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PAPER

Microwave-assisted synthesis of novel 2, 3-disubstituted imidazo[1,2-a]pyridines via one-pot three component reactions

Shaik Karamthulla, Md. Nasim Khan, and Lokman H. Choudhury*

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An expeditious one-pot, metal-free, three-component reaction of arylglyoxals, cyclic 1, 3-dicarbonyls and 2-aminopyridines in the presence of molecular iodine under microwave irradiation is reported. A wide variety of 2, 3-disubstituted imidazo-[1,2-a]pyridine derivatives can be synthesized in good to very good yields using this methodology. The salient features of this methodology are: metal-free, short reaction time, good yields, use of microwave heating and no harmful by-products.

Introduction

Imidazo[1,2-a]pyridines are important molecules both for the organic and medicinal chemists due to their wide range of biological and pharmacological activities.[1] Molecules with the imidazo[1,2-a]pyridine moiety exhibit anticancer,[2] anti-inflammatory,[3] antibacterial,[4] antiprotozonal,[5] antiviral,[6] antiulcer,[7] and antifungal[8] properties. Some of the commercially available drugs such as, alpidem **(I)** used as anxiolytic drug,[9] zolpidem **(II)** used in the treatment of insomnia and some brain disorders,[10] zolimidine **(III)**, used for the treatment of peptic ulcer and gastro esophageal reflux disease,[11] have imidazo[1,2-a]pyridine scaffold. Similarly, olprinone **(IV)** useful for the treatment of acute heart failure[12] and necopidem **(V)** and saripidem **(VI)** are also important sedative and anxiolytic agents[13] (Fig 1) having substituted imidazo[1,2-a]pyridine moiety.

Considering the widespread applications of substituted imidazo[1,2-a]pyridine moiety there is a continuing interest in developing new and efficient synthetic routes.[14] Some of the useful methods for the synthesis of imidazo [1,2-a]pyridines include reaction of α-halo carbonyl compounds with 2-aminopyridines,[15] condensation between 2-aminopyridines and 2-bromo-1,2-diarylethanone,[16] one-pot condensations of aldehydes, isonitriles, and 2-aminopyridines,[17] three-component reactions of aldehydes, alkynes and 2-aminopyridines using copper catalysis,[18] Suzuki-type cross-coupling between 3-halo-2-arylimidazo[1,2-a]pyridines and arylboronic acid,[19] oxidative couplings through C-H activation,[20] and from Morita–Baylis–Hillman acetates of nitroalkenes[21] etc.

Although these methods are useful still some of these methods have limitations such as use of expensive reagents or commercially less available substrates and α -halo carbonyl compounds which have lachrymatory properties.

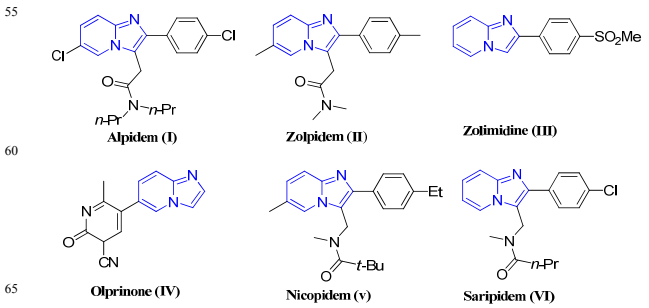
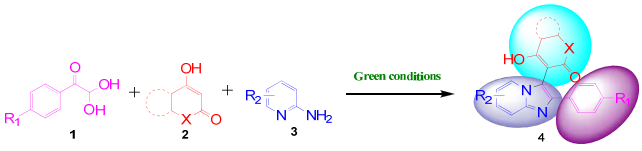


Fig 1. Representative examples of important substituted imidazo[1,2-a]pyridines.

Therefore, we realized the need of a new methodology for the rapid synthesis of substituted imidazo [1,2-a]pyridines from the readily available starting materials under green conditions.

Use of microwave heating technology in organic synthesis has gained tremendous popularity due to its ability to reduce reaction times dramatically, usually in minutes.[22] In addition to its advantage in terms of reaction time; it saves energy, cost as well as provides clean products in good to excellent yields. Multicomponent reactions (MCRs) in tandem with microwave-assisted chemistry offers lot of advantages in terms of selectivity, chemical yield, purity, enhanced reaction rates and simplicity.[23] Considering the merits of microwave assisted MCRs very recently we have explored this strategy for the synthesis of fused heterocycles. [24] In continuation of our work on arylglyoxal-based MCRs[25,26] herein, we report a convenient method to access disubstituted imidazo[1,2-a]pyridine derivatives using three-component reaction of arylglyoxals, cyclic 1, 3-dicarbonyls and 2-aminopyridines under microwave irradiation (Scheme 1).



Scheme 1. Synthesis of 2,3-disubstituted imidazo-[1,2-a]pyridines by three component reactions.

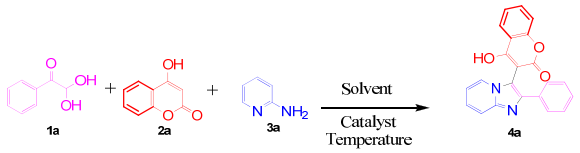
[a]*Department of Chemistry, Indian Institute of Technology Patna, Patna-800013, India..Tel: +91 612 2552038, Fax: +91 612 227 7383*
E-mail: lokman@iitp.ac.in

† Electronic Supplementary Information (ESI) available:: ^{1}H NMR and ^{13}C NMR spectra for all reactions products are available. See DOI: 10.1039/b000000x/

Result and discussion

Initially, phenylglyoxal monohydrate (**1a**), 4-hydroxycoumarin (**2a**) and 2-aminopyridine (**3a**) were selected for the model reaction to check the feasibility of the proposed reaction. In absence of any catalyst the combination of these substrates in ethanol did not provide our desired three component product **4a** both in room as well as reflux temperatures even after 24 hours (Table 1, entries 1-2). Next the same model reaction was tested in the presence of 20-mol% of *p*-toluene sulphonic acid (PTSA) in ethanol under both room temperature and reflux conditions. Unfortunately, in these cases also the desired three component product was not obtained. Changing solvent from ethanol to toluene in the presence of 20-mol% of PTSA under the reflux conditions provided only 6% of desired product **4a** after 24 h. The product **4a** was fully characterized by recording IR, ^{1}H & ^{13}C NMR as well as elemental analysis. Encouraged by this positive result we turned our attention to optimize the reaction condition by varying various parameters such as using microwave heating, changing catalyst, solvent etc. Interestingly, the same model reaction in the presence of 20-mol% PTSA in toluene under the influence of microwave heating at 130 ^{o}C for 15 minutes provided better yield (38%) than the conventional heating conditions (Table 1, entry 6). Next, the same model reaction was tested in the presence of various metal triflates such as $Cu(OTf)_2$, $Sc(OTf)_3$, and $Bi(OTf)_3$ in ethanol under the influence of microwave irradiation and the yields obtained were moderate to good. To further investigate we also performed the same model reaction using readily available Lewis acids $FeCl_3$ and CAN following the similar microwave reaction conditions. Results using these Lewis acids were found not encouraging (Table 1, entries 10 and 11). Then we shifted our attention to use molecular iodine as a catalyst in this MCR. Interestingly using the same amount of (20-mol%) iodine and microwave irradiation for 15 minutes the desired three component product was obtained in 72% yield (Table 1, entry 12). Encouraged by this result as well as considering the non metallic, ready availability and benign nature of molecular iodine.[27] we focused our optimization using iodine by varying the amount of iodine as well as changing solvents. Finally, the optimized condition was obtained using 30-mol% I_2 at 130 ^{o}C in ethanol as solvent (Table 1, entry 13).

Table 1 Optimization of reaction conditions[a]



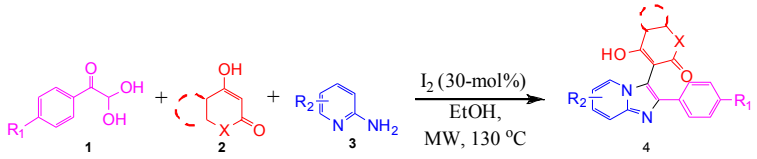
Entry	Solvent	Catalyst (mol %)	Temp (^{o}C)	Time (min/h)	Yield [b](%)
1	EtOH	--------	RT	24 h	0
2	EtOH	---------	Reflux	24 h	0
3	EtOH	PTSA(20)	RT	24 h	0
4	EtOH	PTSA(20)	Reflux	24 h	0
5	Toluene	PTSA(20)	Reflux	24 h	6
6	Toluene	PTSA(20)	130	15 min	38
7	EtOH	$Cu(OTf)_2$(20)	130	15 min	65
8	EtOH	$Sc(OTf)_3$(20)	130	15 min	60
9	EtOH	$Bi(OTf)_3$(20)	130	15 min	69
10	EtOH	$FeCl_3$(20)	130	15 min	45
11	EtOH	CAN(20)	130	15 min	42
12	EtOH	I_2(20)	130	15 min	72
13	**EtOH**	**I_2(30)**	**130**	**15 min**	**82**
14	EtOH	I_2(50)	130	15 min	68
15	EtOH	HI(20)	130	15 min	58
16	CH_3CN	I_2(30)	130	15 min	62
17	THF	I_2(30)	130	15 min	trace
18	DMF	I_2(30)	130	15 min	42
19	H_2O	I_2(30)	130	15 min	51
20	EtOH	I_2(30)	100	15 min	74

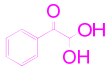
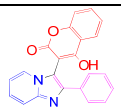
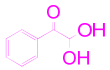
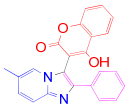
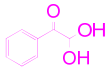
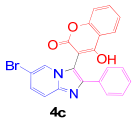
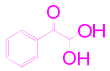
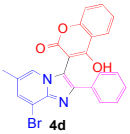
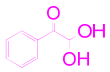
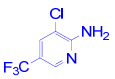
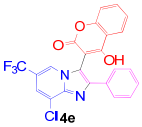
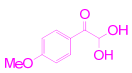
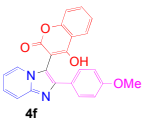
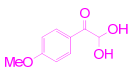
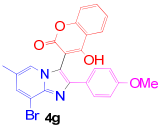
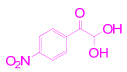
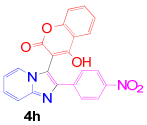
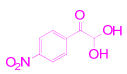
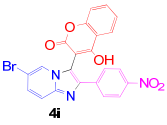
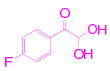
[a]Reaction conditions: phenylglyoxal monohydrate (1 mmol), 4-hydroxycoumarin (1 mmol), and 2-aminopyridine (1 mmol). [b]Isolated yield.

In order to explore the scope of this multicomponent reaction, a wide variety of phenylglyoxal derivatives, cyclic 1,3-dicarbonyl compounds and 2-aminopyridines were reacted under the optimized reaction conditions and the results are summarized in Table 2. Keeping **1a** and **2a** as fixed substrates 2-amino pyridine derivatives tethered with -Me, -Br, -Cl and -CF_3 groups in different positions were reacted under the optimized reaction conditions and in all these cases the observed yields were good to very good (Table 2, entries 2-5). 2-aminopyrdines with trifluoromethyl groups or –Cl /-Br provided lesser yields as compared to the unsubstituted 2-aminopyridine or its derivatives having electron donating groups. Lower yields in these cases may be attributed due to the reduced nucleophilicity of the primary amine or of the endocyclic nitrogen of 2-amino pyridines having electron withdrawing groups. Next the variability of aryl glyoxal was also checked using its derivatives having –OMe, NO_2 and –F substituents. Similar to phenyl glyoxal monohydrate its derivatives having electron donating or withdrawing groups also underwent three component reactions to provide the corresponding 2,3-disubstituted imidazo-[1,2-a]pyridines. Finally, we attempted to check the scope and generality of this MCR by replacing 4-hydroxy coumarin with other cyclic 1, 3-dicarbonyls such as 4-hydroxy-1-methylquinolin-2(1H)-one, 4-hydroxy-6-methyl-2-pyrone and dimedone. Interestingly in all these cases the corresponding expected three component products were obtained in moderate to good yields. All these products were fully characterized by IR, ^{1}H NMR, ^{13}C NMR and by elemental analysis as well as by recording melting points.

This three component reaction can also be extended to 2-aminopyrimidine.Using similar reaction conditions the combination of phenylglyoxal monohydrate, 2-aminopyrimidine and 4-hydroxycoumarine provided 78% yield whereas using 4-hydroxy-1-methylquinolin-2(1H)-one in place of 4-hydroxycoumarine provided 75% yields as shown in Scheme 2.

Table 2 Synthesis of 2,3-disubstituted imidazo[1,2-*a*]pyridines [a] **4**



Entry	Substrate 1	Substrate 2	Substrate 3	Time (min)	Product 4	Yield (%)[b]
1	 1a	 2a	 3a	15	 4a	82
2	 1a	 2a	 3b	15	 4b	84
3	 1a	 2a	 3c	15	 4c	72
4	 1a	 2a	 3d	15	 4d	83
5	 1a	 2a	 3e	15	 4e	65
6	 1b	 2a	 3a	15	 4f	81
7	 1b	 2a	 3d	15	 4g	71
8	 1c	 2a	 3a	15	 4h	83
9	 1c	 2a	 3c	15	 4i	76
10	 1d	 2a	 3a	15	4j	74
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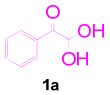
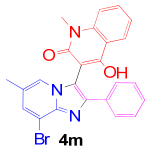
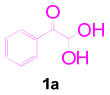
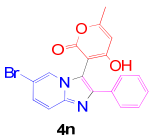
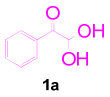
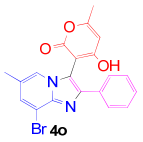
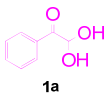
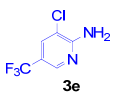
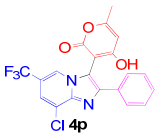
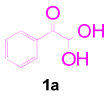
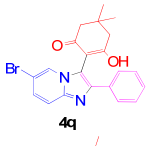
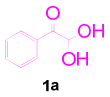
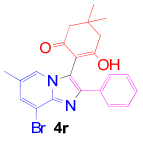
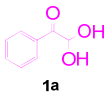
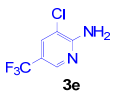
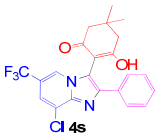
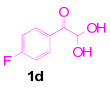
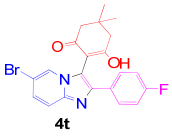
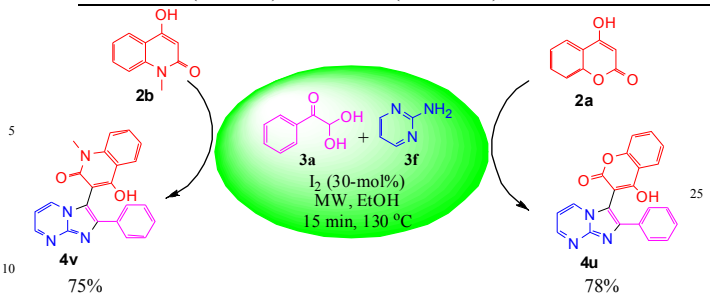
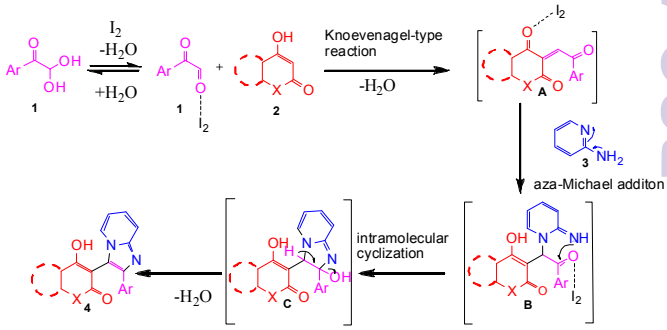
13	 1a	 2b	 3d	15	 4m	67
14	 1a	 2c	 3c	15	 4n	69
15	 1a	 2c	 3d	15	 4o	62
16	 1a	 2c	 3e	15	 4p	58
17	 1a	 2e	 3c	15	 4q	76
18	 1a	 2e	 3d	15	 4r	72
19	 1a	 2e	 3e	15	 4s	68
20	 1d	 2e	 3c	15	 4t	72

[a]Reaction conditions: phenylglyoxal monohydrate or its derivatives (1 mmol), 1, 3-dicarbonyls (1 mmol), 2-aminopyridine derivatives (1 mmol) and iodine (0.3 mmol), in ethanol at 130 °C (MW). [b]Isolated yield.



Scheme 2 Synthesis of 2,3-disubstituted imidazo[1,2-*a*]pyrimidines.

On the basis of the above results a plausible reaction mechanism is shown in Scheme 3. It is believed that this reaction goes via Knoevenagel type reaction to form intermediate **A.** Then the 2-aminopyridine (**3)** undergoes aza-Michael addition to **A** affording intermediate **B** which subsequently undergoes cyclization followed by the loss of water to form desired product **4**. Iodine can either act as Lewis acid[28] or a source of *in situ* HI[29] in organic transformations. To know the actual role of iodine in this three component reaction a few experiments were done.



Scheme 3 Proposed mechanism for the synthesis of **4.**

In one of the experiment the model reaction i.e reaction of **1a**, **2a** and **3a** was tested using 30-mol% HI under the similar reaction conditions, and a moderate yield (55%) of 4a was obtained. To discard the role of *in situ* HI, we have performed another reaction in the presence of iodine (30-mol%) along with 30-mol% base ($NaHCO_3$) keeping solvent and heating conditions same. Interestingly, this combination provided 81% of the desired three component product **4a.** It is expected that if the reaction is catalyzed by HI in that case presence of equimolar amount of base will nullify the acid and will have drastic effect on the net yield obtained. Since in our case this did not happen thus we believe iodine is acting as a Lewis acid to activate carbonyl CO in all the steps as shown in Scheme 3.

Conclusion

In conclusion, we have developed an iodine mediated one-pot three component reaction for the synthesis of 2,3-disubstituted imidazo[1,2-a]pyridines by using 1, 3-binucleophilic nature of 2-aminopyridines. A wide range of cyclic 1, 3-dicarbonyls can be tetrhed in the 3-position of imidazo[1,2-a]pyridine moiety in one-pot by this MCRs. Considering the presence of 4-hydroxycoumarin/ hydroxypyrone or 4-hydroxy-1-methylquinolin-2(1H)-one moiety linked with imidazo[1,2-a]pyridines or imidazo[1,2- a]pyrimidines it is expected that these hybride molecules will exhibit promising medicinal activities. From operatinal point of view, this MCR is easy to perform simply by mixing readily available starting materials under microwave irradiation. All the reactions took place within short times with water as the only benign by-product.

Experimental

General Information

All starting materials were purchased either from Sigma Aldrich or Alfa Aesar and used without further purification. NMR spectra were recorded on 400 or 500 MHz for ^1H and 100 or 125 MHz for ^{13}C in D_2O or DMSO-d6, Chemical shift values were reported in δ values (ppm) downfield from tetramethylsilane. Infrared (IR) spectra were recorded on a Shimadzu IR Affinity-1, FTIR spectrometer. Elemental analyses were carried out using vario MICRO cube elemental analyzer. Microwave irradiation was carried out with Initiator 2.5 Microwave Synthesizers from Biotage, Uppsala, Sweden. Melting points were recorded by using SRS EZ-Melt automated melting point apparatus by capillary methods and uncorrected.

Typical experimental procedure for the synthesis of 1a: A mixture of phenylglyoxal monohydrate (**1a)** (1 mmol), 4-hydroxycoumarin (**2a)** (1 mmol), 2-aminopyridine (**3a)** (1 mmol) and iodine (0.3 mmol) in 3 mL ethanol was introduced in a 2-5 mL reaction vial, the mixture was irradiated for 15 min, keeping temperature at 130 ^oC (Absorption Level: High; Fixed Hold Time and 200 W). The reaction mixture was then cooled to room temperature and the solid was filtered off, and was washed with water-ethanol mixture to get the desired three component product **4a.**

Note: Due to poor solubility of these compounds in common organic solvents, in most of the cases the resultant products were treated with 1.0 equivalent NaOH in 2 ml H_2O to prepare the corresponding sodium enolate of the products and the solvent was removed under vacuum to record NMR in D_2O.

4-hydroxy-3-(2-phenylimidazo[1,2-a]pyridin-3-yl)-2H-chromen-2-one (4a): Yield: 82%, White solid, mp. 133-135 ^oC; IR (KBr): 3400, 3070, 1663, 1597, 1520, 1457, 1407, 1374, 1307, 1267, 1197, 1138, 1109, 1058, 1035, 969, 842, 752, 687, 651 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 8.28 (d, *J* = 7.0 Hz, 1H, Ar-H), 7.94 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.88 (d, *J* = 7.5 Hz, 1H, Ar-H), 7.84(d, *J* = 8.0 Hz, 2H, Ar-H), 7.55 (t, *J* = 8.0 Hz, 1H, Ar-H), 7.49 (t, *J* = 8.0 Hz, 2H, Ar-H), 7.43-7.38 (m, 2H, Ar-H), 7.27-7.22 (m, 2H, Ar-H); **Sodium salt of (4a)** ^{13}C NMR (100 MHz, D_2O-d_2): δ =177.5, 168.1, 167.5, 153.8, 145.4, 142.1, 134.0, 132.6, 128.7, 127.9, 126.8, 126.6, 124.8, 124.5, 124.0, 120.9, 116.7, 115.9, 115.5, 112.9, 89.7; Anal. Calcd for $C_{22}H_{14}N_2O_3$ (354.36): C, 74.57; H, 3.98; N, 7.91; Found: C, 74.59; H, 3.95; N, 7.97.

4-hydroxy-3-(6-methyl-2-phenylimidazo[1,2-a]pyridin-3-yl)-2H-chromen-2-one (4b): Yield: 84%, White solid, mp. 257-259 ^oC; IR (KBr): 3437, 3066, 2620, 1677, 1603, 1534, 1451, 1416, 1373, 1308, 1265, 1203, 1133, 1107, 1057, 967, 898, 852, 810, 759, 690 cm^{-1}; **Sodium salt of (4b)** : ^1H NMR (400 MHz, D_2O-d_2): δ = 7.96 (d, *J* = 7.5, Hz, 1H, Ar-H), 7.83 (d, *J* = 7.4, Hz, 2H, Ar-H), 7. 68 (s, 1H, Ar-H), 7.63 (t, *J* = 7.6 Hz, 1H, Ar-H), 7.54 (d, *J* = 9.1 Hz, 1H, Ar-H), 7.40-7.31 (m, 5H, Ar-H), 7.28 (d, *J* = 9.0 Hz, 1H, Ar-H), 2.25 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.5, 167.5, 153.7, 144.4, 141.9, 134.2, 132.5, 129.6, 128.7, 127.8, 126.8, 124.5, 124.0, 122.9, 122.4, 120.9, 116.7, 115.7, 114.8, 89.9, 17.2; Anal. Calcd for $C_{23}H_{16}N_2O_3$ (368.38): C, 74.99; H, 4.38; N, 7.60; Found: C, 74.95; H, 4.40; N, 7.65.

3-(6-bromo-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4c): Yield: 72 %, Light yellow solid, mp. 273-275 ^oC; IR (KBr): 3381, 1670, 1646, 1600, 1528, 1496, 1456, 1415, 1368, 1323, 1214, 1107, 1082, 1044, 967, 899, 841, 798, 759, 694 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ = 8.61 (s, 1H, Ar-H), 7.92 (d, *J* = 7.5 Hz, 1H, Ar-H), 7.81 (d, *J* = 7.5 Hz, 2H, Ar-H), 7.77-7.74 (m, 2H, Ar-H), 7.65 (t, *J* = 7.5 Hz, 1H, Ar-H), 7.44-7.32 (m, 5H, Ar-H); **Sodium salt of (4c)** ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.5, 167.3, 153.8, 143.8, 142.9, 133.7, 132.6, 129.4, 128.8, 128.1, 126.9, 125.0, 124.5, 124.0, 120.9, 116.7, 116.5, 116.4, 107.0, 89.3; Anal. Calcd for $C_{22}H_{13}BrN_2O_3$ (433.25): C, 60.99; H, 3.02; N, 6.47; Found: C, 60.96; H, 3.05; N, 6.49.

3-(8-bromo-6-methyl-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4d): Yield: 83%, White solid, mp. 274-276 ^oC; IR (KBr): 3410, 3063, 1717, 1612, 1534, 1451, 1391, 1289, 1234, 1104, 1077, 959, 847, 745, 726, 578 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.15 (s, 1H, Ar-H), 7.96 (d, *J* = 5.6 Hz, 1H, Ar-H), 7.81 (d, *J* = 5.2 Hz, 2H, Ar-H), 7.73-7.68 (m, 2H, Ar-H), 7.49-7.20 (m, 5H, Ar-H), 2.28 (s, 3H, CH_3); **Sodium salt of (4d)** ^{13}C NMR (125 MHz, D_2O-d_2): δ = 177.5, 167.3, 166.4, 153.7, 143.1, 142.2, 133.8, 132.6, 131.6, 128.7, 128.0, 127.2, 124.5, 124.0, 123.3, 122.2, 120.9, 117.9, 116.7, 108.3, 89.9, 17.0; Anal. Calcd for $C_{23}H_{15}BrN_2O_3$ (447.28): C, 61.76; H, 3.38; N, 6.26; Found: C, 61.74; H, 3.36; N, 6.29.

3-(8-chloro-2-phenyl-6-(trifluoromethyl)imidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4e): Yield: 65

%, Pale yellow solid, mp. 219-221 °C; IR (KBr): 3465, 1672, 1640, 1596, 1530, 1455, 1367, 1343, 1298, 1218, 1165, 1135, 1092, 1069, 1030, 966, 892, 844, 761, 689, 667 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.99 (s, 1H, Ar-H), 7.96 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.91 (s, 1H, Ar-H), 7.83 (d, *J* = 7.2 Hz, 2H, Ar-H), 7.78-7.74 (m, 1H, Ar-H), 7.52-7.48 (m, 1H, Ar-H), 7.44-7.38 (m, 3H, Ar-H), 7.34-7.31 (m, 1H, Ar-H); **Sodium salt of (4e)** ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.9, 168.6, 167.6, 154.2, 151.4, 148.3, 143.9, 134.2, 133.9, 133.1, 129.2, 128.8, 127.6, 124.9, 124.5, 121.2, 117.2, 115.5, 109.6, 89.1; Anal. Calcd for $C_{23}H_{12}ClF_3N_2O_3$ (456.80): C, 60.47; H, 2.65; N, 6.13; Found: C, 60.45; H, 2.64; N, 6.17.

4-hydroxy-3-(2-(4-methoxyphenyl)imidazo[1,2-a]pyridin-3-yl)-2H-chromen-2-one (4f): Yield: 81%, Yellow solid, mp. 246-248 °C; IR (KBr): 3450, 3079, 2838, 2545, 1669, 1601, 1516, 1457, 1424, 1395, 1372, 1312, 1254, 1180, 1141, 1104, 1028, 954, 897, 835, 756 cm^{-1}; **Sodium salt of (4f)** ^1H NMR (500 MHz, D_2O-d_2): δ = 7.90-7.80 (m, 1H, Ar-H), 7.75-7.60 (m, 3H, Ar-H), 7.58-7.40 (m, 2H, Ar-H), 7.30-7.20 (m, 3H, Ar-H), 6.85-6.70 (m, 3H, Ar-H), 3.16 (s, 3H, OMe); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.5, 167.5, 158.5, 153.8, 145.3, 142.0, 132.6, 128.3, 127.2, 126.4, 124.7, 124.5, 124.0, 121.1, 116.7, 115.4, 114.2, 112.8, 89.6, 55.3; Anal. calcd for $C_{23}H_{16}N_2O_4$ (384.38): C, 71.87; H, 4.20; N, 7.29; Found: C, 71.88; H, 4.22; N, 7.32.

3-(8-bromo-2-(4-methoxyphenyl)-6-methylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4g): Yield: 71%, Pale yellow solid, mp. 261-263 °C; IR (KBr): 3438, 3069, 2571, 1677, 1603, 1529, 1451, 1377, 1271, 1179, 1156, 1105, 1031, 957, 897, 842, 759, 690 cm^{-1}; **Sodium salt of (4g)** ^1H NMR (500 MHz, D_2O-d_2): δ = 7.94 (s, 1H, Ar-H), 7.90 (d, *J* = 7.2 Hz, 1H, Ar-H), 7.72 (d, *J* = 8.8 Hz, 2H, Ar-H), 7.50-7.45 (m, 2H, Ar-H), 7.33-7.22 (m, 2H, Ar-H), 6.84 (d, *J* = 8.8 Hz, 2H, Ar-H), 3.67 (s, 3H, OCH_3), 2.11 (s, 3H, CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ = 177.1, 166.8, 158.7, 153.9, 143.6, 143.0, 132.5, 128.9, 128.6, 127.1, 124.9, 124.8, 124.0, 121.4, 116.7, 116.5, 115.8, 114.2, 106.8, 88.8, 55.3, 29.8; Anal. Calcd for $C_{24}H_{17}BrN_2O_4$ (477.31): C, 60.39; H, 3.59; N, 5.87; Found: C, 60.37; H, 3.61; N, 5.85.

4-hydroxy-3-(2-(4-nitrophenyl)imidazo[1,2-a]pyridin-3-yl)-2H-chromen-2-one (4h): Yield: 83%, Dark Red solid, mp. 297-299 °C; IR (KBr): 3438, 3106, 1672, 1654, 1603, 1525, 1423, 1350, 1289, 1202, 1114, 1054, 962, 870, 764, 713, 690, 653 cm^{-1}; **Sodium salt of (4h)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.18 (d, *J* = 8.0 Hz, 2H, Ar-H), 8.10 (d, *J* = 8.4 Hz, 2H, Ar-H), 8.01 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.91 (d, *J* = 6.8 Hz, 1H, Ar-H), 7.70 (d, *J* = 9.6 Hz, 1H, Ar-H), 7.64 (t, *J* = 7.6 Hz, 1H, Ar-H), 7.45 (t, *J* = 7.6 Hz, 1H, Ar-H), 7.41-7.35 (m, 2H, Ar-H), 7.0 (t, *J* = 6.8 Hz, 1H, Ar-H); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.3, 166.8, 163.9, 154.4, 146.8, 145.6, 142.3, 140.2, 132.5, 127.8, 126.8, 125.2, 125.1, 123.9, 123.8, 121.6, 116.8, 116.2, 113.9, 88.5; Anal. Calcd for $C_{22}H_{13}N_3O_5$ (399.36): C, 66.17; H, 3.28; N, 10.52; Found: C, 66.19; H, 3.25; N, 10.57.

3-(6-bromo-2-(4-nitrophenyl)imidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4i): Yield: 76%, Reddish solid, mp. 315-319 °C; IR (KBr): 3427, 3090, 3060, 1681, 1654, 1603, 1520, 1455, 1423, 1349, 1271, 1183, 1160, 1109, 1040, 957, 897, 865, 759, 690 cm^{-1}; **Sodium salt of (4i)** ^1H NMR (500 MHz, D_2O-d_2): δ = 8.10 (dd, *J* = 9.0, 2.0 Hz, 2H, Ar-H), 8.05 (s, 1H, Ar-H), 7.95 (dd, *J* = 8.5, 1.5 Hz, 2H, Ar-H), 7.90 (dd, *J* = 7.5, 1.5 Hz, 1H, Ar-H), 7.58 (dt, *J* = 9.0, 1.5 Hz, 1H, Ar-H), 7.51 (d, *J* = 9.5 Hz, 1H, Ar-H), 7.42-7.40 (m, 1H, Ar-H), 7.31 (t, *J* = 7.5 Hz, 2H, Ar-H); ^{13}C NMR (125 MHz, D_2O-d_2): δ = 177.2, 166.8, 153.7, 146.4, 143.9, 140.7, 140.6, 132.6, 129.9, 127.4, 125.3, 124.5, 124.0, 123.9, 120.9, 118.6, 116.7, 116.6, 107.4, 88.8; Anal. Calcd for $C_{22}H_{12}BrN_3O_5$ (478.25): C, 55.25; H, 2.53; N, 8.79; Found: C, 55.28; H, 2.54; N, 8.82.

3-(2-(4-fluorophenyl)imidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4j): Yield: 74%, Orange solid, mp. 220-222 °C; IR (KBr): 3438, 3087, 1677, 1605, 1523, 1460, 1423, 1400, 1280, 1239, 1197, 1165, 1165, 1059, 960, 905, 850, 754 cm^{-1}; **Sodium salt of (4j)** ^1H NMR (500 MHz, D_2O-d_2): δ = 7.94 (d, *J* = 7.0 Hz, 1H, Ar-H), 7.82 (d, *J* = 6.0 Hz, 3H, Ar-H), 7.63 (d, *J* = 9.0 Hz, 1H, Ar-H), 7.60 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.41-7.33 (m, 3H, Ar-H), 7.10 (t, *J* = 8.0 Hz, 2H, Ar-H), 6.92 (t, *J* = 6.5 Hz, 1H, Ar-H); ^{13}C NMR (125 MHz, D_2O-d_2): δ = 177.5, 167.4, 163.2, 161.3, 153.8, 145.4, 141.5, 132.6, 130.3, 128.8, 126.6, 124.8, 124.5, 124.1, 121.0, 116.7, 115.6, 115.4, 112.9, 89.5; Anal. Calcd for $C_{22}H_{13}FN_2O_3$ (372.35): C, 70.96; H, 3.52; N, 7.52; Found: C, 70.99; H, 3.54; N, 7.55.

3-(6-bromo-2-(4-fluorophenyl) imidazo[1,2-a]pyridin-3-yl)-4-hydroxy-2H-chromen-2-one (4k): Yield: 68%, Yellow solid, mp. 149-151 °C; IR (KBr): 3428, 3079, 2375, 1676, 1596, 1533, 1503, 1456, 1423, 1375, 1353, 1266, 1231, 1157, 1107, 1039, 955, 897, 846, 764 cm^{-1}; **Sodium salt of (4k)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.12-8.0 (m, 2H, Ar-H), 7.98-7.88 (m, 2H, Ar-H), 7.65 (d, *J* = 9.1 Hz, 2H, Ar-H), 7.47 (d, *J* = 8.8 Hz, 1H, Ar-H), 7.40 (d, *J* = 6.4 Hz, 2H, Ar-H), 7.13 (d, *J* = 6.8 Hz, 2H, Ar-H); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 167.1, 163.3, 161.3, 153.7, 143.7,142.3, 132.5, 129.9, 129.3, 128.8, 125.1, 124.5, 123.9, 120.9, 116.7, 116.3, 115.5, 115.4, 107.0, 89.1; Anal. Calcd for $C_{22}H_{12}BrFN_2O_3$ (451.24): C, 58.56; H, 2.68; N, 6.21; Found: C, 58.58; H, 2.66; N, 6.25.

3-(6-bromo-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-1-methylquinolin-2(1H)-one (4l): Yield: 71 %, White solid, mp. 300-302 °C; IR (KBr): 3460, 3084, 1700, 1650, 1627, 1539, 1496, 1458, 1442, 1417, 1365, 1312, 1198, 1088, 1064, 958, 872, 810, 752, 710, 689 cm^{-1}; **Sodium salt of (4l)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.07 (dd, *J* = 7.9, 1.4 Hz, 1H, Ar-H), 7.94 (d, *J* = 1.2 Hz, 1H, Ar-H), 7.88 (dd, *J* = 8.6, 1.4 Hz, 2H, Ar-H), 7.53 (d, *J* = 9.5 Hz, 1H, Ar-H), 7.46 (t, *J* = 7.5 Hz, 1H, Ar-H), 7.39-7.31 (m, 3H, Ar-H), 7.28-7.21 (m, 3H, Ar-H), 3.44 (s, 3H, NMe); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 173.9, 164.9, 143.6, 142.6, 140.1, 134.0, 131.2, 128.9, 128.6, 127.8, 126.9, 124.8, 121.9, 121.6, 118.7, 116.3, 114.7, 106.8, 96.1, 29.2; Anal. calcd for $C_{23}H_{16}BrN_3O_2$ (446.30): C, 61.90; H, 3.61; N, 9.42; Found: C, 61.92; H, 3.63; N, 9.46.

3-(8-bromo-6-methyl-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-1-methylquinolin-2(1H)-one (4m): Yield: 67%, White solid, mp. 237-239 °C; IR (KBr): 3075, 3028, 2366, 1662, 1635, 1590, 1535, 1506, 1456, 1378, 1310, 1262, 1165, 1091, 1041, 899, 851, 762, 709 cm^{-1}; **Sodium salt of (4m)** ^1H NMR (500 MHz, D_2O-d_2): δ = 8.00 (d, *J* = 7.5 Hz, 1H, Ar-H), 7.69 (d, *J* = 7.5 Hz, 2H, Ar-H), 7.47- 7.43 (m, 2H, Ar-H), 7.42 (s, 1H, Ar-H), 7.30-7.23(m, 3H, Ar-H), 7.22-7.16 (m, 2H, Ar-H), 3.35 (s, 3H, NMe), 2.07 (s, 3H, CH_3); ^{13}C NMR (125 MHz, , D_2O-d_2): δ = 173.8, 165.1, 142.7, 142.0, 140.1, 134.1, 131.4, 128.6, 127.8, 127.1, 124.8, 123.1, 122.1, 121.9, 121.7, 119.9, 114.9, 108.2, 96.9, 29.3, 16.9; Anal. Calcd for $C_{24}H_{18}BrN_3O_2$ (460.32): C, 62.62; H, 3.94; N, 9.13; Found: C, 62.65; H, 3.96; N, 9.15.

3-(6-bromo-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4n): Yield: 69%, White solid, mp. 249-251 °C; IR (KBr): 3066, 2999, 2748, 1709, 1658, 1601, 1555, 1516, 1488, 1449, 1414, 1362, 1305, 1257, 1215, 1177, 1075, 1025, 1005, 987, 892, 827, 788, 754, 716, 690, 528, 513 cm^{-1}; **Sodium salt of (4n)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.08 (s, 1H, Ar-H), 7.83 (d, *J* = 7.5 Hz, 2H, Ar-H), 7.54 (d, *J* = 9.4 Hz, 1H, Ar-H), 7.48-7.47 (m, 3H, Ar-H), 7.45-7.40 (m. 1H, Ar-H), 5.99 (s, 1H, CH), 1.94 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 181.6, 168.5, 162.9, 143.8, 142.8, 133.6, 129.4, 128.8, 128.1, 126.8, 125.0, 116.3, 116.0, 107.3, 107.0, 88.9, 18.8; Anal. Calcd for $C_{19}H_{13}BrN_2O_3$ (397.22): C, 57.45; H, 3.30; N, 7.05; Found: C, 57.47; H, 3.32; N, 7.09.

3-(8-bromo-6-methyl-2-phenylimidazo[1,2-a]pyridin-3-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4o): Yield: 62%, White solid, mp. 257-259 °C; IR (KBr): 3070, 1689, 1643, 1505, 1451, 1411, 1345, 1263, 1162, 1041, 991, 915, 840, 809, 774, 690, 581cm^{-1}; **Sodium salt of (4o)** ^1H NMR (400 MHz, D_2O-d_2): δ = 7.69 (dd, *J* = 8.5, 1.4 Hz, 2H, Ar-H), 7.56 (s, 1H, Ar-H), 7.45 (s, 1H, Ar-H), 7.34 (t, *J* = 7.7 Hz, 2H, Ar-H), 7.29-7.23 (m, 1H, Ar-H), 5.85 (s, 1H, CH), 2.13 (s, 3H, CH_3), 1.81 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 181.6, 168.6, 167.9, 162.9, 142.9, 142.2, 133.7, 131.7, 128.7, 128.1, 127.1, 123.4, 122.1, 117.4, 108.3, 107.4, 89.5, 18.9, 17.0; Anal. Calcd for $C_{20}H_{15}BrN_2O_3$ (411.25): C, 58.41; H, 3.68; N, 6.81; Found: C, 58.43; H, 3.67; N, 6.84.

3-(8-chloro-2-phenyl-6-(trifluoromethyl)imidazo[1,2-a]pyridin-3-yl)-4-hydroxy-6-methyl-2H-pyran-2-one (4p): Yield: 58%, Pale Yellow solid, mp. 249-251 °C; IR (KBr): 3429, 3015, 2903, 2840, 1706, 1675, 1653, 1609, 1569, 1520, 1496, 1459, 1417, 1340, 1304, 1257, 1173, 1153, 1093, 1029, 983, 899, 832, 760, 672, 538 cm^{-1}; **Sodium salt of (4p)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.30 (s, 1H, Ar-H), 7.86 (dd, *J* = 7.0, 1.5 Hz, 2H, Ar-H), 7.78 (s, 1H, Ar-H), 7.52-7.40 (m, 3H, Ar-H), 5.99 (s, 1H, CH), 2.28 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 181.6, 168.4, 163.2, 144.6, 142.5, 133.0, 128.8, 128.5, 127.2, 123.2, 122.1, 120.9, 119.2, 116.9, 116.5, 107.3, 88.5, 18.9; Anal. Calcd for $C_{20}H_{12}ClF_3N_2O_3$ (420.77): C, 57.09; H, 2.87; N, 6.66; Found: C, 57.11; H, 2.85; N, 6.69.

2-(6-bromo-2-phenylimidazo[1,2-a]pyridin-3-yl)-3-hydroxy-5,5-dimethylcyclohex-2-enone (4q): Yield: 76%, Yellow solid, mp. 226-228 °C; IR (KBr): 3530, 3381, 3100, 2962, 2869, 1662, 1592, 1549, 1516, 1408, 1352, 1267, 1173, 1144, 1129, 1089, 1041, 923, 806, 772, 741 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.06 (s, 1H, Ar-H), 7.76 (dd, *J* = 8.4, 1.2 Hz, 2H, Ar-H), 7.59 (d, *J* = 9.6, Hz, 1H, Ar-H), 7.39-7.35 (m, 3H, Ar-H), 7.29 (t, *J* = 7.6 Hz, 1H, Ar-H), 2.68 (d, *J* = 16.8 Hz, 2H, Ar-H), 2.36 (d, *J* = 16.8 Hz, 2H, Ar-H), 1.15 (s, 6H, CH_3); **Sodium salt of (4q)** ^{13}C NMR (100 MHz, D_2O-d_2): δ = 196.9, 181.9, 144.0, 142.9, 134.4, 129.4, 129.1, 128.3, 127.6, 125.2, 118.9, 116.7, 107.2, 102.1, 49.7, 31.8, 28.9, 27.7, 23.7; Anal. Calcd for $C_{21}H_{19}BrN_2O_2$ (411.29): C, 61.33; H, 4.66; N, 6.81; Found: C, 61.35; H, 4.67; N, 6.85.

2-(8-bromo-6-methyl-2-phenylimidazo[1,2-a]pyridin-3-yl)-3-hydroxy-5,5-dimethylcyclohex-2-enone (4r): Yield: 72%, Pale Yellow solid, mp. 230-232 °C; IR (KBr): 3423, 3071, 2945, 2866, 1667, 1608, 1538, 1492, 1399, 1298, 1234, 1186, 1145, 1088, 1028, 894, 818, 717, 629 cm^{-1}; **Sodium salt of (4r)** ^1H NMR (400 MHz, D_2O-d_2): δ = 7.68-7.65 (m, 2H, Ar-H), 7.46-7.45 (m, 2H, Ar-H), 7.39-7.35 (dt, *J* = 7.8, 1.2 Hz, 2H, Ar-H), 7.29 (t, *J* = 7.36, 1H, Ar-H), 2.37 (d, *J* = 16.6 Hz, 2H, Ar-H), 2.25 (d, *J* = 16.8 Hz, 2H, Ar-H), 2.18 (s, 3H, CH_3), 1.10 (s, 3H, CH_3), 1.05 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 196.6, 168.0, 142.8, 142.1, 134.1, 131.3, 128.6, 127.9, 127.4, 123.1, 121.9, 119.6, 108.3, 102.3, 49.3, 31.4, 28.4, 27.3, 17.1; Anal. Calcd for $C_{22}H_{21}BrN_2O_2$ (425.32): C, 62.13; H, 4.98; N, 6.59; Found: C, 62.15; H, 4.99; N, 6.62.

2-(8-chloro-2-phenyl-6-(trifluoromethyl)imidazo[1,2-a]pyridin-3-yl)-3-hydroxy-5,5-dimethylcyclohex-2-enone (4s): Yield: 68%, White solid, mp. 235-237 °C; IR (KBr): 3280, 3070, 2960, 2562, 1661, 1624, 1576, 1545, 1490, 1416, 1317, 1212, 1179, 1142, 1102, 1075, 1031, 893, 855, 775, 694 cm^{-1}; **Sodium salt of (4s)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.04 (s, 1H, Ar-H), 7.74 (d, *J* = 7.5 Hz, 2H, Ar-H), 7.69 (s, 1H, Ar-H), 7.45 (t, *J* = 7.5 Hz, 2H, Ar-H), 7.40-7.38 (m, 1H, Ar-H), 2.47 (d, *J* = 16.5 Hz, 2H, Ar-H), 2.30 (d, *J* = 16.8 Hz, 2H, Ar-H), 1.15 (s, 3H, CH_3), 1.11 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 196.7, 163.9, 144.55, 142.3, 133.4, 128.7, 128.4, 127.6, 124.2, 122.9, 122.1, 121.3, 120.5, 116.4, 116.2, 115.9, 115.6, 101.2, 49.2, 31.4, 28.5, 27.1; Anal. Calcd for $C_{22}H_{18}ClF_3N_2O_2$ (434.84): C, 60.77; H, 4. 17; N, 6.44; Found: C, 60.79; H, 4. 19; N, 6.48.

2-(6-bromo-2-(4-fluorophenyl)imidazo[1,2-a]pyridin-3-yl)-3-hydroxy-5,5-dimethylcyclohex-2-enone (4t): Yield: 72%, Pale Yellow solid, mp. 193-195 °C; IR (KBr): 3401, 3106, 2977, 1658, 1608, 1580, 1515, 1488, 1465, 1428, 1363, 1243, 1179, 1091, 1045, 856, 810, 699 cm^{-1}; **Sodium salt of (4t)** ^1H NMR (400 MHz, D_2O-d_2): δ = 7.92 (s, 1H, Ar-H), 7.79 (d, *J* = 7.0 Hz, 1H, Ar-H), 7.77 (d, *J* = 7.5 Hz, 1H, Ar-H), 7.53 (d, *J* = 11.5 Hz, 1H, Ar-H), 7.46 (d, *J* = 11.0 Hz, 1H, Ar-H), 7.21 (t, *J* = 11.0 Hz, 2H, Ar-H), 2.48 (d, *J* = 21.0 Hz, 2H, CH_2), 2.37 (d, *J* = 21.0 Hz, 2H, CH_2), 1.22 (s, 3H, CH_3), 1.18 (s, 3H, CH_3); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 196.5, 163.5, 143.6, 142.1, 129.1, 129.0, 128.9, 124.9, 118.2, 116.4, 115.5, 115.3, 106.8, 101.5, 49.3, 31.4, 29.6, 28.5, 27.3; Anal. Calcd for $C_{21}H_{18}BrFN_2O_2$ (429.28): C, 58.75; H, 4.23; N, 6.53; Found: C, 58.77; H, 4.25; N, 6.56.

4-hydroxy-3-(2-phenylimidazo[1,2-a]pyrimidin-3-yl)-2H-chromen-2-one (4u): Yield: 78%, White solid, mp. 220-222 °C; IR (KBr): 3435, 1675, 1642, 1598, 1545, 1449, 1418, 1361, 1317, 1264, 1215, 1106, 1063, 1027, 965, 899, 865, 760, 716, 681 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6): δ = 8.81 (bs, 1H, Ar-H), 8.72 (d, *J* = 6.8 Hz, 1H, Ar-H), 7.92 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.86 (d, *J* = 7.6 Hz, 2H, Ar-H), 7.64 (d, *J* = 8.0 Hz, 1H, Ar-H), 7.44 (t, *J* = 7.6 Hz, 2H, Ar-H), 7.38 (d, *J* = 7.6 Hz, 2H, Ar-H), 7.33 (t, *J* = 8.0 Hz, 2H, Ar-H); **Sodium salt of (4u)** ^{13}C NMR (100 MHz, D_2O-d_2): δ = 177.9, 168.5, 154.3, 145.4, 143.1, 133.7, 133.0, 129.2, 127.8, 125.0, 124.5, 123.8, 122.7, 122.1, 121.2, 120.2, 117.2, 89.2; Anal. Calcd for $C_{21}H_{13}N_3O_3$ (355.35): C, 70.98; H, 3.69; N, 11.83; Found: C, 70.95; H, 3.72; N, 11.85.

4-hydroxy-1-methyl-3-(2-phenylimidazo[1,2-a]pyrimidin-3-yl)quinolin-2(1H)-one (4v): Yield: 75%, Yellow solid, mp. 247-249 °C; IR (KBr): 3428, 3070, 2888, 1640, 1616, 1533, 1498, 1417, 1381, 1320, 1251, 1161, 1114, 1084, 959, 878, 808, 768, 716 cm^{-1}; **Sodium salt of (4v)** ^1H NMR (400 MHz, D_2O-d_2): δ = 8.58 (dd, *J* = 4.3, 1.8 Hz, 1H, Ar-H), 8.22 (dd, *J* = 6.8, 1.8 Hz, 1H, Ar-H), 8.11 (dd, *J* = 7.9, 1.2 Hz, 1H, Ar-H), 7.84 (dd, *J* = 8.1, 1.5 Hz, 2H, Ar-H), 7.70 (dt, *J* = 8.5, 1.4 Hz, 1H, Ar-H), 7.56 (d, *J* = 8.5 Hz, 1H, Ar-H), 7.40-7.33 (m, 4H, Ar-H), 7.07 (dd, *J* = 6.7, 4.3 Hz, 1H, Ar-H), 3.60 (s, 3H, NMe); ^{13}C NMR (100 MHz, D_2O-d_2): δ = 173.9, 168.4, 165.1, 150.8, 147.8, 143.2, 140.2, 133.8, 133.6, 131.5, 128.7, 128.2, 127.2, 124.8, 121.9, 117.2,

115.0, 109.1, 95.6, 29.3; Anal. Calcd for $C_{22}H_{16}N_4O_2$ (368.39): C, 71.73; H, 4.38; N, 15.21; Found: C, 71.75; H, 4.37; N, 15.25.

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