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Construction of Dihydropyrido[2,3-*d*]pyrimidine Scaffolds via Aza-Claisen Rearrangement Catalyzed by *N*-Heterocyclic Carbenes

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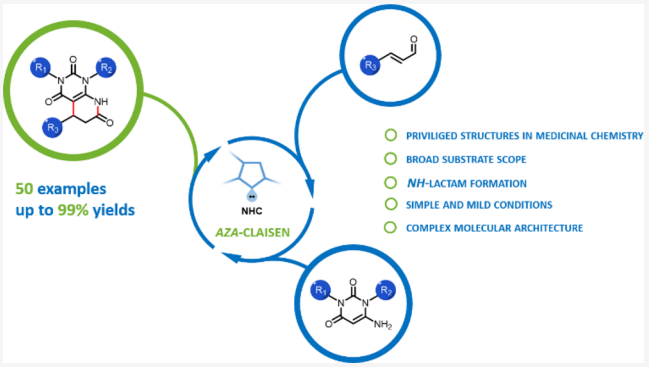
ABSTRACT: *N*-Heterocyclic carbenes (NHCs) catalyzing aza-Claisen rearrangement of α,β-unsaturated enals with cyclic vinylogous amides under oxidative conditions generating potentially biologically active dihydropyridinone-fused uracils have been developed. This strategy represents a unique NHC-activation-based path with the use of 6-aminouracils as stable α,β-diEWG cyclic vinylogous amides for the efficient synthesis of bicyclic *N*-unprotected lactams similar to those in many useful drugs.



INTRODUCTION

The development of new synthetic methodologies for the efficient construction of bioinspired targets constitutes a highly relevant and rapidly developing field in contemporary organic chemistry.[1] Among them, dihydropyridinones and derivatives of bicyclic pyridinone-fused uracils constitute privileged heterocyclic scaffolds found in many natural active molecules displaying a wide range of biological and pharmacological properties. For instance, GSK1120212 **1** and its analogues **2** and **3** were found to be highly potent and selective inhibitors of mitogen-activated protein kinase/extracellular signal-regulated kinase kinase (MEK) as a highly antiproliferative drug candidate for clinical development. Structurally similar dihydropyrido[2,3-*d*]pyrimidine **4** is commonly investigated as a safe and highly effective antileishmanial pharmaceutical (Figure 1).[2] In view of the therapeutic significance of these uracil derivatives, the development of general and concise synthetic strategies that fill the chemical space with diverse heterocyclic structures is highly desirable. Consequently, synthesis of new bicyclic dihydropyridinone-fused uracils gives hope to discover novel biologically active compounds.

N-Heterocyclic carbenes (NHCs) have emerged as versatile organocatalysts for various transformations and constitute convenient methods for carbon−carbon and carbon−heteroatom bond formation.[3] In the recent years, processes in which NHC-catalyzed reactions take place via normal-polarity intermediates have gained importance. There are four well-studied paths of generating α,β-unsaturated acyl azoliums: the reactions of NHCs with α,β-unsaturated enol esters or acyl fluorides,[4] ynals,[5] 2-bromoenals,[6] or stoichiometric oxidation of Breslow intermediates.[7]

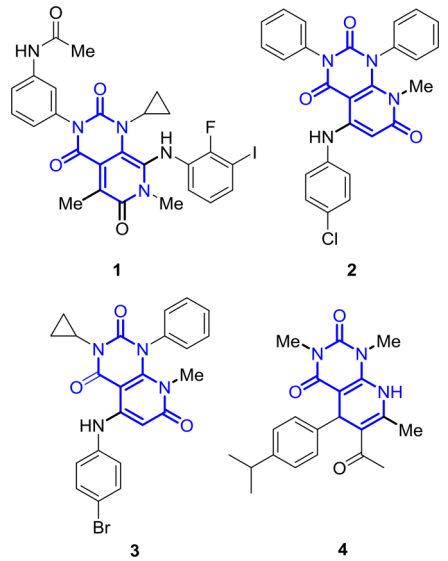


Figure 1. Importance of dihydropyridinone-fused uracil structural motifs.

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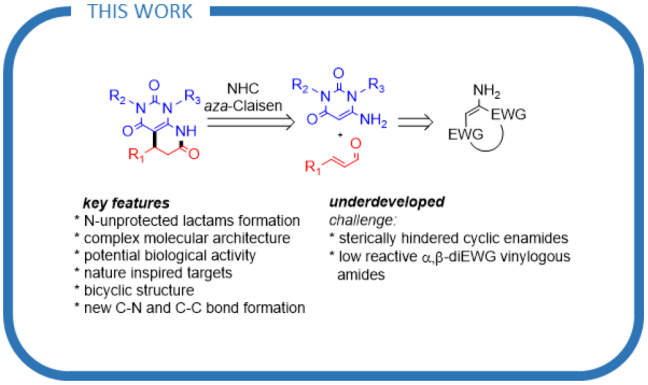
The reactions of α,β-unsaturated acyl azoliums with nucleophiles are unique organocatalytic strategies leading to annulative reactions with single, double, and even triple functionalization via domino/cascade processes.[3d,8] NHC-catalyzed annulations of acyl species with various *N*-protected imine derivatives are well recognized and provide facile entry to diverse heterocyclic motifs.[9] Therefore, many methods have been developed for the construction of *N*-protected dihydropyridinones, including reactions of various precursors of α,β-unsaturated acyl azoliums derived from α-bromoenals or enals under oxidative conditions with different partners such as *N*-Ts-2-aminoacrylates and cyclic and acyclic *N*-sulfonylimines.[10] Very recently, Enders et al. disclosed the use of benzoxazolyl and benzothiozolyl acetates as enamine precursors leading to dihydrobenzoxazolyl- or benzothiozolyl-fused dihydropyridinones with low-to-moderate enantioselectivity.[11] Bode et al. examined the ability of β-electron-withdrawing substituted enamines in the synthesis of useful dihydropyridinones from α,β-unsaturated acyl azoliums.[12] In 2017, Ye et al. utilized indolin-2-imines as enamine precursors allowing the synthesis of nonenantioselective dihydropyridinone-fused indoles.[13] Another interesting approach for their synthesis was the application of azolium dienolate intermediates with hydrazones as reported by Chi and co-workers.[14] The synthetic utility of the immediate annulation products, however, is diminished by its difficulty for use to remove protecting groups. Despite the fact of dynamically developing catalysis involving NHCs in the last few years, organocatalytic aza-Claisen rearrangements are still rare as the reported reactions are strongly limited in scope. To our knowledge, there is no organocatalytic approach for the annulative one-pot strategy for construction of bicyclic *N*-unprotected dihydropyrido[2,3-*d*]pyrimidine derivatives involving the use of sterically hindered α,β-diEWG cyclic vinylogous amides as a template for uracil functionalization (Scheme 1). Our literature survey did not lead to any precedence, either metal- or organocatalyzed, on synthesis 7. Consequently, the development of an effective synthetic method for 7, possibly metal-free and organocatalytic, is highly expected.

Scheme 1. Synthesis of Bicyclic *NH* Heterocycles by an NHC-Catalyzed Annulation Reaction

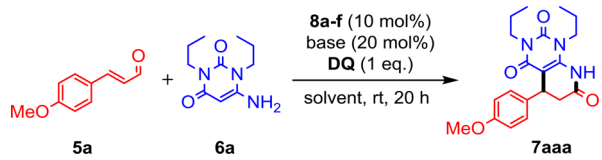


Given the significance of both the bicyclic pyrido[2,3-*d*]pyrimidine derivatives and acyl azolium chemistry, studies on the development of a general synthetic strategy leading to the novel dihydropyridinones bearing a fused uracil moiety were undertaken. Herein, we report our results on this NHC-catalyzed aza-Claisen rearrangement leading to bicyclic dihydropyridinones. The reaction reveals the new reactivity of stable α,β-diEWG cyclic vinylogous amides, which, to date, have not been used as substrates for Claisen rearrangement under oxidative conditions, introducing a new tool for the synthesis of complex bicyclic molecules.

RESULTS AND DISCUSSION

We started the evaluation of our hypothesis by combining 4-methoxycinnamaldehyde **5a** with 1,3-disubstituted 6-aminouracil **6a** using different azolium salts as NHC precursors, tribasic potassium phosphate as a base, and **DQ** as an equimolar oxidant in toluene. To our great delight, all tested NHC precatalysts (**8a**–**8f** displayed remarkable effects on the outcome of the reaction and afforded the desired product in a wide range of yields (Table 1). Gratifyingly, the desired dihydropyrido[2,3-*d*]-pyrimidine **7aaa** could be obtained in a 95% yield when the precatalyst **8c** was employed (Table 1, entry 3). Encouraged by this promising result, various reaction parameters were further examined. All the tested organic and inorganic bases, including NMM, HMPA, and AcOK, gave the desired product, albeit in low yields (entries 7, 8, and 10).

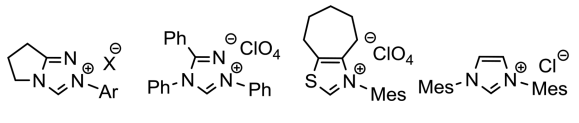
Table 1. Optimization of the Reaction Conditions[a,b,c]



5a 6a 7aaa

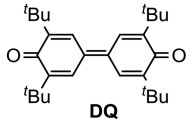
entry	preNHC	solvent	base	yield[c] (%)
1	**8a**	toluene	K_3PO_4	54
2	**8b**	toluene	K_3PO_4	50
3	**8c**	toluene	K_3PO_4	95
4	**8d**	toluene	K_3PO_4	84
5	**8e**	toluene	K_3PO_4	48
6	**8f**	toluene	K_3PO_4	35
7	**8c**	toluene	NMM	30
8	**8c**	toluene	HMPA	21
9	**8c**	toluene	*t*BuOK	88
10	**8c**	toluene	AcOK	54
11	**8c**	toluene	Cs_2CO_3	79
12	**8c**	toluene	P_2-Et	64
13	**8c**	MTBE	K_3PO_4	90
14	**8c**	DCM	K_3PO_4	67
15	**8c**	AcOEt	K_3PO_4	85
16	**8c**	1,4-dioxane	K_3PO_4	43

[a]



8a - Ar = Ph
8b - Ar = C_6F_5
8c - Ar = Mes

8d **8e** **8f**



DQ

[b]Unless otherwise noted, all reactions were carried out with preNHC **8a**–**8f** (10 mol %), base (20 mol %), **DQ** (1.0 equiv), **5a** (0.3 mmol), and **6a** (0.3 mmol) in the solvent (3.0 mL) at rt for 20 h. [c]Isolated yield.

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Interestingly, when the reaction was carried out using bases, such as *tert*-BuOK, KHMDS, and tribasic potassium phosphate, the reactivity increased significantly, and the results indicated that K_3PO_4 was the best choice and furnished the desired product in a 95% yield. Moreover, the change of solvents did not improve the reaction performance, and toluene was proven to be the solvent of choice.

With the optimized reaction conditions in hand, we set out to explore the generality of the procedure in terms of substrates. Initially, a variety of substituted cinnamaldehyde-derived enals including those bearing electron-withdrawing and electron-donating substituents were explored under the optimized conditions. As shown in Scheme 2, a number of aryl-substituted cinnamaldehydes reacted smoothly and cleanly afforded the corresponding annulation products in high yields (**7aaa**−**7iaa**). Notably, the extension of the protocol to alkyl-substituted enals was also successful and gave the desired dihydropyrido[2,3-*d*]pyrimidines in good yields (**7kba**−**7sba**). Unfortunately, (*E*)-3-dimethylaminoacrylaldehyde was unreactive in this model reaction (**7tba**). It is worth noting that aromatic substituents on the enals afforded the expected adducts in greater yields than those of their aliphatic counterparts. Additionally, we also applied modifications in the uracil moiety by replacing the alkyl group in the *N*(3) position with a benzyl substituent in order to make them visible during chromatographic purification with a UV detector.

Encouraged by the above successful aza-Claisen rearrangement, we decided to further increase the attractiveness of the methodology by using unsymmetrical *N*,*N*-disubstituted alkyl−alkyl and alkyl−aryl derived 6-aminouracils (Scheme 3). The influence of the length of the alkyl group in the *N*(3) position was first investigated.

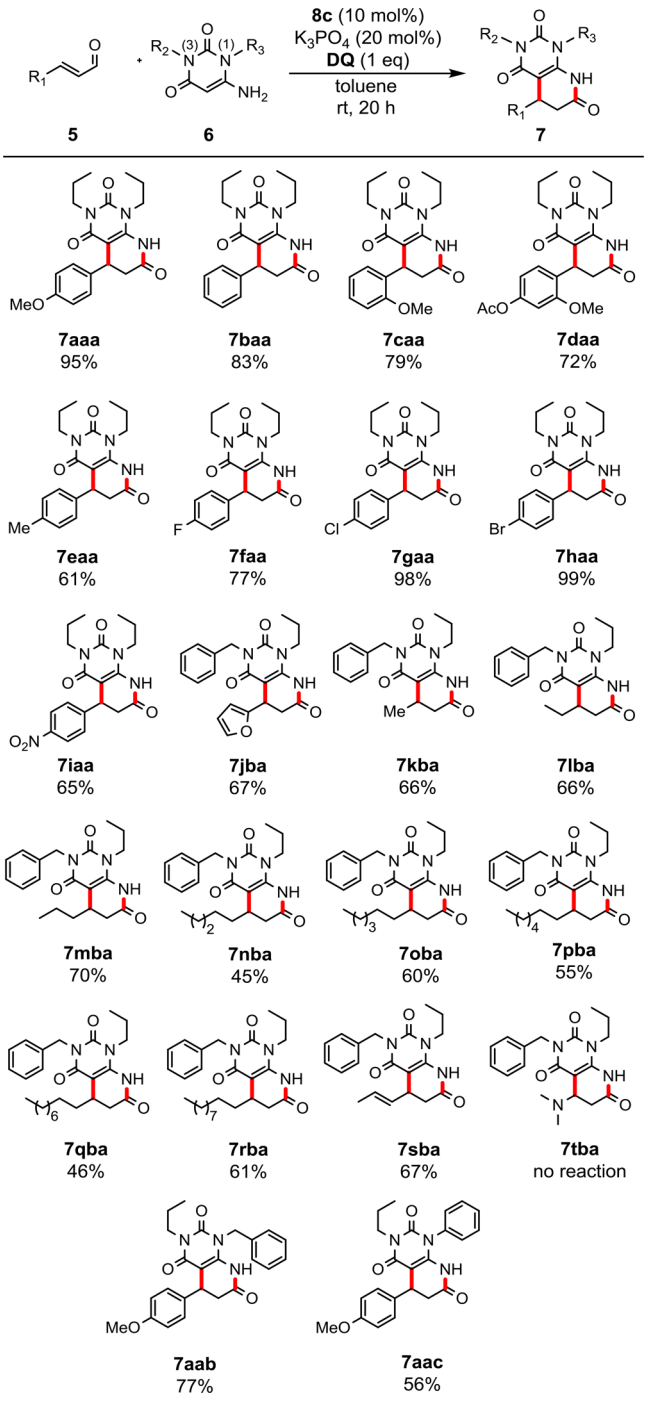
Pleasingly, linear or branched alkyl chains did not affect the reaction outcome, and we were able to isolate the products in good yields (**7afd** and **7aca**−**7aja**). Increasing steric hindrance at the *N*(1) position does not affect the reaction efficiency (**7abc** and **7ajb**). Alkyl chains containing aryl moieties with both electron-poor and electron-rich substituents at various positions of the aromatic ring were well tolerated and gave the desired adducts (**7abb** and **7alb**−**7aub**). The significant influence on the yield was observed for *p*-substituted benzyl groups (products **7anb** and **7aub**).

To demonstrate the scalability of this aza-Claisen transformation, we also performed the reaction in a 50-fold larger scale. Under the optimized reaction condition, the annulation proceeded smoothly and gave the bicyclic heterocycle in a 57% yield (3.17 g) (Scheme 4).

Furthermore, we also were interested in the development of an enantioselective approach to the synthesis of dihydropyridinone-fused uracils (Table 2). After extensive investigations, we found that the chiral pinene-derived NHC precatalyst (**8g**) developed by us gave the best results in terms of enantioselectivity.[15] Unfortunately, the introduction of additives in the form of Lewis acids or Brønsted acids did not have a positive effect on the increase of enantiomeric excess (entries 8−14, Table 2).

The proposed catalytic cycle is presented in Scheme 5. First, the NHC organocatalyst **I** is generated by deprotonation of the triazolium salt **8**. The nucleophilic addition of a free NHC **I** to the enal gives the Breslow intermediate **II**, which is oxidized to form the key α,β-unsaturated acyl azolium **III**. Afterward, the 1,2-addition of cyclic enamine to the acyl azolium gives an *N*-acylation product. The obtained hemiaminal **IV** undergoes aza-Claisen rearrangement via transition state **V**. This rearrangement is followed by the intramolecular lactamization and affords the dihydropyridinone-fused uracil and catalyst turnover. Another possibility for this NHC-catalyzed annulation is nucleophilic addition of uracil enamine to the α,β-unsaturated acyl azolium intermediate as a Michael acceptor in a 1,4-fashion, providing the enol intermediate **VIII**, which undergoes proton transfer and intramolecular acylation to afford the final product. One of the characteristic experimental observations of 1,2-addition is the presence of an amidation side-reaction product. The occurrence of this type of by-product depends on

Scheme 2. Scope of the α,β-Unsaturated Aldehydes and *N*(3)-Substituted 6-Aminouracils



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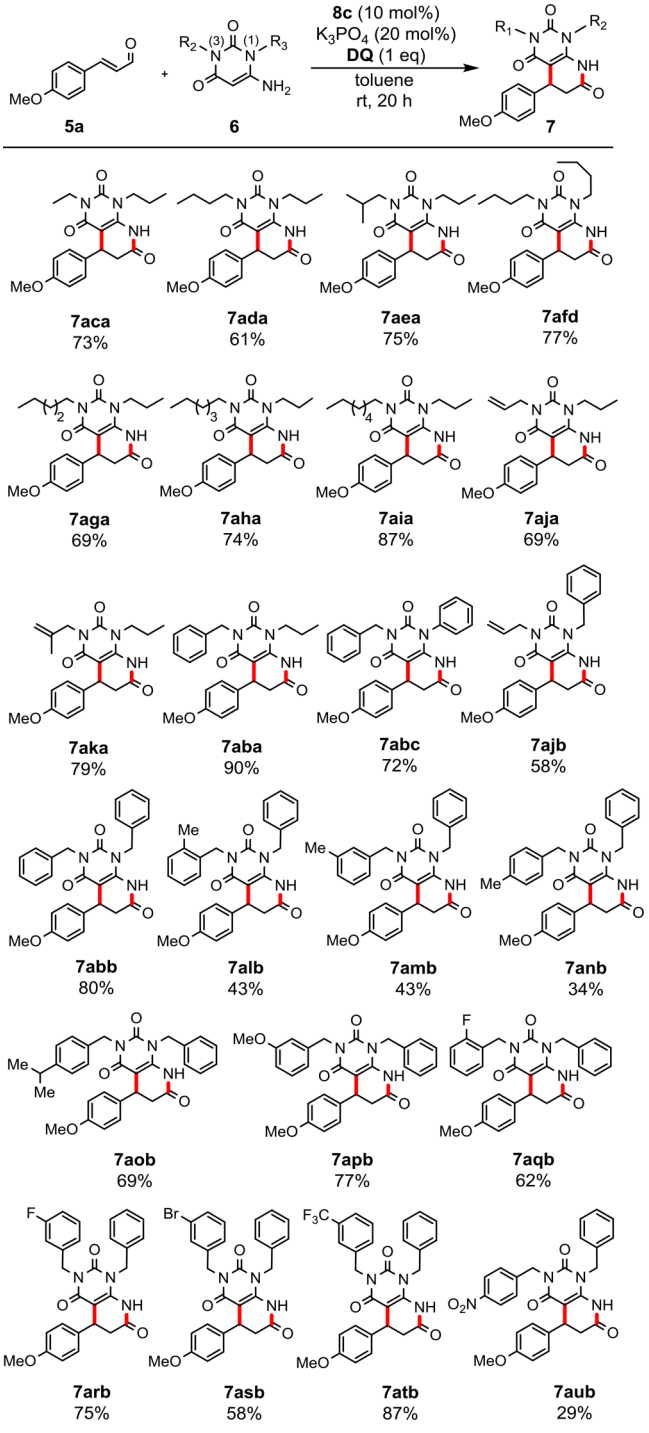
Scheme 3. NHC-Catalyzed Aza-Claisen Rearrangement: $N(1)$ and $N(3)$ Substituent Variation



the construction of the NHC catalyst. In our research, for selected NHC catalysts, amidation side reactions were also observed, which indicates that the course of the reaction is not via nucleophilic 1,4-addition of enamine to the catalytically generated α,β-unsaturated acyl azolium but through 1,2-addition to give an *N*-acylation product. Intensive mechanistic and kinetic investigations for α,β-unsaturated acyl azoliums conducted by Bode and co-workers confirmed that NHC-catalyzed annulation reactions of α,β-unsaturated acyl azoliums proceed through Claisen rearrangement rather than direct Michael addition.[16] Based on theoretical and kinetic studies,

Scheme 4. Scale-Up of the NHC-Catalyzed Aza-Claisen Rearrangement and Enantioselective Approach to the Synthesis of Dihydropyrido[2,3-*d*]pyrimidine 7aaa

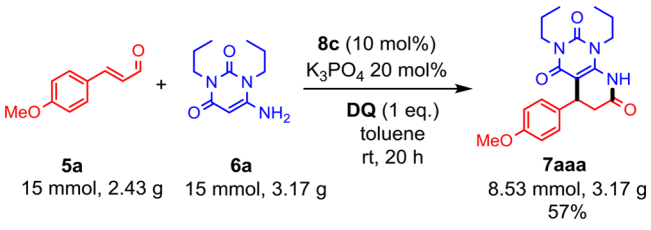
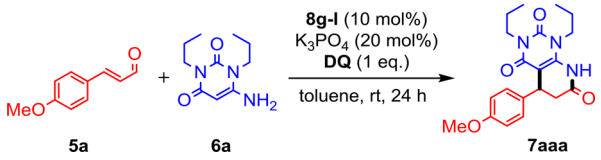
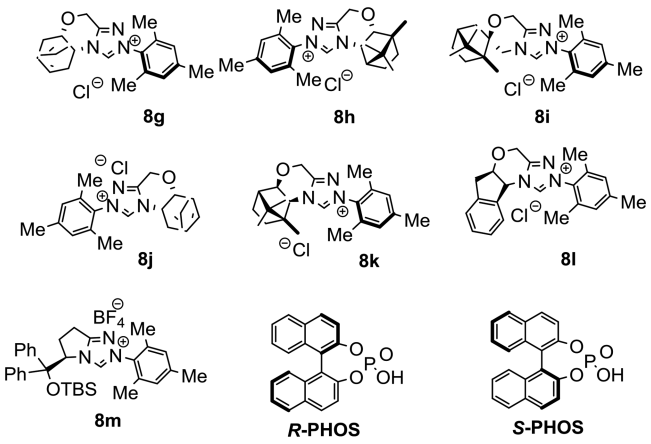


Table 2. Enantioselective Synthesis of Dihydropyridinone 7aaa[a,b,c]



entry	preNHC	additive	yield[c] (%)	ee
1	**8g**		84	67
2	**8h**		76	38
3	**8i**		86	30
4	**8j**		78	16
5	**8k**		43	36
6	**8l**		67	22
7	**8m**		49	30
8	**8g**	$Sc(OTf)_3$		
9	**8g**	LiCl	38	65
10	**8g**	$Ti(OPr^i)_4$	13	
11	**8g**	$Mg(OTf)_2$		
12	**8g**	AcOH	38	67
13	**8g**	*R*-Phos	43	67
14	**8g**	*S*-Phos	45	67

[a]



[b]Unless otherwise noted, all reactions were carried out with preNHC **8g–8l** (10 mol %), K_3PO_4 (20 mol %), **DQ** (1.0 equiv), **5a** (0.3 mmol), and **6a** (0.3 mmol) in toluene (3.0 mL) at rt for 24 h. [c]Isolated yield.

we have adopted the above reaction mechanism as the most likely for our model reaction.[5a2,17] Moreover, it is worth noting that [3 + 3] cycloaddition is rather typical for analogous aminocatalytic reactions.[18]

Finally, the versatile functionalization of the dihydropyrido-[2,3-*d*]pyrimidines was also carried out (Scheme 6). Treating

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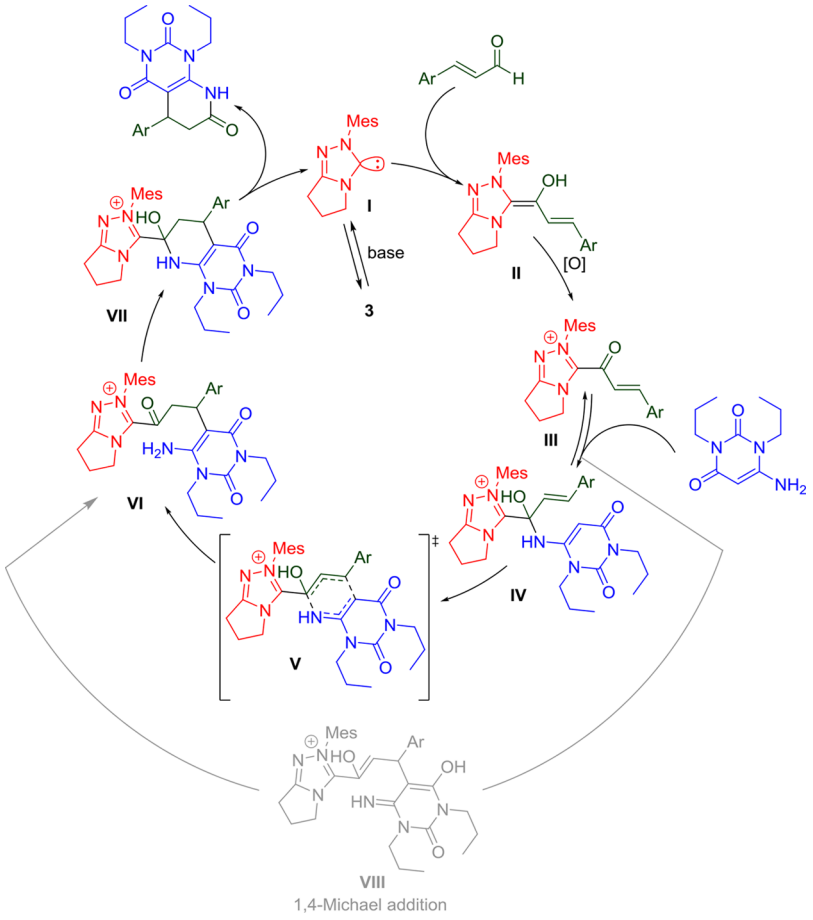
Scheme 5. Proposed Mechanism



7aaa with $LiAlH_4$ led to the corresponding tetrahydropyridine **9** motif. Derivatization of the N−H bond in the lactam motif gave a series of highly interested results. The reaction of **7aaa** with tosyl chloride in pyridine under mild conditions afforded the pyridinium zwitterion **10** in a 20% yield. The replacement of the pyridine with a Hünig base results in tosylation of the amide at the oxygen of the carbonyl group and aromatization of the pyridinone framework. Equally intriguing results were obtained using the benzylation reaction. It turns out that depending on the benzyl halide used, the reaction leads to two different products. The use of benzyl bromide with potassium carbonate in DMF at 80 °C under an inert atmosphere leads to the formation of an *O*-benzylated product (**12**) in a 63% yield. This effect can be explained by the large steric hindrance at the nitrogen atom of the amide group. Nevertheless, the replacement of bromide with benzyl chloride under analogous reaction conditions gives an *O*-benzylation product with simultaneous aromatization of the pyridinone motif **13**. The impacts of factors on aromatization have not yet been fully understood. The influence of the chloride anion may have a decisive role, but further intensive research in this direction is needed.

CONCLUSIONS

In summary, we have developed a very efficient methodology for the synthesis of bicyclic dihydropyridinone-fused uracils from 1,3-disubstituted 6-aminouracils as a α,β-diEWG cyclic vinylogous amide template with various α,β-unsaturated aldehydes using a unique NHC-activation-based approach via aza-Claisen rearrangement. Furthermore, the protocol also allows access to the structural units that are difficult to prepare by traditional strategies. The mild reaction conditions, very broad reaction scope, and readily available substrates make this protocol potentially useful for the construction of dihydropyrido[2,3-*d*]pyrimidine derivatives in good yields. This work contributes the first use of 6-aminouracils as a cyclic enamide template, thus demonstrating the possibility of this novel approach to the design of biologically relevant molecules. Further studies toward a more sustainable synthesis for condensed cyclic heterocycles are currently an area of focus in our laboratory.

EXPERIMENTAL SECTION

General Information. Presented reactions were carried out in dry glassware under an inert atmosphere of argon. Selected reactions were monitored using thin-layer chromatography (TLC), which was visualized under a UV lamp (254 nm). Anhydrous solvents were prepared using an INERT PureSolv solvent purification system. Purification of selected products was performed by column chromatography using a CombiFlash Rf + Lumen system with UV−vis and ELSD detectors. RediSepRf GOLD 4 gram columns were used. NMR spectra were recorded on a Bruker AMX 400 [400 MHz (^{1}H)] spectrometer and Bruker AMX 700 [700 MHz (^{1}H)] spectrometer using $CDCl_3$ and DMSO-d_6 as solvents. Chemical shifts are reported in ppm using the residual solvent peaks as reference: $CDCl_3$ (δ 7.24) or DMSO-d_6 (δ 2.54) for ^{1}H NMR and relative to the central $CDCl_3$ (δ 77.23) or DMSO-d_6 (δ 41.23) resonance for ^{13}C NMR. Coupling constants (J) are provided in Hertz. Infrared spectra were registered on an Alpha FT-IR spectrometer from Bruker with an ATR module. Mass spectra were recorded on an Agilent 6530 Q-TOF

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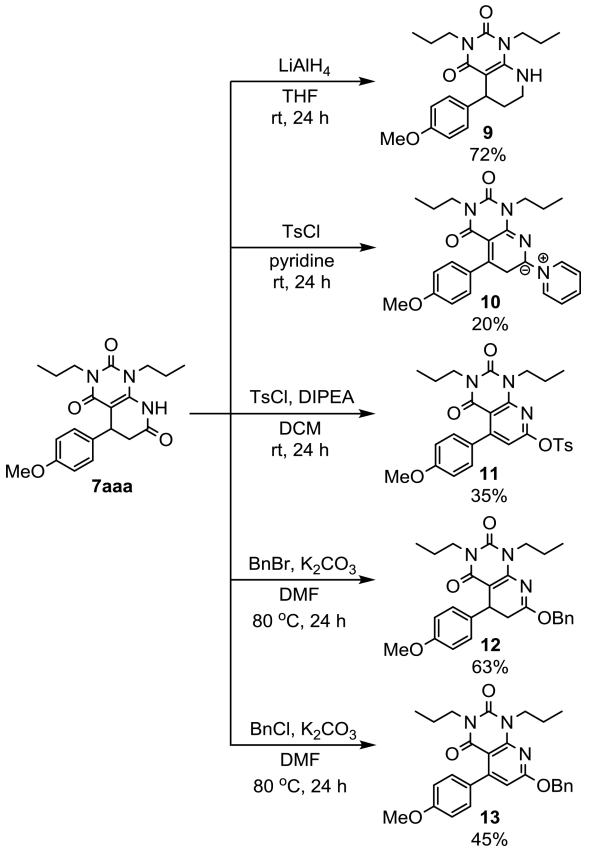
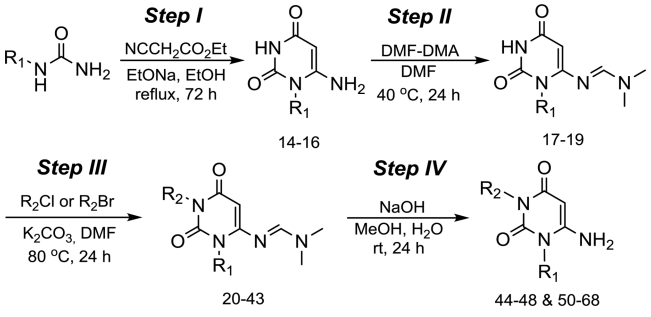
Scheme 6. Synthetic Transformations of the Product 7aaa



LC/MS system coupled with a 1290 Infinity II liquid chromatograph. Melting points of obtained products were measured on a Stuart SMP30 and SMP50 melting point apparatus and were not corrected.

General Procedure for the Preparation of 1,3-Disubstituted 6-Aminouracils. *Step I. General Procedure 1: Synthesis of 3-Unprotected 6-Aminouracils.* Sodium ethoxide solution was prepared from sodium metal (2 equiv) and dry ethanol (2 M solution). To the resulting solution, 1-monosubstituted urea (1 equiv) and ethyl cyanoacetate (1 equiv) were added. The flask with the resulting mixture was immersed in an oil bath and heated under reflux for 72 h. After this time, the solvent was removed using a rotary evaporator, and the residual solid was dissolved in water. The obtained alkaline solution was neutralized by hydrochloric acid, and the precipitated solid was filtered off and washed with water and diethyl ether. The obtained solid was dried under vacuo (0.1 Torr, 2 h).



Step II. General Procedure 2: Synthesis of NH_2-Protected 6-Aminouracils. To the solution of 3-unprotected 6-aminouracil (1 equiv) in DMF (0.6 M solution), DMF-DMA (1.1 equiv) was added. The flask with the resulting mixture was immersed in an oil bath and heated at 40 °C for 24 h. After this time, the solution was cooled down to room temperature, and diethyl ether was added. The precipitated solid was filtered off and washed with diethyl ether. The obtained product was dried under vacuum (0.1 Torr, 2 h).

Step III. General Procedure 3: Synthesis of 1,3-Disubstituted NH_2-Protected 6-Aminouracils. NH_2-protected 6-aminouracil (1 equiv) was dispersed in DMF (0.3 M solution), and potassium carbonate (2 equiv) was added. The corresponding alkyl or benzyl halide was added (10 equiv), and the flask with the resulting mixture was immersed in an oil bath and heated at 80 °C for 24 h. The majority of the solvent and unreacted halide was evaporated. To the residue, water was added, and the precipitated solid was filtered off and washed with diethyl ether. The obtained product was dried in vacuo (0.1 Torr; 2 h).

Step IV. General Procedure 4: Synthesis of 1,3-Disubstituted 6-Aminouracils. 1,3-Disubstituted NH_2-protected 6-aminouracil (1 equiv) was dissolved in methanol (0.2 M solution). To the resulting solution, 2 M sodium hydroxide solution (2 equiv) in water was added. The resulting mixture was stirred at room temperature for 24 h. After this time, methanol was evaporated and water was added. The resulting mixture was stirred at 0 °C for 1 h. The precipitated product was filtered off and washed with water and diethyl ether. The obtained solid was dried under vacuum (0.1 Torr; 2 h).

6-Amino-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (14). A scale of 50 mmol, solid, 5.97 g, isolated yield of 71%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.27 (s, 1H), 6.78 (s, 2H), 4.53 (s, 1H), 3.68 (t, J = 7.6 Hz, 2H), 1.51 (sxt, J = 7.5 Hz, 2H), 0.86 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.9, 156.2, 151.7, 75.7, 42.6, 21.3, 11.2; IR ν_{max}: 3363, 3190, 2962, 1705, 1651, 1574, 1502, 1463, 1386, 1280, 1223, 1170, 1072, 1025, 851, 774, 726, 702, 649, 539, 519, 465 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_7H_{12}N_3O_2$ 170.0930; found: 170.0928; mp 259.3−262.3 °C. The above analysis results correspond to the literature data.[19a]

6-Amino-1-benzylpyrimidine-2,4(1*H*,3*H*)-dione (15). A scale of 57.3 mmol, solid, 10.71 g, isolated yield of 86%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.41 (m, 1H), 7.34 (m, 2H), 7.26 (m, 1H), 7.21 (m, 2H), 6.82 (s, 2H), 5.04 (s, 2H), 4.61 (s, 4H); ^{13}C NMR{^{1}H} (101 MHz, DMSO-d_6) δ 162.8, 156.3, 152.0, 137.1, 128.9, 127.6, 126.8, 76.0, 44.0; IR ν_{max}: 3471, 3329, 3250, 3070, 2954, 2766, 1695, 1628, 1576, 1494, 1401, 1385, 1358, 1278, 1232, 1175, 1124, 941, 891, 822, 729, 692, 592, 538, 510, 434 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{11}H_{12}N_3O_2$ 218.0930; found: 218.0931; mp 270.5−273.1 °C (dec.). The above analysis results correspond to the literature data.[19b]

6-Amino-1-phenylpyrimidine-2,4(1*H*,3*H*)-dione (16). A scale of 50 mmol, solid, 6.65 g, isolated yield of 65%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 10.48 (s, 1H), 7.45−7.57 (m, 3H), 7.27−7.37 (m, 2H), 6.11 (s, 2H), 4.69 (s, 1H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 163.3, 156.1, 151.4, 134.6, 130.2, 129.9, 129.7, 75.5; IR ν_{max}: 3476, 3331, 3086, 2971, 2798, 2776, 1705, 1623, 1583, 1472, 1430, 1384, 1296, 1222, 1144, 875. 804, 777, 761, 702, 566, 531, 510 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{10}H_{10}N_3O_2$ 204.0773; found: 204.0773; mp 321.9−323.7 °C (dec.). The above analysis results correspond to the literature data.[19c]

***N*′-(2,6-Dioxo-3-propyl-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (17).** A scale of 30 mmol, solid, 5.48 g, isolated yield of 81%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 10.58 (s, 1H), 8.05 (s, 1H), 4.96 (s, 1H), 3.81 (t, J = 7.4 Hz, 2H), 3.10 (s, 3H), 2.97 (s, 3H), 1.52 (sxt, J = 7.4 Hz, 2H), 0.82 (t, J = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 163.3, 160.6, 155.9, 151.7, 82.3, 42.8, 40.3, 34.4, 21.6, 11.2; IR ν_{max}: 3152, 3016, 2965, 1659, 1627, 1557, 1445, 1421, 1395, 1356, 1331, 1230, 1111, 865, 604, 535, 432 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{10}H_{17}N_4O_2$ 225.1352; found: 225.1350; mp 217.6−220.7 °C. The above analysis results correspond to the literature data.[19d]

***N*′-(3-Benzyl-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (18).** A scale of 48.7 mmol, solid, 10.84 g, isolated yield of 82%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.07 (s, 1H), 7.19−7.33 (m, 5H), 5.10 (s, 2H), 5.04 (s, 1H), 3.09 (s, 3H), 2.94 (s, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 163.7, 160.8, 156.5, 152.3, 139.0, 128.7, 127.6, 127.3, 82.6, 44.7, 40.8, 35.0; IR ν_{max}: 2976, 2784, 1694, 1646, 1611, 1549, 1491, 1452, 1427, 1389, 1329,

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1256, 1215, 1152, 1121, 1088, 1060, 1019, 904, 876, 813, 787, 760, 741, 701, 600, 583, 516, 462, 431 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{17}N_4O_2$ 273.1352; found: 273.1351; mp 221.7−223.4 °C. The above analysis results correspond to the literature data.[19d]

***N′*-(2,6-Dioxo-3-phenyl-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (19).** A scale of 24.2 mmol, solid, 5.54 g; isolated yield of 89%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 10.79 (s, 1H), 7.96 (s, 1H), 7.36−7.40 (m, 2H), 7.30−7.34 (m, 1H), 7.14−7.17 (m, 2H), 5.09 (d, J = 1.9 Hz, 1H), 2.98 (s, 3H), 2.51 (s, 2H); ^{13}C{1H} NMR (176 MHz, DMSO-d_6) δ 163.6, 161.0, 155.2, 151.5, 137.1, 129.5, 128.2, 127.5, 82.3, 40.1, 33.9; IR ν_{max}: 2963, 2808, 1698, 1650, 1615, 1541, 1446, 1425, 1373, 1337, 1261, 1210, 1130, 1087, 879, 787, 736, 698, 588, 568, 532, 458, 424 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{13}H_{15}N_4O_2$ 259.1195; found: 259.1196; mp 276.1−279.0 °C. The above analysis results correspond to the literature data.[19d]

***N′*-(3-Benzyl-2,6-dioxo-1-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (20).** A scale of 21 mmol, used halide: iodide, solid, 4.86 g, isolated yield of 74%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.09 (s, 1H), 7.18−7.33 (m, 7H), 5.18 (s, 1H), 5.16 (s, 2H), 3.73 (t, J = 7.3 Hz, 2H), 3.09 (s, 3H), 2.94 (s, 3H), 1.51 (sxt, J = 7.4 Hz, 3H), 0.83 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.5, 159.2, 156.5, 152.4, 138.9, 128.7, 127.6, 127.3, 82.2, 45.6, 42.0, 40.8, 35.0, 21.2, 11.6; IR ν_{max}: 2961, 1690, 1648, 1618, 1569, 1496, 1448, 1414, 1363, 1224, 1125, 1096, 996, 810, 765, 693, 602, 577, 524 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{17}H_{23}N_4O_2$ 315.1821; found: 315.1821; mp 130.2−132.8 °C. The above analysis results correspond to the literature data.[19d]

***N′*-(2,6-Dioxo-3-phenyl-1-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (21).** A scale of 19.3 mmol, used halide: iodide, solid, 3.17 g, isolated yield of 55%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.99 (s, 1H), 7.37−7.40 (m, 3H), 7.18 (dd, J = 3.7, 1.5 Hz, 2H), 5.25 (s, 1H), 3.74 (t, J = 7.3 Hz, 2H), 2.99 (s, 6H), 1.54 (sxt, J = 7.4 Hz, 2H), 0.85 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.9, 159.8, 155.6, 152.1, 137.9, 129.9, 129.8, 128.7, 82.2, 42.0, 34.3, 21.3, 11.7; IR νmax: 2953, 1696, 1651, 1612, 1572, 1424, 1406, 1375, 1341, 1116, 1079, 1005, 877, 798, 764, 700, 577, 536 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{16}H_{21}N_4O_2$ 301.1665; found: 301.1662; mp 183.7−186.7 °C. The above analysis results correspond to the literature data.[19d]

***N′*-(1-Ethyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (22).** A scale of 4.46 mmol, used halide: iodide, solid, 0.35 g, isolated yield of 31%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.07 (s, 1H), 5.11 (s, 1H), 3.85−3.93 (m, 2H), 3.79 (q, J = 6.9 Hz, 2H), 3.11 (s, 3H), 2.99 (s, 3H), 1.49−1.61 (m, 2H), 1.06 (t, J = 7.1 Hz, 3H), 0.84 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.3, 159.4, 156.3, 151.9, 82.3, 44.1, 40.7, 35.4, 34.8, 21.9, 13.5, 11.5; IR ν_{max}: 3088, 2965, 2934, 2875, 1687, 1648, 1614, 1566, 1448, 1416, 1362, 1337, 1233, 1116, 1037, 1001, 887, 815, 767, 666, 571, 551 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{12}H_{21}N_4O_2$ 253.1665; found: 253.1666; mp 142.0−144.3 °C.

***N′*-(1-Butyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (23).** A scale of 4.46 mmol, used halide: bromide, solid, 0.36 g, isolated yield of 29%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.08 (s, 1H), 5.11 (s, 1H), 3.85−3.93 (m, 2H), 3.75 (t, J = 7.1 Hz, 2H), 3.11 (s, 3H), 2.99 (s, 3H), 1.42−1.61 (m, 4H), 1.25 (dq, J = 14.9, 7.4 Hz, 2H), 0.88 (t, J = 7.3 Hz, 3H), 0.83 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.5, 159.4, 156.4, 152.1, 82.3, 44.1, 40.7, 34.8, 30.1, 21.9, 20.1, 14.2, 11.6; IR ν_{max}: 2960, 2932, 2873, 1687, 1645, 1611, 1568, 1494, 1439, 1412, 1365, 1348, 1226, 1181, 1112, 1048, 987, 886, 815, 766, 572, 550 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{25}N_4O_2$ 281.1978; found: 281.1977; mp 129.0−130.2 °C.

***N′*-(1-Isobutyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (24).** A scale of 4.46 mmol, used halide: bromide, solid, 0.74 g, isolated yield of 59%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.09 (s, 1H), 5.11 (s, 1H), 3.90 (t, J = 7.2 Hz, 2H), 3.60 (d, J = 7.3 Hz, 2H), 3.11 (s, 3H), 2.99 (s, 3H), 2.00 (dt, J = 13.8, 6.9 Hz, 1H), 1.55 (sxt, J = 7.3 Hz, 2H), 0.79−0.85 (m, 9H), ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.8, 159.34, 156.4, 152.4, 82.2, 47.2, 44.1, 40.7, 34.8, 27.1, 21.9, 20.4, 11.5; IR ν_{max}: 2961, 2932, 2874, 1685, 1611, 1566, 1441, 1412, 1364, 1346, 1323, 1226, 1114, 1056, 992, 888, 806, 767, 577, 550 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{25}N_4O_2$ 281.1978; found: 281.1980; mp 133.5−136.7 °C.

***N′*-(2,6-Dioxo-1-pentyl-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (25).** A scale of 4.46 mmol, used halide: bromide, solid, 1.00 g, isolated yield of 76%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.07 (s, 1H), 5.10 (s, 1H), 3.87 (t, J = 7.5 Hz, 2H), 3.72 (t, J = 1.0 Hz, 2H), 3.10 (s, 3H), 2.97 (s, 3H), 1.50−1.56 (m, 2H), 1.47 (dt, J = 15.0, 7.4 Hz, 2H), 1.23−1.30 (m, 4H), 0.80−0.85 (m, 6H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 162.1, 159.0, 156.0, 151.7, 81.8, 67.4, 43.7, 40.3, 34.4, 28.6, 27.1, 21.9, 21.5, 13.9, 11.2; IR ν_{max}: 2956, 2925, 2871, 1691, 1643, 1613, 1566, 1449, 1415, 1501, 1367, 1349, 1258, 1112, 1053, 994, 885, 807, 767, 728, 577, 553, 437 cm^{-1}. HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{15}H_{27}N_4O_2$ 295.2134; found: 295.2134; mp 80.1−86.9 °C.

***N′*-(1-Hexyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (26).** A scale of 4.46 mmol, used halide: bromide, solid, 0.39 g, isolated yield 28%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.07 (s, 1H), 5.10 (s, 1H), 3.86−3.90 (m, 2H), 3.71−3.74 (m, 2H), 3.10 (s, 3H), 2.98 (s, 3H), 1.51−1.56 (m, 2H), 1.45−1.47 (m, 2H), 1.27−1.32 (m, 6 H), 0.81−0.85 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.5, 159.4, 156.3, 152.1, 82.3, 44.1, 34.8, 31.4, 31.1, 27.8, 26.5, 22.4, 21.9, 14.3, 11.5; IR ν_{max}: 3448, 3292, 3171, 2955, 2927, 1693, 1646, 1614, 1569, 1451, 1417, 1354, 1280, 1235, 1184, 1112, 1054, 999, 942, 887, 813, 789, 727, 717, 694, 555 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{16}H_{29}N_4O_2$ 309.2291; found: 309.2294; mp 95.5−99.3 °C.

***N′*-(1-Heptyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (27).** A scale of 4.46 mmol, used halide: bromide, solid, 0.61 g, isolated yield 42%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.07 (s, 1H), 5.10 (s, 1H), 3.11 (s, 3H), 2.99 (s, 2H), 1.51−1.58 (m, 2H), 1.44−1.51 (m, 2H), 1.26−1.31 (m, 8H), 0.81−0.86 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.5, 159.4, 156.3, 152.2, 82.3, 44.1, 34.8, 31.7, 28.6, 27.9, 26.8, 22.5, 21.9, 14.4, 11.6; IR ν_{max}: 2954, 2926, 2857, 1692, 1641, 1618, 1573, 1456, 1416, 1366, 1347, 1260, 1224, 1112, 1060, 993, 888, 807, 766, 572, 549 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{17}H_{31}N_4O_2$ 323.2447; found: 323.2446; mp 70.0−75.9 °C.

***N′*-(1-allyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (28).** A scale of 4.46 mmol, used halide: bromide, solid, 0.36 g, isolated yield of 31%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.09 (s, 1H), 5.79 (ddt, J = 17.2, 10.3, 5.2 Hz, 1H), 5.13 (s, 1H), 5.04 (dd, J = 10.3, 1.3 Hz, 1H), 4.99 (dd, J = 17.2, 1.5 Hz, 1H), 4.35 (d, J = 5.2 Hz, 2H), 3.88 (t, J = 7.3 Hz, 2H), 3.11 (s, 3H), 2.98 (s, 3H), 1.54 (sxt, J = 7.4 Hz, 2H), 0.82 (t, J = 7.4 Hz, 3H). ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.8, 159.2, 156.1, 151.5, 133.4, 115.8, 81.7, 43.8, 42.0, 40.4, 34.4, 21.5, 11.2; IR ν_{max}: 2961, 2932, 2874, 1686, 1617, 1570, 1446, 1418, 1366, 1229, 1116, 994, 811, 767, 552 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{13}H_{21}N_4O_2$ 265.1665; found: 265.1663; mp: 151.1−153.0 °C.

***N*,*N*-Dimethyl-*N′*-(1-(2-methylallyl)-2,6-dioxo-3-propyl-1,2,3,6-tetrahydropyrimidin-4-yl)formimidamide (29).** A scale of 4.46 mmol, used halide: chloride, solid, 0.96 g, isolated yield of 77%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.10 (s, 1H), 5.14 (s, 1H), 4.68−4.74 (m, 1H), 4.46 (s, 1H), 4.28 (s, 2H), 3.90 (t, J = 7.9 Hz, 2H), 3.12 (s, 3H), 3.00 (s, 3H), 1.67 (s, 3H), 1.55 (dq, J = 14.8, 7.4 Hz, 2H), 0.82 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.3, 159.6, 156.4, 152.0, 141.0, 109.2, 82.0, 45.1, 44.1, 34.8, 21.9, 20.8, 11.5; IR ν_{max}: 3088, 2961, 2934, 2875, 1688, 1645, 1609, 1566, 1497, 1443, 1418, 1393, 1360, 1335, 1322, 1227, 1261, 1281, 1204, 1182, 1115, 1067, 1017, 993, 904, 881, 806, 764, 583, 552, 423 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{23}N_4O_2$ 2279.1821; found: 279.1822; mp 87.4−90.3 °C.

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***N′*-(1-Benzyl-2,6-dioxo-3-propyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (30).** A scale of 7 mmol, used halide: chloride, solid, 1.78 g, 81% yield; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.11 (s, 1H), 7.26−7.29 (m, 2H), 7.19−7.24 (m, 3H), 5.18 (s, 1H), 4.94 (s, 2H), 3.87−3.90 (m, 2H), 3.11 (s, 3H), 2.99 (s, 1H), 1.54 (sxt, J = 7.4 Hz, 2H), 0.82 (t, J = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 162.1, 159.3, 156.1, 151.8, 138.2, 128.3, 127.4, 126.9, 81.7, 43.9, 43.1, 40.4, 34.4, 21.5, 11.2; IR ν_{max}: 2964, 2927, 2870, 1747, 1687, 1640, 1608, 1561, 1497, 1451, 1420, 1349, 1321, 1257, 1226, 1181, 1113, 1062, 1030, 989, 922, 885, 815. 691, 552 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{17}H_{23}N_4O_2$ 315.1821; found: 315.1820; mp 116.6−118.1 °C.

***N′*-(1-Benzyl-2,6-dioxo-3-phenyl-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (31).** A scale of 1.6 mmol, used halide: chloride, solid, 0.37 g, isolated yield of 65%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.02 (s, 1H), 7.17-7.42 (m, 10H), 5.32 (s, 1H), 4.98 (s, 2H), 3.00 (s, 3H), 2.53 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.8, 160.0, 155.8, 152.2, 138.4, 137.8, 129.8, 128.7, 128.0, 128.0, 127.4, 82.1, 43.7, 34.3; IR ν_{max}: 2917, 2951, 1695, 1652, 1613, 1591, 1569, 1490, 1416, 1398, 1377, 1352, 1332, 1302, 1266, 1233, 1137, 1102, 1072, 1060, 1025, 998, 947, 890, 808, 778, 744, 731, 711, 694, 574, 531, 468 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H) calcd for $C_{20}H_{21}N_4O_2$ 349.1665; found: 349.1668; mp 158.2−164.2 °C.

***N′*-(1-Allyl-3-benzyl-2,6-dioxo-1,2,3,6-tetrahydro-pyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (32).** A scale of 3.8 mmol, used halide: bromide, solid, 1.10 g, isolated yield of 96%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.11 (s, 1H), 7.20−7.30 (m, 5H), 5.81 (ddt, J = 17.2, 10.4, 5.1, 5.1 Hz, 1H), 5.21 (s, 1H), 5.16 (s, 2H), 5.04 (dq, J = 2.9, 1.6 Hz, 1H), 4.37 (dt, J = 5.1, 1.5 Hz, 2H), 3.95 (d, J = 7.3 Hz, 1H), 3.09 (s, 3H), 2.94 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.2, 159.4, 156.6, 152.2, 138.8, 133.7, 128.7, 127.6, 127.3, 116.3, 82.0, 45.7, 42.5, 40.8, 35.0; IR ν_{max}: 3080, 3033, 2946, 1689, 1649, 1613, 1567, 1494, 1443, 1417, 1391, 1365, 1323, 1257, 1210, 1122, 1093, 993, 925, 900, 818, 765, 740, 692, 642, 601, 523, 565 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H) calcd for $C_{17}H_{21}N_4O_2$ 313.1665; found: 313.1664; mp 135.0−138.5 °C.

***N′*-(1,3-Dibenzyl-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (33).** A scale of 3.8 mmol, used halide: chloride, solid, 0.88 g, isolated yield of 66%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.13 (s, 1H), 7.17−7.35 (m, 10H), 5.26 (s, 1H), 5.17 (s, 2H), 4.97 (s, 2H), 3.10 (s, 3H), 2.95 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.4, 159.5, 156.7, 152.5, 138.8, 138.4, 128.7, 128.7, 127.8, 127.6, 127.3, 82.1, 45.8, 43.6, 40.8, 35.0; IR ν_{max}: 2965, 2934, 2875, 1687, 1613, 1567, 1448, 1415, 1362, 1338, 1233, 1116, 1001, 815, 767, 570, 551 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{21}H_{23}N_4O_2$ 363.1821; found: 363.1820; mp: 142.3−143.4 °C. The above analysis results correspond to the literature data.[19e]

***N′*-(3-Benzyl-1-(2-methylbenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (34).** A scale of 3.8 mmol, used halide: chloride, solid, 1.25 g, 91%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.16 (s, 1H), 7.19−7.30 (m, 6H), 7.04−7.12 (m, 2H), 6.77 (s, 1H), 5.29 (s, 1H), 5.17 (s, 2H), 4.93 (s, 2H), 3.10 (s, 3H), 2.95 (s, 2H), 2.32 (s, 3H). ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.5, 159.6, 156.7, 152.5, 138.8, 136.1, 135.4, 130.3, 128.7, 127.5, 127.3, 126.8, 126.2, 124.9, 82.1, 45.8, 41.4, 35.0, 19.2 IR ν_{max}: 3030, 2955, 2922, 1694, 1642, 1615, 1562, 1492, 1448, 1423, 1368, 1324, 1258, 1215, 1125, 1094, 998, 903, 813, 759, 741, 694, 603, 574, 525, 475 cm^{-1}. HRMS (ESI-TOF): (M + H)$^+$ calcd for $C_{22}H_{25}N_4O_2$ 377.1978; found: 377.1980; mp 139.4−142.7 °C.

***N′*-(3-Benzyl-1-(3-methylbenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (35).** A scale of 3.8 mmol, used halide: chloride, solid, 0.74 g, isolated yield of 54%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.12 (s, 1H), 7.26−7.30 (m, 2H), 7.19−7.25 (m, 3H), 7.13−7.18 (m, 1H), 6.99−7.04 (m, 3H), 5.24 (s, 1H), 5.16 (s, 2H), 4.92 (s, 2H), 3.09 (s, 3H), 2.94 (s, 3H), 2.24 (s, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 162.0, 159.0, 156.3, 152.1, 138.4, 138.0, 137.3, 128.3, 128.2, 127.8, 127.6, 127.2, 127.0, 124.4, 81.7, 45.3, 43.2, 40.5, 34.6, 21.1; IR ν_{max}: 3061, 3032, 2925, 1692, 1621, 1562, 1490, 1446, 1423, 1386, 1364, 1337, 1280, 1254, 1210, 1128, 1094, 1067, 1015, 919, 899, 792, 764, 699, 597, 570, 522, 468 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{25}N_4O_2$ 377.1978; found: 377.1978; mp 131.9−133.2 °C.

***N′*-(3-Benzyl-1-(4-methylbenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (36).** A scale of 3.8 mmol, used halide: chloride, solid, 1.00 g, isolated yield of 72%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.12 (s, 1H), 7.19−7.34 (m, 5H), 7.05−7.17 (m, 4H), 5.25 (s, 1H), 5.16 (s, 2H), 4.92 (s, 2H), 3.09 (s, 3H), 2.95 (s, 3H), 2.25 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.4, 159.4, 156.7, 152.5, 138.8, 136.4, 135.4, 129.2, 128.7, 127.9, 127.6, 127.3, 82.1, 45.7, 43.4, 40.8, 35.0, 21.1; IR ν_{max}: 2965, 2934, 2875, 1688, 1614, 1566, 1448, 1416, 1363, 1233, 1117, 1001, 814, 767, 570, 551 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{25}N_4O_2$ 377.1978; found: 377.1977; mp 156.6−159.7 °C.

***N′*-(3-Benzyl-1-(4-isopropylbenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (37).** A scale of 3.8 mmol, used halide: chloride, solid, 1.47 g, isolated yield of 99%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.12 (s, 1H), 7.26 (spt, J = 7.1 Hz, 6H), 7.16 (d, J = 1.2 Hz, 3H), 5.25 (s, 1H), 5.16 (s, 2H), 4.93 (s, 2H), 3.09 (s, 3H), 2.95 (s, 3H), 2.82 (s, 1H), 1.17 (d, J = 6.8 Hz, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ (ppm) = 162.4, 159.4, 156.7, 152.5, 147.5, 138.8, 135.8, 133.6, 128.7, 127.9, 127.6, 126.6, 82.1, 45.8, 43.4, 40.8, 35.0, 33.6, 24.4; IR ν_{max}: 2957, 1694, 1638, 1610, 1560, 1510, 1445, 1421, 1407, 1356, 1346, 1295, 1256, 1216, 1125, 1092, 1056, 910, 899, 805, 768, 738, 698, 600, 589, 523 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{24}H_{29}N_4O_2$ 405.2291; found: 405.2294; mp: 129.2−130.6 °C.

***N′*-(3-Benzyl-1-(3-methoxybenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (38).** A scale of 3.8 mmol, used halide: chloride, solid, 0.80 g, isolated yield of 56%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.14 (s, 1H), 7.17−7.32 (m, 6H), 6.75−6.84 (m, 3H), 5.26 (s, 1H), 5.18 (s, 2H), 4.95 (s, 2H), 3.70 (s, 3H), 3.10 (s, 3H), 2.96 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.4, 159.7, 159.5, 156.7, 152.5, 140.0, 138.8, 129.7, 128.7, 127.6, 127.3, 119.8, 113.4, 112.6, 82.1, 55.4, 45.7, 43.6, 40.8, 35.0; IR ν_{max}: 2928, 2832, 1690, 1614, 1561, 1487, 1449, 1425, 1408, 1367, 1335, 1283, 1250, 1211, 1150, 1123, 1092, 1055, 921, 897, 805, 764, 699, 599, 571, 522, 476 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{25}N_4O_3$ 393.1927; found: 393.1927; mp 122.5−123.5 °C.

***N′*-(3-Benzyl-1-(2-fluorobenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (39).** A scale of 3.8 mmol, used halide: chloride, solid, 0.86 g, isolated yield of 61%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.14−8.16 (m, 1H), 7.25−7.30 (m, 3H), 7.19−7.25 (m, 3H), 7.14−7.18 (m, 1H), 7.08−7.12 (m, 1H), 6.98−7.02 (m, 1H), 5.28 (s, 1H), 5.16 (s, 2H), 5.02 (s, 2H), 3.10 (s, 3H), 2.95 (s, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.9, 160.6, 159.2, 156.4, 152.0, 138.3, 128.7 (d, J = 8.2 Hz), 128.3, 127.8 (d, J = 4.1 Hz), 127.2, 127.0, 124.7 (d, J = 14.3 Hz), 124.4 (d, J = 3.3 Hz), 115.2 (d, J = 21.3 Hz), 81.6, 45.4, 40.5, 37.2 (d, J = 5.3 Hz), 34.6; IR ν_{max}: 1693, 1645, 1614, 1579, 1560, 1490, 1441, 1421, 1360, 1327, 1218, 1125, 1093, 813, 756, 701, 600, 524, 427 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{21}H_{22}FN_4O_2$ 381.1727; found: 381.1725; mp 113.0−116.0 °C.

***N′*-(3-Benzyl-1-(3-fluorobenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (40).** A scale of 3.8 mmol, used halide: chloride, solid, 0.99 g, isolated yield of 71%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.14 (s, 1H), 7.19−7.38 (m, 6H), 6.99−7.11 (m, 3H), 5.27 (s, 1H), 5.17 (s, 2H), 4.98 (s, 2H), 3.10 (s, 3H), 2.96 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 163.8, 162.3, 159.6, 156.8, 152.5, 141.4 (d, J = 7.2 Hz), 138.8, 130.7 (d, J = 7.9 Hz), 128.7, 127.6, 127.4, 123.8 (d, J = 2.4 Hz), 114.5 (d, J = 21.5 Hz), 114.2 (d, J = 20.7 Hz), 82.0, 45.8, 43.3, 40.9, 35.0; IR ν_{max}: 1699, 1646, 1614, 1568, 1447, 1415, 1365, 1323, 1246, 1219, 1135, 1093, 985, 941, 878, 811, 765, 691, 603, 525, 465 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H) calcd for $C_{21}H_{22}FN_4O_2$ 381.1727; found: 381.1726; mp 135.8−141.6 °C.

***N′*-(3-Benzyl-1-(3-bromobenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (41).** A scale

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of 3.8 mmol, used halide: chloride, solid, 1.53 g, isolated yield of 94%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.13 (s, 1H), 7.41−7.44 (m, 1H), 7.37−7.39 (m, 1H), 7.19−7.30 (m, 7H), 5.26 (s, 1H), 5.16 (s, 2H), 4.95 (s, 2H), 3.09 (s, 3H), 2.94 (s, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.9, 159.2, 156.4, 152.0, 140.8, 138.3, 130.6, 130.0, 129.9, 128.4, 127.1, 127.0, 126.6, 121.6, 81.6, 45.4, 42.7, 40.5; IR ν_{max}: 2961, 1691, 1643, 1618, 1567, 1495, 1443, 1412, 1354, 1315, 1258, 1220, 1128, 1096, 1066, 987, 926, 842, 803, 765, 692, 613, 602, 520, 459 cm^{-1}. HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{21}H_{22}BrN_4O_2$ 441.0926; found: 441.0928; mp: 182.5−185.1 °C.

N′-(3-Benzyl-2,6-dioxo-1-(3-(trifluoromethyl)benzyl)-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylform-imidamide (42). A scale of 3.8 mmol, used halide: chloride, solid, 1.17 g, isolated yield of 74%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.14 (s, 1H), 7.60 (s, 1H), 7.55 (m, 3H), 7.19−7.29 (m, 5H), 5.28 (s, 1H), 5.17 (s, 2H), 5.05 (s, 2H), 3.09 (s, 3H), 2.94 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.3, 159.6, 156.8, 152.5, 139.9, 138.7, 132.0, 129.9, 129.5 (q, J = 31.8 Hz), 128.7, 127.5, 127.4, 124.0−124.3 (m), 82.0, 45.8, 43.3, 40.9, 35.0; IR ν_{max}: 2962, 1692, 1643, 1619, 1566, 1445, 1415, 1365, 1320, 1196, 1145, 1114, 1095, 1071, 990, 923, 889, 807, 768, 702, 695, 661, 601, 521 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{22}F_3N_4O_2$ 431.1695; found: 431.1694; mp 188.6−191.7 °C.

N′-(3-Benzyl-1-(4-nitrobenzyl)-2,6-dioxo-1,2,3,6-tetrahydropyrimidin-4-yl)-*N*,*N*-dimethylformimidamide (43). A scale of 3.8 mmol, used halide: chloride, solid, 1.27 g, isolated yield of 85%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.14−8.20 (m, 3H), 7.49 (d, J = 8.8 Hz, 2H), 7.20−7.32 (m, 5H), 5.30 (s, 1H), 5.17 (s, 2H), 5.09 (s, 2H), 3.11 (s, 3H), 2.96 (s, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.3, 159.7, 156.8, 152.5, 147.0, 146.4, 138.7, 128.8, 127.6, 127.4, 124.2, 124.0, 81.9, 45.9, 43.4, 40.9, 35.0; IR ν_{max}: 1695, 1644, 1621, 1562, 1515, 1447, 1424, 1367, 1339, 1221, 1129, 1096, 990, 901, 856, 809, 765, 750, 692, 603, 577, 524 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{21}H_{22}N_5O_4$ 408.1672; found: 408.1670; mp 176.8−182.7 °C.

6-Amino-1-benzyl-3-propylpyrimidine-2,4(1*H*,3*H*)-dione (44). A scale of 12.7 mmol, solid, 2.89 g, isolated yield of 88%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.31−7.38 (m, 2H), 7.23−7.30 (m, 1H), 7.16−7.22 (m, 2H), 6.73−6.83 (m, 2H), 5.04−5.11 (m, 2H), 4.72 (s, 1H), 3.69 (t, J = 7.1 Hz, 2H), 1.49 (sxt, J = 7.3 Hz, 2H), 0.82 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.4, 154.5, 151.6, 136.6, 128.5, 127.2, 126.4, 75.4, 44.6, 41.5, 20.9, 11.2; IR ν_{max}: 3405, 3341, 3194, 2963, 1633, 1582, 1490, 1451, 1432, 1405, 1366, 1274, 1218, 1125, 1083, 809, 721, 685, 548, 517, 459 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{14}H_{18}N_3O_2$ 260.1399; found: 260.1400; mp 198.5−199.9 °C. The above analysis results correspond to the literature data.[19f]

6-Amino-1-phenyl-3-propylpyrimidine-2,4(1*H*,3*H*)-dione (45). A scale of 9.8 mmol, solid, 1.07 g, isolated yield of 45%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 7.46−7.54 (m, 3H), 7.29−7.34 (m, 2H), 6.10 (s, 2H), 4.79 (s, 1H), 3.66 (t, J = 7.3 Hz, 2H), 1.49 (sxt, J = 7.4 Hz, 2H), 0.82 (t, J = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 162.1, 154.6, 151.6, 135.0, 130.1, 129.8, 129.7, 75.3, 41.8, 21.4, 11.7; IR ν_{max}: 3342, 3531, 3176, 2972, 1694, 1663, 1612, 1587, 1477, 1437, 1404, 1359, 1453, 1299, 1154, 1101, 1064, 1019, 797, 761, 695, 573, 534 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{13}H_{16}N_3O_2$ 246.1243; found: 246.1243; mp 187.7−189.5 °C. The above analysis results correspond to the literature data.[19g)]

6-Amino-3-ethyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (46). A scale of 1.3 mmol, solid, 70 mg, isolated yield of 28%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 6.77 (s, 2H), 4.65 (s, 1H), 3.69−3.79 (m, 4H), 1.53 (sxt, J = 7.5 Hz, 2H), 1.02 (t, J = 7.0 Hz, 3H), 0.86 (t, J = 7.3 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.5, 154.7, 151.6, 75.6, 43.5, 40.7, 35.2, 21.3, 13.7; IR ν_{max}: 3459, 3356, 3131, 2966, 2934, 2875, 1688, 1612, 1569, 1494, 1451, 1410, 1363, 1269, 1117, 1095, 787, 695, 593, 533 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_9H_{16}N_3O_2$ 198.1243; found: 198.1242; mp 78.7−83.5 °C. The above analysis results correspond to the literature data.[19h)]

6-Amino-3-butyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (47). A scale of 1.3 mmol, solid, 0.12 g, isolated yield of 41%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 6.76 (s, 2H), 4.65 (s, 1H), 3.65−3.78 (m, 4H), 1.53 (sxt, J = 7.5 Hz, 4H), 1.43 (quin, J = 7.4 Hz, 3H), 1.23 (dq, J = 14.9, 7.4 Hz, 6H), 0.81−0.91 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.7, 154.7, 151.8, 75.5, 43.5, 30.2, 21.3, 20.1, 14.2, 11.2; IR ν_{max}: 3437, 3114, 2962, 2931, 2874, 1698, 1654, 1604, 1503, 1464, 1436, 1411, 1364, 1284, 1270, 1198, 1180, 1044, 788, 760, 526 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{11}H_{20}N_3O_2$ 226.1556; found: 226.1554; mp 91.4−98.1 °C.

6-Amino-3-isobutyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (48). A scale of 2.6 mmol, solid, 0.21 g, isolated yield of 36%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 6.77 (s, 2H), 4.64 (s, 1H), 3.73 (t, J = 1.0 Hz, 2H), 3.54 (d, J = 7.3 Hz, 2H), 1.95 (dquin, J = 13.8, 6.9, 6.9, 6.9, 6.9 Hz, 1H), 1.51 (sxt, J = 7.4 Hz, 2H), 0.84 (t, J = 7.4 Hz, 3H), 0.78 (d, J = 6.7 Hz, 6H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.6, 154.3, 151.7, 75.1, 46.6, 43.1, 26.7, 20.9, 20.0, 10.8; IR ν_{max}: 3436, 3113, 2962, 1656, 1604, 1504, 1463, 1435, 1408, 1386, 1271, 1170, 1101, 1047, 788, 757, 548, 537 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{11}H_{20}N_3O_2$ 226.1556; found: 226.1556; mp 107.9−110.8 °C.

6-Amino-1,3-dibutylpyrimidine-2,4(1*H*,3*H*)-dione (49). 1,3-Dibutylurea (11.61 mmol; 2.00 g) and cyanoacetic acid (12,77 mmol; 1.09 g) were dissolved in acetic anhydride (12 mL). The resulting mixture was heated at 70 °C for 16 h. After this time, acetic anhydride was evaporated to give a red viscous oil. The residue was dissolved in NaOH solution in water (15 mL; 20%). The precipitated solid was filtered off, washed with water (3 × 10 mL) and diethyl ether (3 × 10 mL), and dried in vacuo (0.1 Torr, 2 h). The expected product (2.31 g) was obtained with an 83% yield; ^{1}H NMR (700 MHz, DMSO-d_6) δ 6.76 (s, 2H), 4.64 (s, 1H), 3.75 (t, J = 7.7 Hz, 2H), 3.68 (t, J = 7.3 Hz, 2H), 1.44−1.49 (m, 2H), 1.39−1.44 (m, 2H), 1.19−1.30 (m, 4H), 0.84−0.89 (m, 6H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.3, 154.3, 151.3, 75.2, 41.6, 29.8, 29.7, 19.7, 19.3, 13.8, 13.8; IR ν_{max}: 3434, 3125, 2957, 2931, 2873, 1656, 1604, 1505, 1460, 1411, 1289, 790, 770, 530 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{12}H_{22}N_3O_2$ 240.1712; found: 240.1710; mp 96.5−102.2 °C.

6-Amino-3-pentyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (50). A scale of 3.4 mmol, solid, 0.53 g, isolated yield 65%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 6.76 (s, 2H), 4.65 (s, 1H), 3.65−3.77 (m, 4H), 1.41−1.58 (m, 4H), 1.17−1.32 (m, 4H), 0.82−0.89 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.7, 154.7, 151.8, 75.6, 43.5, 29.0, 27.6, 22.3, 21.3, 14.3, 11.2; IR ν_{max}: 3439, 3352, 3117, 2959, 2930, 1646, 1603, 1500, 1456, 1411, 1368, 1271, 1191, 1113, 1048, 790, 769, 547 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{12}H_{22}N_3O_2$ 240.1712; found: 240.1715; mp 92.2−95.5 °C.

6-Amino-3-hexyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (51). A scale of 1.3 mmol, solid, 0.17 g, isolated yield of 53%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 6.76 (s, 2H), 4.64 (s, 1H), 3.65−3.77 (m, 4H), 1.38−1.61 (m, 4H), 1.24 (s, 6H), 0.81−0.89 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.7, 154.7, 151.8, 75.5, 43.5, 31.4, 27.9, 26.5, 22.4, 21.3, 14.3, 11.2; IR ν_{max}: 3439, 3118, 2962, 2931, 1650, 1603, 1500, 1410, 1370, 1270, 1192, 788, 759, 545 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{13}H_{24}N_3O_2$ 254.1869; found: 254.1868; mp 89.9−92.5 °C.

6-Amino-3-heptyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (52). A scale of 1.9 mmol, solid, 0.15 g, isolated yield of 29%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 6.77 (s, 2H), 4.63 (s, 1H), 3.72 (t, J = 7.7 Hz, 2H), 3.67 (d, J = 7.3 Hz, 2H), 1.51 (sxt, J = 7.5 Hz, 2H), 1.43 (quin, J = 7.4 Hz, 2H), 1.17−1.28 (m, 8H), 0.84 (td, J = 7.2, 2.6 Hz, 6H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.3, 154.3, 151.4, 75.1, 43.1, 31.3, 28.5, 27.6, 26.4, 22.1, 20.9, 14.0, 10.8; IR ν_{max}: 3438, 3138, 2961, 2928, 2855, 1651, 1603, 1501, 1462, 1436, 1410, 1367, 1270, 1192, 1085, 789, 762, 544 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{14}H_{26}N_3O_2$ 268.2025; found: 268.2022; mp 71.2−75.2 °C.

3-Allyl-6-amino-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (53). A scale of 1.4 mmol, solid, 0.13 g, isolated yield of 46%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 6.83 (s, 2H), 5.78 (ddt, J = 17.1, 10.3, 5.2, 5.2 Hz, 1H), 4.94−5.06 (m, 2H), 4.67 (s, 1H), 4.31 (d, J = 5.1 Hz, 1H), 3.74 (t, J = 7.8 Hz, 2H), 1.53 (dq, J = 15.0, 7.5 Hz, 2H), 0.86 (t, J = 7.3 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.3,

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154.9, 151.6, 134.1, 116.0, 75.3, 43.6, 42.2, 21.2, 11.2; IR ν_{max}: 3452, 3355, 3116, 2969, 1694, 1605, 1498, 1403, 1424, 1323, 1295, 1270, 1189, 1081, 1047, 994, 938, 776, 744, 669, 634, 546, 502, 463, 408 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{10}H_{16}N_3O_2$ 210.1243; found: 210.1244; mp: 97.9−103.9 °C.

6-Amino-3-(2-methylallyl)-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (54). A scale of 3.4 mmol, solid, 0.37 g, isolated yield of 48%; ^1H NMR (700 MHz, DMSO-d_6) δ 6.84 (s, 2H), 4.67−4.68 (m, 1H), 4.66 (s, 1H), 4.43 (dd, J = 1.6, 1.0 Hz, 1H), 4.21 (s, 2H), 3.73 (t, J = 1.0 Hz, 2H), 1.64 (d, J = 0.4 Hz, 3H), 1.52 (sxt, J = 7.5 Hz, 2H), 0.84 (t, J = 7.4 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.0, 154.5, 151.2, 140.8, 108.6, 74.8, 44.4, 43.2, 20.9, 20.4, 10.8; IR ν_{max}: 3438, 3114, 2974, 1653, 1604, 1502, 1423, 1398, 1294, 1270, 1194, 1096, 886, 779, 746, 567, 543 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{11}H_{18}N_3O_2$ 224.1399; found: 224.1400; mp 93.0−97.0 °C.

6-Amino-3-benzyl-1-propylpyrimidine-2,4(1*H*,3*H*)-dione (55). A scale of 4.6 mmol, solid, 1.01 g, isolated yield of 84%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.24−7.28 (m, 2H), 7.18−7.23 (m, 3H), 6.88 (s, 2H), 4.89 (s, 2H), 4.70 (s, 1H), 3.70−3.75 (m, 2H), 1.51 (sxt, J = 7.5 Hz, 2H), 0.84 (t, J = 7.4 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.2, 154.6, 151.5, 138.4, 128.2, 127.4, 126.8, 74.9, 43.3, 43.0, 20.9, 10.8; IR ν_{max}: 3479, 3067, 2964, 2876, 1688, 1646, 1607, 1583, 1490, 1453, 1428, 1402, 1343, 1288, 1258, 1190, 926, 897, 784, 757, 730, 700, 672, 646, 600, 547, 511 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{18}N_3O_2$ 260.1399; found: 260.1397; mp 147.3−151.9 °C.

6-Amino-3-benzyl-1-phenylpyrimidine-2,4(1*H*,3*H*)-dione (56). A scale of 1.1 mmol, solid, 0.24 g, isolated yield of 77%; ^1H NMR (400 MHz, DMSO-d_6) δ 7.48−7.56 (m, 3H), 7.20−7.37 (m, 7H), 6.22 (s, 2H), 4.93 (s, 2H), 4.89 (s, 1H); $^{13}C\{^1H\}$ NMR (101 MHz, DMSO-d_6) δ 162.1, 154.9, 151.8, 138.6, 134.8, 130.2, 129.8, 128.6, 128.0, 127.3, 75.2, 43.4; IR ν_{max}: 3449, 3293, 3229, 3178, 1702, 1635, 1610, 1578, 1469, 1450, 1421, 1397, 1350, 1287, 1182, 1153, 1077, 1049, 1021, 942, 789, 772, 754, 716, 691, 645, 599, 569, 522, 507, 452 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{17}H_{16}N_3O_2$ 294.1243; found: 294.1243; mp 183.3−186.6 °C.

3-Allyl-6-amino-1-benzylpyrimidine-2,4(1*H*,3*H*)-dione (57). A scale of 4.5 mmol, solid, 0.51 g, isolated yield of 44%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.31−7.35 (m, 2H), 7.23−7.27 (m, 1H), 7.17−7.20 (m, 2H), 6.84 (s, 2H), 5.78 (ddt, J = 17.2, 10.3, 5.2, 5.2 Hz, 1H), 5.07 (s, 2H), 5.03 (dq, J = 10.3, 1.5 Hz, 2H), 4.99 (dq, J = 17.2, 1.6 Hz, 1H), 4.73 (s, 1H), 4.33 (dt, J = 5.2, 1.5 Hz, 2H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 160.9, 154.6, 151.4, 136.6, 133.6, 128.5, 127.2, 126.4, 115.7, 75.2, 44.6, 42.0; IR ν_{max}: 3420, 3336, 3182, 1632, 1607, 1579, 1490, 1450, 1418, 1270, 1129, 993, 925, 789, 762, 733, 692, 635, 593, 530, 437 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{14}H_{16}N_3O_2$ 258.1243; found: 258.1244; mp 198.1−203.8 °C.

6-Amino-1,3-dibenzylpyrimidine-2,4(1*H*,3*H*)-dione (58). A scale of 1.2 mmol, solid, 0.26 g, isolated yield of 72%; ^1H NMR (400 MHz, DMSO-d_6) δ 7.12−7.42 (m, 10H), 6.90 (s, 2H), 5.09 (s, 2H), 4.95 (s, 2H), 4.80 (s, 1H); $^{13}C\{^1H\}$ NMR (101 MHz, DMSO-d_6) δ 161.6, 155.1, 152.1, 138.7, 136.9, 128.9, 128.6, 127.7, 127.6, 127.3, 126.8, 75.7, 45.2, 43.5; IR ν_{max}: 3459, 2966, 1688, 1612, 1568, 1496, 1450, 1414, 1363, 1282, 1118, 813, 755, 738, 694, 593, 534, 507 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{18}H_{18}N_3O_2$ 308.1399; found: 308.1399; mp 87.4−90.3 °C. The above analysis results correspond to the literature data.[19e]

6-Amino-1-benzyl-3-(2-methylbenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (59). A scale of 3.3 mmol, solid, 0.65 g, isolated yield of 61%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.33 (t, J = 7.5 Hz, 2H), 7.25 (t, J = 7.3 Hz, 1H), 7.18−7.22 (m, 2H), 7.13 (d, J = 6.9 Hz, 1H), 7.08 (quind, J = 7.0, 7.0, 7.0, 7.0, 1.4 Hz, 2H), 6.96 (s, 2H), 6.78−6.82 (m, 1H), 5.09 (s, 2H), 4.90 (s, 2H), 4.82 (s, 1H), 2.31 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.3, 154.8, 151.8, 136.6, 136.0, 134.9, 129.9, 128.5, 127.2, 126.4, 125.8, 124.6, 75.2, 44.8, 40.9, 18.8; IR ν_{max}: 3425, 3344, 1618, 1582, 1488, 1450, 1417, 1277, 1197, 1028, 948, 790, 746, 728, 703, 648, 593, 549, 503, 430 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{19}H_{20}N_3O_2$ 322.1556; found: 322.1557; mp 191.3−197.5 °C.

6-Amino-1-benzyl-3-(3-methylbenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (60). A scale of 2.0 mmol, solid, 0.50 g, isolated yield of 79%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.31−7.35 (m, 2H), 7.23−7.27 (m, 1H), 7.17−7.20 (m, 2H), 7.13−7.17 (m, 1H), 6.98−7.04 (m, 3H), 6.90 (s, 2H), 5.07 (s, 2H), 4.89 (s, 2H), 4.77 (s, 1H), 2.24 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.2, 154.7, 151.7, 138.2, 137.3, 136.6, 128.5, 128.2, 127.7, 127.5, 127.2, 126.4, 124.4, 75.2, 44.7, 43.0, 21.1; IR ν_{max}: 3466, 3437, 3146, 1702, 1642, 1604, 1494, 1452, 1426, 1402, 1282, 936, 790, 757, 727, 689, 589, 535, 527, 464, 427 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{19}H_{20}N_3O_2$ 322.1556; found: 322.1556; mp 90.0−93.2 °C.

6-Amino-1-benzyl-3-(4-methylbenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (61). A scale of 2.5 mmol, solid, 0.65 g, isolated yield of 81%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.30−7.34 (m, 2H), 7.22−7.27 (m, 1H), 7.18 (d, J = 7.5 Hz, 2H), 7.05−7.13 (m, 4H), 6.89 (s, 2H), 5.06 (s, 2H), 4.88 (s, 2H), 4.77 (s, 1H), 2.24 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.2, 154.6, 136.5, 136.0, 135.2, 128.8, 128.5, 127.4, 126.4, 79.3, 75.2, 44.7, 42.8, 20.7; IR ν_{max}: 2965, 2933, 2875, 1687, 1612, 1568, 1449, 1416, 1362, 1233, 1116, 814, 767, 570, 551, 471 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{19}H_{20}N_3O_2$ 322.1556; found: 322.1555; mp 83.6−90.2 °C.

6-Amino-1-benzyl-3-(4-isopropylbenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (62). A scale of 3.6 mmol, solid, 0.97 g, isolated yield of 76%; ^1H NMR (400 MHz, DMSO-d_6) δ 7.13−7.34 (m, 9H), 6.87 (s, 2H) 5.08 (s, 2H), 4.89 (s, 2H), 4.76 (s, 1H), 2.84−2.82 (m, 1H), 1.17 (d, J = 7.1 Hz, 6H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 162.0, 159.0, 156.3, 152.1, 147.1, 138.4, 135.4, 128.3, 127.5, 127.2, 126.2, 81.7, 45.3, 43.0, 33.2, 24.0; IR ν_{max}: 3179, 2958, 2869, 1608, 1491, 1451, 1422, 1400, 1351, 1279, 1209, 1188, 1117, 1054, 1018, 930, 783, 726, 694, 595, 529 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{21}H_{24}N_3O_2$ 350.1869; found: 350.1868; mp 89.5−93.2 °C.

6-Amino-1-benzyl-3-(3-methoxybenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (63). A scale of 2.0 mmol, solid, 0.45 g, isolated yield of 65%; ^1H NMR (400 MHz, DMSO-d_6) δ 7.17−7.37 (m, 6H), 6.91 (s, 2H), 6.72−6.82 (m, 3H), 5.09 (s, 2H), 4.92 (s, 2H), 4.80 (s, 1H), 3.70 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.2, 159.3, 154.7, 151.7, 139.8, 136.5, 129.3, 128.5, 127.3, 126.4, 119.4, 113.0, 112.2, 75.2, 55.0, 44.7, 43.0; IR ν_{max}: 3446, 3355, 3137, 1702, 1645, 1599, 1490, 1451, 1428, 1403, 1344, 1283, 1259, 1160, 1038, 938, 786, 776, 757, 728, 688, 591, 530, 502 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{19}H_{20}N_3O_3$ 338.1505; found: 338.1505; mp 89.4−91.2 °C.

6-Amino-1-benzyl-3-(2-fluorobenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (64). A scale of 2.3 mmol scale, solid, 0.49 g, isolated yield of 66%; ^1H NMR (700 MHz, DMSO-d_6) δ 7.31−7.35 (m, 2H), 7.23−7.28 (m, 2H), 7.19 (d, J = 7.1 Hz, 2H), 7.12−7.17 (m, 1H), 7.10 (td, J = 7.5, 1.0 Hz, 1H), 6.91−7.03 (m, 3H), 5.08 (s, 2H), 4.99 (s, 2H), 4.80 (s, 1H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.1, 159.2, 154.9, 151.7, 136.5, 128.6 (d, J = 8.2 Hz), 128.6, 127.8 (d, J = 4.5 Hz), 127.2, 126.3, 125.0 (d, J = 14.3 Hz), 124.4 (d, J = 3.7 Hz), 115.2 (d, J = 21.3 Hz), 75.1, 44.8, 37.0 (d, J = 5.3 Hz); IR ν_{max}: 3464, 3319, 3190, 1692, 1608, 1580, 1487, 1453, 1418, 1360, 1274, 1221, 1193, 834, 778, 758, 734, 693, 523 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{18}H_{17}FN_3O_2$ 326.1305; found: 326.1307; mp 176.9−181.6 °C.

6-Amino-1-benzyl-3-(3-fluorobenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (65). A scale of 2.6 mmol, solid 0.64 g, isolated yield of 75%; 1H NMR (700 MHz, DMSO-d_6) δ 7.30−7.35 (m, 3H), 7.24−7.27 (m, 1H), 7.18 (d, J = 7.3 Hz, 2H), 7.03−7.06 (m, 2H), 6.98−7.01 (m, 1H), 6.95 (s, 2 H), 5.07 (s, 2H), 4.93 (s, 2H), 4.79 (s, 1H); $^{13}C\{^1H\}$ NMR (176 MHz, DMSO-d_6) δ 161.5, 161.1, 154.8, 151.7, 141.2 (d, J = 7.4 Hz), 136.5, 130.3 (d, J = 8.2 Hz), 128.5, 127.3, 126.4, 123.3 (d, J = 2.5 Hz), 114.0 (d, J = 21.7 Hz), 113.7 (d, J = 20.9 Hz), 75.1, 44.8, 42.7; IR ν_{max}: 3461, 3371, 3135, 1697, 1609, 1498, 1428, 1409, 1269, 1253, 1116, 932, 793, 775, 761, 713, 684, 660, 596, 534 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{18}H_{17}FN_3O_2$ 326.1305; found: 326.1305; mp 90.3−95.1 °C.

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6-Amino-1-benzyl-3-(3-bromobenzyl)pyrimidine-2,4-(1*H*,3*H*)-dione (66). A scale of 3.5 mmol, solid, 0.93 g, isolated yield of 69%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.43 (dt, J = 6.3, 2.1 Hz, 1H), 7.39 (s, 1H), 7.31−7.37 (m, 2H), 7.23−7.30 (m, 3H), 7.20 (d, J = 7.3 Hz, 2H), 6.96 (s, 2H), 5.10 (s, 2H), 4.94 (s, 2H), 4.81 (s, 1H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.5, 155.2, 152.1, 141.5, 136.9, 130.9, 130.4, 130.2, 128.9, 127.7, 126.9, 126.7, 122.0, 75.6, 45.2, 43.0; IR ν_{max}: 3435, 3133, 1702, 1640, 1604, 1493, 1427, 1402, 1281, 1215, 1070, 935, 880, 784, 768, 725, 692, 666, 588, 534, 507, 462, 423 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{18}H_{17}BrN_3O_2$ 386.0504; found: 386.0502; mp 86.6−88.5 °C.

6-Amino-1-benzyl-3-(3-trifluoromethyl)benzyl)-pyrimidine-2,4(1*H*,3*H*)-dione (67). A scale of 2.7 mmol, solid, 0.68 g, isolated yield of 67%; ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.50−7.64 (m, 4H), 7.23−7.37 (m, 3H), 7.13−7.22 (m, 2H), 6.91−7.04 (m, 2H), 5.09 (s, 2H), 5.02 (s, 2H), 4.80 (s, 1H); ^{13}C{^{1}H} NMR (101 MHz, DMSO-d_6) δ 161.5, 155.2, 152.2, 140.1, 136.8, 132.0, 129.8, 129.5 (q, J = 31.8 Hz), 128.9, 127.7, 126.7, 123.8−124.4 (m), 124.7 (q, J = 271.8 Hz), 75.6, 45.2, 43.2; IR ν_{max}: 3434, 3138, 1643, 1605, 1497, 1450, 1406, 1326, 1281, 1157, 1122, 1073, 938, 790, 728, 695, 659, 535, 509 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{17}F_3N_3O_2$ 376.1273; found: 376.1274; mp 95.8−102.3 °C.

6-Amino-1-benzyl-3-(4-nitrobenzyl)pyrimidine-2,4(1*H*,3*H*)-dione (68). A scale of 3.1 mmol, solid, 0.64 g, isolated yield of 58%; ^{1}H NMR (700 MHz, DMSO-d_6) δ 8.16 (d, J = 8.4 Hz, 2H), 7.46 (d, J = 8.6 Hz, 2H), 7.33 (t, J = 7.5 Hz, 2H), 7.25 (t, J = 7.3 Hz, 1H), 7.18 (d, J = 7.5 Hz, 2H), 7.00 (s, 2H), 5.06 (d, J = 18.9 Hz, 4H), 4.81 (s, 1H); ^{13}C{^{1}H} NMR (176 MHz, DMSO-d_6) δ 161.0, 154.9, 151.7, 146.6, 146.2, 136.4, 128.6, 128.2, 127.3, 126.3, 123.6, 75.1, 44.8, 42.9; IR ν_{max}: 3428, 1689, 1643, 1602, 1518, 1493, 1451, 1405, 1343, 1285, 1215, 1208, 1110, 855, 804, 745, 693, 639, 598, 548, 522, 453 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{18}H_{17}N_4O_4$ 353.1250; found: 353.1247; mp 153.0−158.5 °C.

General Procedure for the Catalytic Reactions. The corresponding 1,3-disubstituted 6-aminouracil (0.3 mmol), NHC precatalyst **8c** (0.03 mmol; 8 mg), Kharach oxidant **DQ** (0.3 mmol; 122 mg), and α,β-unsaturated aldehyde (0.3 mmol) were dispersed in anhydrous toluene (3 mL). Anhydrous potassium phosphate tribasic (0.06 mmol; 13 mg) as a base was added, and the mixture was stirred at room temperature for 24 h. The final product was isolated by flash column chromatography using gradient elution (0−100% AcOEt in hexane).

5-(4-Methoxyphenyl)-1,3-dipropyl-5,8-dihydro-pyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aaa). Solid, 106 mg, isolated yield of 95%.; ^{1}H NMR (700 MHz, $CDCl_3$) δ 8.61 (s, 1H), 7.14−7.18 (m, 2H), 6.79−6.83 (m, 2H), 4.43 (d, J = 6.56 Hz, 1H), 3.85−3.97 (m, 4H), 3.76 (s, 3H), 2.94−3.00 (m, 1H), 2.87 (dd, J = 1.40, 16.46 Hz, 1H), 1.61−1.73 (m, 5H), 0.96 (t, J = 7.37 Hz, 3H), 0.92 (t, J = 7.48 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.5, 161.2, 158.8, 150.8, 143.8, 133.5, 127.5, 114.3, 93.1, 55.2, 44.2, 43.4, 37.8, 33.0, 22.1, 21.0, 11.3, 10.7; IR ν_{max}: 3156.1, 2965.2, 2923.6, 1686.7, 1645.9, 1632.8, 1608.0, 1509.5, 1491.6, 1454.8, 1440.2, 1417.7, 1375.4, 1353.9, 1302.6, 1257.8, 1235.4, 1203.8, 1175.4, 1153.3, 1031.4, 998.3, 831.8, 776.1, 760.1, 739.0, 717.2, 687.0, 649.5, 584.3, 554.9, 530.0, 518.6, 496.6, 462.0 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{20}H_{26}N_3O_4$ 372.1923; found: 372.1924; mp 174.2−176.7 °C.

5-Phenyl-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7baa). Solid, 113 mg, isolated yield of 83%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 9.37 (s, 1H), 7.25−7.30 (m, 2H), 7.20−7.24 (m, 3H), 4.48 (d, J = 6.99 Hz, 1H), 3.98 (ddd, J = 6.10, 10.00, 14.90 Hz, 1H), 3.83−3.93 (m, 3H), 3.00 (dd, J = 8.01, 16.51 Hz, 1H), 2.87 (dd, J = 1.02, 16.51 Hz, 1H), 1.57−1.69 (m, 4H), 0.91 (dt, J = 2.04, 7.42 Hz, 6H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.4, 161.2, 150.8, 144.0, 141.4, 128.9, 127.3, 126.4, 92.7, 44.2, 43.4, 37.7, 33.8, 22.1, 21.0, 11.3, 10.7; IR ν_{max}: 3246.8, 3217.6, 2963.0, 2933.5, 1687.6, 1654.9, 1632.6, 1504.3, 1480.4, 1471.1, 1441.4, 1417.7, 1353.8, 1300.8, 1281.2, 1242.6, 1227.0, 1173.6, 1137.5, 759.8, 741.3, 695.9, 683.2, 646.5, 565.7, 552.8, 496.2, 441.7 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{24}N_3O_3$ 342.1818; found: 342.1819; mp 173.6−175.0 °C.

5-(2-Methoxyphenyl)-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7caa). Solid, 149 mg, isolated yield of 79%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 8.10 (s, 1H), 7.18−7.23 (m, 1H), 7.05 (dd, J = 1.72, 7.53 Hz, 1H), 6.83−6.87 (m, 2H), 4.63 (d, J = 7.96 Hz, 1H), 3.81−3.97 (m, 4H), 3.79 (s, 3H), 2.93 (dd, J = 9.09, 16.83 Hz, 1H), 2.80 (dd, J = 1.02, 16.73 Hz, 1H), 1.61 (dt, J = 1.40, 7.53 Hz, 4H), 0.98 (t, J = 7.42 Hz, 3H), 0.89 (t, J = 7.42 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.5, 161.0, 157.0, 150.9, 144.6, 128.8, 128.5, 128.0, 120.5, 110.8, 90.9, 54.8, 44.1, 43.2, 36.6, 30.8, 22.1, 21.0, 11.3, 10.7; IR ν_{max}: 3203.0, 2963.4, 2934.6, 1719.4, 1691.7, 1635.0, 1612.6, 1510.8, 1487.6, 1475.9, 1455.0, 1424.9, 1359.0, 1334.2, 1284.8, 1235.7, 1177.0, 1109.6, 1028.1, 754.1, 657.3, 488.8 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{20}H_{26}N_3O_4$ 372.1923; found: 372.1922; mp 219.0−220.3 °C.

3-Methoxy-4-(2,4,7-trioxo-1,3-dipropyl-1,2,3,4,5,6,7,8-octahydropyrido[2,3-*d*]pyrimidin-5-yl)phenyl Acetate (7daa). Solid, 124 mg, isolated yield of 72%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.04 (d, J = 2.04 Hz, 1H), 6.91 (d, J = 8.17 Hz, 1H), 6.76 (d, J = 8.28 Hz, 1H), 4.50 (dd, J = 3.44, 6.02 Hz, 1H), 3.84−3.95 (m, 4H), 3.78 (s, 3H), 2.96 (d, J = 6.24 Hz, 2H), 2.29 (s, 3H), 1.60−1.75 (m, 4H), 0.99 (t, J = 7.42 Hz, 3H), 0.93 (t, J = 7.48 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.4, 169.1, 161.3, 151.3, 150.7, 143.9, 140.3, 138.9, 122.8, 117.6, 111.7, 92.8, 55.8, 44.3, 43.4, 37.0, 33.4, 22.1, 21.0, 20.6, 11.3, 10.7; IR ν_{max}: 2957.9, 2926.2, 1767.7, 1689.5, 1637.6, 1603.8, 1504.8, 1477.7, 1458.7, 1419.6, 1365.1, 1355.3, 1332.1, 1293.3, 1269.2, 1258.3, 1233.5, 1191.0, 1176.8, 1160.9, 1135.2, 1120.3, 1033.0, 1012.0, 892.4, 757.3, 743.8, 678.4, 502.3, 473.7, 457.7 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{28}N_3O_6$ 430.1978; found: 430.1980; mp 176.3−177.4 °C.

1,3-Dipropyl-5-(*p*-tolyl)-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7eaa). Solid, 85 mg, isolated yield of 61%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 8.07 (s, 1H), 7.11−7.16 (m, 2H), 7.07−7.10 (m, 2H), 4.45 (d, J = 6.67 Hz, 1H), 3.83−3.94 (m, 4H), 2.98 (dd, J = 7.80, 16.60 Hz, 1H), 2.89 (dd, J = 1.10, 16.50 Hz, 1H), 2.29 (s, 3H), 1.60−1.75 (m, 4H), 0.98 (t, J = 7.42 Hz, 3H), 0.92 (t, J = 7.48 Hz, 3H); ^{13}C{^{1}H} NMR (176 MHz, $CDCl_3$) δ 170.7, 160.6, 150.3, 143.1, 138.0, 136.6, 129.2, 125.9, 92.6, 43.8, 43.0, 37.0, 33.1, 21.8, 20.6, 20.6, 10.9, 10.5; IR ν_{max}: 3167.0, 2967.6, 2933.9, 1686.8, 1648.7, 1633.7, 1493.5, 1457.1, 1353.7, 1238.8, 774.0, 495.5 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{20}H_{26}N_3O_3$ 356.1974; found: 356.1974; mp 169.3−171.5 °C.

5-(4-Fluorophenyl)-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7faa). Solid, 110 mg, isolated yield of 77%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.18−7.22 (m, 2H), 6.95−6.99 (m, 2H), 4.47 (d, J = 6.99 Hz, 1H), 3.85−3.97 (m, 4H), 3.00 (dd, J = 8.01, 16.51 Hz, 1H), 2.86 (dd, J = 1.40, 16.56 Hz, 1H), 1.61−1.75 (m, 4H), 0.97 (t, J = 7.42 Hz, 3H), 0.92 (t, J = 7.42 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.0, 163.2, 161.1, 160.8, 150.7, 143.9, 137.1 (d, J = 3.18 Hz), 128.0 (d, J = 7.95 Hz), 115.8 (d, J = 21.50 Hz), 92.6, 44.3, 43.4, 37.7, 33.2, 22.1, 21.0, 11.3, 10.7; IR ν_{max}: 3242.4, 3187.6, 2967.5, 2929.6, 1687.3, 1636.8, 1605.1, 1506.1, 1464.8, 1438.4, 1409.7, 1380.9, 1353.2, 1299.3, 1268.4, 1229.0, 1158.3, 1021.1, 833.3, 781.8, 754.2, 738.5, 684.3, 664.2, 575.4, 551.2, 496.2, 460.8 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{23}FN_3O_3$ 360.1723; found: 360.1722; mp 166.3−168.7 °C.

5-(4-Chlorophenyl)-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7gaa). Solid, 148 mg, isolated yield of 98%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.22−7.28 (m, 2H), 7.13−7.18 (m, 2H), 4.45 (d, J = 6.78 Hz, 1H), 4.00 (ddd, J = 6.10, 10.00, 15.10 Hz, 1H), 3.85−3.93 (m, 3H), 3.00 (dd, J = 8.07, 16.56 Hz, 1H), 2.83 (dd, J = 0.86, 16.56 Hz, 1H), 1.58−1.69 (m, 4H), 0.90−0.93 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.1, 161.1, 150.7, 144.1, 139.8, 133.2, 129.1, 127.9, 92.3, 44.3, 43.4, 37.5, 33.3, 22.1, 21.0, 11.3, 10.7; IR ν_{max}: 3245.9, 2958.4, 2925.1, 1688.4, 1632.8, 1506.6, 1492.8, 1470.9, 1435.8, 1422.8, 1347.7, 1299.1, 1233.4, 835.1, 784.5, 750.2, 723.0, 681.4, 669.6, 504.5, 491.7, 460.0

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cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{23}ClN_3O_3$ 376.1428; found: 376.1430; mp 128.0−134.0 °C.

5-(4-Bromophenyl)-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7haa). Solid, 167 mg, isolated yield of 99%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 9.15 (s, 1H), 7.43−7.37 (m, 2H), 7.13−7.08 (m, 2H), 4.44 (d, J = 6.8 Hz, 1H), 4.02−3.95 (m, 1H), 3.93−3.86 (m, 3H), 3.00 (dd, J = 8.1, 16.6 Hz, 1H), 2.83 (dd, J = 0.9, 16.6 Hz, 1H), 1.70−1.60 (m, 4H), 0.97−0.88 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 171.8, 161.0, 150.7, 144.0, 140.4, 132.0, 128.2, 121.3, 92.2, 44.3, 43.4, 37.4, 33.4, 22.1, 21.0, 11.3, 10.8; IR ν_{max}: 2965.6, 2930.8, 1688.2, 1643.4, 1632.5, 1490.7, 1463.8, 1453.9, 1441.8, 1422.8, 1351.3, 1316.0, 1298.8, 1240.2, 1142.3, 1075.5, 1011.2, 1000.4, 836.5, 782.4, 755.3, 743.0, 555.0, 495.0 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{23}BrN_3O_3$ 420.0923; found: 420.0923; mp 72.0−76.0 °C.

5-(4-Nitrophenyl)-1,3-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7iaa). Solid, 75 mg, isolated yield 65%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.49 (s, 1H), 8.14−8.21 (m, 2H), 7.41−7.48 (m, 2H), 4.61 (d, J = 6.8 Hz, 1H), 3.87−4.03 (m, 4H), 3.10 (dd, J = 16.9, 8.1 Hz, 1H), 2.92 (dd, J = 16.9, 1.5 Hz, 1H), 1.61−1.70 (m, 4H), 1.00 (t, J = 7.3 Hz, 3H), 0.94 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.5, 160.9, 150.5, 148.8, 147.3, 144.1, 127.5, 124.2, 91.4, 44.4, 43.5, 36.9, 33.9, 22.2, 21.0, 11.3, 10.9; IR ν_{max}: 3244.7, 3192.1, 2967.7, 2937.4, 2876.6, 1685.1, 1633.1, 1513.8, 1470.1, 1346.7, 1308.4, 1269.3, 1238.0, 1174.7, 1152.2, 1111.0, 856.0, 787.5, 749.8, 704.4, 685.5, 61.6, 485.8 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{23}N_4O_5$ 387.1668; found: 387.1669; mp 194.4−197.9 °C.

3-Benzyl-5-(furan-2-yl)-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7jba). Solid, 76 mg, isolated yield of 67%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 9.00 (s, 1H), 7.47 (d, J = 6.9 Hz, 1H), 7.21−7.33 (m, 4H), 6.25 (dd, J = 3.2, 1.9 Hz, 1H), 6.09 (dt, J = 3.2, 0.8 Hz, 1H), 5.12 (dd, J = 18.1, 13.8 Hz, 2H), 4.53 (d, J = 6.7 Hz, 1H), 3.91 (dddd, J = 38.7, 15.1, 10.1, 6.0 Hz, 2H), 2.94 (dd, J = 16.4, 1.3 Hz, 1H), 2.86 (dd, J = 16.4, 7.5 Hz, 1H), 1.61−1.69 (m, 2H), 0.95 (t, J = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 171.4, 160.7, 153.7, 150.8, 144.4, 142.1, 136.9, 129.1, 128.4, 127.6, 110.3, 105.7, 90.5, 44.9, 44.4, 35.2, 28.6, 22.1, 10.7; IR ν_{max}: 3164.7, 2964.7, 2933.0, 2876.4, 1688.1, 1635.7, 1500.2, 1477.0, 1350.2, 1322.5, 1290.8, 1239.0, 1147.8, 1133.3, 1012.6, 776.6, 753.5, 734.0, 714.0, 692.7, 657.4, 598.7, 542.7, 480.6 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{21}H_{22}N_3O_4$ 380.1610; found: 380.1608; mp 166.8−168.9 °C.

3-Benzyl-5-methyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7kba). Solid, 65 mg, isolated yield of 66%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.62 (s, 1H), 7.47−7.53 (m, 2H), 7.28−7.37 (m, 3H), 5.15 (s, 2H), 3.85−3.97 (m, 2H), 3.35 (quind, J = 7.1, 1.6 Hz, 1H), 2.73 (dd, J = 16.4, 7.3 Hz, 1H), 2.50 (dd, J = 16.4, 1.2 Hz, 1H), 1.59−1.79 (m, 2 H), 1.14 (d, J = 7.1 Hz, 3H), 1.00 (t, J = 7.5 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 171.9, 160.9, 150.9, 143.0, 137.0, 129.1, 128.4, 127.6, 94.5, 44.8, 44.3, 37.7, 24.2, 22.1, 18.8, 10.9; IR ν_{max}: 3237.4, 3182.1, 2964.9, 2935.5, 2876.2, 1690.0, 1627.3, 1502.8, 1475.1, 1433.8, 1357.6, 1328.2, 1298.5, 1242.0, 1183.4, 1069.8, 1027.8, 770.7, 751.7, 698.5, 595.1, 495.7, 459.3 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{18}H_{22}N_3O_3$ 328.1661; found: 328.1663; 79.8−82.5 °C.

3-Benzyl-5-ethyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7lba). Solid, 68 mg, isolated yield of 66%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88 (s, 1H), 7.47−7.52 (m, 2H), 7.28−7.35 (m, 3H), 5.15 (dd, J = 17.1, 13.4 Hz, 2H), 3.87 (t, J = 7.8 Hz, 2H), 3.17 (m, J = 8.7, 8.7, 5.4 Hz, 1H), 2.64−2.68 (m, 2H), 1.60−1.80 (m, 3H), 1.43 (m, J = 8.3 Hz, 1H), 0.92−1.04 (m, 6H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.2, 161.1, 150.9, 143.3, 137.1, 129.1, 128.4, 127.6, 93.6, 44.8, 44.3, 34.9, 30.4, 26.0, 22.1, 11.0, 10.8; IR ν_{max}: 3190.1, 2962.0, 2931.9, 2874.7, 1689.2, 1624.8, 1502.7, 1472.1, 1434.2, 1362.0, 1297.2, 1235.6, 1182.9, 1073.1, 768.4, 750.9, 698.1, 493.9 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{19}H_{24}N_3O_3$ 342.1818; found: 342.1819; mp 82.0−90.8 °C.

3-Benzyl-1,5-dipropyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7mba). Solid, 74 mg, isolated yield of 70%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.69 (s, 1H), 7.45−7.54 (m, 2H), 7.28−7.36 (m, 3H), 5.11−5.20 (m, 2H), 3.84−4.00 (m, 2H), 3.19−3.29 (m, 1H), 2.58−2.70 (m, 2H), 1.64−1.77 (m, 2H), 1.26−1.55 (m, 4H), 1.00 (t, J = 7.5 Hz, 3H), 0.93 (t, J = 6.8 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 172.3, 161.1, 150.9, 143.3, 137.1, 129.0, 128.4, 127.6, 94.0, 44.8, 44.3, 35.3, 35.2, 28.7, 22.1, 19.8, 13.9, 10.8; IR ν_{max}: 3244.9, 3193.4, 2963.8, 2934.6, 2876.4, 1690.8, 1627.5, 1512.2, 1475.7, 1433.0, 1357.0, 1300.8, 1246.3, 1184.0, 1069.9, 755.7, 699.8, 596.9, 548.4, 497.8, 460.5 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{20}H_{26}N_3O_3$ 356.1974; found: 356.1974; 133.5−141.8 °C.

3-Benzyl-5-pentyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7nba). Solid, 52 mg, isolated yield of 45%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.41 (s, 1H), 7.42−7.55 (m, 2H), 7.20−7.37 (m, 3H), 5.16 (dd, J = 21.0, 13.7 Hz, 2H), 3.96 (dddd, J = 36.0, 14.9, 9.3, 6.6 Hz, 2H), 3.15−3.28 (m, 1H), 2.56−2.70 (m, 2H), 1.60−1.76 (m, 2H), 1.46−1.56 (m, 1H), 1.20−1.44 (m, 7H), 0.99 (t, J = 7.3 Hz, 3H), 0.88 (t, J = 6.8 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 173.1, 161.2, 150.9, 143.5, 137.1, 129.0, 128.4, 127.5, 94.0, 44.8, 44.3, 35.3, 33.0, 31.7, 28.8, 26.2, 22.5, 22.1, 14.0, 10.7; IR ν_{max}: 3235.1, 3186.1, 2960.6, 2928.1, 2854.0, 1687.6, 1634.0, 1503.3, 1468.8, 1432.3, 1360.8, 1295.2, 1242.2, 1177.0, 1072.7, 750.7, 701.5, 595.0, 493.6 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{22}H_{30}N_3O_3$ 384.2287; found: 384.2288; mp 107.5−108.9 °C.

3-Benzyl-5-hexyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7oba). viscous oil, 72 mg, isolated yield of 60%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.39 (s, 1H), 7.45−7.52 (m, 2H), 7.22−7.36 (m, 3H), 5.16 (dd, J = 21.0, 13.7 Hz, 2H), 3.96 (dddd, J = 36.2, 15.2, 9.5, 6.6 Hz, 2H), 3.16−3.27 (m, 1H), 2.56−2.71 (m, 2H), 1.58−1.78 (m, 2H), 1.45−1.58 (m, 1H), 1.16−1.45 (m, 9H), 0.99 (t, J = 7.3 Hz, 3H), 0.88 (t, J = 6.8 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 173.1, 161.2, 150.9, 143.4, 137.1, 129.0, 128.4, 127.5, 94.0, 44.8, 44.3, 35.4, 33.0, 31.7, 29.2, 28.8, 26.5, 22.5, 22.1, 14.0, 10.7; IR ν_{max}: 3223.8, 3174.9, 2949.7, 2926.6, 2855.4, 1682.8, 1635.1, 1496.6, 1475.1, 1431.1, 1358.8, 1325.1, 1295.9, 1240.1, 1184.4, 769.6, 751.8, 723.1, 694.3, 664.4, 594.2, 489.1 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{23}H_{32}N_3O_3$ 398.2444; found: 398.2442.

3-Benzyl-5-heptyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7pba). Viscous oil, 67 mg, isolated yield of 55%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 9.61 (s, 1H), 7.49 (d, J = 6.8 Hz, 2H), 7.22−7.36 (m, 3H), 5.16 (dd, J = 21.3, 13.9 Hz, 2H), 3.97 (dddd, J = 38.4, 14.9, 9.5, 7.1 Hz, 2H), 3.15−3.28 (m, 1H), 2.55−2.71 (m, 2H), 1.59−1.77 (m, 2H), 1.45−1.57 (m, 1H), 1.17−1.45 (m, 11H), 0.99 (t, J = 7.3 Hz, 3H), 0.88 (t, J = 6.8 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 173.4, 161.2, 150.9, 143.5, 137.1, 129.0, 128.4, 127.6, 94.0, 44.8, 44.3, 35.3, 33.0, 31.8, 29.5, 29.2, 28.8, 26.6, 22.6, 22.1, 14.1, 10.7; IR ν_{max}: 3234.8, 3176.0, 2953.6, 2922.4, 2851.1, 1681.9, 1639.1, 1495.1, 1475.2, 1429.8, 1360.7, 1317.2, 1289.9, 1242.4, 1185.3, 1137.5, 1090.6, 1050.2, 951.5, 751.2, 693.0, 663.9, 653.6, 593.5, 485.9, 439.9 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{24}H_{34}N_3O_3$ 412.2600; found: 412.2601.

3-Benzyl-5-nonyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7qba). Viscous oil, 61 mg, isolated yield of 46%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 9.53 (s, 1H), 7.43−7.50 (m, 2H), 7.21−7.32 (m, 3H), 5.14 (dd, J = 30.3, 13.8 Hz, 2H), 3.99 (ddd, J = 14.6, 10.3, 6.2 Hz, 1H), 3.90 (ddd, J = 15.0, 9.9, 5.5 Hz, 1H), 3.15−3.23 (m, 1H), 2.56−2.66 (m, 2H), 1.58−1.71 (m, 2H), 1.44−1.52 (m, 1H), 1.21−1.38 (m, 15H), 0.97 (t, J = 7.3 Hz, 3H), 0.87 (t, J = 7.2 Hz, 3H); ^{13}C{^{1}H} NMR (176 MHz, $CDCl_3$) δ 173.0, 160.8, 150.5, 143.1, 136.7, 128.6, 128.0, 127.2, 93.6, 44.4, 43.9, 34.9, 32.6, 31.5, 29.2, 29.2, 29.1, 28.9, 28.4, 26.2, 22.3, 21.7, 13.7, 10.3; IR ν_{max}: 3234.2, 3182.2, 2924.0, 2853.0, 1690.7, 1631.9, 1503.9, 1472.5, 1435.3, 1361.0, 1299.2, 1242.4, 1183.4, 751.6, 697.8, 594.5, 493.8 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{26}H_{37}N_3O_3$ 440.2835; found: 440.2835.

3-Benzyl-5-decyl-1-propyl-5,8-dihydropyrido[2,3-*d*]-pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7rba). Viscous oil, 83 mg, isolated yield of 61%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 9.19 (s, 1H), 7.45−7.49

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(m, 2H), 7.28−7.32 (m, 2H), 7.23−7.25 (m, 1H), 5.13 (dd, J = 27.6, 13.8 Hz, 2H), 3.92−3.99 (m, 1H), 3.88 (ddd, J = 14.6, 9.9, 5.4 Hz, 1H), 3.16−3.22 (m, 1H), 2.57−2.66 (m, 2H), 1.58−1.72 (m, 2H), 1.43−1.52 (m, 1H), 1.23−1.38 (m, 17H), 0.97 (t, J = 7.4 Hz, 3H), 0.87 (t, J = 7.2 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 173.2, 160.8, 150.5, 143.1, 136.7, 128.6, 128.0, 127.2, 93.7, 44.4, 44.0, 34.9, 32.6, 31.5, 29.2, 29.2, 29.2, 28.9, 28.4, 26.2, 22.3, 21.7, 13.7, 10.3; IR ν_{max}: 3234.3, 3191.5, 2923.2, 2852.7, 1691.7, 1632.6, 1504.1, 1472.8, 1435.5, 1361.6, 1299.7, 1241.7, 1183.8, 1074.1, 752.1, 698.0, 594.8, 493.5 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{27}H_{40}N_3O_3$ 454.3070; found: 454.3069.

(*E*)-3-Benzyl-5-(prop-1-en-1-yl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7sba). Solid, 71 mg, isolated yield of 67%; ^1H NMR (400 MHz, $CDCl_3$ δ 9.20 (s, 1H), 7.44−7.56 (m, 2H), 7.20−7.39 (m, 3H), 5.47−5.61 (m, 1H), 5.42 (ddd, J = 15.4, 5.9, 1.5 Hz, 1H), 5.15 (s, 2H), 3.95 (dddd, J = 27.1, 14.9, 9.5, 5.9 Hz, 2H), 3.83 (td, J = 5.7, 1.3 Hz, 1H), 2.62−2.78 (m, 2H), 1.58−1.80 (m, 5H), 1.00 (t, J = 7.3 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 172.4, 160.9, 150.9, 143.6, 137.0, 129.1, 128.6, 128.4, 127.6, 125.9, 92.3, 44.8, 44.4, 36.0, 31.1, 22.1, 17.8, 10.8; IR-ATR ν_{max}: 3192.3, 2965.1, 2935.2, 2876.9, 1688.9, 1625.8, 1502.4, 1472.3, 1435.0, 1354.0, 1289.4, 1231.4, 1179.6, 1073.6, 964.6, 751.4, 697.6, 490.9, 466.0 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{20}H_{24}N_3O_3$ 354.1818; found: 354.1818; mp 58.9−61.5 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aab). Solid, 97 mg, isolated yield of 77%; ^1H NMR (700 MHz, $CDCl_3$) δ 7.96 (s, 1H), 7.32−7.39 (m, 3H), 7.20−7.23 (m, 2H), 7.07−7.11 (m, 2H), 6.76−6.79 (m, 2H), 5.42 (d, J = 16.78 Hz, 1H), 5.07 (d, J = 16.78 Hz, 1H), 4.40 (dd, J = 1.18, 7.85 Hz, 1H), 3.94 (dddd, J = 6.30, 8.60, 12.80, 21.70 Hz, 2H), 3.75 (s, 3H), 2.89 (dd, J = 8.00, 16.40 Hz, 1H), 2.78 (dd, J = 1.08, 16.35 Hz, 1H), 1.68 (dddt, J = 1.24, 5.08, 6.28, 7.47 Hz, 2H), 0.94 (t, J = 7.42 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 170.5, 161.0, 158.8, 151.2, 144.0, 134.5, 133.3, 129.6, 128.7, 127.5, 126.2, 114.3, 93.7, 55.3, 46.0, 43.6, 37.7, 33.4, 21.0, 11.3; IR ν_{max}: 3234, 3186, 2957, 1688, 1643, 1633, 1510, 1457, 1439, 1350, 1299, 1248, 1186, 1154, 1119, 1035, 848, 781, 756, 732, 692, 672, 576, 525, 505, 477, 456 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{24}H_{26}N_3O_4$ 420.1923; found: 420.1925; mp 167.4−176.8 °C.

5-(4-Methoxyphenyl)-1-phenyl-3-propyl-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aac). Solid, 68 mg, isolated yield of 56%; ^1H NMR (700 MHz, $CDCl_3$) δ 7.57−7.62 (m, 3H), 7.30−7.35 (m, 2H), 7.21−7.24 (m, 2H), 6.82−6.86 (m, 2H), 6.74 (s, 1H), 4.48 (dd, J = 8.0, 1.5 Hz, 1H), 3.86−3.95 (m, 2H), 3.78 (s, 3H), 3.00 (dd, J = 16.7, 8.1 Hz, 1H), 2.87 (dt, J = 16.4, 1.4 Hz, 1H), 1.67 (ddt, J = 7.6, 6.3, 1.2, 1.2 Hz, 2H), 0.93 (t, J = 7.5 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 169.1, 160.8, 158.4, 150.1, 142.4, 133.4, 131.9, 130.5, 130.5, 130.4, 128.9, 128.6, 127.2, 113.9, 92.0, 54.9, 43.0, 37.0, 33.0, 20.6, 11.0; IR ν_{max}: 2961.3, 1699.4, 1638.3, 1485.1, 1241.9, 1178.4, 1029.4, 831.4, 751.9, 685.9, 520.4 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{23}H_{24}N_3O_4$ 406.1767; found: 406.1770; mp 94.9−102.3 °C.

3-Ethyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aca). Solid, 78 mg, isolated yield of 73%; ^1H NMR (400 MHz, $CDCl_3$) δ 8.83 (s, 1H), 7.15−7.20 (m, 2H), 6.80−6.86 (m, 2H), 4.46 (dd, J = 7.7, 1.3 Hz, 2H), 3.90−4.07 (m, 4H), 3.78 (s, 3H), 2.99 (dd, J = 16.4, 7.8 Hz, 1H), 2.88 (dd, J = 16.4, 1.5 Hz, 1H), 1.64−1.73 (m, 2H), 1.23 (t, J = 7.1 Hz, 3H), 0.97 (t, J = 7.3 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 172.0, 160.9, 158.8, 150.6, 143.6, 133.4, 127.5, 114.3, 93.2, 55.3, 44.2, 37.7, 37.0, 33.1, 22.2, 12.9, 10.8; IR ν_{max}: 3234.5, 3190.7, 2965.0, 1688.8, 1642.4, 1508.2, 1462.4, 1346.1, 1299.6, 1234.6, 1179.7, 1028.0, 1014.0, 839.2, 742.3, 661.0, 523.2, 474.3 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{19}H_{24}N_3O_4$ 358.1767; found: 358.1765; mp 80.9−83.7 °C.

3-Butyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7ada). Solid, 71 mg, isolated yield of 61%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.63 (d, J = 4.9 Hz, 1H), 7.14−7.17 (m, 2H), 6.80−6.83 (m, 2H), 4.43 (d, J = 6.7 Hz, 1H), 3.85−3.97 (m, 4H), 3.76 (s, 3H), 2.97 (dd, J = 16.3, 8.0 Hz, 1H), 2.86 (d, J = 15.7 Hz, 1H), 1.56−1.64 (m, 4H), 1.35 (sxt, J = 7.5 Hz, 2H), 0.96 (t, J = 7.4 Hz, 3H), 0.92 (t, J = 7.3 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 171.4, 160.7, 158.4, 150.3, 143.2, 133.0, 127.1, 113.9, 92.7, 54.9, 43.8, 41.3, 37.3, 32.7, 29.4, 21.8, 19.8, 13.4, 10.4; IR ν_{max}: 3171.5, 2959.1, 2930.8, 2872.1, 1683.6, 1636.1, 1509.2, 1475.1, 1438.0, 1355.2, 1330.7, 1296.3, 1249.9, 1234.7, 1175.5, 1147.8, 1087.2, 1031.8, 834.3, 779.2, 755.0, 659.5, 529.0, 495.1, 474.1 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{21}H_{28}N_3O_4$ 386.2080; found: 386.2082; mp 146.3−147.7 °C.

3-Isobutyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aea). Solid, 87 mg; isolated yield of 75%; ^1H NMR (400 MHz, $CDCl_3$) δ 8.97 (s, 1H), 7.14−7.19 (m, 2H), 6.80−6.85 (m, 2H), 4.46 (d, J = 6.4 Hz, 1H), 3.88−4.04 (m, 2 H), 3.80 (t, J = 7.1 Hz, 2 H), 3.77 (s, 3H), 3.00 (dd, J = 16.6, 7.8 Hz, 1H), 2.88 (dd, J = 16.4, 1.5 Hz, 1H), 2.13 (spt, J = 7.0 Hz, 1H), 1.61−1.73 (m, 2H), 0.88−0.99 (m, 9H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 172.1, 161.4, 158.8, 151.0, 143.6, 133.4, 127.5, 114.3, 93.0, 55.2, 48.6, 44.2, 37.7, 33.1, 27.1, 22.1, 20.2, 20.1, 10.8; IR ν_{max}: 3165.2, 2959.4, 2874.0, 2833.6, 1683.2, 1636.2, 1509.4, 1477.4, 1332.3, 1295.6, 1250.2, 1233.1, 1175.2, 1147.3, 1086.9, 1034.0, 834.2, 779.2, 754.2, 658.3, 474.2 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{21}H_{28}N_3O_4$ 386.2080; found: 386.2081; mp 153.8−156.9 °C.

1,3-Dibutyl-5-(4-methoxyphenyl)-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7afd). Solid, 92 mg, isolated yield of 77%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.10−8.16 (m, 1H), 7.17−7.21 (m, 2H), 6.83−6.87 (m, 2H), 4.47 (d, J = 6.5 Hz, 1H), 3.90−4.00 (m, 4H), 3.79 (s, 3H), 3.00 (dd, J = 16.6, 7.7 Hz, 1H), 2.91 (dd, J = 16.7, 1.4 Hz, 1H), 1.61−1.63 (m, 4H), 1.35−1.47 (m, 4H), 1.00 (t, J = 7.3 Hz, 3H), 0.96 (t, J = 7.4 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 172.0, 162.0, 159.8, 151.7, 144.4, 134.5, 128.5, 115.3, 94.2, 56.3, 43.7, 42.7, 38.6, 34.2, 31.9, 30.8, 21.2, 20.9, 14.7, 14.6; IR ν_{max}: 3223.5, 3176.3, 2958.4, 2931.7, 2872.3, 1684.9, 1634.4, 1505.3, 1472.4, 1356.9, 1297.7, 1247.4, 1224.6, 1175.6, 1148.0, 1030.4, 831.5, 780.8, 755.3, 685.4, 658.2, 556.6, 529.2 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{22}H_{30}N_3O_4$ 400.2236; found: 400.2236; mp 95.7−98.9 °C.

5-(4-Methoxyphenyl)-3-pentyl-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aga). Solid, 82 mg, isolated yield of 69%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.52 (s, 1H), 7.13−7.18 (m, 2H), 6.79−6.84 (m, 2H), 4.44 (dd, J = 7.9, 1.4 Hz, 1H), 3.85−3.96 (m, 4H), 3.76 (s, 3H), 2.94−3.00 (m, 1H), 2.87 (dd, J = 16.3, 0.9 Hz, 1H), 1.56−1.66 (m, 4H), 1.27−1.35 (m, 4H), 0.96 (t, J = 7.4 Hz, 3H), 0.88 (t, J = 7.1 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 171.5, 161.0, 158.8, 150.7, 143.5, 133.5, 127.5, 114.3, 93.2, 55.3, 44.2, 41.9, 37.6, 33.1, 29.1, 27.3, 22.4, 22.2, 14.0, 10.9; IR ν_{max}: 2958.8, 2932.9, 1687.4, 1634.4, 1506.1, 1474.2, 1355.6, 1296.4, 1234.6, 1176.7, 1032.2, 831.5, 780.9, 755.6 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{22}H_{30}N_3O_4$ 400.2236; found: 400.2235; mp 124.6−126.5 °C.

3-Hexyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aha). Solid, 92 mg, isolated yield of 74%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.84 (s, 1H), 7.13−7.17 (m, 2H), 6.79−6.83 (m, 2H), 4.43 (dd, J = 7.9, 1.4 Hz, 1H), 3.84−3.99 (m, 4H), 3.75 (s, 3H), 2.97 (dd, J = 16.4, 8.2 Hz, 1H), 2.86 (dd, J = 16.6, 1.3 Hz, 1H), 1.55−1.73 (m, 4H), 1.25−1.35 (m, 6 H), 0.95 (t, J = 7.4 Hz, 3H), 0.86 (t, J = 7.1 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 171.6, 160.7, 158.3, 150.3, 143.2, 133.0, 127.1, 113.9, 92.7, 54.9, 43.8, 41.5, 37.3, 32.7, 31.1, 27.2, 26.2, 22.2, 21.8, 13.6, 10.4; IR ν_{max}: 3246.3, 2953.0, 2927.2, 1690.1, 1637.3, 1508.9, 1465.6, 1353.0, 1299.5, 1239.0, 1176.2, 1033.4, 833.4, 754.2, 735.9, 664.9, 557.4, 510.3 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{23}H_{32}N_3O_4$ 414.2393; found: 414.2390; mp 131.9−134.7 °C.

3-Heptyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aia). Solid, 111 mg, isolated yield of 87%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.92 (s, 1H), 7.13−7.17 (m, 2H), 6.79−6.83 (m, 2H), 4.43 (dd, J = 7.9, 1.4 Hz, 1H), 3.86−3.99 (m, 4H), 3.75 (s, 3H), 2.97 (dd, J = 16.6, 8.0 Hz, 1H), 2.86 (dd, J = 16.5, 1.2 Hz, 1H), 1.56−1.67 (m, 4H), 1.21−1.34

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(m, 8H), 0.95 (t, J = 7.4 Hz, 3H), 0.86 (t, J = 7.1 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 171.7, 160.7, 158.4, 150.3, 143.2, 133.0, 127.1, 113.9, 92.7, 54.9, 43.8, 41.5, 37.3, 32.8, 31.8, 28.6, 27.3, 26.5, 22.2, 21.8, 13.7, 10.4; IR ν_{max}: 3234.1, 3184.0, 2963.2, 2927.4, 2854.5, 1686.7, 1633.6, 1510.2, 1472.7, 1348.2, 1294.0, 1239.4, 1180.7, 1034.4, 826.3, 781.4, 756.4, 738.2, 485.0 cm^{-1}; HRMS (ESI-TOF): $(M + H)^+$ calcd for $C_{24}H_{34}N_3O_4$ 428.2549; found: 428.2550; mp 129.9−131.6 °C.

3-Allyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aja). Solid, 80 mg; isolated yield of 69%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.43 (s, 1H), 7.13−7.17 (m, 2H), 6.80−6.83 (m, 2H), 5.82−5.91 (m, 1H), 5.24 (dq, J = 17.1, 1.4 Hz, 1H), 5.17 (dq, J = 10.2, 1.3 Hz, 1H), 4.54 (ddt, J = 19.7, 5.9, 1.3, 1.3 Hz, 2H), 4.44 (dd, J = 7.9, 1.4 Hz, 1H), 3.91 (m, J = 18.7, 9.7, 5.8 Hz, 2H), 3.76 (s, 3H), 2.97 (dd, J = 16.4, 8.0 Hz, 1H), 2.87 (dd, J = 16.5, 1.0 Hz, 1H), 1.62−1.74 (m, 2H), 0.97 (t, J = 7.4 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 171.1, 160.4, 158.4, 150.1, 143.3, 132.9, 131.4, 127.1, 117.6, 113.9, 92.7, 54.9, 43.9, 43.4, 37.2, 32.7, 21.7, 10.5; IR ν_{max}: 3171.5, 2936.3, 1688.1, 1651.3, 1509.0, 1464.7, 1348.0, 1303.2, 1237.4, 1174.2, 1031.4, 934.6, 831.6, 778.3, 758.0, 686.9, 557.1, 518.2, 461.1 cm^{-1}; HRMS (ESI-TOF): $(M + H)^+$ calcd for $C_{20}H_{24}N_3O_4$ 370.1767; found: 370.1766; mp 137.6−140.0 °C.

5-(4-Methoxyphenyl)-3-(2-methylallyl)-1-propyl-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aka). Solid, 90 mg, isolated yield of 79%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.76 (s, 1H), 7.13−7.17 (m, 2H), 6.79−6.82 (m, 2H), 4.82−4.84 (m, 1H), 4.65 (s, 1H), 4.46−4.52 (m, 2H), 3.94−4.01 (m, 1H), 3.87−3.93 (m, 1H), 3.75 (s, 3H), 2.99 (dd, J = 16.6, 8.0 Hz, 1H), 2.88 (d, J = 16.6 Hz, 1H), 1.75 (d, J = 0.4 Hz, 3H), 1.62−1.73 (m, 2H), 0.95 (t, J = 7.4 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 172.1, 160.6, 158.4, 150.3, 143.6, 139.2, 132.9, 127.1, 113.9, 110.0, 92.6, 54.9, 46.0, 43.9, 37.4, 32.6, 21.7, 20.2, 10.3; IR ν_{max}: 3165.1, 2965.4, 2936.3, 1683.8, 1639.4, 1508.3, 1478.2, 1436.4, 1354.4, 1296.9, 1249.5, 1232.4, 1174.4, 1148.5, 1032.4, 906.4, 839.2, 779.3, 753.4, 659.2, 528.9 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{21}H_{26}N_3O_4$ 384.1923; found: 384.1923; 154.4−156.6 °C.

3-Benzyl-5-(4-methoxyphenyl)-1-propyl-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aba). Solid, 113 mg, isolated yield of 90%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.73 (s, 1H), 7.45 (d, J = 7.10 Hz, 2H), 7.27−7.32 (m, 2H), 7.22−7.25 (m, 1H), 7.15 (d, J = 8.60 Hz, 2H), 6.81 (d, J = 8.71 Hz, 2H), 5.11 (dd, J = 13.70, 20.40 Hz, 2H), 4.44 (d, J = 7.10 Hz, 1H), 3.83−3.96 (m, 2H), 3.76 (s, 3H), 2.95 (dd, J = 8.00, 16.60 Hz, 1H), 2.85 (d, J = 15.38 Hz, 1H), 1.59−1.64 (m, 2H), 0.93 (t, J = 7.42 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 172.5, 161.2, 158.8, 150.9, 144.0, 137.0, 133.3, 129.0, 128.4, 127.6, 127.5, 114.3, 93.1, 55.3, 44.9, 44.4, 37.8, 33.1, 22.1, 10.7; IR ν_{max}: 3244.0, 2965.1, 2934.1, 1688.0, 1634.7, 1506.8, 1472.9, 1454.4, 1436.5, 1360.9, 1306.0, 1237.4, 1180.2, 1031.9, 828.3, 745.0, 694.4, 598.4, 550.7, 523.2, 494.7, 461.0 cm^{-1}; HRMS (ESI-TOF) $(M + H)^+$ m/z: calcd for $C_{24}H_{26}N_3O_4$ 420.1923; found: 420.1923; mp 164.9−177.1 °C.

3-Benzyl-5-(4-methoxyphenyl)-1-phenyl-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7abc). Solid, 98 mg, isolated yield of 72%; ^1H NMR (400 MHz, $CDCl_3$) δ 7.57−7.64 (m, 3H), 7.50−7.56 (m, 2H), 7.29−7.36 (m, 5H), 7.22−7.26 (m, 2H), 6.84−6.90 (m, 2H), 6.79 (s, 1H), 5.14 (dd, J = 21.0, 13.4 Hz, 3H), 4.51 (dd, J = 7.8, 1.2 Hz, 2H), 3.80 (s, 3H), 3.00 (dd, J = 16.6, 7.8 Hz, 2H), 2.87 (dd, J = 16.3, 0.7 Hz, 2H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 169.4, 161.2, 158.9, 150.6, 143.1, 136.8, 133.7, 132.2, 130.9, 130.9, 130.8, 129.6, 129.3, 129.0, 128.4, 127.7, 127.6, 114.4, 55.3, 44.9, 37.5, 33.5; IR ν_{max}: 2928.5, 1700.2, 1637.0, 1484.3, 1239.2, 1141.9, 1029.5, 938.6, 832.6, 746.3, 685.5 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{27}H_{24}N_3O_4$ 454.1767; found: 454.1769; mp 93.1−98.9 °C.

3-Allyl-1-benzyl-5-(4-methoxyphenyl)-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7ajb). Solid, 73 mg, isolated yield of 58%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.00 (s, 1 H), 7.36−7.40 (m, 2H), 7.33−7.36 (m, 1H), 7.20−7.24 (m, 2H), 7.07−7.11 (m, 2H), 6.77−6.80 (m, 2H), 5.91 (ddt, J = 17.1, 10.2, 5.8, 5.8 Hz, 1H), 5.44 (d, J = 17.0 Hz, 1H), 5.27 (dq, J = 17.1, 1.4 Hz, 1H), 5.20 (dq, J = 10.3, 1.2 Hz, 1H), 5.08 (d, J = 16.8 Hz, 1H), 4.60 (ddt, J = 18.3, 5.8, 1.4, 1.4 Hz, 2H), 4.41 (dd, J = 8.0, 1.5 Hz, 1H), 3.74−3.76 (m, 3H), 2.90 (dd, J = 16.3, 8.0 Hz, 1H), 2.79 (dd, J = 16.3, 1.5 Hz, 1H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 170.2, 160.3, 158.4, 150.6, 143.8, 133.9, 132.8, 131.3, 129.6, 129.2, 128.3, 127.1, 125.8, 117.7, 113.9, 54.9, 45.7, 43.6, 37.3, 33.0; IR ν_{max}: 3236.9, 3182.4, 1689.9, 1631.7, 1509.6, 1452.5, 1349.4, 1299.4, 1246.9, 1185.8, 1154.0, 1032.9, 933.5, 722.9, 692.5, 589.6, 522.0 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{24}H_{24}N_3O_4$ 418.1767; found: 418.1765; mp 143.6−145.9 °C.

1,3-Dibenzyl-5-(4-methoxyphenyl)-5,8-dihydropyrido-[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7abb). Solid, 112 mg, isolated yield of 80%; ^1H NMR (700 MHz, $CDCl_3$) δ 7.48−7.50 (m, 3H), 7.38−7.42 (m, 2H), 7.36 (d, J = 7.5 Hz, 1H), 7.29−7.33 (m, 2H), 7.26−7.29 (m, 1H), 7.21−7.24 (m, 2H), 7.07−7.10 (m, 2H), 6.76−6.80 (m, 2H), 5.50 (d, J = 16.8 Hz, 1H), 5.19 (d, J = 13.8 Hz, 1H), 5.13 (d, J = 13.8 Hz, 1H), 4.91−4.97 (m, 1H), 4.42 (dd, J = 8.0, 1.5 Hz, 1H), 3.75 (s, 3H), 2.89 (dd, J = 16.3, 8.0 Hz, 1H), 2.78 (dd, J = 16.3, 1.5 Hz, 1H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 169.8, 160.6, 158.4, 151.0, 143.8, 136.4, 133.8, 132.8, 129.3, 128.7, 128.4, 128.1, 127.3, 127.1, 125.7, 114.0, 93.3, 54.9, 45.9, 44.7, 37.3, 33.1; IR ν_{max}: 1693.6, 1626.6, 1505.6, 1454.0, 1435.1, 1351.9, 1295.8, 1231.1, 1177.3, 1147.3, 1029.5, 830.2, 730.4, 696.6, 602.5, 525.7, 490.6 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{28}H_{26}N_3O_4$ 468.1923; found: 468.1922; mp 93.4−100.2 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-(2-methylbenzyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7alb). Solid, 62 mg, isolated yield of 43%; ^1H NMR (700 MHz, $CDCl_3$) δ 8.38 (s, 1H), 7.30−7.36 (m, 3H), 7.09−7.19 (m, 7H), 7.03 (d, J = 7.3 Hz, 1H), 6.76−6.83 (m, 2H), 5.34 (d, J = 16.6 Hz, 1H), 5.12−5.23 (m, 3H), 4.44 (d, J = 6.7 Hz, 1H), 3.76 (s, 3H), 2.91 (dd, J = 16.5, 7.9 Hz, 1H), 2.76−2.84 (m, 1H), 2.42 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 170.5, 160.7, 158.4, 151.0, 144.0, 135.5, 134.2, 134.1, 132.7, 130.0, 129.1, 128.2, 127.1, 126.8, 125.8, 125.6, 125.5, 114.1, 93.2, 54.9, 45.6, 42.2, 37.3, 33.0, 19.0; IR ν_{max}: 3235.8, 3176.8, 1684.1, 1643.2, 1508.4, 1460.7, 1301.5, 1247.5, 1230.6, 117.0, 1147.7, 1028.5, 833.1, 739.6, 700.4, 473.6 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{29}H_{28}N_3O_4$ 482.2080; found: 482.2079; mp 198.9−203.2 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-(3-methylbenzyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7amb). Solid, 62 mg, isolated yield 43%; ^1H NMR (700 MHz, $CDCl_3$) δ 7.75 (s, 1H), 7.33−7.39 (m, 3H), 7.26−7.29 (m, 2H), 7.18−7.23 (m, 3H), 7.06−7.11 (m, 3H), 6.76−6.80 (m, 2H), 5.46 (d, J = 16.8 Hz, 1H), 5.15 (d, J = 14.0 Hz, 1H), 5.11 (d, J = 13.8 Hz, 1H), 5.00 (d, J = 16.8 Hz, 1H), 4.42 (dd, J = 7.7, 1.3 Hz, 1H), 3.75 (s, 3H), 2.89 (dd, J = 16.5, 7.9 Hz, 1H), 2.78 (dd, J = 16.6, 1.3 Hz, 1H), 2.32 (s, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 169.8, 160.6, 158.4, 151.0, 143.8, 137.7, 136.3, 133.8, 132.8, 129.3, 129.2, 128.4, 128.1, 128.0, 127.1, 125.7, 125.6, 113.9, 93.3, 54.9, 45.9, 44.7, 37.3, 33.1, 21.0; IR ν_{max}: 1693.6, 1621.5, 1505.8, 1454.6, 1435.1, 1350.0, 1296.9, 1230.6, 1177.4, 1147.1, 1030.3, 830.8, 780.0, 755.7, 729.6, 695.8, 525.6, 467.4 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{29}H_{28}N_3O_4$ 482.2080; found: 482.2079; mp 82.2−85.5 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-(4-methylbenzyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7anb). Solid, 50 mg, isolated yield of 34%; ^1H NMR (700 MHz, $CDCl_3$) δ 7.37 (d, J = 8.0 Hz, 2H), 7.29−7.33 (m, 4H), 7.16 (dd, J = 7.2, 2.3 Hz, 2H), 7.07−7.12 (m, 4H), 6.76−6.79 (m, 2H), 5.09−5.15 (m, 5H), 4.39 (dd, J = 7.7, 1.3 Hz, 1H), 3.74 (s, 3H), 2.83 (dd, J = 16.4, 8.0 Hz, 1H), 2.74 (dd, J = 16.4, 1.7 Hz, 1H), 2.31 (s, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 170.1, 161.0, 158.8, 151.4, 144.2, 137.4, 134.3, 133.9, 133.2, 129.7, 129.1, 129.1, 128.8, 127.5, 126.1, 114.4, 93.7, 55.3, 46.3, 44.9, 37.7, 33.5, 21.2; IR ν_{max}: 1693.6, 1621.6, 1506.4, 1453.3, 1435.5, 1351.6, 1296.8, 1232.7, 1178.7, 1148.0, 1029.8, 831.2, 782.1, 729.7, 697.0, 526.0, 472.9 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{29}H_{28}N_3O_4$ 482.2080; found: 482.2080; mp 72.1−79.6 °C.

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1-Benzyl-3-(4-isopropylbenzyl)-5-(4-methoxyphenyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aob). Solid, 105 mg, isolated yield of 69%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.34−7.52 (m, 6H), 7.25 (d, J = 7.6 Hz, 2H), 7.19 (d, J = 8.1 Hz, 2H), 7.08−7.15 (m, 2H), 6.77−6.83 (m, 2H), 5.52 (d, J = 16.9 Hz, 1 H), 5.19 (d, J = 13.7 Hz, 1 H), 5.12 (d, J = 13.7 Hz, 1H), 4.94 (d, J = 16.9 Hz, 1H), 4.44 (d, J = 5.9 Hz, 1H), 3.77 (s, 3H), 2.91 (dd, J = 13.8, 6.7 Hz, 1H), 2.76−2.84 (m, 1H), 1.25 (d, J = 6.8 Hz, 6H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.3, 161.0, 158.8, 151.4, 148.3, 144.2, 134.3, 134.2, 133.2, 129.6, 129.2, 128.7, 127.5, 126.5, 126.2, 114.4, 93.8, 55.3, 46.2, 44.9, 37.7, 33.8, 33.5, 24.0, 24.0; IR ν_{max}: 2957.8, 1694.3, 1628.9, 1506.6, 1459.9, 1352.1, 1294.7, 1231.9, 1178.0, 1147.0, 1031.0, 830.5, 780.9, 730.4, 697.1, 526.6 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{31}H_{32}N_3O_4$ 510.2393; found: 510.2392; mp 93.1−96.5 °C.

1-Benzyl-3-(3-methoxybenzyl)-5-(4-methoxyphenyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7apb). Solid, 115 mg, isolated yield of 77%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.37−7.45 (m, 4H), 7.22−7.27 (m, 3H), 7.03−7.13 (m, 4H), 6.78−6.86 (m, 3H), 5.52 (d, J = 16.9 Hz, 1H), 5.12−5.22 (m, 2H), 4.95 (d, J = 16.9 Hz, 1H), 4.44 (dd, J = 7.7, 1.6 Hz, 1H), 3.79 (s, 3H), 3.77 (s, 3H), 2.92 (dd, J = 16.6, 7.6 Hz, 1H), 2.81 (dd, J = 16.6, 1.0 Hz, 1H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.1, 160.9, 159.7, 158.8, 151.4, 144.2, 138.3, 134.2, 133.2, 129.7, 129.4, 128.8, 127.5, 126.1, 121.2, 114.4, 114.2, 113.4, 93.7, 55.3, 55.2, 46.3, 45.0, 37.7, 33.5; IR ν_{max}: 1694.3, 1632.8, 1506.6, 1454.4, 1351.2, 1286.8, 1232.4, 1148.3, 1032.1, 953.8, 831.4, 780.6, 757.3, 730.8, 694.8, 526.1, 481.6 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{29}H_{28}N_3O_5$ 498.2029; found: 498.2031; 82.5−86.8 °C.

1-Benzyl-3-(2-fluorobenzyl)-5-(4-methoxyphenyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aqb). Solid, 91 mg, isolated yield of 62%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.58 (s, 1H), 7.35−7.47 (m, 3H), 7.21−7.33 (m, 6H), 7.02−7.16 (m, 4H), 6.76−6.85 (m, 2H), 5.49 (d, J = 16.6 Hz, 1H), 5.29 (dd, J = 17.6, 14.7 Hz, 2H), 5.01 (d, J = 16.6 Hz, 1H), 4.45 (dd, J = 7.7, 1.6 Hz, 1H), 3.77 (s, 3H), 2.94 (dd, J = 16.4, 7.8 Hz, 1H), 2.82 (dd, J = 16.4, 1.0 Hz, 1H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.4, 162.0, 160.9, 159.6, 158.8, 151.2, 144.4, 134.3, 133.1, 129.6, 129.4 (d, J = 4.0 Hz), 129.1 (d, J = 7.9 Hz), 128.7, 127.5, 126.2, 124.1 (d, J = 3.2 Hz), 123.7 (d, J = 14.3 Hz), 115.5 (d, J = 21.5 Hz), 114.4, 93.6, 55.3, 46.2, 37.7, 33.4; IR ν_{max}: 2834.9, 1694.6, 1627.3, 1505.2, 1493.2, 1454.2, 1351.8, 1296.4, 1228.6, 1177.2, 1147.2, 1094.1, 1030.5, 831.3, 751.2, 729.6, 696.5, 654.7, 525.9, 507.4, 432.3 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{28}H_{25}FN_3O_4$ 486.1829; found: 486.1828; mp 84.1−90.9 °C.

1-Benzyl-3-(3-fluorobenzyl)-5-(4-methoxyphenyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7arb). Solid, 109 mg, isolated yield of 75%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.93 (s, 1H), 7.34−7.44 (m, 3H), 7.16−7.33 (m, 5H), 7.07−7.15 (m, 2H), 6.92−7.03 (m, 1H), 6.77−6.86 (m, 2H), 5.45 (d, J = 16.6 Hz, 1H), 5.17 (dd, J = 23.0, 13.9 Hz, 2H), 5.06 (d, J = 16.6 Hz, 1H), 4.44 (dd, J = 7.7, 1.6 Hz, 1H), 3.77 (s, 3H), 2.92 (dd, J = 16.4, 7.8 Hz, 1H), 2.81 (dd, J = 16.4, 1.7 Hz, 1H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.3, 164.0, 161.2 (d, J = 75.5 Hz), 158.9, 151.3, 144.4, 139.2 (d, J = 7.9 Hz), 134.2, 133.1, 129.9 (d, J = 8.7 Hz), 129.7, 128.8, 127.5, 126.1, 124.6, 115.8 (d, J = 22.3 Hz), 114.6 (d, J = 21.5 Hz), 114.4, 93.7, 55.3, 46.3, 44.6, 37.7, 33.5; IR ν_{max}: 3148.5, 3058.8, 3005.0, 2955.0, 2836.6, 1686.5, 1632.4, 1508.8, 1462.3, 1439.7, 1359.0, 1309.5, 1241.9, 1184.5, 1153.5, 1137.5, 1026.5, 965.5, 948.8, 832.9, 781.2, 755.5, 689.1, 519.4, 486.7 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{28}H_{25}FN_3O_4$ 486.1829; found: 486.1829; mp 173.3−174.5 °C.

1-Benzyl-3-(3-bromobenzyl)-5-(4-methoxyphenyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7asb). Solid, 95 mg, isolated yield of 58%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (s, 1H), 7.63 (t, J = 1.7 Hz, 1H), 7.35−7.46 (m, 5H), 7.16−7.26 (m, 3H), 7.07−7.14 (m, 2H), 6.77−6.86 (m, 2H), 5.46 (d, J = 16.9 Hz, 1H), 5.14 (dd, J = 22.0, 13.9 Hz, 2H), 5.04 (d, J = 16.6 Hz, 1H), 4.44 (d, J = 6.1 Hz, 1H), 3.77 (s, 3H), 2.93 (dd, J = 16.4, 7.8 Hz, 1H), 2.81 (dd, J = 16.4, 1.5 Hz, 1H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) δ 170.1, 160.8, 158.9, 151.3, 144.4, 139.0, 134.2, 133.1, 131.8, 130.9, 130.0, 129.7, 128.8, 127.7, 127.4, 126.1, 122.5, 114.4, 93.7, 55.3, 46.3, 44.5, 37.6, 33.5; IR ν_{max}: 3156.6, 2961.8, 2833.7, 1686.5, 1633.2, 1509.8, 1459.2, 1439.2, 1357.5, 1231.6, 1177.1, 1035.4, 779.3, 756.1, 695.5, 668.2, 650.1, 479.0 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{28}H_{25}BrN_3O_4$ 546.1028; found: 546.1031; mp 197.6−200.0 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-(3-(trifluoromethyl)-benzyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7atb). Solid, 139 mg, isolated yield of 87%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.72 (s, 1H), 7.67 (d, J = 7.7 Hz, 1H), 7.53 (d, J = 8.0 Hz, 1H), 7.51 (s, 1H), 7.34−7.47 (m, 4H), 7.20−7.25 (m, 2H), 7.07−7.11 (m, 2H), 6.75−6.82 (m, 2H), 5.48 (d, J = 16.8 Hz, 1H), 5.21 (dd, J = 25.2, 14.0 Hz, 2H), 4.97 (d, J = 16.6 Hz, 1H), 4.43 (dd, J = 7.9, 1.4 Hz, 1H), 3.75 (s, 3H), 2.92 (dd, J = 16.6, 8.0 Hz, 1H), 2.81 (dd, J = 16.5, 1.4 Hz, 1H); ^{13}C{^{1}H} NMR (176 MHz, $CDCl_3$) δ 169.4, 160.4, 158.5, 150.9, 144.0, 137.3, 133.6, 132.6, 132.2, 129.4, 128.6, 128.5, 127.0, 125.7, 125.2 (q, J = 4.1 Hz), 124.2 (q, J = 3.7 Hz), 114.0, 93.2, 54.9, 46.0, 44.2, 37.2, 33.0; IR ν_{max}: 1693.8, 1629.9, 1506.1, 1453.0, 1325.5, 1233.6, 1158.4, 1116.3, 1072.4, 1030.6, 830.9, 781.4, 730.0, 698.8, 657.0, 525.6, 477.5 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H): calcd for $C_{29}H_{25}F_3N_3O_4$ 536.1797; found: 536.1797; mp: 78.7−86.4 °C.

1-Benzyl-5-(4-methoxyphenyl)-3-(4-nitrobenzyl)-5,8-dihydropyrido[2,3-*d*]pyrimidine-2,4,7(1*H*,3*H*,6*H*)-trione (7aub). Solid, 44 mg, isolated yield of 29%; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.64−7.61 (m, 2H), 7.54 (s, 1H), 7.43−7.36 (m, 4 H), 7.24−7.20 (m, 2H), 7.10−7.05 (m, 2H), 6.82−6.77 (m, 2H), 5.50 (d, J = 16.8 Hz, 1H), 5.23 (dd, J = 14.1, 36.0 Hz, 2H), 4.96 (d, J = 16.7 Hz, 1H), 4.41 (dd, J = 1.3, 7.9 Hz, 1H), 3.76 (s, 3H), 2.92 (dd, J = 8.0, 16.5 Hz, 1H), 2.80 (dd, J = 1.3, 16.5 Hz, 1H); ^{13}C{^{1}H} NMR (176 MHz, $CDCl_3$) δ 169.3, 160.3, 158.5, 150.9, 147.1, 144.2, 143.5, 133.4, 132.6, 129.5, 129.4, 128.7, 127.0, 125.7, 123.3, 114.0, 93.2, 54.9, 46.1, 44.1, 37.2, 33.2; IR ν_{max}: 1693.7, 1620.2, 1508.9, 1341.6, 1292.0, 1229.4, 1175.4, 1147.6, 1109.6, 1030.9, 834.3, 807.5, 696.0, 527.4, 507.7 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{28}H_{25}N_4O_6$ 513.1774; found: 513.1772; mp 150.7−159.2 °C.

5-(4-Methoxyphenyl)-1,3-dipropyl-5,6,7,8-tetrahydropyrido[2,3-*d*]pyrimidine-2,4(1*H*,3*H*)-dione (9). A solution of **7aaa** (0.5 mmol; 0.19 g) in THF (5 mL) was added slowly to the solution of $LiAlH_4$ (0.5 mL; 1 M) cooled down to the temperature of 0 °C. The reaction was carried out at the same temperature for 24 h. The reaction mixture was diluted with ethyl acetate (10 mL), and the precipitated solid was filtered off. The crude product was purified by flash chromatography using gradient elution (0−100% AcOEt in hexane). The expected product was obtained as solid (194 mg) with a 72% yield; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.03−7.07 (m, 2H), 6.79−6.83 (m, 2H), 4.54 (s, 1H), 4.19 (d, J = 5.0 Hz, 1H), 3.77−3.93 (m, 4H), 3.76 (s, 3H), 3.29 (d, J = 12.0 Hz, 1H), 3.13 (dt, J = 2.5, 12.3 Hz, 1H), 1.97 (tt, J = 5.0, 13.0 Hz, 1H), 1.87 (dd, J = 2.3, 13.0 Hz, 1H), 1.73 (sxt, J = 7.6 Hz, 2H), 1.59−1.66 (m, 2H), 1.01 (t, J = 7.4 Hz, 3H), 0.88 (t, J = 7.4 Hz, 3H); ^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$) 161.5, 158.0, 151.4, 148.9, 137.7, 128.4, 113.8, 85.6, 55.3, 43.3, 42.7, 37.5, 34.4, 28.8, 21.6, 21.3, 11.4, 11.2; IR ν_{max}: 3301.0, 2960.2, 2932.8, 2873.3, 1685.9, 1590.3, 1538.2, 1507.9, 1443.3, 1347.7, 1336.0, 1264.4, 1241.4, 1170.1, 1032.4, 831.6, 770.3, 757.8, 549.7, 532.4, 485.6 cm^{-1}; HRMS (ESI-TOF) m/z: (M + H)$^+$ calcd for $C_{20}H_{28}N_3O_3$ 358.2131; found: 358.2131; mp 69.3−81.4 °C.

5-(4-Methoxyphenyl)-2,4-dioxo-1,3-dipropyl-7-(pyridin-1-ium-1-yl)-1,2,3,4,6,7-hexahydropyrido[2,3-*d*]pyrimidin-7-ide (10). To the solution of **7aaa** (1 mmol; 0,37 g) in pyridine (1 mL), tosyl chloride (1.1 mmol; 0.21 g) was added at a temperature of 0 °C. The reaction was then continued at room temperature for 24 h. After the addition of 5 mL of water, the product was extracted with ethyl acetate. The crude product was purified by flash chromatography using gradient elution (0−100% AcOEt in hexane). The expected compound was obtained as solid (86 mg) with a 20% yield; ^{1}H NMR (700 MHz, $CDCl_3$) δ 7.25−7.22 (m, 2H), 7.10 (s, 2H), 6.96−6.93 (m, 2H), 6.37 (s, 1H), 5.08 (dtd, J = 1.1, 3.6, 7.4 Hz, 2H), 4.29−4.25 (m, 2H), 3.91−3.87 (m, 2H), 3.85−3.84 (m, 3H), 2.99 (tt, J = 1.7, 3.6 Hz, 2H), 1.80 (tsxt, J = 1.8, 7.4 Hz, 2H), 1.62 (tsxt, J = 2.1, 7.5

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Hz, 2H), 1.01 (t, J = 7.4 Hz, 3H), 0.89 (t, J = 7.5 Hz, 3H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 160.0, 159.5, 155.8, 152.3, 151.7, 151.3, 132.3, 129.3, 123.7, 113.2, 107.3, 104.9, 100.8, 55.2, 44.6, 43.0, 23.2, 21.2, 21.1, 11.5, 11.3; IR ν_{max}: 2958.3, 1703.4, 1656.8, 1582.8, 1544.2, 1514.1, 1426.3, 1390.3, 1361.6, 1260.2, 1236.3, 1179.2, 1105.6, 1086.4, 1026.9, 969.3, 957.8, 893.1, 820.5, 755.8, 735.4, 705.7, 676.8, 574.9, 562.4, 488.5, 453.8 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{25}H_{29}N_4O_3$ 433.2240; found: 433.2243; mp 76.6−83.8 °C.

5-(4-Methoxyphenyl)-2,4-dioxo-1,3-dipropyl-1,2,3,4-tetrahydropyrido[2,3-*d*]pyrimidin-7-yl 4-methylbenzene-sulfonate (11). To the solution of **7aaa** (0.5 mmol; 0,19 g) in DIPEA (1 mL) and DCM (1 mL), tosyl chloride (0.55 mmol; 0.10 g) was added at a temperature of 0 °C. The reaction was then continued at room temperature for 24 h. After the addition of 5 mL of water, the product was extracted with DCM. The crude product was purified by flash chromatography using gradient elution (0−100% AcOEt in hexane). The expected compound was obtained as solid (91 mg) with a 35% yield; ^1H NMR (700 MHz, $CDCl_3$) δ 7.92−7.89 (m, 2H), 7.41−7.37 (m, 2H), 7.24−7.21 (m, 2H), 6.96−6.93 (m, 2 H), 6.70 (s, 1H), 4.03−3.99 (m, 2H), 3.91−3.87 (m, 2H), 3.85 (s, 4H), 2.47 (s, 3H), 1.61−1.57 (m, 4H), 0.90−0.87 (m, 6H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 159.7, 159.1, 158.9, 157.4, 151.0, 150.1, 145.3, 133.7, 129.9, 129.5, 129.1, 127.8, 113.0, 111.9, 106.2, 54.8, 44.4, 43.0, 21.4, 20.5, 20.5, 10.8, 10.7; IR ν_{max}: 2961.6, 2932.6, 2874.3, 1713.0, 1664.7, 1580.4, 1555.5, 1514.4, 1457.7, 1378.8, 1339.4, 1247.1, 1222.7, 1193.8, 1177.2, 1156.5, 1088.8, 1031.8, 946.3, 904.5, 830.4, 804.1, 754.5, 721.1, 665.2, 577.2, 546.1 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{27}H_{30}N_3O_6S$ 524.1855; found: 524.1855; mp 76.6−83.8 °C.

7-(Benzyloxy)-5-(4-methoxyphenyl)-1,3-dipropyl-5,6-dihydropyrido[2,3-*d*]pyrimidine-2,4(1*H*,3*H*)-dione (12). **7aaa** (0.25 mmol; 93 mg), benzyl bromide (1.25 mmol; 149 μL), and potassium carbonate (1.25 mmol; 0.16 g) were mixed in anhydrous DMF (1 mL) at 80 °C (oil bath) for 24 h. The reaction mixture was diluted with water (5 mL), and the product was extracted with ethyl acetate. The crude product was purified by flash chromatography using gradient elution (0−100% AcOEt in hexane). The expected compound was obtained as solid (73 mg) with a 63% yield; ^1H NMR (700 MHz, $CDCl_3$) δ 7.39−7.31 (m, 5H), 7.04−7.01 (m, 2H), 6.77−6.74 (m, 2H), 5.41 (d, J = 13.4 Hz, 1H), 5.36 (d, J = 12.3 Hz, 1H), 4.26 (d, J = 9.3 Hz, 1H), 4.11−4.06 (m, 1 H), 4.04−3.99 (m, 1H), 3.88 (ddd, J = 3.8, 6.5, 8.7 Hz, 2H), 3.75 (s, 3H), 2.89 (dd, J = 9.4, 16.8 Hz, 1H), 2.75 (dd, J = 1.0, 16.8 Hz, 1H), 1.71−1.61 (m, 4H), 0.95−0.89 (m, 6H); $^{13}C\{^1H\}$ NMR (101 MHz, $CDCl_3$) δ 172.5, 162.2, 158.6, 151.6, 150.7, 135.3, 134.1, 128.7, 128.5, 127.8, 127.6, 114.1, 95.5, 69.5, 55.2, 44.7, 43.0, 33.4, 33.4, 22.3, 21.0, 11.4, 11.3; IR ν_{max}: 2962.7, 2934.6, 2875.5, 1696.3, 1640.6, 1587.8, 1508.9, 1460.3, 1398.5, 1389.7, 1339.8, 1272.6, 1243.0, 1222.7, 1174.7, 1099.4, 1033.2, 1024.0, 951.8, 909.5, 892.6, 830.5, 785.7, 750.6, 697.9, 594.5, 559.2, 527.7 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{27}H_{32}N_3O_4$ 462.2393; found: 462.2392; mp 99.4−106.6 °C.

7-(Benzyloxy)-5-(4-methoxyphenyl)-1,3-dipropyl-pyrido-[2,3-*d*]pyrimidine-2,4(1*H*,3*H*)-dione (13). **7aaa** (0.25 mmol; 93 mg), benzyl chloride (1.25 mmol; 144 μL), and potassium carbonate (1.25 mmol; 0.16 g) were mixed in anhydrous DMF (1 mL) at 80 °C (oil bath) for 24 h. The reaction mixture was diluted with water (5 mL), and the product was extracted with ethyl acetate. The crude product was purified by flash chromatography using gradient elution (0−100% AcOEt in hexane). The expected compound was obtained as a viscous oil (52 mg) with a 45% yield; ^1H NMR (700 MHz, $CDCl_3$) δ 7.45−7.42 (m, 2H), 7.41−7.38 (m, 2H), 7.36−7.33 (m, 1H), 7.27−7.23 (m, 2H), 6.97−6.93 (m, 2H), 6.48 (s, 1H), 5.49 (s, 2H), 4.28−4.24 (m, 2H), 3.93−3.89 (m, 2H), 3.85 (s, 3H), 1.77−1.72 (m, 2H), 1.66−1.60 (m, 2H), 1.00 (t, J = 7.4 Hz, 2H), 0.90 (t, J = 7.5 Hz, 3H); $^{13}C\{^1H\}$ NMR (176 MHz, $CDCl_3$) δ 164.1, 159.6, 156.5, 150.6, 136.1, 131.1, 128.9, 128.2, 127.8, 127.1, 113.1, 112.8, 109.0, 101.8, 68.0, 44.4, 42.8, 20.9, 20.6, 11.1, 10.9; IR ν_{max}: 3466.3, 2960.6, 2932.4, 2874.1, 1705.8, 1658.7, 1594.5, 1553.0, 1514.8, 1491.8, 1453.2, 1394.4, 1341.4, 1288.2, 1246.3, 1225.4, 1209.4, 1174.8, 1105.0, 1086.2, 1032.4, 1016.2, 996.0, 828.8, 803.4, 752.7, 731.4, 695.9, 670.9, 569.4, 526.0 cm^{-1}; HRMS (ESI-TOF) m/z: $(M + H)^+$ calcd for $C_{27}H_{30}N_3O_4$ 460.2236; found: 460.2235.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.joc.0c00657.

NMR and IR spectra (PDF)

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Notes

The authors declare no competing financial interest.

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