared from **1** in the presence of ammonium acetate in ethanol under reflux in high yields (up to 95%, Scheme 2). With substrates **2a**−**g** in hand, we selected **2a** as a model compound to examine its reaction behavior under Vilsmeier conditions.

Scheme 2. Preparation of 3-Aminopropenamides 2



1 **2**

Thus, the reaction of **2a** with Vilsmeier reagent $POCl_3$/DMF (2.0 equiv) was first attempted at room temperature. Unfortunately, no predominant product was formed even after prolonged reaction time (up to 8.0 h) as indicated by TLC. When the mixture was heated to 60 °C and stirred for 3.0 h, the reaction furnished a product after workup and purification of the resulting mixture by column chromatography. From the spectral and analytical data, the product was characterized as 5-(1-chlorovinyl)-3-*p*-tolylpyrimidin-4(3*H*)-one **3a1** (Scheme 3).

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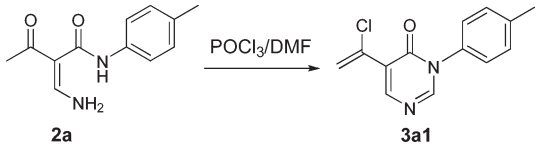
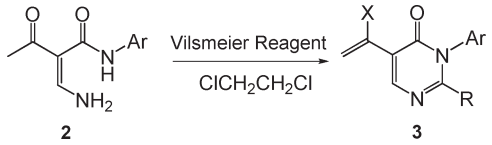
Scheme 3. Reaction of 2a with Vilsmeier Reagent $POCl_3$/DMF



The reaction conditions, including reaction temperature, solvent, and the ratio of $POCl_3$ and DMF to **2a**, were investigated. Low conversion was observed when the reaction was performed at temperatures below 60 °C even with prolonged reaction time. It seemed that the increase of the ratio of $POCl_3$ to **2a** had no significant influence on the reaction. But ratios of $POCl_3$ to **2a** lower than 2/1 resulted in low conversion and prolonged reaction time. It should be noted that increase of the ratio of DMF to **2a** to more than 4/1 led to a complex mixture. A series of experiments revealed that 2.5 equiv of $POCl_3$ and 1.5 equiv of DMF was sufficient for the synthesis of pyrimidin-4(3*H*)-one, and the optimal results were obtained when the reaction of **2a** was performed with $POCl_3$ (3.0 equiv) and DMF (2.0 equiv) in dichloroethane at 70 °C for 1.5 h. Hence product **3a1** was obtained in 74% yield (Table 1, entry 1) .

Table 1. Synthesis of Substituted Pyrimidin-4(3*H*)-ones 3 via Vilsmeier Reaction[a]



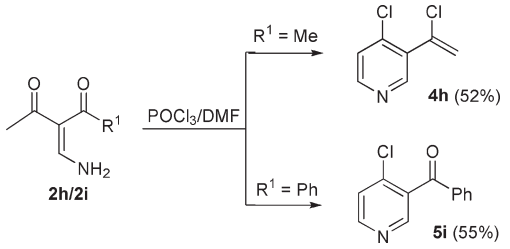
entry	2	Ar	R	X	3	yields (%)[b]
1	**2a**	4-MeC_6H_4	H	Cl	**3a1**	74
2	**2b**	4-$MeOC_6H_4$	H	Cl	**3b1**	67
3	**2c**	4-ClC_6H_4	H	Cl	**3c1**	56
4	**2d**	2,4-$Me_2C_6H_3$	H	Cl	**3d1**	57
5	**2e**	2-$MeOC_6H_4$	H	Cl	**3e1**	63
6	**2f**	5-Cl-2-$MeOC_6H_3$	H	Cl	**3f1**	65
7	**2g**	C_6H_5	H	Cl	**3g1**	70
8	**2a**	4-MeC_6H_4	H	Br	**3a2**	85
9	**2b**	4-$MeOC_6H_4$	H	Br	**3b2**	68
10	**2a**	4-MeC_6H_4	CH_3	Cl	**3a3**	81
11	**2d**	2,4-$Me_2C_6H_3$	CH_3	Cl	**3d3**	54

[a] Reagents and conditions: (i) for entries 1−7, $POCl_3$/DMF (3.0/2.0 equiv), $ClCH_2CH_2Cl$, 70 °C, 1.0−2.0 h; (ii) for entries 8 and 9, PBr_3/DMF (3.0/2.0 equiv), $ClCH_2CH_2Cl$, 70 °C, 1.5−2.0 h; (iii) for entries 10 and 11 $POCl_3$/DMAC (3.0/2.0 equiv), $ClCH_2CH_2Cl$, 75 °C, 12.0−15.0 h. [b] Isolated yields.

Under conditions identical to the ones used to form **3a1** in Table 1 (entry 1), 3-aminopropenamides **2b−g** afforded the corresponding substituted pyrimidin-4(3*H*)-one **3b1−g1** in good yields (Table 1, entries 2−7). Pyrimidin-4(3*H*)-ones were also obtained by subjecting **2a** and **2b** to Vilsmeier reagent PBr_3/DMF under otherwise indentical conditions (Table 1, entries 8 and 9). The versatility of this pyrmidin-4(3*H*)-one synthesis was further evaluated by performing **2a** and **2d** with Vilsmeier reagent $POCl_3$/*N*,*N*-dimethylacetamide (DMAC), respectively (Table 1, entries 10 and 11). The results demonstrated the efficiency and synthetic interest of the cyclization reaction with respect to varied Vilsmeier reagents and substrates **2** bearing variable arylamide groups. Therefore, we provided a facile approach for the synthesis of pyrimidin-4(3*H*)-ones of type **3**.

To expand the scope of the reaction, we then examined the reaction of 3-(aminomethylene)pentane-2,4-dione **2h** toward Vilsmeier reagent $POCl_3$/DMF under identical conditions. The reaction proceeded smoothly, as indicated by TLC, and furnished a product, which was characterized as 4-chloro-3-(1-chlorovinyl)pyridine **4h** (yield: 52%, Scheme 4). Further experiment revealed that the cyclization proved to be suitable for 2-(aminomethylene)-1-phenylbutane-1,3-dione **2i** to afford the corresponding 3-benzoyl-4-chloropyridine **5i** in 55% yield (Scheme 4).

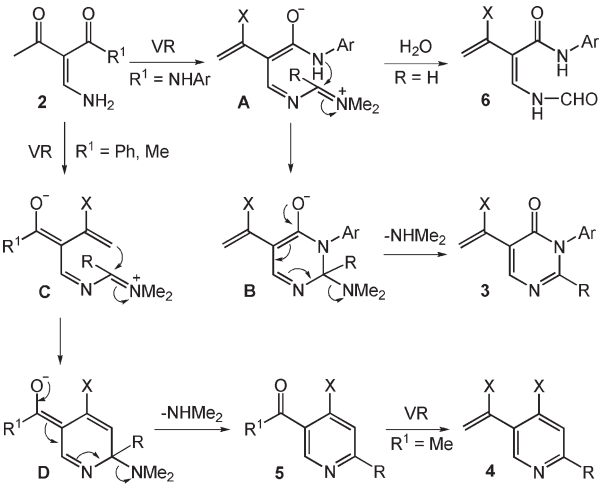
Scheme 4. Reaction of 2h with Vilsmeier Reagent $POCl_3$/DMF



To gain insight into the mechanism of the cyclization of 3-aminopropenamides **2**, a separate experiment with **2a** was conducted. When substrate **2a** was subjected to $POCl_3$ (2.0 equiv) and DMF (1.2 equiv) in dichloroethane at 40 °C for 1.0 h, 2-(formamidomethylene)-3-oxo-*N*-*p*-tolylbutanamide **6a** was obtained in 26% yield. The result suggested that amino and acyl groups of 3-aminopropenamide **2** were more reactive than amide group under Vilsmeier conditions.

On the basis of all the results obtained together with our previous studies, a plausible mechanism for the cyclization of 3-aminopropenamides **2** is presented in Scheme 5. Under Vilsmeier conditions, 3-aminopropenamide **2** bearing an amide group

Scheme 5. Plausible Mechanism of the Reaction of 3-Aminopropenamides 2 under Vilsmeier Conditions



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undergoes formylation of the amino group and halogenation of the acyl group to give intermediate **A**, which can be transformed into 3-halo-2-(formamidomethylene)-*N*-aryl-3-enamide **6** upon treatment with water. An intramolecular cyclization of **A** occurs to form intermediate **B**, which looses dimethylamine to afford pyrimidin-4(3*H*)-one **3**. In the same fashion, enaminone **2h** or **2i** undergoes formylation and halogenation reactions mediated by Vilsmeier reagent to give intermediate **C**. Intramolecular cyclization of **C**, followed by aromatization of intermediate **D** with the elimination of dimethylamine, furnishes pyridine of type **5**. When R^1 is a methyl group, pyridine **5** undergoes further halogenation to give rise to pyridine **4**.

In summary, a facile one-pot synthesis of pyrimidin-4(3*H*)-one of type **3** was developed via a Vilsmeier reaction of aminopropenamides **2**, which involved sequential formylation, halogenation, and intramolecular nucleophilic cyclization reactions. The protocol was extended to the synthesis of halogenated pyridine of types **4** and **5**. The simple execution, readily available substrates, mild conditions, and wide range of synthetic potential of the products make this protocol very attractive.

EXPERIMENTAL SECTION

Typical Procedure for the Synthesis of Substituted Pyrimidin-4(3*H*)-ones 3 (with 3a1 as an example). To a mixture of anhydrous DMF (4.0 mmol) and CH_2ClCH_2Cl (20 mL) was added $POCl_3$ (6.0 mmol) at 0 °C under stirring. The mixture was stirred for 15 min, to which point **2a** (0.44 g, 2.0 mmol) was added in one portion. Then, the mixture was heated to 70 °C and stirred for 2.0 h. After **2a** was consumed as monitored by TLC, the resulting mixture was poured into saturated aqueous NaCl (50 mL) and extracted with dichloromethane (3 × 20 mL). The combined organic phases were washed with water (3× 20 mL), dried over $MgSO_4$, filtered and concentrated in vacuo. The crude product was purified by flash chromatography (silica gel, petroleum ether: diethyl ether = 8:1) to give a 74% yield of **3a1**: white solid; mp 112−114 °C; ^{1}H NMR (300 MHz, $CDCl_3$) δ 2.44 (s, 3H), 5.85 (s, 1H), 6.88 (s, 1H), 7.24 (d, J = 8.0 Hz, 2H), 7.35 (d, J = 8.0 Hz, 2H), 8.17 (s, 1H), 8.52 (s, 1H); ^{13}C NMR (75 MHz, $CDCl_3$) δ 21.6, 121.3, 123.3, 126.9, 130.7, 131.4, 134.4, 140.3, 150.5, 152.7, 158.8; IR (KBr, neat) 3140, 3064, 2923, 1679, 1579, 1512, 1371, 1232, 892, 820 cm^{-1}. Anal. Calcd for $C_{13}H_{11}ClN_2O$: C, 63.29; H, 4.49; N, 11.36. Found: C, 62.91; H, 4.29; N, 11.08.

ASSOCIATED CONTENT

Supporting Information. Experimental details, spectral and analytical data, and copies of ^{1}H NMR and ^{13}C NMR spectra for compounds **2**−**6**. This material is available free of charge via the Internet at http://pubs.acs.org.

AUTHOR INFORMATION

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