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Ir(I)-Catalyzed Synthesis of (*E*)-4-Benzylidenylacridines/(*E*)-2-StyrylQuinoline-3-carboxamide via Sequential Suzuki-Miyaura /Dehydrogenative Friedlander reaction/sp^3 C-H activation

Soda Prameela, Fazlur-Rahman Nawaz Khan*.

Abstract. An efficient one-pot strategy evolved in the synthesis of (E)-4-benzylidenylacridin-1(2H)-ones, (**4** or **9**) (E)-2-styryl quinoline-3-carboxamides (**5** or **10**) by sequential Suzuki-Miyaura coupling-dehydrogenative Friedlander-sp^3 C-H activation. The 2-amino-5-chloro benzhydrol, **1** aromatic alcohol, **2** or **6** ketones, 3 or 3` and phenylboronic acid, **7** underwent smooth reaction in basic deep eutectic solvent K_2CO_3: ethylene glycol (1:1) (DES-1). DES-1 could enable Pd catalyzed Suzuki coupling reaction of **6**, as well as the rapid oxidation of primary, **2** secondary, **1** and Suzuki coupled primary alcohol, **8** with iridium catalyst/1, 10-phenanthroline in a proficient way. A while later, the acidic DES-2, Dimethyl urea: Tartaric acid (7:3), assists the Friedlander annulation, subsequent sp^3 C-H functionalization resulting in (E)-4-benzylidenylacridin-1(2H)-ones, (**4** or **9**) (E)-2-styryl quinoline-3-carboxamides (**5** or **10**). This protocol has several advantages, such as DES-1; Suzuki coupling reaction working without a base and additive could dehydrogenate the primary and secondary alcohols in the presence of iridium catalyst DES-2; Friedlander reaction-sp^3 C-H functionalization without further addition of a catalyst.

Introduction

Acridine and quinolines are ubiquitous heterocycles, occur in natural products, mainly in alkaloids.[1] Moreover, they are useful in the design of several synthetic moieties, unique chemical structure and diverse biological activities with medicinal benefits. Besides, found their applications in pharmaceuticals, flavors, agrochemicals, dyes, and industrial applications.[2] Several acridines, quinolines, and its derivatives are known to possess biological activities, as summarized in figure.1: For instances, antiprotozoal (A_1),[3] antimalarial (A_2),[4] (A_{13}),[5] anticancer (A_3),[6] (A_{14}),[7]DNA-targeting acridines (A_4),[8] antimicrobial (A_5),[9] antitumour (A_6),[10] (A_{10}),[11] Anti HIV (A_7),[12] (A_{15}),[13] antibacterial (A_8),[6] antiproliferative (A_9),[14] antileishmanial (A_{11}),[5] antifungal (A_{12}),[15] antitubercular(A_{16}),[16] antiplatelet (A_{17})[17] activity. There are various methodologies developed for the synthesis of quinolines and acridines in past few decades, such as Friedlander synthesis,[18] Skraup synthesis,[19] Doebner-von Miller synthesis.[20] And other methods for these heteroarenes synthesis consuming different substrates have also been reported in recent years.[21] So far, all these methods involved in the synthesis of these heteroarenes by straightforward, easy Friedlander annulation[22] of *o*-amino benzaldehydes/ketones involving condensation with carbonyl compounds. Though it has some limitations, for example, harsh reaction conditions (high temperature), o-amino benzaldehydes/ketones are highly unstable, and limited availability of substitutions. To devoid the drawback, the corresponding alcohol utilized instead of o-amino benzaldehydes/ketones through oxidative cyclization for heteroarenes synthesis (Scheme 1. **A^1** [(i) (ii)]).[23]

An Indirect dehydrogenative Friedlander reaction has become a powerful tool for the benign construction of complex organic molecules,There is no use of stoichiometric amount of oxidants, and here water and hydrogen only the non-toxic by-products. Already there are catalytic reactions have been accomplished with various metals such as Ru,[23a, 23c] Ir, [24] Rh,[25] Co,[26] Mn,[23b] Fe,[27] Pd,[28] with suitable ligands.All these protocols contain harsh reaction conditions, more reaction time and hazardous solvents. There is indeed an increasing for the green synthetic process, to simplify and to avoid toxic solvents.

Recently, a solvent free multicomponent synthesis of quinolines is reported by Xinhua Xu and coworkers[29] using an air-stable zirconium (IV)-salophen perfluorooctanesulfonate complex as catalyst. Likewise, 1,4-dihydropyridine and polyhydroquinoline compounds reported by Schiff base zirconium perfluorooctanesulfonate Lewis acid catalysis. [29b] Accordingly, green protocol is reported using Deep eutectic solvent (DES), These DESs are composed of two or more inexpensive, available and safe components[30] and which avoid the volatility, flammability, hazards, and toxicity also possess water bio renewability and biodegradability.[31] Besides, in many chemical processes, DES was used as green and sustainable reaction media, also catalysts.[32]

Recently, our group focused on the green solvents, such as DESs in a base-rree Suzuki-Miyaura/Sonogashira Cross-Coupling reactions.[33] Likewise, a sequential Friedländer reaction/Pd-Catalyzed sp^3 C-H functionalization of Methyl Ketones reported.[23f] Subsequently, developed Chimanine B analogues with a Ru-Catalyzed sequential dehydrogenative Friedlander reaction/sp^3 C–H Activation/Knovenagel condensation cascade reaction in one-pot. (Scheme 1. A^2).[23j] In this context, in the present study (E)-4-benzylidenylacridin-1(2H)-ones, (**4**) (E)-2-styryl quinoline-3-carboxamides (**5**) (Scheme 1. B^1), (Scheme 1. B^2) developed in the presence of Ir catalyst.

Presently, the [IrCl(COD)]$_2$/1,10-phenanthroline combination facilitated an acceptorless dehydrogenation of both primaty[34a] and secondary alcohols.[34b] For instances, 2-amino-5-chloro benzhydrol, **1** benzyl alcohols, **2** dehydrogenative in K_2CO_3: Ethylene glycol(1:1)[35] (DES-1) in the presence of Ir catalyst /1,10-phenanthroline additive without a base in the pH of 11. Then, modified to acidic pH of 1 by the addition of DMU: Tartaric acid[36] (DES-2), then added diketones, **3** for the formation of acridines, **4** and quinolines, **5** *via* Friedlander reaction/ sp^3 C–H Activation. Moreover, a base and ligand-free Suzuki-Miyaura cross coupling heterocyclic styrenes, **9**, **10** have been developed in a comfortable and clean process.

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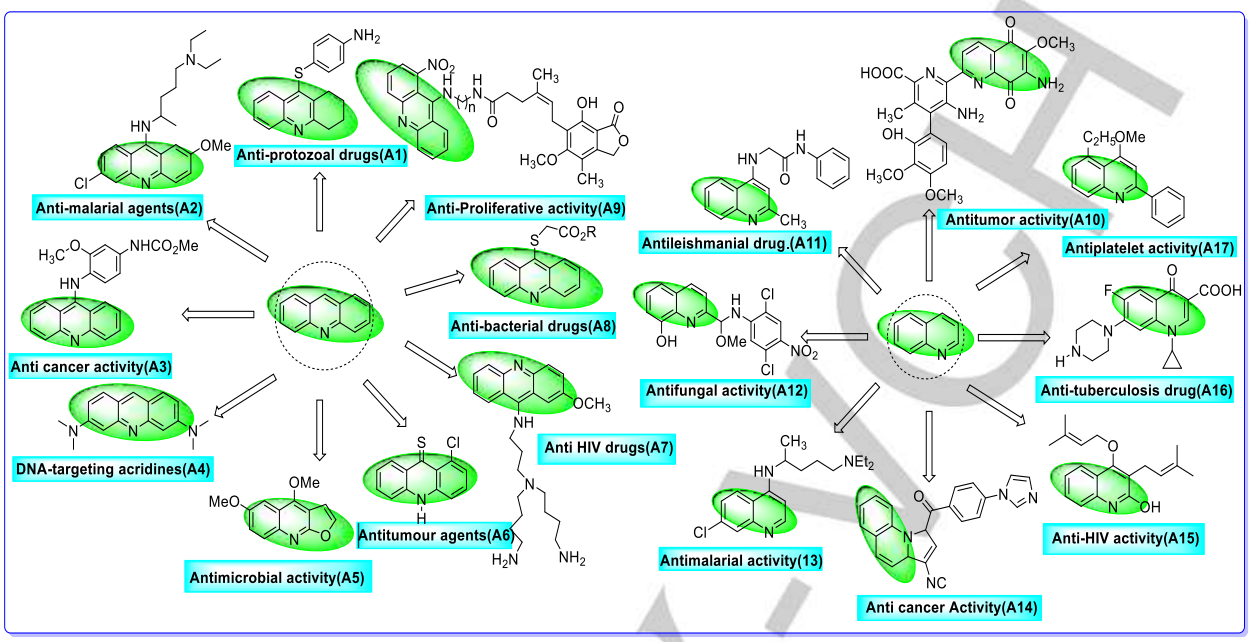
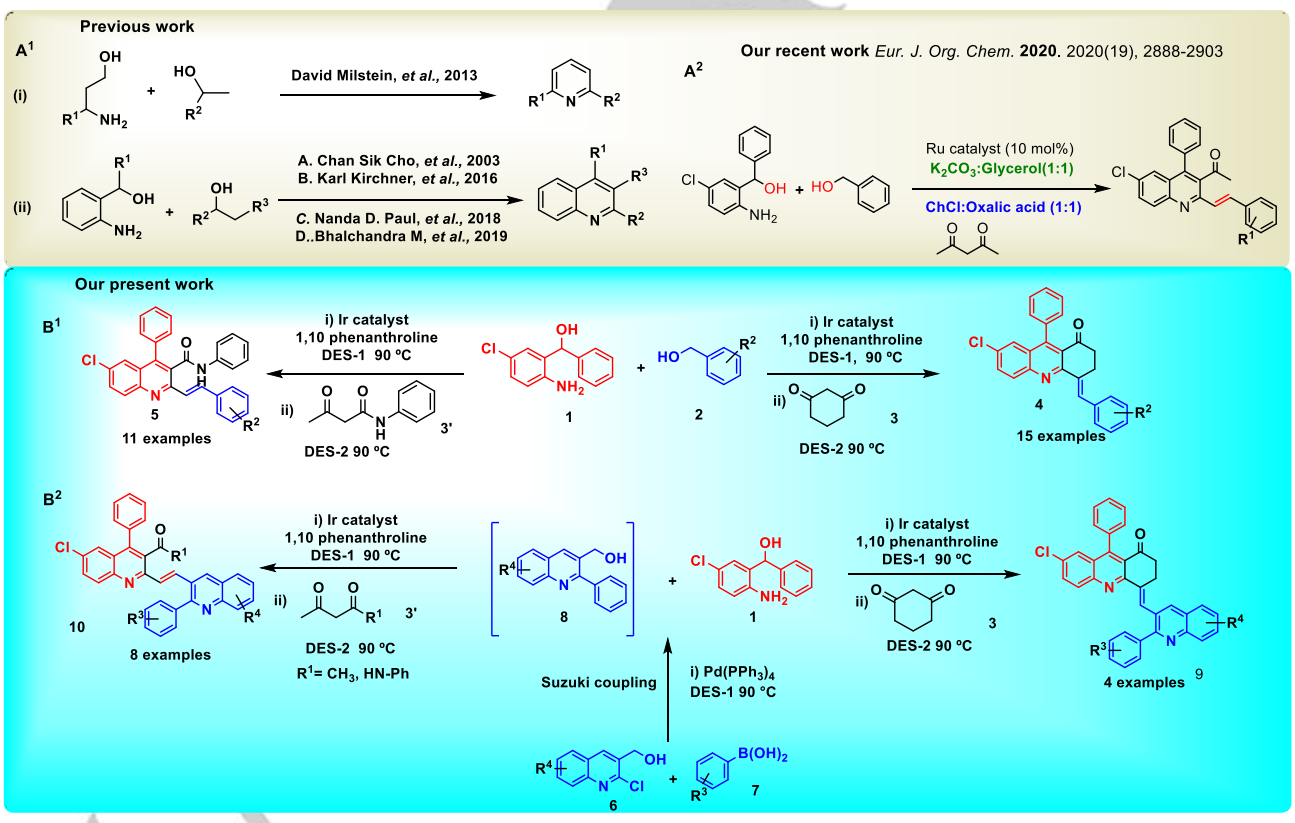


Figure.1 Biologically significant heteroarene

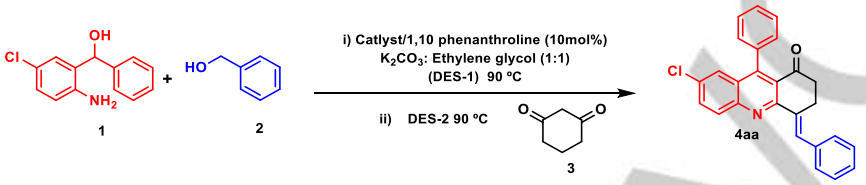


Scheme 1. **A**[1] [(i) (ii)] Previous work: Synthesis strategy of quinoline Motifs. (**A**[2]) Our recent work: Synthesis of Chimanine B analogues sequential dehydrogenative Friendlander/sp[3] C-H functionalization. (**B**[1]) This work: Synthesis of Quinoline and acridine stilbene motifs: sequential dehydrogenative, cyclization, and sp[3] C-H functionalization (**B**[2]) Suzuki-Miyaura cross-coupled quinoline and acridine stilbene formation.

Results and Discussion

Firstly, the oxidation of 2-amino benzhydrol, **1** benzyl alcohol, **2** were carried out with an iridium catalyst, 1,10-phenanthroline in basic DES-1 to yield the corresponding 2-amino-5-chlorobenzophenone **1a** and benzaldehyde **2a** at 90 °C. Then, changed to acidic reaction media with DES-2, added 1,3-cyclohexadione, 3 continued heating to provide (E)-4-benzylidene-7-chloro-9-phenyl-3,4-dihydroacridin-1(2H)-one, **4aa** in quantitative yields and purity, through sequential Friedlander reactions/sp^3C-H bond activation.To determine the best optimization condition for heteroaryltyrenes, studies performed by screening with different DES, catalysts, and catalytic ratios and temperatures. Our initial study focused on

Table 1. Optimization of the reaction conditions[a,b,c,d]



Entry	Reaction media DES-2 for step (ii) (molar ratio)	Catalyst (mol%)	Temp (°C)	Time (h)	Yield (%)[b]
1	DMU/D-Glucose (7:3)	$[IrCl(COD)]_2$ (10)	100	3	Trace
2	DMU/Malonic acid (7:5)	$[IrCl(COD)]_2$ (10)	100	5	12
3	DMU/Citric acid (6:4)	$[IrCl(COD)]_2$ (10)	100	5	40
4	DMU/Succinic acid (7:3)	$[IrCl(COD)]_2$ (10)	100	3	60
5	DMU/PTSA (7:3)	$[IrCl(COD)]_2$ (10)	100	4	50
6	DMU/Sorbitol (7:3)	$[IrCl(COD)]_2$ (10)	100	4	Nil
7	DMU/Sorbitol/NH_4Cl (2:7:1)	$[IrCl(COD)]_2$ (10)	100	4	Nil
8	DMU/Oxalic acid (7:3)	$[IrCl(COD)]_2$ (10)	100	2	88
9	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (10)	100	2	90
10	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (10)	90	2	90
11	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (10)	80	2	60
12	DMU/Tartaric acid (7:3)	$Pd(OAc)_2$ (10)	90	2	55
13	DMU/Tartaric acid (7:3)	$Cu(OAc)_2$ (10)	90	2	50
14	DMU/Tartaric acid (7:3)	$Co(OAc)_2$ (10)	90	2	48
15	DMU/Tartaric acid (7:3)	$Zn(OAc)_2$ (10)	90	2	40
16	DMU/Tartaric acid (7:3)	$Ni(OAc)_2$ (10)	90	2	37
17	DMU/Tartaric acid (7:3)	$RuCl_2(p\text{-}Cym)_2$ (10)	90	2	49
18[c]	DMU/Tartaric acid (7:3)	$RuCl_2(p\text{-}Cym)_2$ (10)	90	2	30
19[c]	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (10)	90	2	Nil
20[d]	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (10)	90	2	45
21	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (5)	90	2	65
22	DMU/Tartaric acid (7:3)	$[IrCl(COD)]_2$ (15)	90	2	90

[a] **Reaction condition:** Dehydrogenation: (2-amino-5-chlorophenyl)(phenyl)methanol, 1a (1.0 mmol), benzyl alcohol, 2a (1.0 mmol), catalyst (10 mol%), additive 1,10-phenanthroline (10 mol%), K_2CO_3: ethylene glycol (1:1) (DES-1 200 mg), heated (at indicated temperature) for time 20 min. Then added 1,3-diketone **3** (1 mmol), DES-2 (200mg), continued heating till overall time as indicated. [b]Isolated yields, [c]without additive, [d] 5 mol% additive.

a sequential one-pot reaction of 2-aminobenzhydrol, **1** (1mmol) benzyl alcohol, **2** (1mmol) at 100 °C using 10 mol% of Ir catalyst, 10 mol% 1,10 phenanthroline additive in K_2CO_3: Ethylene glycol(1:1) DES-1 for 20 min. Then, added cyclohexadione, **3** (1mmol) DMU/ D-Glucose (7:3) as reaction media, and continued heating, which provided the desired product **4aa** in trace amount (Table 1, entry1). With DMU/Malonic acid (7:5), an increased yield detected (Table1, entry 2). Later, with DMU/citric acid (6:4), DMU/Succinic acid (7:3) and DMU/PTSA (7:3) slightly increased to moderate yields observed (Table1, entry 3-5).

The reaction failed to proceed with DMU/Sorbitol (7:3) or DMU/Sorbitol/NH_4Cl (2:7:1) (Table1, entry 6,7). Interestingly, the DMU/Oxalic acid (7:3) or DMU/ Tartaric acid (7:3) emerged as an active reaction media for the desired product formation (Table1, entry 8, 9). Dropping the temperature to 90 °C did not affect the yield (Table1, entry 10), while the yield decreased at 80 °C (Table1, entry 11). Further, when optimized with a different catalyst, it resulted in low yields of the products (37-55%) (Table1, entry 12-16). When explored with $RuCl_2(p\text{-}Cym)_2$ catalyst in the presence or absence of an additive (Table1, entry 17, 18), gave decreased yield compared to Ir catalyst. The reaction is failed without an additive, implied the necessity of additive 10 mol% 1,10-phenanthroline (Table1, entry 19[c]) with less mol% of additive and catalyst resulted in a low yield of product (Table1, entry 20[d],21). A 10 mol% catalyst is essential for product formation (Table1, entry 22).

Having established the optimal reaction condition, (Table1, entry 10), applied to alicyclic ketones (cyclohexanone), 3,4-dihydronaphthalen-2(1H)-one as well as open-chain aliphatic ketones, underwent efficient sequential dehydrogenative reaction towards the targeted heterocyclic motif. Later, these heteroarenes reacts with differently substituted benzyl alcohols smoothly yielded the corresponding acridiny stilbenes, **4aa-aj, 4ba-bc**, **4ca-cb**, irrespective of the electronic nature of the alcohol group shown in Table.2.

Likewise, with 3-oxo-N-phenylbutanamides substrates, the quinolinylstilbene , **5da-dk** successfully achieved. Electron-donating and electron-withdrawing alcohols well-tolerated and underwent smooth oxidation, sequential alkenylation reactions to the desired products (EDG **4aa-4ca**, **5da-5de**, EWG **4ah-4cb**, **5df-5di**) in 73-90% yields. Di- and trisubstituted electron-donating alcohols (**4ab**, **4af**, **5db**, **5dd**) provided the desired product in good yields of 88-90%. Furthermore, the polycyclic and α, β unsaturated alcohol converted to designed products **4ag**, **5dj** in 90%, 89% yields respectively. In the case of heteroaromatic alcohols were also tolerated, the corresponding product **5dk** obtained in 73% yield.

Control Experiments: To understand the critical role and importance of DESs in the reaction mechanistic pathway, conducted a series of control experiments in the sequential dehydrogenative Friedlander-sp^3 C-H activation reaction between **1**, **2** and **3** and contrasted with a reaction between **1a'**, **2a'** and **3**.

When a reaction of **1**, **2** carried out in standard established optimization condition in DES-1, observed the dehydrogenated products (**1a**', 94% and **2a**', 97% as evidenced from ^{1}H NMR spectrum, supporting information) as in scheme 2C(i). In the next 1h, the intermediate 2-amino-5-chlorobenzophenone **1a'** condensed well with cyclohexane-1,3-dione **3** in DES-2 by a Friedlander annulation to provide **8a**" (Scheme 2C(i) confirmed by the ^{1}H NMR analysis provided in supporting information). Meanwhile, benzaldehyde, **2a'** formed duly reacted with an intermediate **8a**" to provide the acridine, **4aa**, in 48% yield (Scheme 2C(i)). ^{1} H NMR revealed the carbonyl compounds **1a'**, **2a'** and acridine **8a"** intermediates formation. Further, after 40 min, the reaction completely gets converted to **4aa** in 90% yield (Scheme 2C(ii)).

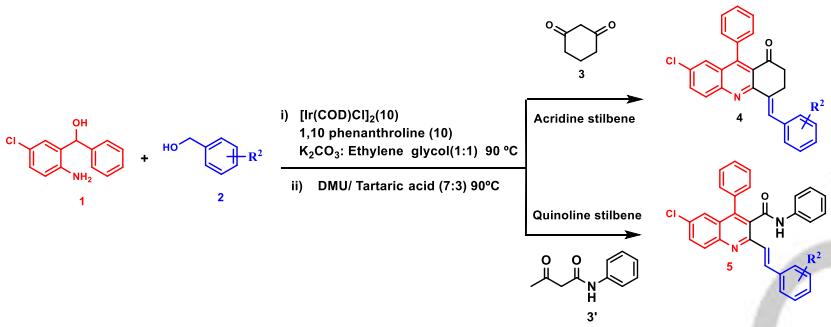
Later, to evolve the necessity of different DESs in the sequential formation of acridine styrenes in one-pot, control experiments were conducted. The substrates **1, 2** and **3** when reacted with $[IrCl(COD)]_2$ catalyst, 1,10 phenanthroline additive in acidic DES-2, (pH =1) (see Scheme 2(C) (iii) the desired product **4aa** and intermediates **1a'**, **2a'**not observed. However, the same reaction when explored in basic DES-1 (pH=11, see Scheme 2(C),(v)) exclusively dehydrogenation products **1a'**, **2a'** obtained, demonstrating the role of DES-1 in the dehydrogenation of alcohols.

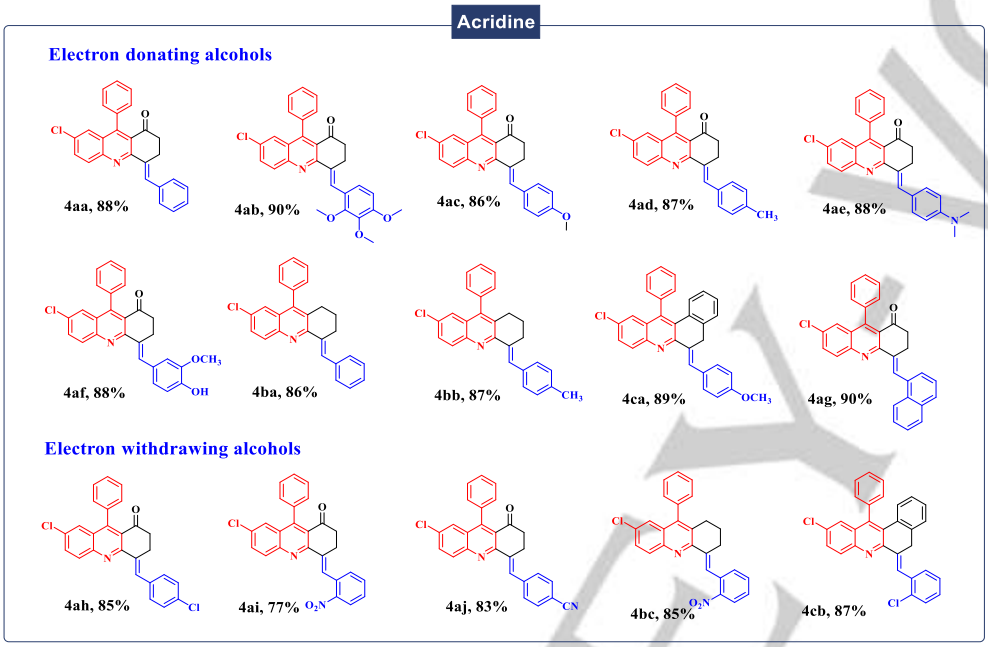
Then, the sequential Friedlander/sp^3 C-H activation explored with substrates **1a'**, **2a'** and **3** in both the DESs, however acidic DES-2 successfully provided the acridine **4aa** (Scheme 2(C) (vii)) while the basic DES-1, failed to provide the targeted product, **4aa** (Scheme 2(C) (viii)). This observation revealed that DES-1 played a role in dehydrogenation reaction, while DES-2 in the seqeuntial Friedlander/sp^3 C-H functionalization reactions (Scheme 2(C) (iv).

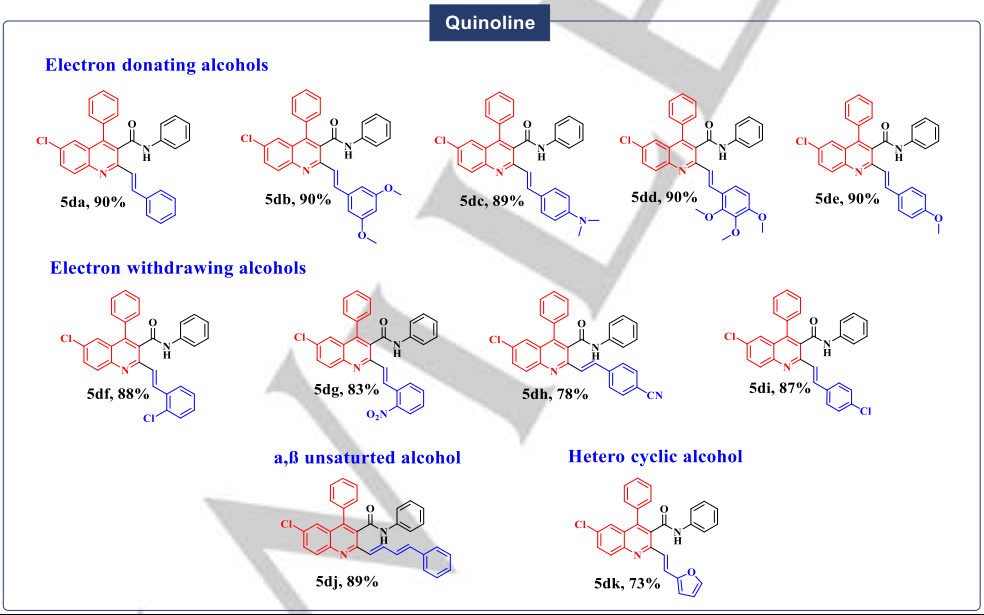
Application in the scale-up synthesis

Then practical efficacy, scalability of the sequential synthesis of (E)-4-benzylidenylacridin-1(2H)-ones, **4**, examined with standard reaction conditions. For instance, a gram scale reaction between **1**, **2** and **3** performed, and the reaction proceeded smoothly to the desired product **4aa** in 87% yield (scheme (2D)). Then, the reduction of the carbonyl group of (E)-4-benzylidenylacridin-1(2H)-ones, **4** was done efficiently to provide **4aa'** in 75% yield. In 2006 Konig *et al*.,[37] reported mannitol-DMU-NH_4Cl (DES) mediated Suzuki-Miyaura coupling reaction of biaryls using a palladium catalyst and Na_2CO_3 as a base. Besides, literature does available for the Suzuki cross-coupling reactions in DES,[38] Very recently, in 2019, Saavedra *et al*.[39] reported the Suzuki coupling reaction in ChCl: glycerol (1:2), utilizing the bipyridine-palladium complex as a catalyst, $NaHCO_3$, as a base.

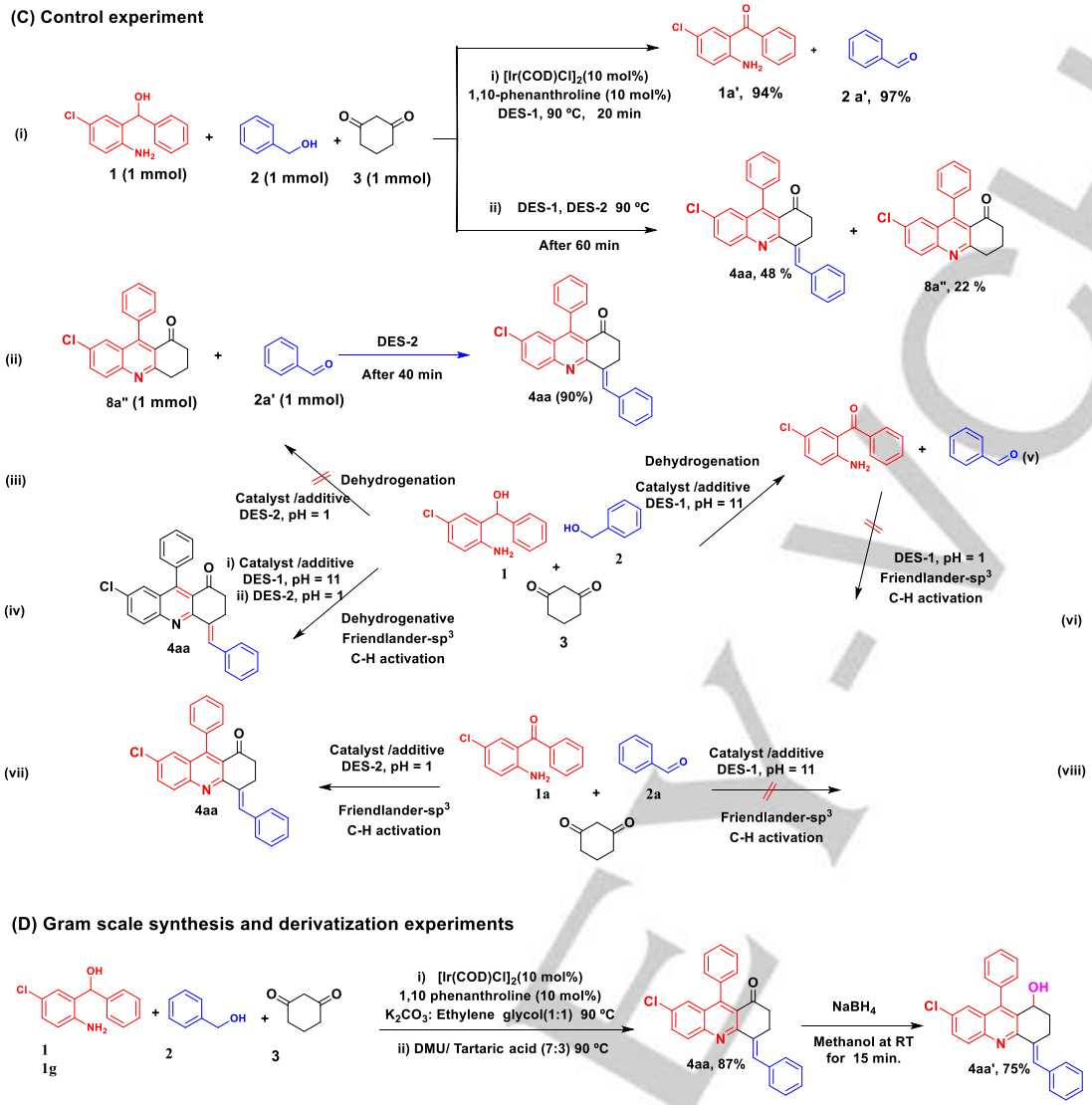
Table 2. Substrate Scope[a,b]







[a]**Reaction condition:** Dehydrogenation: (2-amino-5-chlorophenyl)(phenyl)methanol, **1** (1.0 mmol), benzyl alcohol, **2** (1.0mmol), $[IrCl(COD)]_2$ (10 mol%), 1,10-phenanthroline (10 mol%), K_2CO_3: ethylene glycol (1:1) (DES-1 200 mg), time 20 min; **Acridines**: added 1,3-diketone 3 (1 mmol), DMU/ Tartaric acid (7:3) (200 mg), time 120 min. **Quinoline**: added 3-oxo-*N*-phenylbutanamide 4 (1.0 mmol), DMU/ Tartaric acid (7:3) (200 mg), time 120 min. [b]Isolated yields.

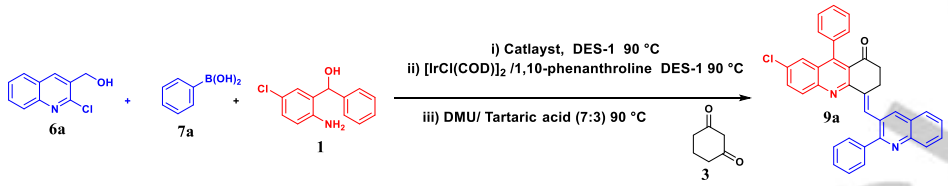


Scheme.2 (C) Control experiment, (D) Gram scale synthesis

Subsequently, in this present study in our continued interest[40] a green chemical approach utilizing a base free Suzuki reaction of (2-chloroquinolin-3-yl)methanol, **6** with phenylboronic acid **7**, sequential dehydrogenative-Friedlander reaction/sp^3 C-H functionalization (Table.3) established C-C bond formation in DES as reaction media, to provide the desired product smoothly in 80-84%. Herein the transition metal-catalyzed Suzuki C-C coupling reaction in K_2CO_3: Ethylene glycol (1:1- DES-1) by excluding base and ligand established. Further, established subsequent Friedlander annulation, sp^3 C-H functionalization in DES-2. The optimization of Suzuki-Miyaura coupling reactions screened using different DES, the results denoted in Table 3.

Initially, when a reaction between **6a** and **7a** checked with 5 mol % of $PdCl_2(PPh_3)_2$ or $Pd(OAc)_2$, and 5mol% of PPh_3 in DES-1, provided a moderate yield of 55-57% of **9a**, (Table 3, entry1, 2), while with 5 mol% of $Pd(PPh_3)_4$, provided 82% yield, (Table 3, entry 3). The yield was not affected by 2 mol% of $Pd(PPh_3)_4$ (Table 3, entry 4). Then with xanthphos as additive resulted in 75% yield (Table 3, entry 5). Interestingly, the absence of additive did not show an effect on the yield of the desired product (Table 3, entry 6,7). Reaction, when conducted in K_2CO_3: glycerol (1:1), slightly decreased the yield (Table 3 entry 8). Attempts with different K_2CO_3: Ethylene glycol (1:3), and K_2CO_3: Ethylene glycol (1:6) by reducing the base quantity, resulting in decreased yield and took more reaction time (Table 3, entry 9,10).

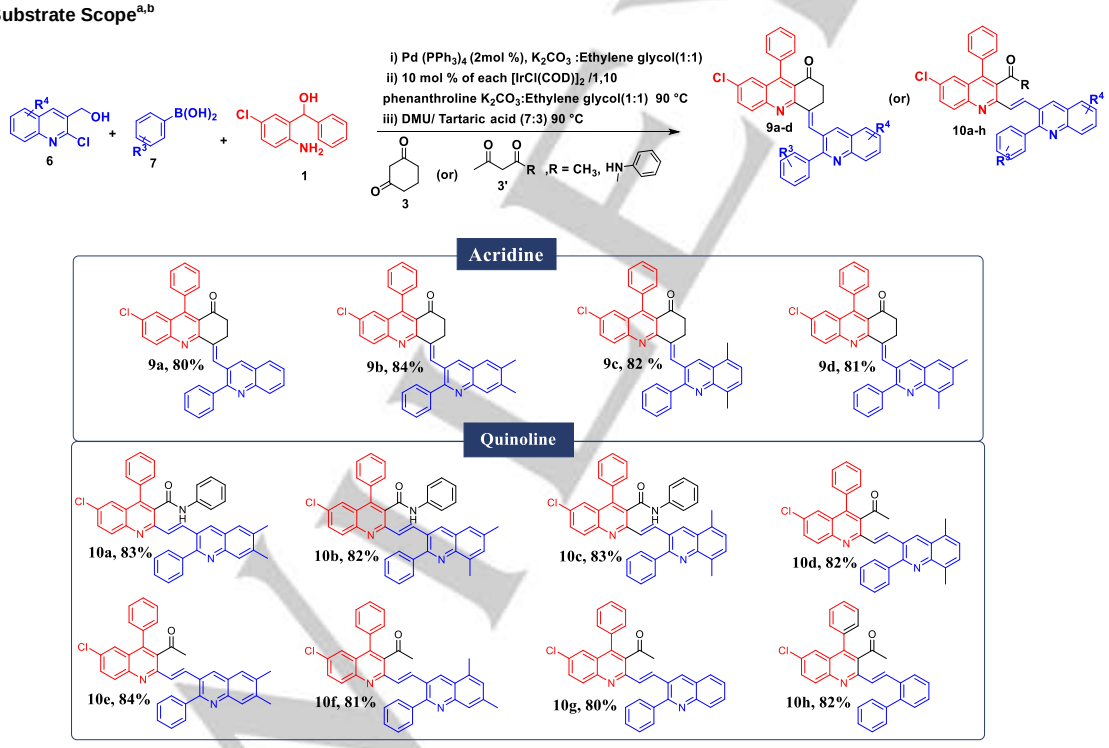
Table 3. Optimization of the reaction condition[a,b]



Entry	DES-1 (molar ratio)	Catalyst (mol %)	additive (mol %)	Time (h)	Yield (%)
1	K_2CO_3: Ethylene glycol(1:1)	$PdCl_2(PPh_3)_2$(5)	PPh_3(5)	2.5	55
2	K_2CO_3: Ethylene glycol(1:1)	$Pd(OAc)_2$(5)	PPh_3(5)	3	57
3	K_2CO_3: Ethylene glycol(1:1)	$Pd(PPh_3)_4$(5)	PPh_3(5)	4	82
4	K_2CO_3: Ethylene glycol(1:1)	$Pd(PPh_3)_4$(2)	PPh_3(5)	2.5	82
5	K_2CO_3: Ethylene glycol(1:1)	$Pd(PPh_3)_4$(2)	Xanthphos (5)	2.5	70
6	K_2CO_3: Ethylene glycol(1:1)	$Pd(PPh_3)_4$(2)	PPh_3(2)	2.5	82
7	K_2CO_3: Ethylene glycol(1:1)	$Pd(PPh_3)_4$(2)	.....	2.5	82
8	K_2CO_3: glycerol(1:1)	$Pd(PPh_3)_4$(2)	.....	3	74
9	K_2CO_3: Ethylene glycol(1:3)	$Pd(PPh_3)_4$(2)	.....	4	73
10	K_2CO_3: Ethylene glycol(1:6)	$Pd(PPh_3)_4$(2)	.....	5	74

[a]**Reaction condition**: For Suzuki reaction: DES-1 (200mg), (2-chloroquinolin-3-yl)methanol, **6** (1.0 mmol) phenylboronic acid, **7** (1.2 mmol), catlayst as indicated, at 90 °C time 15 min; For dehydrogenation: added (2-amino-5-chlorophenyl)(phenyl)methanol, **1** (1 mmol), in the presence of $[Ir(COD)Cl]_2$(10 mol%), 1,10-phenanthroline (10 mol%), 90 °C, 20 min. Then added 1,3-diketone **3** (1 mmol), DMU/Tartaric acid (7:3) (200 mg), time 120 min. [b]Isolated yields.

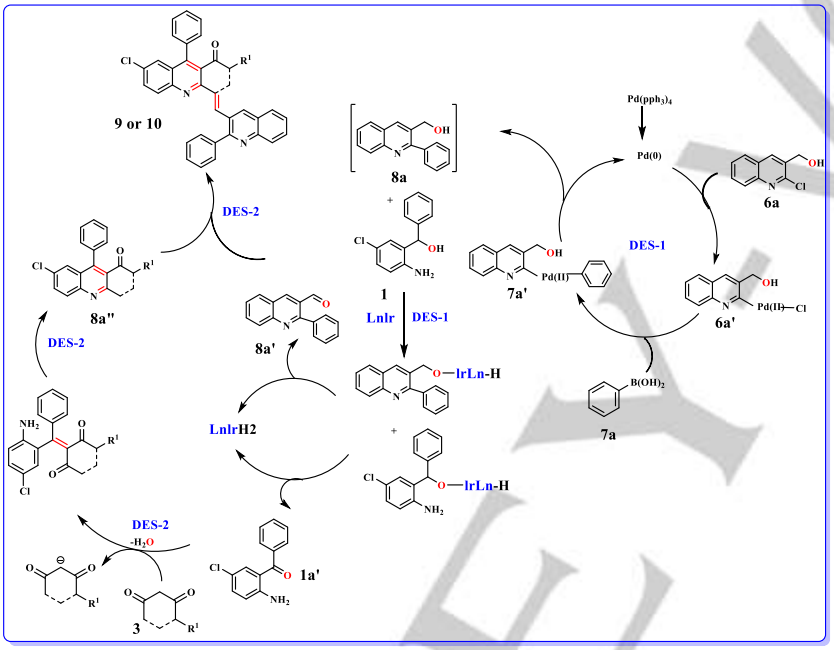
Table 4. Substrate Scope[a,b]



[a]**Reaction condition:** (2-chloroquinolin-3-yl)methanol, 6 (1.0 mmol), phenyl boronic acid, 7 (1.2 mmol) $Pd(PPh_3)_4$, (2 mol%) K_2CO_3: Ethylene glycol (1:1) (100mg: 100mg), 15 min, 90 °C; then added (2-amino-5-chlorophenyl)(phenyl)methanol, 1 (1 mmol), $[Ir(COD)Cl]_2$ (10 mol%), 1,10-phenanthroline (10 mol%), 90 °C, 20 min. For Acridines: added 1,3 diketone, 3 (1 mmol), DMU/ Tartaric acid (7:3) (200 mg), time 120 min. For Quinolines: added pentane-2,4-dione (1.0 mmol) or 3-oxo-N-phenylbutanamide, 3' (1.0 mmol), DMU/ Tartaric acid (7:3) (200 mg), 120 min. [b]Isolated yields.

Based on this optimization condition (Table 3), the cross-coupling reaction of various aryl chloride alcohols with phenylboronic acids and then the sequential dehydrogenative Friedlander/sp^3 C-H activation towards Suzuki reaction coupled (E)-4-benzylidenylacridin-1(2H)-ones, 9 attempted. Additionally, the Suzuki coupled (E)-2-styryl quinoline-3-carboxamides, 10, explored, as denoted in Table 4.

Based on controlled experiment studies, a probable reaction mechanism depicted the formation of C-C single and double bond formations in the one-pot tandem reaction as in scheme 3. At first, oxidative addition of palladium to halogenated compound, **6a** provided organopalladium species **6a'** which then reacts with boronic acid, **7** to yield the boronate complex **7b**. Reductive elimination of **7c** completes the catalytic cycle of the Suzuki reaction. Later, the dehydrogenation of (2-amino-5- chlorophenyl)(phenyl)methanol **1**, and (2-phenyl quinolin-3-yl)methanol, **8a** provided the corresponding carbonyl compound **1a'**, **8a'** *via* a β-hydride elimination in the presence of $[IrCl(COD)]_2$,[23d] Finally, with the modified acidic DES-2 dehydrogenative product **1a'** reacted well with various keto compounds **3** to yield the heteroarene **8a"**. And then **8a"** condensed with **8a'** to yield the compound **9** or **10** through sp^3 C-H activation.



Scheme 3. Proposed Reaction Mechanism

Conclusion

In summary, an efficient one-pot sequence reaction towards (E)-4-benzylidenylacridin-1(2H)-ones, **4aa-cb** as well as (E)-2-styryl quinoline-3-carboxamides **5da-dk** is reported. The 2-amino-5-chloro benzhydrol, **1** and benzyl alcohols **2** underwent dehydrogenation with $[IrCl(COD)]_2$ in K_2CO_3: Ethylene glycol (1:1) DES-1 and then sequential Fridelander-sp^3 C-H with ketones (3) in DMU: Tartaric acid (7:3) based DES-2. Besides, DES-1 enables the sequential Suzuki-Miyaura C-C coupling reaction of **6** with **7**- dehydrogenative Friedlander and subsequent sp^3 C-H activation to provide the (E)-4-benzylidenylacridin-1(2H)-ones, **9** and (E)-2-styryl quinoline-3-carboxamides, **10**. Gram scale synthesis and synthetic utility have been established.

Experimental Section

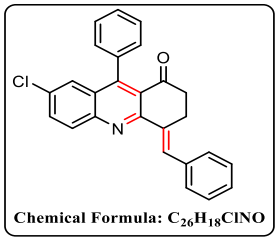
The typical preparation procedure for DESs:

Dimethyl urea: Tartaric acid (7:3)(70 mg:30 mg) was stirred at 90 °C for 1h to obtain a homogeneous clear melt. Tartaric acid form hydrogen bonds with dimethyl urea, to yield the DMU: Tartaric acid(7:3) DES. Likewise, K_2CO_3: Ethylene glycol(1:1) formed initially white viscous gel, and afterward, a clear liquid DES at 90 °C, The hydrogen atom of the ethylene glycol forms a hydrogen bond with the oxygen atom of the potassium carbonate.

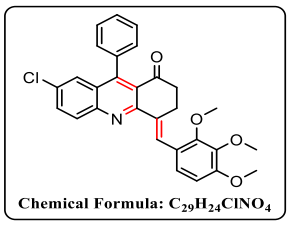
Synthetic procedure of (*E*)-4-benzylidenylacridin-1(2H)-ones, 4 and (*E*)-2-styryl quinoline-3-carboxamides, 5

A mixture of K_2CO_3: ethylene glycol (100mg: 100mg) stirred at 90 °C for 1h to attain DES-1. Then, added 2-amino-5-chloro benzhydrol, **1** (1.0 mmol) and benzyl alcohols, 2 (1.0 mmol) and 10 mol% of each $[IrCl(COD)]_2$, 1,10 phenanthroline, continued heating for 20 min at 90 °C. After confirming the dehydrogenative products **1a**, **2a** by thin-layer chromatography (TLC), ketone cyclohexane-1,3-dione **3** (1.0 mmol) or 3-oxo-N-phenylbutanamide 3` (1.0 mmol). Subsequently, DES-2 Dimethyl urea: Tartaric acid (7:3 -70 mg:30 mg) added, continued the reaction at 90 °C for 2hr to yield the (E)-4-benzylidenylacridin-1(2H)-ones**, 4** and (E)-2-styryl quinoline-3-carboxamides**, 5.** After the reaction completion, the crude reaction mixture purified by recrystallization using methanol to yield pure products **4**, **5**.

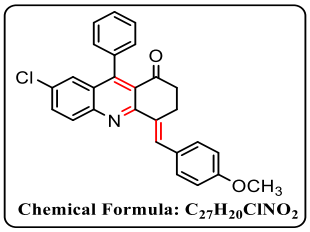
***(E)-4-benzylidene-7-chloro-9-phenyl-3,4-dihydroacridin-1(2H)-one (4aa)*:** 88% yield, Yellow solid, Mp:165-167 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ δ 8.32 (s, 1H), 8.09 (d, *J* = 9.0 Hz, 1H), 7.69 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.54 – 7.46 (m, 5H), 7.47 – 7.39 (m, 3H), 7.34 (t, *J* = 7.2 Hz, 1H), 7.19 (dd, *J* = 7.0, 2.1 Hz, 2H), 3.26 (dd, *J* = 7.1, 5.9 Hz, 2H), 2.78 – 2.72 (t, 2H)**.** ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.6, 156.9, 149.9, 147.5, 136.9, 133.9, 132.6, 132.4, 131.0, 129.6, 128.4, 128.3, 128.1, 127.9, 127.8, 126.6, 123.9, 40.4, 25.9. FT-IR (KBr): v= 3053, 2920, 2850, 1693, 1600, 1527, 1467, 1442, 1392, 1332, 1220, 1149, 1097, 1074, 991, 939, 875, 840, 748, 696, 663, 609, 540, 503 cm^{-1}. $C_{26}H_{18}ClNO$: m/z -395.88[M$^+$], found 396.07 [M+1]$^+$.



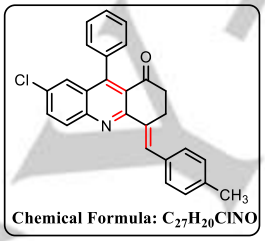
***(E)-7-chloro-9-phenyl-4-(2,3,4-trimethoxybenzylidene)-3,4-dihydroacridin-1(2H)-one (4ab)*:** 90% yield, Yellow solid**,** Mp: 183-185 °C**;** ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.29 (s, 1H), 8.09 (d, *J* = 9.0 Hz, 1H), 7.67 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.56 – 7.44 (m, 3H), 7.39 (d, *J* = 2.2 Hz, 1H), 7.23 – 7.15 (m, 2H), 7.08 (d, *J* = 8.6 Hz, 1H), 6.72 (d, *J* = 8.7 Hz, 1H), 4.01 – 3.87 (m, 9H), 3.18 (dd, *J* = 7.1, 5.7 Hz, 2H), 2.74 (dd, *J* = 8.0, 6.0 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.8, 157.2, 153.8, 152.8, 149.8, 147.6, 142.3, 136.9, 133.5, 132.5, 132.4, 131.1, 128.3, 128.3, 128.1, 127.9, 127.8, 126.5, 124.8, 123.9, 123.7, 106.8, 61.3, 61.0, 56.0, 40.6, 26.3. FT-IR (KBr): v = 2920, 2850, 1695, 1595, 1535, 1492, 1436, 1303, 1263, 1217, 1168, 1093, 1037, 1006, 943, 883, 827, 796, 758, 702, 663, 597, 540, 495, 426 cm^{-1}. $C_{29}H_{24}ClNO_4$: m/z -485.96 [M$^+$], found 486.10 [M+1]$^+$.



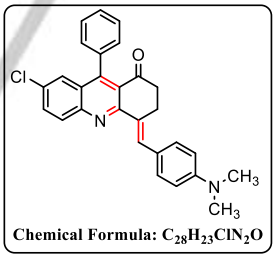
***(E)-7-chloro-4-(4-methoxybenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (4ac)*:** 86% yield, light yellow solid, Mp:196-198 °C**;** ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.27 (s, 1H), 8.07 (d, *J* = 9.0 Hz, 1H), 7.67 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.57 – 7.31 (m, 6H), 7.21 – 7.15 (m, 2H), 6.97 (d, *J* = 8.7 Hz, 2H), 3.86 (s, 3H), 3.35 – 3.06 (m, 2H), 2.75 (dd, *J* = 7.9, 6.1 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.8, 159.3, 157.2, 149.7, 147.6, 136.8, 132.5, 132.4, 132.1, 132.1, 131.2, 131.0, 129.5, 128.3, 128.2, 128.1, 127.9, 126.6, 123.9, 113.9, 55.3, 40.4, 26.0. FT-IR (KBr): v = 2997, 2899, 2835, 1695, 1600, 1529, 1504, 1419, 1327, 1301, 1247, 1161, 1078, 1031, 985, 937, 881, 831, 756, 698, 611, 561, 526, 493 cm^{-1}. $C_{27}H_{20}ClNO_2$: m/z -425.91 [M$^+$], found 426.08 [M+1]$^+$.



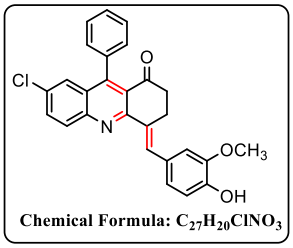
***(E)-7-chloro-4-(4-methylbenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (4ad)*:** 87% yield, Dark yellow solid, Mp: 180-182 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.29 (s, 1H), 8.08 (d, *J* = 9.0 Hz, 1H), 7.68 (d, *J* = 9.0 Hz, 1H), 7.60 – 7.47 (m, 3H), 7.39 (d, *J* = 7.0 Hz, 3H), 7.25 (d, *J* = 7.8 Hz, 2H), 7.19 (d, *J* = 7.9 Hz, 2H), 3.26 (t, *J* = 7.0 Hz, 2H), 2.78 – 2.68 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.8, 193.1, 157.1, 149.8, 147.5, 137.9, 136.8, 134.0 133.1, 132.6, 132.5, 132.4, 131.0, 129.6, 129.1, 128.3, 128.1, 127.9, 126.6, 123.9, 40.4, 25.9, 21.4. FT-IR (KBr): v = 3024 2920, 2850, 1699, 1606, 1533, 1508, 1440, 1390, 1328, 1288, 1222, 1165, 1087, 985, 943, 987, 833, 759, 700, 667, 613, 530, 505, 464 cm^{-1}. $C_{27}H_{20}ClNO$: m/z -409.91 [M$^+$], found 410.09 [M+1]$^+$.



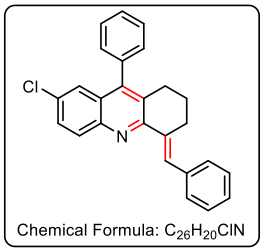
***(E)-7-chloro-4-(4-(dimethylamino)benzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4ae)*:** 88% yield, Yellow solid, Mp:215-217 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 8.05 (d, *J* = 9.0 Hz, 1H), 7.65 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.56 – 7.47 (m, 3H), 7.45 (d, *J* = 8.7 Hz, 2H), 7.37 (d, *J* = 2.3 Hz, 1H), 7.19 (dd, *J* = 7.2, 2.0 Hz, 2H), 6.76 (d, *J* = 8.8 Hz, 2H), 3.30 (dd, *J* = 7.0, 6.0 Hz, 2H), 3.03 (s, 6H), 2.78 – 2.70 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 198.3, 157.6, 150.0, 149.4, 147.7, 136.9, 132.4, 132.0, 131.3, 130.9, 129.8, 128.3, 128.2, 128.0, 127.8, 126.5, 124.9, 123.9, 111.7, 40.4, 40.3, 26.2. FT-IR (KBr): v = 3051, 2900, 2800, 1703, 1598, 1554, 1516, 1442, 1359, 1327, 1222, 1190, 1163, 1128, 1085, 987, 941, 906, 819, 731, 705, 653, 613, 532, 497 cm^{-1}. $C_{28}H_{23}ClN_2O$: m/z -438.95 [M$^+$], found 439.11 [M+1]$^+$.



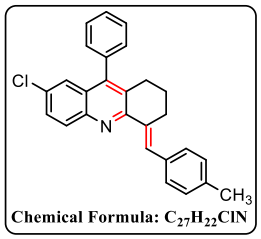
(E)-7-chloro-4-(4-hydroxy-3-methoxybenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4af) : 88% yield, light brown solid, Mp:181-183 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 8.07 (d, *J* = 9.0 Hz, 1H), 7.68 (dd, *J* = 9.0, 1.3 Hz, 1H), 7.52 (d, *J* = 5.4 Hz, 3H), 7.40 (s, 1H), 7.19 (d, *J* = 7.3 Hz, 2H), 7.05 (d, *J* = 8.1 Hz, 1H), 7.02 – 6.94 (m, 2H), 3.94 (s, 3H), 3.28 (t, *J* = 6.9 Hz, 2H), 2.83 – 2.67 (m, 2H).^{13}C NMR (101 MHz, $CDCl_3$) δ 197.9, 157.1, 149.7, 147.5, 146.3, 145.6, 136.8, 132.5, 132.5, 132.4, 132.1, 130.9, 129.2, 128.3, 128.2, 128.1, 127.9, 126.6, 123.9, 123.2, 114.4, 112.3, 56.0, 40.4, 26.0. FT-IR (KBr): v = 3305, 2920, 2850, 1697, 1587, 1533, 1510, 1444, 1390, 1332, 1261, 1213, 1161, 1122, 1093, 1031, 995, 939, 906, 835, 785, 698, 651, 611, 565, 540, 497. cm^{-1}. $C_{27}H_{20}ClNO_3$: m/z - 441.91 [M$^+$], found 442.08 [M+1]$^+$.



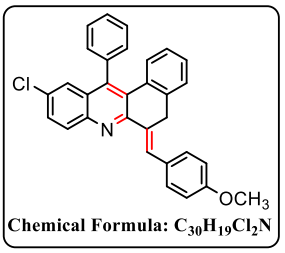
(E)-7-chloro-4-(4-(dimethylamino)benzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4ba) **:** 86% yield, Yellow solid, Mp:147-149 °C**;** ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.23 (s, 1H), 8.05 (d, *J* = 9.0 Hz, 1H), 7.53 (ddd, *J* = 12.2, 6.5, 3.8 Hz, 5H), 7.40 (t, *J* = 7.6 Hz, 2H), 7.29 (t, *J* = 6.0 Hz, 2H), 7.24 (s, 3H), 2.98 (t, *J* = 5.5 Hz, 2H), 2.67 (t, *J* = 6.2 Hz, 2H), 1.86 – 1.74 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 154.5, 145.4, 145.1, 137.8, 136.5, 136.2, 131.5, 131.1, 129.9, 129.8, 129.8, 129.4, 129.2, 128.8, 128.1, 128.1, 127.7, 127.1, 124.5, 28.5, 28.1, 22.9. FT-IR (KBr): v = 3026, 2953, 2846, 1601, 1563, 1537, 1475, 1442, 1392, 1339, 1262, 1187, 1158, 1076, 1031, 961, 926, 832, 761, 747, 699, 659, 615, 567, 508, 466, 439, cm^{-1}**.** $C_{26}H_{20}ClN$: m/z -381.90 [M$^+$], found 382.09 [M+1]$^+$.



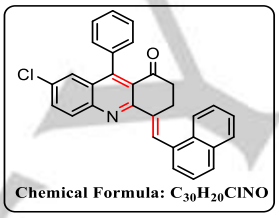
(E)-7-chloro-4-(4-methylbenzylidene)-9-phenyl-1,2,3,4-tetrahydroacridine (4bb)**:** 87% yield, Yellow solid, Mp: 151-153 °C**;** ^{1}H NMR ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.20 (s, 1H), 8.04 (d, *J* = 9.0 Hz, 1H), 7.57 – 7.46 (m, 4H), 7.42 (d, *J* = 7.9 Hz, 2H), 7.32 – 7.17 (m, 5H), 2.97 (t, *J* = 5.5 Hz, 2H), 2.66 (t, *J* = 6.2 Hz, 2H), 2.39 (s, 3H), 1.87 – 1.72 (m, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ ^{13}C NMR (101 MHz, $CDCl_3$) δ 154.7, 145.3, 145.1, 137.0, 136.5, 135.4, 134.9, 131.4, 131.0, 130.0, 129.8, 129.7, 129.3, 129.2, 128.9, 128.8, 128.1, 127.6, 124.5, 28.5, 28.2, 22.9, 21.3. FT-IR (KBr): v = 3029, 2960, 2937, 2837, 1614, 1566, 1540, 1507, 1474, 1442, 1317, 1269, 1230, 1163, 1145, 1073, 1029, 959, 909, 880, 818, 782, 754, 707, 657, 614, 565, 541, 508, 466, 439 cm^{-1}. $C_{27}H_{22}ClN$: m/z -395.93 [M$^+$], found 396.11 [M+1]$^+$.



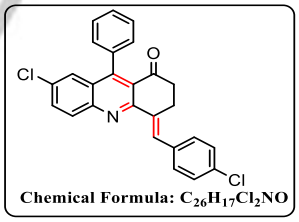
(E)-10-chloro-6-(4-methoxybenzylidene)-12-phenyl-5,6-dihydrobenzo[a]acridine(4ca) **:** 89% yield, Yellow solid, Mp: 184-186 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.15 – 8.07 (m, 1H), 7.92 (s, 1H), 7.66 – 7.58 (m, 2H), 7.56 – 7.50 (m, 3H), 7.49 – 7.44 (m, 2H), 7.38 (dt, *J* = 4.9, 3.9 Hz, 2H), 7.35 – 7.25 (m, 2H), 7.13 (d, *J* = 7.3 Hz, 1H), 7.09 – 7.01 (m, 1H), 6.84 – 6.77 (m, 2H), 3.99 (s, 2H).^{13}C NMR (101 MHz, $CDCl_3$) δ 156.4, 145.4, 143.4, 137.6, 136.9, 136.7, 135.3, 134.8, 132.2, 131.8, 131.2, 131.0, 130.4, 130.0, 129.6, 129.2, 128.7, 128.6, 128.4, 127.8, 127.7, 127.0, 126.3, 126.2, 125.9, 125.2, 33.2. FT-IR (KBr):v = 3048, 2964, 2934, 2835, 1640, 1602, 1543, 1507, 1470, 1440, 1341, 1293, 1245, 1180, 1107, 1072, 1029, 959, 879, 839, 763, 697, 672, 604, 567, 509, 446, 406 cm^{-1}. $C_{30}H_{19}Cl_2N$: m/z -459.97 [M$^+$], found 460.10 [M+1]$^+$.



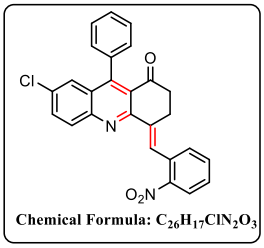
(E)-7-chloro-4-(naphthalen-1-ylmethylene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (4ag)**:** 90% Yield, light yellow solid, Mp:185-187 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.78 (s, 1H), 8.17 – 8.11 (m, 2H), 7.96 – 7.89 (m, 1H), 7.87 (d, *J* = 7.4 Hz, 1H), 7.71 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.58 – 7.47 (m, 7H), 7.44 (d, *J* = 2.2 Hz, 1H), 7.24 – 7.18 (m, 2H), 3.11 (td, *J* = 6.8, 1.2 Hz, 2H) 2.70 (dd, *J* = 8.0, 5.9 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.6, 156.8, 150.2, 147.6, 136.9, 135.7, 134.3, 133.6, 132.7 132.6, 132.2, 131.2, 130.5, 128.6, 128.5, 128.3, 128.1, 127.9, 126.7, 126.6, 126.3, 126.1, 125.2, 125.0, 123.8, 40.6, 26.2. FT-IR (KBr): v **=** 3018, 2920, 1695, 1600, 1533, 1469, 1390, 1319, 1274, 1219, 1182, 1095, 1072, 1010, 989, 939, 877, 827, 779, 700, 657, 611, 536, 495, 428 cm^{-1}. $C_{30}H_{20}ClNO$: m/z -445.94 [M$^+$], found 446.09 [M+1]$^+$.



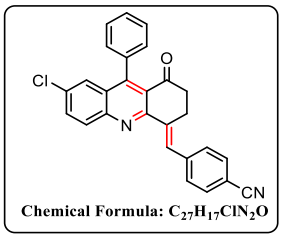
(E)-7-chloro-4-(4-chlorobenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (4ah)**:** 85% Yield Yellow solid, Mp: 250-252 °C; ^{1}H NMR 400 MHz, $CDCl_3$**)** δ 8.26 (s, 1H), 8.08 (d, *J* = 9.0 Hz, 1H), 7.70 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.52 (dd, *J* = 5.1, 1.9 Hz, 3H), 7.41 (s, 5H), 7.21 – 7.15 (m, 2H), 3.22 (td, *J* = 6.9, 1.4 Hz, 2H), 2.75 (dd, *J* = 8.0, 6.0 Hz, 2H) ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.4, 156.6, 150.0, 147.5, 136.6, 135.3, 134.5, 133.6, 132.8, 132.7, 131.0, 130.8, 128.7, 128.4, 128.3, 128.1, 128.0, 126.6, 123.9, 40.2, 25.8. FT-IR (KBr): v **=** 3059, 2922, 2850, 1699, 1533, 1483, 1440, 1390, 1323, 1284, 1217, 1176, 1085, 1010, 983, 941, 906, 829, 759, 700, 663, 613, 561, 507 cm^{-1}**.** $C_{26}H_{17}Cl_2NO$: m/z -430.32 [M$^+$], found 430.03 [M$^+$].



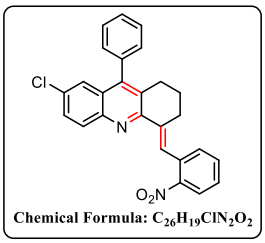
***(E)-7-chloro-4-(2-nitrobenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (4ai)*:** 77% yield, light yellow solid, Mp: 222-224 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.46 (s, 1H), 8.20 – 8.14 (m, 1H), 8.10 (d, *J* = 9.0 Hz, 1H), 7.74 – 7.63 (m, 2H), 7.59 – 7.45 (m, 5H), 7.42 (d, *J* = 2.2 Hz, 1H), 7.23 – 7.16 (m, 2H), 3.06 – 2.94 (m, 2H), 2.74 (dd, *J* = 8.1, 5.8 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.0, 156.0, 150.3, 148.6, 147.5, 136.7, 135.5, 133.1, 133.0, 132.8, 132.7, 131.6, 131.2, 128.7, 128.7, 128.3, 128.3, 128.0, 128.0, 126.6, 125.0, 123.7, 40.3, 25.9. FT-IR (KBr): v = 3059, 2978, 1695, 1602, 1523, 1471, 1438, 1390, 1346, 1327, 1220, 1168, 1130, 1091, 1072, 985, 941, 875, 839, 765, 715, 704, 665, 611, 569, 532, 499, 459 cm^{-1}. $C_{26}H_{17}ClN_2O_3$: m/z -440.88 [M$^+$], found 441.06 [M+1]$^+$.



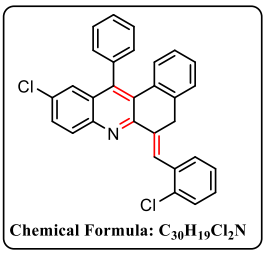
***(E)-7-chloro-4-(4-hydroxy-3-methoxybenzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4aj)* :** 83% yield, light yellow solid, Mp:261-263 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.31 (s, 1H), 8.10 (d, *J* = 9.0 Hz, 1H), 7.76 – 7.68 (m, 3H), 7.58 (d, *J* = 8.2 Hz, 2H), 7.53 (dd, *J* = 5.0, 1.8 Hz, 3H), 7.43 (d, *J* = 2.2 Hz, 1H), 7.22 – 7.13 (m, 2H), 3.22 (td, *J* = 6.9, 1.3 Hz, 2H), 2.77 (dd, *J* = 8.0, 6.0 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 196.9, 155.9, 150.2, 147.4, 141.6, 136.7, 136.5, 133.2, 132.9, 132.2, 131.1, 130.1, 130.1, 128.6, 128.4, 128.1, 126.7, 123.9, 118.8, 111.1, 40.1, 25.8. FT-IR (KBr): v = 2960, 2218, 1693, 1598, 1533, 1471, 1440, 1388, 1325, 1284, 1219, 1159, 1082, 1012, 943, 881, 829, 756, 698, 609, 543, 497, 447 cm^{-1}. $C_{27}H_{17}ClN_2O$: m/z -420.89 [M$^+$], found 421.07 [M+1]$^+$.



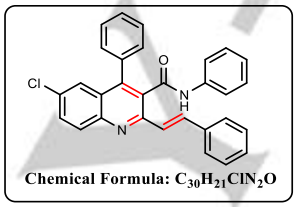
***(E)-7-chloro-4-(4-(dimethylamino)benzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4bc)* :** 85% yield, Dark yellow solid, Mp:175-177 °C; ^{1}H NMR 400 MHz, $CDCl_3$ δ 8.38 (s, 1H), 8.06 (dd, *J* = 14.6, 8.6 Hz, 2H), 7.62 (t, *J* = 7.3 Hz, 1H), 7.58 – 7.41 (m, 6H), 7.29 (d, *J* = 2.2 Hz, 1H), 7.26 – 7.19 (m, 2H), 2.68 (dd, *J* = 12.6, 6.4 Hz, 4H), 1.85 – 1.74 (m, 2H).^{13}C NMR (101 MHz, $CDCl_3$) δ ^{13}C NMR (101 MHz, $CDCl_3$) δ 153.3, 148.8, 145.9, 145.1, 138.1, 136.4, 133.6, 132.6, 131.9, 131.8, 131.3, 129.5, 129.5, 129.1, 128.8, 128.1, 127.9, 125.6, 124.6, 124.4, 28.6, 27.9, 22.9. FT-IR (KBr): v = 3360, 3056, 2943, 2917, 2858, 1730, 1603, 1566, 1522, 1476, 1438, 1346, 1270, 1173, 954, 916, 824, 768, 714, 659, 619, 566, 538, 515 cm^{-1}. $C_{26}H_{19}ClN_2O_2$: m/z -426.90 [M$^+$], found 427.08 [M+1]$^+$.



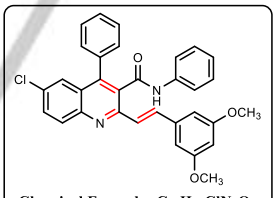
***(E)-7-chloro-4-(4-(dimethylamino)benzylidene)-9-phenyl-3,4-dihydroacridin-1(2H)-one(4cb)* :** 87% yield, light yellow solid, Mp: 215-217 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.15 – 8.07 (m, 1H), 7.92 (s, 1H), 7.66 – 7.58 (m, 2H), 7.56 – 7.50 (m, 3H), 7.49 – 7.44 (m, 2H), 7.38 (dt, *J* = 4.9, 3.9 Hz, 2H), 7.35 – 7.25 (m, 2H), 7.13 (d, *J* = 7.3 Hz, 1H), 7.09 – 7.01 (m, 1H), 6.84 – 6.77 (m, 2H), 3.99 (s, 2H).^{13}C NMR (101 MHz, $CDCl_3$) δ 156.4, 145.4, 143.4, 137.6, 136.9, 136.7, 135.3, 134.8, 132.2, 131.8, 131.2, 131.0, 130.4, 130.0, 129.6, 129.2, 128.7, 128.6, 128.4, 127.8, 127.7, 127.0, 126.3, 126.2, 125.9, 125.2, 33.2. FT-IR (KBr): v = 3053, 2918, 1598, 1539, 1471, 1415, 1342, 1321, 1205, 1166, 1076, 1031, 987, 939, 875, 827, 794, 754, 704, 605, 536, 447 cm^{-1}. $C_{30}H_{19}Cl_2N$: m/z - 464.38 [M$^+$], found 464.05 [M$^+$].



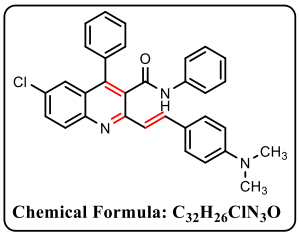
***(E)-6-chloro-N,4-diphenyl-2-styrylquinoline-3-carboxamide(5da)* :** 90% yield, light yellow solid, Mp: 146-148 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.01 – 7.92 (m, 2H), 7.65 – 7.58 (m, 1H), 7.53 (s, 1H), 7.47 (d, *J* = 8.0 Hz, 3H), 7.38 (t, *J* = 6.0 Hz, 3H), 7.35 – 7.26 (m, 5H), 7.25 – 7.13 (m, 4H), 7.09 (t, *J* = 7.1 Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.8, 151.7, 146.4, 144.9, 137.0, 136.9, 136.2 134.4, 132.5, 131.3, 131.0, 130.0, 129.2, 129.0, 128.9, 128.8, 128.8, 128.7, 127.7, 126.3, 125.3, 125.2, 123.7, 120.8. FT-IR (KBr): v =3265, 3062, 1732, 1639, 1598, 1552, 1479, 1444, 1394, 1332, 1251, 1149, 1076, 958, 835, 752, 690, 657, 601, 543, 509, 468 cm^{-1}. $C_{30}H_{21}ClN_2O$: m/z – 460.96 [M$^+$], found 461.09 [M+1]$^+$.



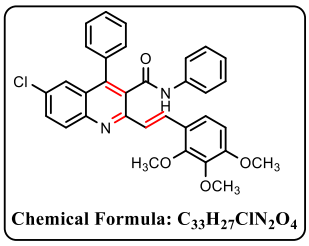
***(E)-6-chloro-2-(3,5-dimethoxystyryl)-N,4-diphenylquinoline-3-carboxamide (5db)* :** 90% yield, Yellow solid, Mp: 216-218 °C; ^{1}H NMR (400 MHz, $CDCl_3$) 8.02 (dd, *J* = 14.3, 12.5 Hz, 2H), 7.65 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.54 – 7.43 (m, 4H), 7.39 (d, *J* = 5.7 Hz, 3H), 7.25 (d, *J* = 5.6 Hz, 4H), 7.16 – 7.09 (m, 2H), 7.02 (s, 1H), 6.82 (d, *J* = 8.3 Hz, 1H), 3.89 (s, 3H), 3.82 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.9, 152.1, 149.9, 148.9, 146.5, 144.9, 137.0, 136.9, 134.5 132.4, 131.4, 130.9, 130.0, 129.3, 129.3, 129.0, 129.0, 128.8, 126.2, 125.3, 125.2, 121.9, 121.3, 120.6, 111.0, 109.9, 55.9, 55.8. FT-IR (KBr): v = 3354, 2999, 2929, 2835, 1739, 1676, 1633, 1597, 1525, 1510, 1436, 1388, 1317, 1255, 1238, 1139, 1076, 1022, 956, 885, 846, 804, 756, 692, 642, 605, 586, 542, 447 cm^{-1}. $C_{32}H_{25}ClN_2O_3$: m/z -521.01 [M$^+$], found 521.11 [M$^+$].



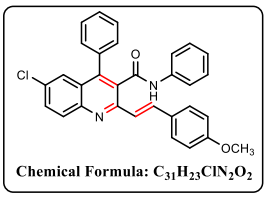
(E)-6-chloro-2-(4-(dimethylamino)styryl)-N,4-diphenylquinoline-3-carboxamide (5dc) **:** 89% yield, Yellow solid, Mp: 215-217 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (t, *J* = 10.4 Hz, 2H), 7.64 (d, *J* = 9.1 Hz, 1H), 7.54 – 7.41 (m, 8H), 7.28 (d, *J* = 7.7 Hz, 2H), 7.21 (d, *J* = 8.2 Hz, 2H), 7.11 (t, *J* = 7.1 Hz, 2H), 6.67 (d, *J* = 8.2 Hz, 2H), 3.00 (s, 6H). δ ^{13}C NMR (101 MHz, $CDCl_3$) δ 150.9, 137.5, 130.8, 129.3, 129.2, 128.9, 128.8, 125.2, 120.7, 119.0, 112.0, 40.2. FT-IR (KBr): ν **=** 3244, 3057, 2922, 2800, 1653, 1598, 1560, 1519, 1440, 1355, 1325, 1251, 1180, 1145, 1070, 956, 829, 804, 748, 690, 648, 601, 509, 478 cm^{-1}. $C_{32}H_{26}ClN_3O$ m/z -504.03 [M$^+$], found 504.20 [M$^+$].



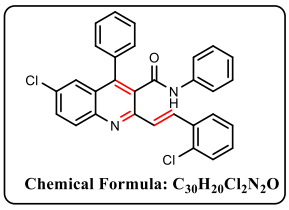
(E)-6-chloro-N,4-diphenyl-2-(2,3,4-trimethoxystyryl)quinoline-3-carboxamide (5dd) **:** 90% yield, Dark yellow solid, Mp: 183-185 °C; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.25 (d, *J* = 15.7 Hz, 1H), 8.07 (d, *J* = 9.0 Hz, 1H), 7.64 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.46 (ddd, *J* = 12.2, 5.6, 2.7 Hz, 6H), 7.30 – 7.21 (m, 7H), 6.65 (d, *J* = 8.8 Hz, 1H), 3.90 (s, 3H), 3.88 (d, *J* = 2.9 Hz, 6H). **δ** ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.9, 154.3, 152.9, 152.5, 146.6, 144.8, 136.9, 134.6, 132.3, 132.1, 131.2, 131.1, 130.1, 129.3, 129.0, 128.9, 128.8, 126.2, 125.2, 125.1, 123.5, 123.4, 123.1, 120.7, 107.5, 61.3, 60.9, 56.0. FT-IR (KBr): **ν =** 3381, 2933, 2823, 1680, 1595, 1523, 1492, 1440, 1388, 1317, 1286, 1244, 1147, 1083, 1031, 983, 896, 806, 756, 688, 663, 559, 501, 476 cm^{-1}. $C_{33}H_{27}ClN_2O_4$ m/z -551.03 [M$^+$], found 551.19 [M$^+$].



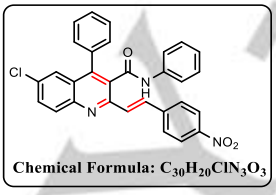
(E)-6-chloro-2-(4-methoxystyryl)-N,4-diphenylquinoline-3-carboxamide(5de) Yellow solid, Yield-90% Mp:201-203 °C**;** ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 – 8.02 (m, 2H), 7.66 (dd, *J* = 9.0, 2.2 Hz, 1H), 7.35 (s, 1H), 7.29 – 7.21 (m, 4H), 7.20 (d, *J* = 7.8 Hz, 3H), 7.12 (t, *J* = 7.3 Hz, 1H), 6.88 (d, *J* = 8.6 Hz, 2H), 3.83 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.8, 160.3, 146.6, 144.9, 136.8, 136.7, 134.6, 132.3, 131.4, 131.0, 129.2, 129.2, 129.1, 129.0, 129.0, 128.9, 126.1, 125.3, 125.2, 121.6, 120.7, 114.1, 55.3. FT-IR (KBr): ν **=** 3441, 3257, 2980, 1647, 1602, 1533, 1440, 1382, 1317, 1242, 1174, 1145, 1068, 1022, 958, 883, 837, 783, 744, 688, 657, 578, 516 cm^{-1}. $C_{31}H_{23}ClN_2O_2$ m/z -490.98 [M$^+$], found 491.17 [M+1]$^+$.



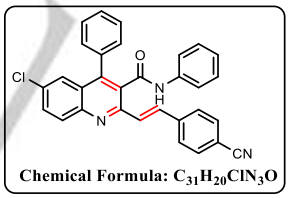
(E)-6-chloro-2-(2-chlorostyryl)-N,4-diphenylquinoline-3-carboxamide (5df) **:** 88% yield, Yellow solid, Mp:170-172 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.41 (d, *J* = 15.6 Hz, 1H), 8.03 (d, *J* = 9.0 Hz, 1H), 7.63 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.58 (dd, *J* = 7.4, 1.8 Hz, 1H), 7.49 (d, *J* = 2.2 Hz, 1H), 7.44 (dt, *J* = 13.7, 8.3 Hz, 4H), 7.35 (ddd, *J* = 6.8, 6.0, 1.6 Hz, 3H), 7.26 – 7.14 (m, 6H), 7.09 (dd, *J* = 9.5, 4.4 Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165, 151, 146, 145, 136, 134, 134, 134, 133, 132, 131, 131, 130, 129, 129, 129, 129, 128, 128, 127, 126, 126, 126, 125, 125, 120. FT-IR (KBr): ν **=** 3282, 3024, 2916, 2848, 1735, 1654, 1598, 1556, 1473, 1442, 1319, 1257, 1165, 1078, 1043, 960, 906, 827, 742, 692, 651, 597, 511, 439 cm^{-1}. $C_{30}H_{20}Cl_2N_2O$ m/z -495.40 [M$^+$], found 495.05 [M$^+$].



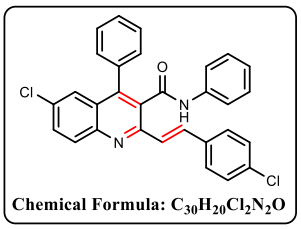
(E)-6-chloro-2-(4-nitrostyryl)-N,4-diphenylquinoline-3-carboxamide (5dg) **:** 83% yield, Yellow solid, Mp:206-208 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ 8.53 (d, *J* = 15.3 Hz, 1H), 8.13 (d, *J* = 9.0 Hz, 1H), 8.00 (d, *J* = 8.1 Hz, 1H), 7.77 (d, *J* = 7.8 Hz, 1H), 7.71 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.63 – 7.56 (m, 2H), 7.55 – 7.50 (m, 2H), 7.47 (t, *J* = 6.4 Hz, 4H), 7.23 (d, *J* = 7.5 Hz, 2H), 7.19 – 7.07 (m, 4H).^{13}C NMR (101 MHz, $CDCl_3$) δ 165.6, 150.9, 147.8, 146.5, 145.6, 145.1, 134.5, 133.3, 132.4, 132.2, 131.6, 131.6, 129.2, 129.1, 129.0 128.9, 128.8, 125.3, 125.2, 124.7, 120.6. FT-IR (KBr): ν **=** 3437, 3041, 1639, 1600, 1548, 1519, 1477, 1442, 1313, 1253, 1149 1076, 958, 833, 779, 742, 698, 653, 603, 511 cm^{-1}. $C_{30}H_{20}ClN_3O$: m/z – 505.95 [M$^+$], found 506.07 [M+1]$^+$.



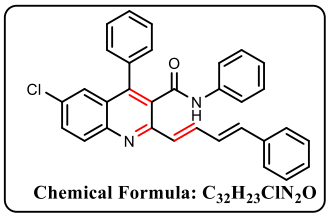
(E)-6-chloro-2-(4-cyanostyryl)-N,4-diphenylquinoline-3-carboxamide(5dh) **:** 78% yield, Dark yellow solid, Mp: 226-228 °C; ^{1}H NMR 400 MHz, $CDCl_3$) δ δ 8 09 (dd, *J* = 10.3, 6.2 Hz, 2H), 7.71 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.65 (q, *J* = 8.5 Hz, 3H), 7.59 (d, *J* = 2.4 Hz, 1H), 7.56 (dd, *J* = 8.8, 4.0 Hz, 1H), 7.53 – 7.48 (m, 2H), 7.45 (dd, *J* = 14.0, 6.2 Hz, 3H), 7.32 – 7.23 (m, 3H), 7.22 – 7.11 (m, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.5, 140.7, 136.7, 134.5, 134.4, 134.0, 133.3, 133.0, 132.5, 132.3, 131.7, 131.4, 131.2, 129.9, 129.3, 129.3, 129.1, 129.1, 129.0, 128.4, 128.0, 127.9, 126.6, 126.5, 125.4, 125.4, 120.6 FT-IR (KBr): ν **=** 3242, 3064, 1600, 1442, 1315, 1253, 1149, 1076, 960, 835, 781, 690, 653, 513 cm^{-1}. $C_{31}H_{20}ClN_3O$:m/z -485.97 [M$^+$], found 486.08 [M+1]$^+$.



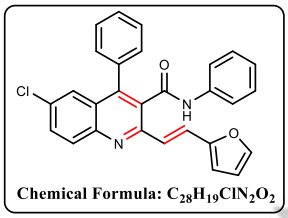
(E)-6-chloro-2-(4-chlorostyryl)-N,4-diphenylquinoline-3-carboxamide (5di) **:** 87% Yellow solid, Mp:190-192 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 – 8.05 (m, 2H), 7.69 (d, *J* = 9.0 Hz, 1H), 7.58 (s, 1H), 7.52 (t, *J* = 7.1 Hz, 4H), 7.46 (dd, *J* = 10.0, 3.2 Hz, 4H), 7.33 (d, *J* = 8.3 Hz, 2H), 7.27 (s, 1H), 7.18 (d, *J* = 7.8 Hz, 2H), 7.13 (t, *J* = 7.6 Hz, 1H), 7.03 (d, *J* = 10.5 Hz, 1H). δ ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.9, 152.1, 149.9, 148.9, 146.5, 144.9, 137.0, 129.2, 129.1, 129.0, 128.9, 128.8, 128.5, 125.3, 122.6, 120.6, 119.6**.** FT-IR (KBr): v **=** 3354, 3030, 2924, 1649, 1597, 1552, 1485, 1442, 1396, 1342, 1259, 1151, 1093, 1010, 962, 829, 748, 690, 644, 572, 507, 474 cm^{-1}. $C_{30}H_{20}Cl_2N_2O$: m/z -495.40 [M$^+$], found 495.12 [M$^+$].



6-chloro-N,4-diphenyl-2-((1E,3E)-4-phenylbuta-1,3-dien-1-yl)quinoline-3-carboxamide (5dj) **:** 89% yield, light yellow solid, Mp: 217-219 °C-; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.05 (d, *J* = 9.0 Hz, 1H), 7.92 (dd, *J* = 14.8, 10.9 Hz, 1H), 7.70 – 7.62 (m, 1H), 7.53 (d, *J* = 2.2 Hz, 1H), 7.47 (dd, *J* = 11.8, 7.2 Hz, 14H), 7.43 – 7.37 (m, 6H), 7.33 (d, *J* = 7.7 Hz, 6H), 7.29 – 7.21 (m, 19H), 7.17 – 7.10 (m, 6H), 7.07 – 6.98 (m, 6H), 6.89 (d, *J* = 15.6 Hz, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.77, 152.01, 143.74, 137.52, 137.42, 136.91, 134.58, 132.54, 131.44, 131.10, 129.28, 129.12, 129.01, 128.93, 128.73, 128.48, 128.30, 127.73, 126.90, 125.31, 120.64. FT-IR (KBr): v = 3221.12, 3024.38, 2922.16, 2850.79, 1649.14, 1595.13, 1533.41, 1473.62, 1442.75, 1392.61, 1313.52, 1257.59, 1136.07, 1072.42, 989.48, 958.62, 873.75, 835.18, 790.81, 748.38, 688.59, 630.72, 603.72, 569.00, 501.49 cm^{-1}. $C_{32}H_{23}ClN_2O$ m/z -486.99 [M$^+$], found 487.17 [M+1]$^+$.



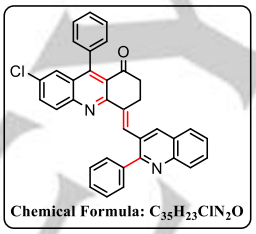
(E)-6-chloro-2-(2-(furan-2-yl)vinyl)-N,4-diphenylquinoline-3-carboxamide (5dk) Brown solid, Yield-73% Mp:198-200 °C**,** ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.03 (d, *J* = 9.0 Hz, 1H), 7.88 (d, *J* = 15.3 Hz, 1H), 7.66 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.53 (d, *J* = 2.2 Hz, 1H), 7.45 (dd, *J* = 11.4, 7.1 Hz, 5H), 7.34 (d, *J* = 15.3 Hz, 1H), 7.25 (d, *J* = 7.6 Hz, 3H), 7.22 – 7.18 (m, 2H), 7.11 (t, *J* = 7.2 Hz, 1H), 6.56 (d, *J* = 3.3 Hz, 1H), 6.45 (d, *J* = 1.8 Hz, 1H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.7, 152.7, 151.7, 146.5, 143.5, 136.8, 134.6, 131.4, 131.0, 129.2, 129.0, 128.9, 128.9, 126.3, 125.3, 125.2, 123.9, 121.8, 120.7, 112.5, 112.0. FT-IR (KBr): v **=** 3263, 1639, 1598, 1554, 1388, 1336, 1255, 1128, 1076, 979, 931, 823, 732, 692, 648, 590, 509 cm^{-1}. $C_{28}H_{19}ClN_2O_2$ m/z -450.92 [M$^+$], found 451.14 [M+1]$^+$.



Synthesis procedure for Suzuki coupled (E)-4-benzylidenylacridin-1(2H)-ones, 9 and (E)-2-styryl quinoline-3-carboxamides, 10

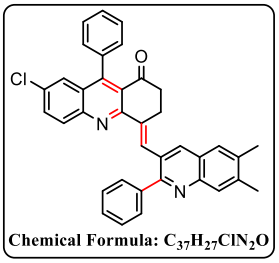
A mixture of K_2CO_3: ethylene glycol (100 mg:100 mg) was stirred for 1h at 90 °C to attain the DES-1. Then added (2-chloroquinolin-3-yl)methanol, **6** (1.0 mmol), phenylboronic acid, **7** (1.0 mmol) 2 mol% $Pd(PPh_3)_4$, heated at 90 °C for 20 min to provide the Suzuki coupling product. Subsequently, added 2-amino-5-chloro benzhydrol, **1** (1.0 mmol) and ketones (**3** or **3`**) and continued dehydrogenative Friedlander annulation/sp^3 C-H activation as per the above procedure) to yield the Suzuki coupled (E)-4-benzylidenylacridin-1(2H)-ones, **9** and (E)-2-styryl quinoline-3-carboxamides, **10.** The crude reaction mixtures of 10a,10b,10c was passed through column chromatography, with petroleum ether/ethyl acetate (8/2) solvents. And the remaining crude mixtures purified by recrystallization using methanol.

(E)-7-chloro-9-phenyl-4-((2-phenylquinolin-3-yl)methylene)-3,4-dihydroacridin-1(2H)-one (9a)



light yellow solid, Yield-80% Mp:135-17 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.32 (s, 1H), 8.27 – 8.17 (m, 2H), 7.99 (d, *J* = 9.0 Hz, 1H), 7.89 – 7.81 (m, 3H), 7.76 (s, 1H), 7.65 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.58 (s, 1H), 7.52 (t, *J* = 2.2 Hz, 3H), 7.48 – 7.38 (m, 4H), 7.24 – 7.14 (m, 2H), 3.12 (dd, *J* = 7.1, 5.8 Hz, 2H), 2.65 (dd, *J* = 7.9, 6.0 Hz, 2H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.2, 159.0, 156.3, 150.1, 147.5, 147.4, 140.2, 137.1, 136.8, 135.0, 132.8, 132.6, 131.2, 130.3, 130.0, 129.8, 129.5, 129.1, 128.8, 128.5, 128.3, 128.3, 128.1, 128.0, 127.5, 126.9, 126.6, 126.5, 123.5, 39.9, 25.9. FT-IR (KBr): v = 3053, 2966, 1693, 1535, 1475, 1440, 1392, 1321, 1269, 1219, 1180, 1078, 1010, 943, 910, 835, 792, 754, 696, 613, 542, 474, 406 cm^{-1}. $C_{35}H_{23}ClN_2O$ m/z -523.03 [M$^+$], found 523.17 [M$^+$].

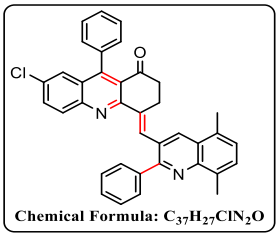
(E)-7-chloro-4-((6,7-dimethyl-2-phenylquinolin-3-yl)methylene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (9b)



Yellow solid, Yield-84% Mp:236-238 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, *J* = 15.5 Hz, 2H), 8.01 (d, *J* = 9.0 Hz, 1H), 7.87 – 7.76 (m, 3H), 7.66 (dd, *J* = 9.0, 2.0 Hz, 1H), 7.53 (d, *J* = 6.0 Hz, 3H), 7.47 – 7.35 (m, 3H), 7.26 (s, 2H), 7.21 – 7.14 (m, 2H), 3.10 (t, *J* = 6.8 Hz, 2H), 2.69 (s, 3H), 2.63 – 2.57 (m, 2H), 2.55 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.3, 158.3, 156.5, 150.1, 148.0, 147.5, 140.4, 140.1, 136.8, 134.7, 134.0,

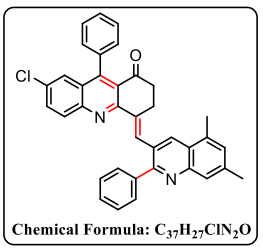
133.8, 132.7, 132.6, 131.2, 130.9, 129.8, 129.7, 128.6, 128.5, 128.3, 128.3, 128.0, 127.9, 127.7, 126.7, 126.6, 124.0, 123.5, 39.9, 25.9, 21.9, 18.6. FT-IR (KBr): v = 2916, 2850, 1695, 1610, 1535, 1469, 1381, 1313, 1267, 1180, 1074, 983, 918, 844, 767, 700, 663, 542, 513 cm^{-1}. $C_{37}H_{27}ClN_2O$ m/z -551.08 [M^+], found 551.30 [M^+].

(E)-7-chloro-4-((5,8-dimethyl-2-phenylquinolin-3-yl)methylene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (9c)



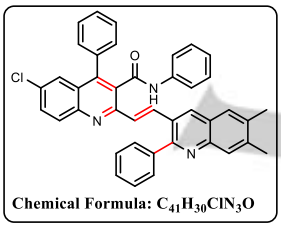
Yellow solid, Yield-82% Mp:255-257 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, *J* = 16.0 Hz, 2H), 8.00 (d, *J* = 9.0 Hz, 1H), 7.86 – 7.73 (m, 3H), 7.66 (d, *J* = 9.0 Hz, 1H), 7.52 (d, *J* = 5.8 Hz, 3H), 7.42 (dd, *J* = 12.6, 4.4 Hz, 4H), 7.26 (s, 1H), 7.19 (d, *J* = 7.6 Hz, 2H), 3.10 (t, *J* = 6.8 Hz, 2H), 2.69 (s, 3H), 2.63 – 2.57 (m, 2H), 2.55 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.3, 158.3, 156.5, 150.1, 148.0, 147.5, 140.4, 140.1, 136.8, 134.7, 134.0, 133.8, 132.7, 132.6, 131.2, 130.9, 129.8, 129.7, 128.6, 128.5, 128.4, 128.3, 128.0, 127.9, 127.7, 126.7, 126.6, 124.0, 123.5, 39.9, 25.9, 21.9, 18.6. FT-IR (KBr): v = 3055, 2914, 2852, 2337, 1695, 1618, 1533, 1469, 1379, 1323, 1267 1182, 1126, 1076, 983, 918, 844, 767, 700, 663, 613, 563, 542, 453 cm^{-1}. $C_{37}H_{27}ClN_2O$ m/z -551.08 [M^+], found 551.15 [M^+].

(E)-7-chloro-4-((5,7-dimethyl-2-phenylquinolin-3-yl)methylene)-9-phenyl-3,4-dihydroacridin-1(2H)-one (9d)

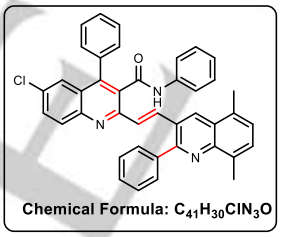


Yellow solid, Yield-81% Mp:251-253 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, *J* = 12.0 Hz, 2H), 8.01 (d, *J* = 9.0 Hz, 1H), 7.82 (dd, *J* = 8.6, 7.1 Hz, 3H), 7.73 – 7.60 (m, 1H), 7.58 – 7.49 (m, 3H), 7.48 – 7.34 (m, 3H), 7.26 (s, 2H), 7.19 (dd, *J* = 7.2, 2.2 Hz, 2H), 3.08 (d, *J* = 7.2 Hz, 2H), 2.69 (s, 3H), 2.62 – 2.58 (m, 2H), 2.55 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 197.2, 173.7, 158.3, 156.5, 147.5, 140.0, 136.8, 134.6, 134.0, 133.7, 132.6, 131.2, 130.9, 129.8, 129.7, 128.6, 128.3, 128.2, 128.0, 127.9, 127.7, 126.7, 126.5, 124.0, 39.9, 25.9, 21.9, 18.5. FT-IR (KBr): v = 3053, 2980, 1695, 1618, 1533, 1469, 1381, 1323, 1265, 1219, 1182, 1076, 983, 918, 844, 798, 736, 700, 671, 613, 563, 542, 451 cm^{-1}. $C_{37}H_{27}ClN_2O$ m/z -551.08 [M^+], found 551.21 [M^+].

(E)-6-chloro-2-(2-(6,7-dimethyl-2-phenylquinolin-3-yl)vinyl)-N,4-diphenylquinoline-3-carboxamide (10a) Dark yellow solid, Yield-83% Mp:152-154 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.35 (d, *J* = 12.2 Hz, 2H), 7.97 (d, *J* = 8.9 Hz, 1H), 7.83 (d, *J* = 7.7 Hz, 2H), 7.64 (d, *J* = 9.1 Hz, 1H), 7.52 – 7.43 (m, 8H), 7.26 (s, 5H), 7.17 (s, 2H), 7.10 (d, *J* = 16.2 Hz, 2H), 2.80 (s, 3H), 2.49 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 161.0, 141.8, 129.6, 128.0, 126.6, 125.2, 125.0, 124.5, 124.4, 124.2, 124.2, 123.9, 123.5, 121.7, 120.5, 120.4, 115.9, 17.2, 13.7. FT-IR (KBr): v = 3664, 3047, 2912, 1653, 1600, 1546, 1498, 1444, 1382, 1315, 1255, 1147, 1074, 1018, 960, 904, 831, 756, 698, 653, 592, 509 cm^{-1}. $C_{41}H_{30}ClN_3O$ m/z -616.16 [M^+], found 617.22 [M+1]$^+$.

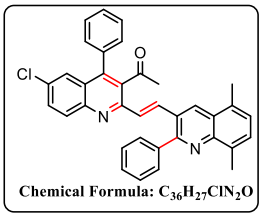


(E)-6-chloro-2-(2-(5,7-dimethyl-2-phenylquinolin-3-yl)vinyl)-N,4-diphenylquinoline-3-carboxamide (10b) Dark yellow solid, Yield-82% Mp:155-157 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (s, 1H), 8.26 (d, J = 15.5 Hz, 1H), 7.97 (d, *J* = 9.0 Hz, 1H), 7.78 (s, 1H), 7.76 – 7.67 (m, 2H), 7.63 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.48 (ddt, *J* = 9.4, 4.7, 2.6 Hz, 9H), 7.27 (d, *J* = 6.9 Hz, 3H), 7.18 (dd, *J* = 6.3, 2.4 Hz, 4H), 7.11 (s, 1H), 2.62 (s, 3H), 2.51 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.7, 151.7, 146.5, 144.9, 136.8, 135.4, 134.3, 132.8, 131.4, 130.0, 129.2, 129.0, 128.9, 128.3, 126.3, 125.3, 120.6, 21.9, 18.4. FT-IR (KBr): v = 3475, 3049, 2924, 1651, 1598, 1548, 1498, 1477, 1442, 1371, 1315, 1255, 1213, 1151, 1074, 960, 860, 781, 756, 702, 690, 596, 563, 509 cm^{-1}. $C_{41}H_{30}ClN_3O$ m/z -616.16 [M^+], found 617.22 [M+1]$^+$.



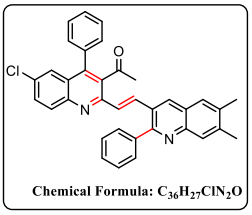
(E)-6-chloro-2-(2-(5,8-dimethyl-2-phenylquinolin-3-yl)vinyl)-N,4-diphenylquinoline-3-carboxamide (10c) Dark yellow solid, Yield-83% Mp:213-215 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (s, 1H), 8.26 (d, *J* = 15.5 Hz, 1H), 7.97 (d, *J* = 9.0 Hz, 1H), 7.78 (s, 1H), 7.75 – 7.67 (m, 2H), 7.63 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.53 (ddd, *J* = 13.9, 6.4, 4.0 Hz, 3H), 7.49 – 7.38 (m, 5H), 7.30 – 7.21 (m, 3H), 7.18 (dd, *J* = 6.3, 2.4 Hz, 4H), 7.11 (d, *J* = 7.3 Hz, 1H), 2.62 (s, 3H), 2.51 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 165.75, 146.59, 145.59, 136.84, 134.36, 132.82, 131.41, 130.02, 129.27, 129.02, 128.99, 128.31, 126.39, 125.31, 120.64, 21.96, 18.49. FT-IR (KBr): v = 3049, 2922, 2852, 1670, 1597, 1529, 1473, 1440, 1375, 1319, 1247, 1205, 1149, 1070, 989, 958, 906, 858, 829, 746, 688, 638, 561, 505 cm^{-1}. $C_{41}H_{30}ClN_3O$ m/z -616.16 [M^+], found 616.23 [M^+].

(E)-1-(6-chloro-2-(2-(5,8-dimethyl-2-phenylquinolin-3-yl)vinyl)-4-phenylquinolin-3-yl)ethan-1-one (10d) Yellow solid, Yield-82% Mp:226-228 °C ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (s, 1H), 8.18 (d, *J* = 15.5 Hz, 1H), 7.98 (d, J = 9.0 Hz, 1H), 7.80 (s, 1H), 7.75 – 7.68 (m, 2H), 7.67 – 7.60 (m, 1H), 7.54 (q, *J* = 4.0 Hz, 4H), 7.48 (dd, *J* = 13.0, 7.2 Hz, 3H), 7.36 (s, 2H), 7.25 (d, *J* = 8.4

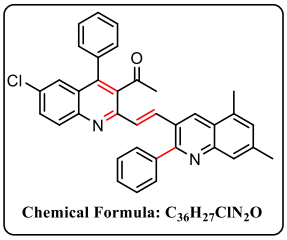


Hz, 2H), 7.15 (d, *J* = 15.4 Hz, 1H), 2.74 (s, 3H), 2.53 (s, 3H), 2.03 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 205.4, 158.6, 150.3, 148.3, 146.3, 143.6, 140.2, 140.2, 135.7, 135.0, 134.4, 134.3, 132.8, 131.3, 131.2, 131.1, 129.9, 129.9, 129.7, 129.2, 128.9, 128.5, 128.3, 127.8, 126.6, 126.3, 126.2, 124.9, 124.6, 32.6, 21.9, 18.6. FT-IR (KBr): v = 2916, 1689, 1618, 1537, 1473, 1354, 1193, 1147, 1074, 977, 906, 856, 827, 765, 698, 561, 447, 414, cm^{-1}. $C_{36}H_{27}ClN_2O$ m/z -539.07 [M+], found 539.15 [M+].

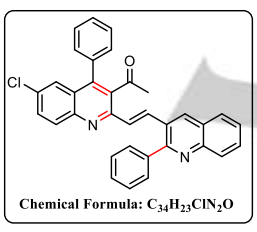
(E)-1-(6-chloro-2-(2-(6,7-dimethyl-2-phenylquinolin-3-yl)vinyl)-4-phenylquinolin-3-yl)ethan-1-one (10e) Yellow solid, Yield-94% Mp:210-212 °C- ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (s, 1H), 8.18 (d, *J* = 15.4 Hz, 1H), 7.98 (d, *J* = 9.0 Hz, 1H), 7.80 (s, 1H), 7.73 (d, *J* = 7.3 Hz, 3H), 7.63 (d, *J* = 8.9 Hz, 1H), 7.56 – 7.42 (m, 4H), 7.36 (dd, *J* = 7.9, 4.9 Hz, 3H), 7.25 (d, *J* = 7.9 Hz, 2H), 7.15 (d, *J* = 15.4 Hz, 1H), 2.74 (s, 3H), 2.53 (s, 3H), 2.03 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 205.4, 158.6, 150.3, 148.3, 146.3, 143.6, 140.2, 140.2, 135.7, 135.0, 134.4, 134.3, 132.8, 131.3, 131.2, 131.1, 129.9, 129.9, 129.7, 129.2, 128.9, 128.5, 128.4, 127.8, 126.7, 126.3, 126.1, 124.9, 124.6, 32.7, 22.0, 18.6. FT-IR (KBr): v = 3269, 2916, 2848, 1730, 1689, 1618, 1537, 1473, 1379, 1352, 1303, 1178, 1124, 1074, 975, 908, 856, 827, 767, 698, 651, 588, 542, 503, 451 cm^{-1}. $C_{36}H_{27}ClN_2O$ m/z -539.07 [M$^+$], found 539.15 [M$^+$].



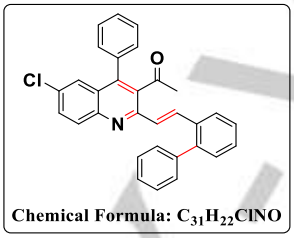
(E)-1-(6-chloro-2-(2-(5,7-dimethyl-2-phenylquinolin-3-yl)vinyl)-4-phenylquinolin-3-yl)ethan-1-one (10f) Yellow solid, Yield-94% Mp:203-205 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.54 (s, 1H), 8.18 (d, *J* = 15.4 Hz, 1H), 7.98 (d, *J* = 9.0 Hz, 1H), 7.80 (s, 1H), 7.76 – 7.68 (m, 2H), 7.63 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.51 (ddd, *J* = 20.2, 9.6, 5.6 Hz, 6H), 7.35 (dd, *J* = 6.5, 2.9 Hz, 2H), 7.25 (d, *J* = 8.1 Hz, 2H), 7.14 (d, *J* = 15.4 Hz, 1H), 2.74 (s, 3H), 2.53 (s, 3H), 2.03 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 205.3, 158.6, 150.3, 148.3, 146.3, 143.6, 140.2, 135.7, 134.5, 134.3, 132.8, 131.3, 131.2, 131.1, 129.9, 129.9, 129.7, 129.2, 128.9, 128.5, 128.3, 127.8, 126.7, 126.3, 126.2, 124.8, 32.6, 21.9, 18.6. FT-IR (KBr): v = 2945, 1689, 1616, 1537, 1475, 1381, 1313, 1193, 1147, 1074, 966, 910, 856, 829, 767, 700, 588, 505 cm^{-1}. $C_{36}H_{27}ClN_2O$ m/z -539.07 [M$^+$], found 540.20 [M+1]$^+$.



(E)-1-(6-chloro-4-phenyl-2-(2-(2-phenylquinolin-3-yl)vinyl)quinolin-3-yl)ethan-1-one (10g) light yellow solid, Yield-80% Mp:221-223 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.48 (s, 1H), 8.22 (d, *J* = 15.1 Hz, 1H), 8.16 (d, *J* = 8.5 Hz, 1H), 7.97 (d, *J* = 9.0 Hz, 1H), 7.92 (d, *J* = 7.9 Hz, 1H), 7.78 – 7.68 (m, 4H), 7.63 (dd, *J* = 9.0, 2.3 Hz, 1H), 7.55 (d, *J* = 2.6 Hz, 5H), 7.41 – 7.34 (m, 3H), 7.27 – 7.17 (m, 2H), 2.06 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 205.5, 159.2, 150.0, 147.7, 146.3, 143.7, 140.0, 135.4, 135.0, 134.4, 134.4, 132.9, 131.3, 131.3, 130.1, 130.0, 129.4, 129.3, 129.1, 129.0, 128.6, 128.4, 128.0, 127.8, 127.2, 126.9, 126.4, 126.3, 124.8, 32.7 ppm. FT-IR (KBr): v = 3439, 3059, 1687, 1608, 1537, 1475, 1388, 1348, 1311, 1193, 1143, 1076, 1012, 972, 906, 831, 767, 698, 644, 588, 507, 464 cm^{-1}. $C_{34}H_{23}ClN_2O$ m/z -511.02 [M$^+$], found 511.11 [M$^+$].



(E)-1-(2-(2-([1,1'-biphenyl]-4-yl)vinyl)-6-chloro-4-phenylquinolin-3-yl)ethan-1-one(10h) light yellow solid, Yield-82% Mp:176-178 °C, ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.06 (d, *J* = 15.5 Hz, 1H), 7.93 (d, *J* = 9.0 Hz, 1H), 7.79 – 7.73 (m, 1H), 7.62 – 7.56 (m, 1H), 7.55 – 7.49 (m, 4H), 7.48 – 7.36 (m, 8H), 7.33 (dd, *J* = 6.5, 2.9 Hz, 2H), 7.07 (d, J = 15.5 Hz, 1H), 2.02 (s, 3H). ^{13}C NMR (101 MHz, $CDCl_3$) δ 205.3, 150.6, 146.3, 143.4, 142.2, 140.7, 136.3, 135.1, 134.5, 134.5, 132.5, 131.3, 131.0, 130.5, 130.0, 129.9, 129.1, 128.9, 128.7, 128.2, 127.6, 127.2, 126.9, 126.24, 125.1, 124.8, 32.6 ppm. FT-IR (KBr): v = 3062, 1699, 1625, 1539, 1471, 1440, 1390, 1352, 1301, 1193, 1143, 1076, 974, 881, 825, 756, 702, 669, 615, 540, 495, 451 cm^{-1}. $C_{31}H_{22}ClNO$ m/z – 459.97 [M$^+$], found 459.07 [M$^+$].



Gram-scale synthesis (E)-4-benzylidene-7-chloro-9-phenyl-3,4-dihydroacridin-1(2H)-one(4aa) its synthetic utility towards (E)-4-benzylidene-7-chloro-9-phenyl-1,2,3,4-tetrahydroacridin-1-ol (4aa`)

The gram-scale reaction performed with 2-amino-5-chloro benzhydrol, **1** (4.2 mmol), benzyl alcohol, **2** (4.0 mmol) 10% mol of each $[IrCl(COD)]_2$ and 1,10 phenanthroline, continued heating at 90 °C in K_2CO_3: ethylene glycol (400 mg:400 mg) for 20 min, After dehydrogenation, added cyclohexane-1,3-dione, **3** (4.2 mmol), DMU: Tartaric acid (560 mg : 240 mg), continued heating for 2h, monitored by TLC, purified with methanol to obtained pure **4aa** (87%).

Later. **4aa** (3.0 mmol) was dissolved in methanol, and small portions of $NaBH_4$ (10.0 mmol) added, stirred for 15 min at room temperature. Monitored the reaction by TLC, after completion of the reaction, diluted the reaction mixture with water, extracted into CH_2Cl_2, the solvent evaporated to obtain the crude product. Purified compound **4aa`**, by column chromatography using ethyl acetate and petroleum ether as a solvent system (EtOAc petroleum ether/0.5:9.5 v/v).

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Keywords: *Deep eutectic solvents • Dehydrogenation • Friendlander annulation • sp3 C-H functionalization • Suzuki-mayura coupling*

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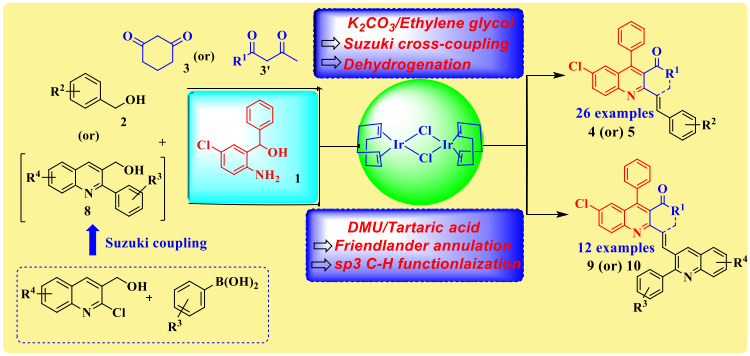

Sequential Suzuki-Miyaura/Dehydrogenative Friedlander reaction/sp^3 C-H activation

A diiridium(I) -catalysed DESs mediated three (or) four component one pot assembly of (*E*)-4-benzylidenylacridin-1(2H)-ones, (**4** or **9**) (*E*)-2-styryl quinoline-3-carboxamides (**5** or **10**) is described. The method involve the consecutive three Carbon-Carbon and one carbon- nitrogen bond formation in single reaction vessel. Moreover,Gram scale synthesis and detailed mechanistic study have been established