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Fast and convenient synthesis of new symmetric pyrano[2,3-*d*:6,5-*d′*]dipyrimidinones by an organocatalyzed annulation reaction

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A B S T R A C T

A fast and facile one-pot procedure for the preparation of symmetric 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d′*]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives by two-component reaction of *N*-methylbarbituric acid and arylglyoxalmonohydrates catalyzed by DABCO in ethanol at 50 ºC is described. This protocol has the advantages of environmental friendless, very simple operation, high regio- and chemoselectivity and moderate to excellent yields.

1. Introduction

Fused heterocyclic scaffolds have attracted the attention of chemists due to their unique characteristics and wide applications in medicinal chemistry and material science.[1] For example, fused-pyran derivatives are an important class of heterocyclic scaffolds demonstrates a broad range of biological and pharmacological activities (Fig 1).[2] Among different fused-pyran derivatives, pyranopyrimidines are of significant importance in terms of their bioactivities (Fig 2).[3]

One-pot multicomponent reactions are highly efficient methods for the synthesis of natural and unnatural products due to their great advantages in environmental friendless.[4]

Green chemistry[5] emphasizes on the use of catalysts with specific properties such as high activity, cost-effective preparation, high stability and safety and also high selectivity.[6] In recent years, organocatalysis[7] has enhanced its importance as a tool for the synthesis of heterocyclic compounds.[8] 1,4-diazabicyclo[2.2.2]octane (DABCO) has emerged as an efficient organic base which has been successfully used for various organic transformations like Baylis-Hillman reaction,[9] *o*-alkylations of

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phenols,[10] synthesis of glycidic amidester,[11] cross-coupling reactions[12] and heterocyclic compound synthesis.[13]

As part of an ongoing investigation on the synthesis of heterocyclic compounds,[14] especially pyrano[2,3-*d*:6,5-*d*′]dipyrimidine scaffolds,[15] herein we wish to report a fast and convenient one-pot two-component process for the regio- and chemoselective synthesis of 5-aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d*′]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives from the reaction between *N*-methylbarbituric acid and arylglyoxalmonohydrates in ethanol medium at 50 ºC in the presence of DABCO as green base-organocatalyst (Scheme 1).

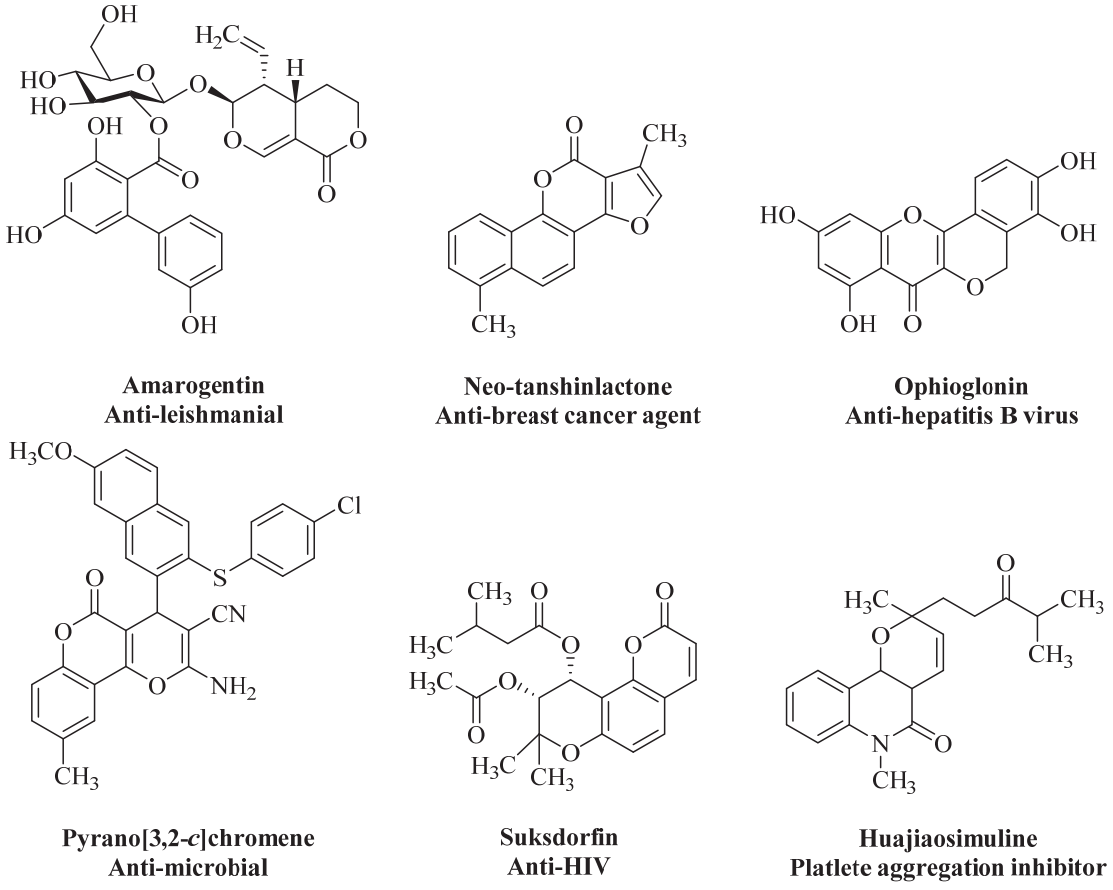


Fig 1. Examples of the bioactive compounds bearing pyran-annulated scaffolds.

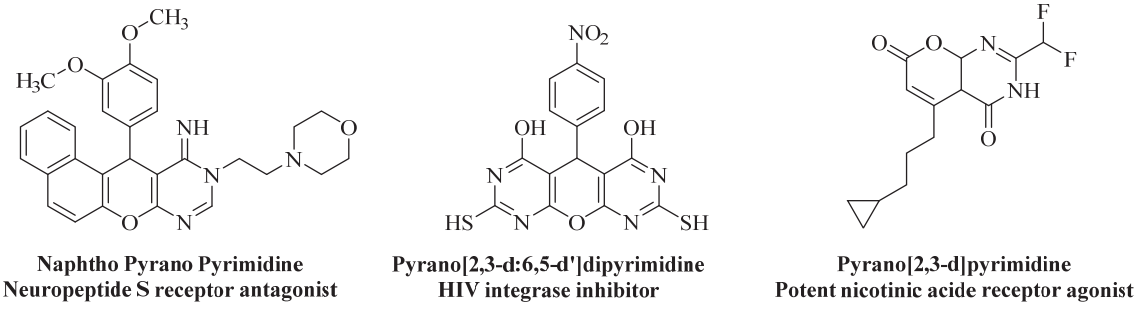
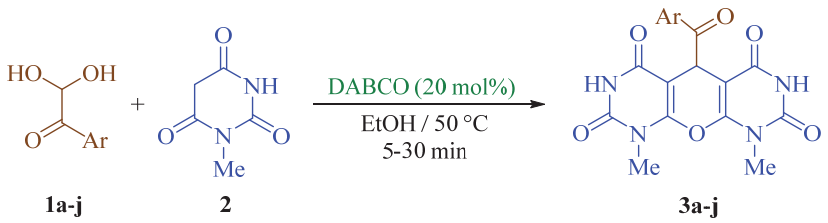


Fig 2. Biologically active pyranopyrimidine derivatives.



1a-j **2** **3a-j**

Ar = C_6H_5, 4-BrC_6H_4, 4-ClC_6H_4, 4-FC_6H_4, 4-$NO_2C_6H_4$, 4-$MeOC_6H_4$, 3-$MeOC_6H_4$, 3-BrC_6H_4, 3,4-$(MeO)_2C_6H_3$, 3,4-$(OCH_2O)C_6H_3$

Scheme1. One-pot two-component synthesis of pyrano[2,3-*d*:6,5-*d′*]dipyrimidinederivatives catalyzed by DABCO

2. Results and discussion

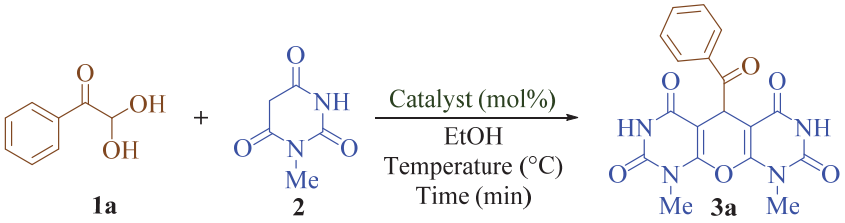
Firstly, we have started our study with the one-pot condensation of phenylglyoxalmonohydrate (**1a**) and *N*-methylbarbituric acid (**2**) in the presence of different basic catalysts such as 1,4-diazabicyclo[2.2.2]octane (DABCO),1,5-Diazabicyclo[4.3.0]non-5-ene (DBN), 1,8-Diazabicyclo[5.4.0]undec-7-ene (DBU), pyridine, dimethylamine (Me_2NH), potassium hydroxide (KOH), sodium hydroxide (NaOH), Potassium carbonate (K_2CO_3)and also acidic catalysts such as zirconium (IV) oxydichloride octahydrate ($ZrOCl_2.8H_2O$) and ammonium acetate (NH_4OAc). To further optimize the reaction conditions, the reaction was studied in different solvents such as ethanol, water, H_2O-EtOH (1:1), H_2O-EtOH (2:1), H_2O-EtOH (1:2), dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$), dimethylformamide (DMF), tetrahydrofuran (THF) and acetonitrile (CH_3CN). The effects of catalysts, solvents and temperatures were evaluated for this reaction and the results are summarized in Table 1. It was observed that 20 mol% of DABCO in ethanol at 50 ºC provided the best result in term of yields and time (Table 1, entry 8). We have attempted different ratios of DABCO (10, 15, 20 and 30 mol%) and observed that The increase and or decrease in the molar ratio of DABCO did not improve the yield.

As shown in Table 2, we investigated the reaction with a wide range of arylglyoxalmonohydrates with electron donating and electron withdrawing groups. Both electron rich and electron-deficient arylglyoxalmonohydrates worked well and give moderate to excellent yields of products under the optimization reaction conditions.

The structures of all products were secured on the basis of their spectral data. With surveys conducted on the spectrum data (especially ^{1}H NMR and FT IR data) determined that no exist any tautomeric forms (such as lactam-lactim or keto-enol tautomeric forms) in the structure of all the obtained 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d′*]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives (Scheme 3). For example, in the ^{1}H NMR spectrum of **3a** which is obtained as a sole product, the C_5-H proton of the pyran ring appears as a singlet at a δ= 5.92 ppm and also the singlet pick in the region of 9.53 ppm belong to the two NH protons.

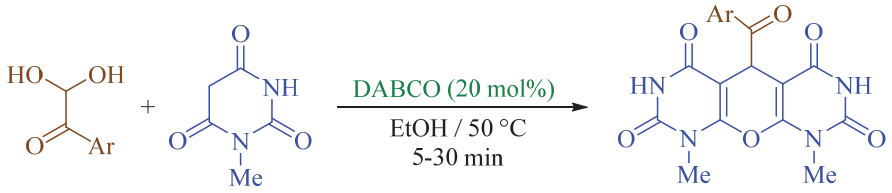
A proposed mechanism for the one-pot two-component regio- and chemoselective synthesis of new pyrano[2,3-*d*:6,5-*d′*]dipyrimidine derivatives from *N*-methylbarbituric acid (**2**) and arylglyoxalmonohydrates (**1a-j**) catalyzed by DABCO is shown in scheme 4. Firstly, DABCO as a green base-organocatalyst take off an acidic proton of *N*-methylbarbituric acid (**2**). Then, regioselective condensation of **5** with formyl group of arylglyoxal (**6a-j**) leads to intermediate **7** with elimination of water. The subsequent base-promoted Michael addition of **5** with Knoevenagel adduct (**7**) and then intra-molecular heterocyclization of **8** that leads to the formation of 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d′*]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives (**3a-j**).

Table 1. Optimization reaction conditions for the synthesis of **9a**

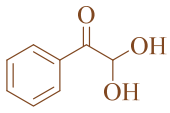
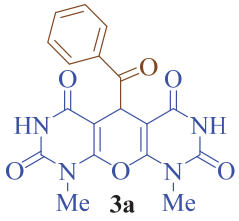
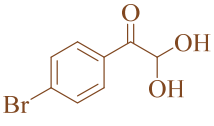
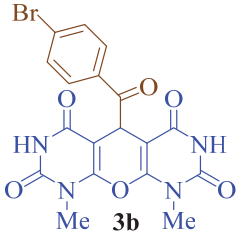
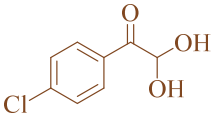
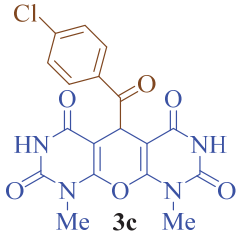
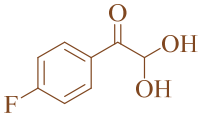
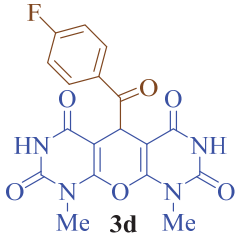


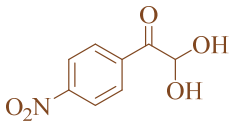
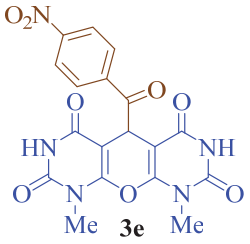
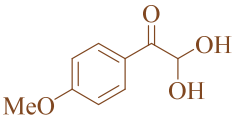
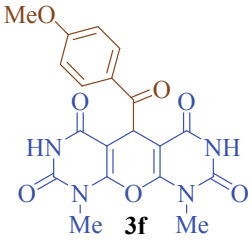
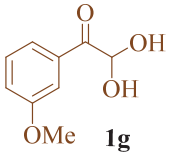
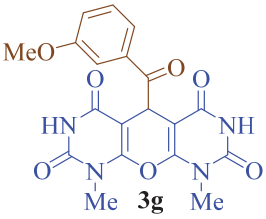
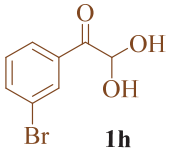
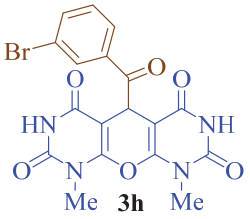
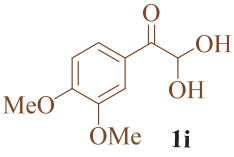
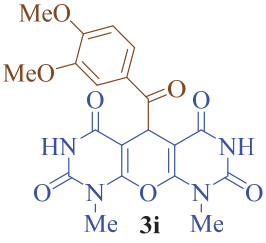
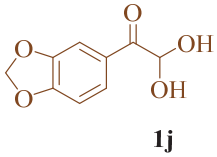
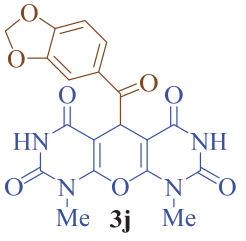
Entry	Solvent	Catalyst (mol%)	Temperature (˚C)	Time	Yield (%)
1	EtOH	-	rt	720	-
2	EtOH	-	50	720	-
3	EtOH	-	Reflux	720	-
4	EtOH	DABCO (5)	50	480	-
5	EtOH	DABCO (10)	50	180	30
6	EtOH	DABCO (15)	50	180	50
7	EtOH	DABCO (20)	rt	480	-
8	EtOH	DABCO (20)	50	10	88
9	EtOH	DABCO (20)	65	30	55
10	EtOH	DABCO (20)	Reflux	180	-
11	EtOH	DABCO (30)	50	10	88
12	H_2O	DABCO (20)	50	720	-
13	H_2O-EtOH	DABCO (20)	50	720	-
14	H_2O-EtOH	DABCO (20)	50	720	-
15	H_2O-EtOH	DABCO (20)	50	720	-
16	CH_2Cl_2	DABCO (20)	50	720	-
17	$CHCl_3$	DABCO (20)	50	720	-
18	DMF	DABCO (20)	50	720	-
19	THF	DABCO (20)	50	720	-
20	CH_3CN	DABCO (20)	50	720	-
21	EtOH	DBN (20)	50	20	75
22	EtOH	DBU (20)	50	10	85
23	EtOH	Pyridine (50)	50	480	-
24	EtOH	Me_2NH (50)	50	480	-
25	EtOH	NaOH (100)	50	480	-
26	EtOH	KOH (100)	50	480	-
27	EtOH	K_2CO_3 (10)	50	480	-
28	EtOH	$ZrOCl_2.8H_2O$ (10)	50	180	-
29	H_2O	$ZrOCl_2.8H_2O$ (10)	50	180	-
30	EtOH	$ZrOCl_2.8H_2O$ (20)	50	180	-
31	H_2O	$ZrOCl_2.8H_2O$ (20)	Reflux	180	-
32	EtOH	$ZrOCl_2.8H_2O$ (20)	Reflux	180	-
33	H_2O	$ZrOCl_2.8H_2O$ (20)	50	180	-
34	H_2O	NH_4OAC (100)	50	480	-
35	EtOH	NH_4OAC (100)	50	480	-

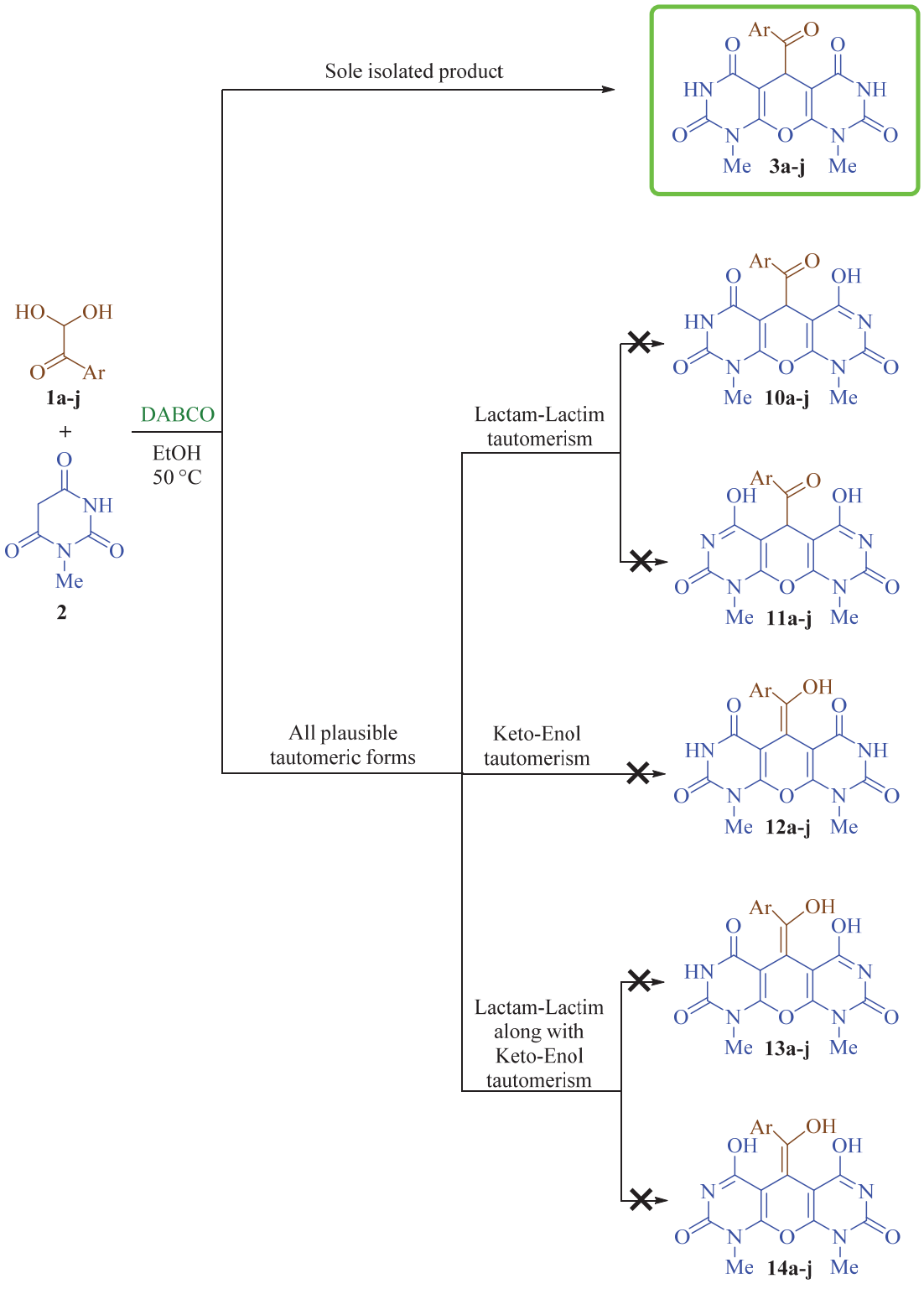
Table 2. Substrate scope for the synthesis of 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d'*]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives.



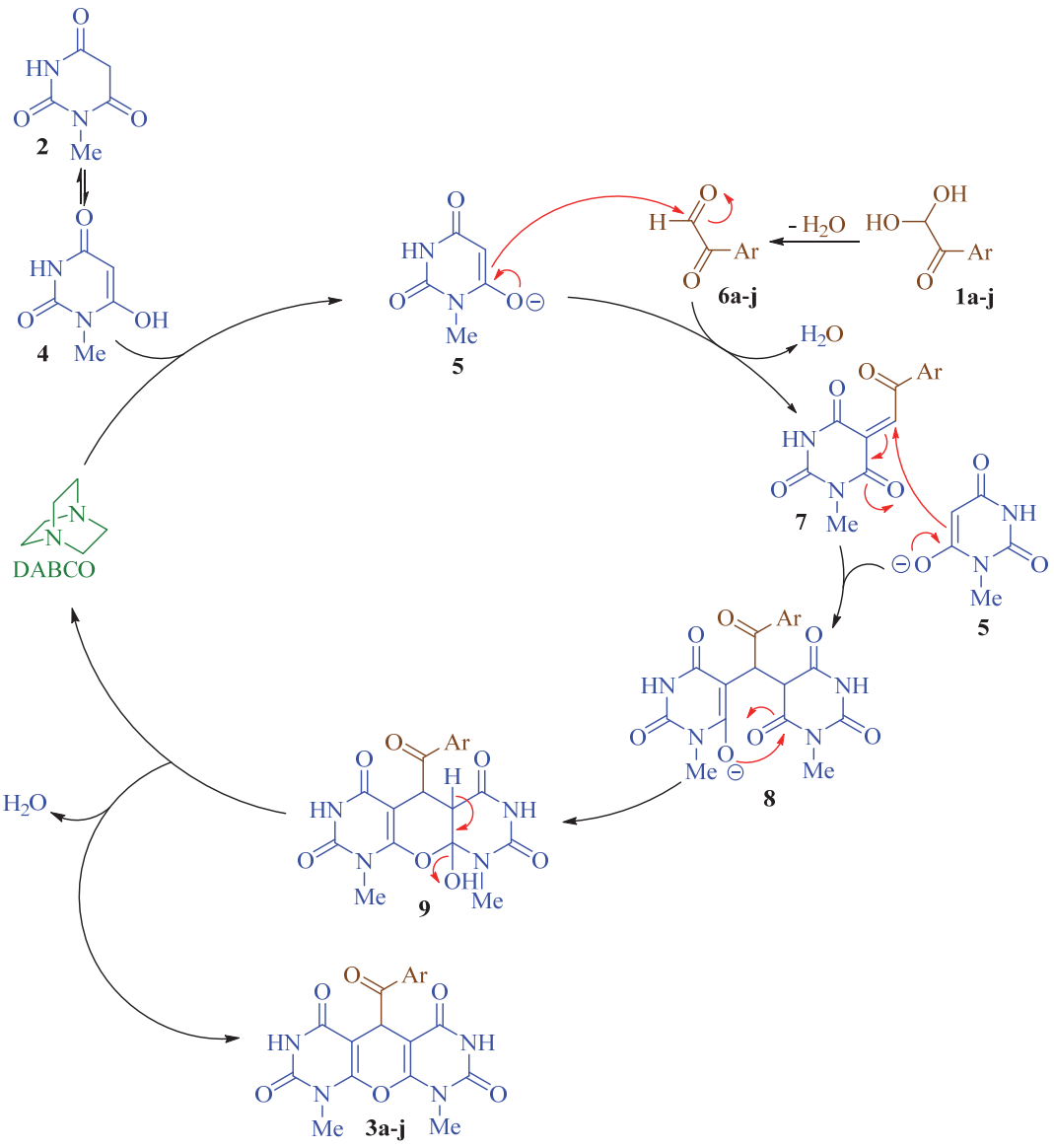
1a-j **2** **3a-j**

Entry	Arylglyoxalmonohydrate	Product	Time (min)	Yield (%)
1	 **1a**	 **3a**	10	88
2	 **1b**	 **3b**	10	60
3	 **1c**	 **3c**	10	60
4	 **1d**	 **3d**	5	90

5	 **1e**	 **3e**	5	95
6	 **1f**	 **3f**	15	76
7	 **1g**	 **3g**	5	93
8	 **1h**	 **3h**	5	91
9	 **1i**	 **3i**	30	50
10	 **1j**	 **3j**	30	50



Scheme 2. Possible structures of pyranodipyrimidine derivatives



Scheme 3. Plausible mechanism for synthesis of pyrano[2,3-*d*:6,5-*d'*]dipyrimidine derivatives catalyzed by DABCO.

3. Experimental

3.1. General

Melting points were determined on an Electrothermal 9200 apparatus. ^{1}H (300 MHz) and ^{13}C (75.5 MHz) NMR spectra were recorded on a BRUKER DRX-300 AVANCE spectrometer in DMSO-d_6 with tetramethylsilane as internal standard. Infrared spectra were recorded on a Perkin Elmer Spectrum Two FT-infrared spectrophotometer, measured as KBr disks. Microanalyses were performed on a Leco Analyzer *932*.

3.2. General procedure for the preparation of 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone derivatives

A mixture of arylglyoxalmonohydrates (1 mmol) and *N*-methylbarbituric acid (142 mg, 1 mmol) was stirred for 5-30 minutes in ethanol at 50 ºC in the presence of DABCO (22 mg, 20 mol%). After

completion of the reaction, the reaction mixture was cooled to room temperature and solid product was separated by just filtration and washed with excess cool ethanol (10 mL) and then washed with hot methanol (10 mL) to afford the pure products.

3.3 Physical and spectral data

*5-benzoyl-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3a**)* white, amorphous solid (169 mg, 88%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 2.94 (s, 6H, 2×N-CH$_3$), 5.92 (s, 1H, CH), 7.38 (t, *J* = 7.2 Hz, 2H, Ar), 7.50 (t, *J* = 7.2 Hz, 1H, Ar), 7.99 (d, *J* = 7.2 Hz, 2H, Ar), 9.53 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ: 26.4, 69.4, 86.6, 127.9, 128.5, 133.1, 135.6, 152.6, 162.9, 163.7, 200.6 ppm. FT-IR (KBr) ν_{max}: 3179, 3066, 2992, 2889, 1693, 1664, 1606, 1577, 1376, 1241, 779 cm^{-1}.Anal. Calcd. For $C_{18}H_{14}N_4O_6$: C, 56.55; H, 3.69; N, 14.65; Found: C, 56.58; H, 3.70; N, 14.85.

*5-(4-bromobenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3b**)* pink, amorphous solid (139 mg, 60%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 3.18 (s, 6H, 2×N-CH$_3$), 6.13 (s, 1H, CH), 7.55 (d, *J* = 8.1 Hz, 2H, Ar), 7.63 (d, *J* = 8.1 Hz, 2H, Ar), 10.36 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ: 27.2, 44.14, 88.9, 125.9, 129.8, 131.3, 136.8, 151.3, 162.3, 164.6, 198.9 ppm. FT-IR (KBr) ν_{max}: 3172, 3062, 2985, 2891, 1695, 1624, 1587, 1461, 1362, 1245, 1102, 769 cm^{-1}. Anal. Calcd. for $C_{18}H_{13}BrN_4O_6$: C, 46.87; H, 2.84; N, 12.15; Found: C, 46.89; H, 2.81; N, 12.30.

*5-(4-chlorobenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3c**)* white, amorphous solid (126 mg, 60%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 2.95 (s, 6H, 2×N-CH$_3$), 5.79 (s, 1H, CH), 7.44 (d, *J* = 8.4 Hz, 2H, Ar), 7.92 (d, *J* = 8.4 Hz, 2H, Ar), 9.55 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ: 26.5, 62.9, 83.4, 128.6, 129.9, 134.8, 137.8, 152.7, 163.2, 164.0, 197.6 ppm. FT-IR (KBr) ν_{max}: 3221, 3060, 2980, 2902, 1661, 1628, 1596, 1347, 1251, 1073, 826 cm^{-1}. Anal. Calcd. for $C_{18}H_{13}ClN_4O_6$: C, 51.87; H, 3.14; N, 13.44; Found: C, 51.84; H, 3.13; N, 13.44.

*5-(4-fluorobenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3d**)* pink, amorphous solid (181 mg, 90%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 3.02 (s, 6H, 2×N-CH$_3$), 6.15 (s, 1H, CH), 7.12-7.20 (m, 2H, Ar), 7.70-7.81 (m,2H, Ar), 10.40 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ:27.2, 44.7, 89.2, 115.4, 129.8, 131.0, 134.1, 135.8, 137.5, 151.2, 162.9, 165.2, 198.2 ppm. FT-IR (KBr) ν_{max}:3198, 3062, 2969, 2904, 1688, 1630, 1595, 1507, 1362, 1238, 1158, 771 cm^{-1}. Anal. Calcd. for $C_{18}H_{13}FN_4O_6$: C, 54.01; H, 3.27; N, 14.00; Found:C, 54.00; H, 3.26; N, 14.14.

*5-(4-nitrobenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3e**)* orange, amorphous solid (203 mg, 95%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 3.03 (s, 6H, 2×N-CH$_3$), 6.20 (s, 1H, CH), 7.87 (d, *J* = 8.1 Hz, 2H, Ar), 8.19 (d, *J* = 8.1 Hz, 2H, Ar), 10.46 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ: 27.3, 43.7, 87.9, 124.0, 128.7, 143.5, 149.4, 151.2, 162.6, 164.6, 199.1 ppm. FT-IR (KBr) ν_{max}: 3253, 3045, 2981, 2901, 1688, 1603, 1524, 1457, 1347, 1055, 1011, 769 cm^{-1}. Anal. Calcd. for $C_{18}H_{13}N_5O_8$: C, 50.59; H, 3.07; N, 16.39; Found: C, 50.62; H, 3.03; N, 16.60.

*5-(4-methoxybenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3f**)* cream, amorphous solid (157 mg, 76%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 3.03 (s, 6H, 2×N-CH$_3$), 3.76 (s, 1H, OCH$_3$),6.13 (s, 1H, CH), 6.86 (d, *J* = 8.4 Hz, 2H, Ar), 7.71(d, *J* = 8.4 Hz, 2H, Ar), 10.36 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, CDCl$_3$) δ: 27.2, 44.5, 55.6, 89.0, 113.5, 126.1, 130.1, 137.2, 151.3, 162.3, 164.6, 197.9 ppm. FT-IR (KBr) ν_{max}: 3162, 3066, 2991, 2897,

1702, 1679, 1628, 1585, 1364, 1267, 1174, 793 cm^{-1}. Anal. Calcd. for $C_{19}H_{16}N_4O_7$: C, 55.34; H, 3.91; N, 13.59; Found: C, 55.36; H, 3.88; N, 13.59.

*5-(3-methoxybenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3g**)* white, amorphous solid (192 mg, 93%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 2.95 (s, 6H, 2×N-CH_3), 3.74 (s, 1H, OCH_3),5.80 (s, 1H, CH), 7.06 (d, *J* = 8.1 Hz, 1H, Ar), 7.29(t, *J* = 8.1 Hz, 1H, Ar), 7.49-7.62 (m, 2H, Ar), 10.43 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, $CDCl_3$) δ: 26.5, 55.5, 62.9, 83.7, 112.9, 119.1, 120.4, 129.3, 129.6, 137.3, 152.7, 159.3, 163.2, 198.2 ppm. FT-IR (KBr) ν_{max}: 3223, 3061, 2974, 1661, 1625, 1594, 1346, 1269, 1096, 791, 679 cm^{-1}. Anal. Calcd. for $C_{19}H_{16}N_4O_7$: C, 55.34; H, 3.91; N, 13.59; Found: C, 55.36; H, 3.88; N, 13.75.

*5-(3-bromobenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3h**)* pale brown, amorphous solid (210 mg, 91%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 2.99 (s, 6H, 2×N-CH_3), 6.48 (s, 1H, CH), 7.32 (t, *J* = 7.5 Hz, 1H, Ar), 7.63-7.68 (m, 2H, Ar),7.81 (s, 1H, Ar), 10.43 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, $CDCl_3$) δ: 27.1, 44.1, 88.7, 122.4, 128.5, 131.9, 137.6, 149.8, 152.3, 161.5, 166.2, 198.1 ppm. FT-IR (KBr) ν_{max}: 3213, 3036, 2954, 2829, 1689, 1621, 1580, 1372, 1235, 1055, 896, 770 cm^{-1}. Anal. Calcd. for $C_{18}H_{13}BrN_4O_6$: C, 46.87; H, 2.84; N, 12.15; Found: C, 46.90; H, 2.83; N, 12.28.

*5-(3,4-dimethoxybenzoyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3i**)* pink, amorphous solid (111 mg, 50%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 3.03 (s, 6H, 2×N-CH_3), 3.68 (s, 1H, OCH_3),3.76 (s, 3H, OCH_3), 6.15 (s, 1H, CH), 6.91(d, *J* = 7.8 Hz, 1H, Ar), 7.37-7.47 (m, 2H, Ar), 10.41 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, $CDCl_3$) δ: 27.3, 44.0, 55.7, 56.0, 89.1, 111.3, 121.9, 129.8, 130.1, 147.9, 151.3, 152.1, 163.2, 164.7, 197.7 ppm. FT-IR (KBr) ν_{max}: 3258, 3078, 2954, 2904, 1710, 1699, 1609, 1365, 1267, 1021, 803, 782 cm^{-1}. Anal. Calcd. for $C_{20}H_{18}N_4O_8$: C, 54.30; H, 4.10; N, 12.66; Found: C, 54.32; H, 4.12; N, 12.87.

*5-(benzo[d][1,3]dioxole-5-carbonyl)-1,9-dimethyl-5,9-dihydro-2H-pyrano[2,3-d:6,5-d']dipyrimidine-2,4,6,8(1H,3H,7H)-tetraone (**3j**)* pink, amorphous solid (107 mg, 50%). ^{1}H NMR (300 MHz, DMSO-*d*$_6$) δ: 2.99 (s, 6H, 2×N-CH_3), 6.04 (s, 2H, CH_2), 6.14 (s, 1H, CH), 6.87 (d, *J* = 8.1 Hz, 1H, Ar), 7.21 (s, 1H, Ar), 7.36 (d, *J* = 8.1 Hz, 1H, Ar), 10.43 (s, 2H, 2×NH) ppm. ^{13}C NMR (75.5 MHz, $CDCl_3$) δ: 27.3, 43.9, 87.0, 102.0, 107.8, 123.5, 131.7, 138.1, 147.4, 150.6, 151.2, 162.8, 164.7, 197.4 ppm. FT-IR (KBr) ν_{max}: 3218, 040, 2974, 2009, 1687, 1606, 1504, 1444, 1361, 1254, 1038, 878, 803, 768 cm^{-1}. Anal. Calcd. for $C_{19}H_{14}N_4O_8$: C, 53.53; H, 3.31; N, 13.14; Found: C, 53.70; H, 3.31; N, 13.14.

4. Conclusions

In summary, we have developed a fast, green and very simple methodology for regio- and chemoselective synthesis of 5-Aryloyl-1,9-dimethyl-5,9-dihydro-2*H*-pyrano[2,3-*d*:6,5-*d'*]dipyrimidine-2,4,6,8(1*H*,3*H*,7*H*)-tetraone derivatives by one-pot reaction of *N*-methylbarbituric acid and arylglyoxalmonohydrates in the presence of DABCO as green base-organocatalyst in ethanol at 50 ºC. This method have advantages such as being inexpensive reagents, moderate to excellent yields, high atom economy and easy work-up.

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