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FULL PAPER

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Pd-NHC catalysed Carbonylative Suzuki coupling reaction and its application towards the synthesis of biologically active 3-aroylquinolin-4 (1*H*)-one and acridone scaffolds

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We have unfolded a convenient and mild protocol for the synthesis of diaryl ketones *via* Pd- NHC catalysed carbonylative Suzuki coupling reaction. Notably, this method offers advantages like no use of toxic CO gas, shorter reaction time, high yield, and broad substrate scope. Several sensitive functional groups (like-COMe, -COOMe, -F, -Cl, -Br, -NH_2, -CN) are well tolerated in this reaction. In addition, we have also demonstrated a new efficient route for the synthesis of biologically active and pharmaceutically important 2-substituted 3-Aroylquinolin-4(1*H*)-ones and acridone scaffolds.

KEYWORDS
3-aroyl quinolin-4(1*H*)one, Acridone, Carbonylative Suzuki coupling, Diaryl ketones, Pd-NHC

1 | INTRODUCTION

Numerous photosensitizers, natural products, advanced organic materials and pharmaceutically apposite agents are comprised of diaryl ketone units.[1] Some of the top-selling drugs, for instance, Evista,[2] Tricor[3] and Sector[4] contain diaryl ketone core unit. On other hand, heterocycles are important structural motifs found in many natural products, functional molecules and bioactive compounds.[5a] Thus, the hybrid compounds, comprising of both diaryl ketones and heterocyles, are of the interest in medicinal chemistry. Few reported compounds containing such hybrid framework are represented in Figure 1.[5b, c] Among them, chromone derivatives, especially 4-quinolones and acridones, comprised a large segment of the biologically active compounds reported.[6] Recently, Botta and Goggiamani unfolded the synthesis of 2-Substituted 3-Aroylquinolin-4(1*H*)-ones *via* Pd-catalysed carbonylative cyclization of N-(2-iodoaryl) enaminones.[7] The 2-Substituted 3-Aroylquinolin-4(1*H*)-one derivatives has proved itself as inhibitor of Hedgehog (Hh) signaling pathway in vitro and suppress the growth of MB cells which opened a new avenue for the discovery of anticancer lead compounds. Development of newer methodologies for the synthesis of diaryl ketone motifs is of great importance in view of medicinal chemistry.

Traditionally, diaryl ketones are easily prepared by Friedel-Crafts acylations, acylative Suzuki couplings, and nucleophilic additions of an organometallic to a carbonyl moiety. In 1997, Bumagin *et al*. synthesized the aryl ketones *via* Pd-catalysed cross-coupling reactions of acid chlorides.[8] Afterwards, this methodology was extended towards the acylative Suzuki coupling with esters[9], anhydrides[10] and carboxylic acid in the presence of an activating agents.[10c, 11] Liebeskind and Srogl used catalytic amount of Pd(0) salt and stoichiometric amount of expensive Cu(I)TC [Copper(I)-thiophene-2-carboxylate to synthesize the diaryl ketones *via* thiolester–boronic acid coupling.[12a–d] Very recently, Das *et al*. reported a method for the synthesis of diaryl ketones *via* thiolester-boronic acid coupling in the presence of Cu and Ag salts.[12e]

However, the traditional transition metal catalysed carbonylative cross-coupling reactions between aryl electrophiles, carbon monoxide and organometallic reagent has been substantiated to be the more convenient route for the synthesis of wide range of diaryl ketones. Several aryl metal reagents such as silicon,[13] aluminium,[14] tin[15] and magnesium[16] were frequently used for this

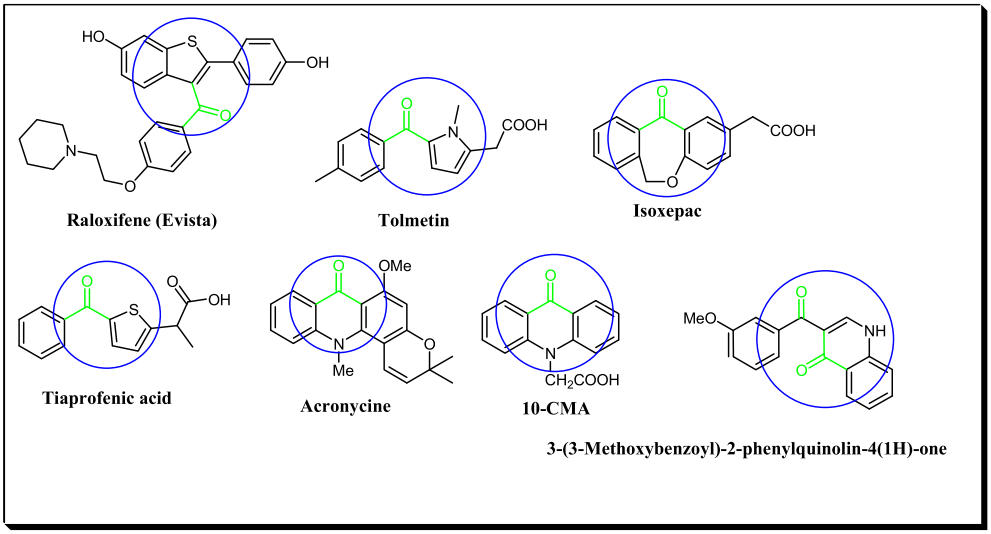


FIGURE 1 Some biologically active Diaryl ketone compounds

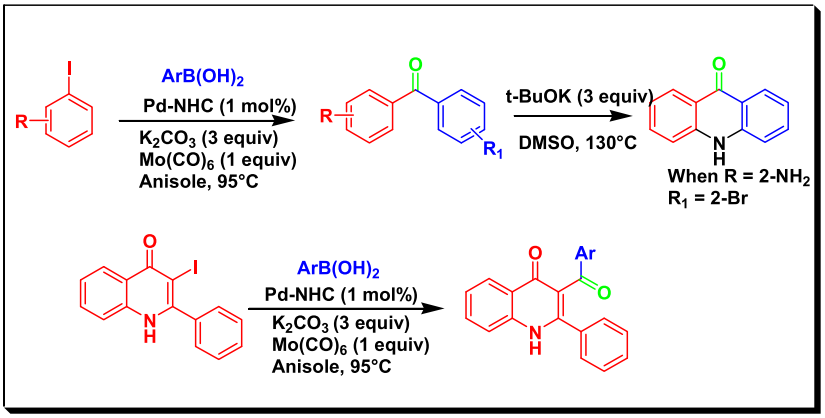
case. The main disadvantage of this carbonylative cross-coupling reaction is the formation diaryl by products, especially in the case of electron deficient aryl halides.[17] Suzuki *et al*. introduced a facile protocol for synthesizing the diaryl ketones from arylboronic acid, carbon monoxide and aryl halides in presence of palladium catalyst.[18] Though this method was quite effective due to the versatile nature of boronic acid as it is non-toxic, bench-stable and easily available, yet the essential involvement of toxic CO gas rendered the method narrowed down its scope. After that, several research groups studied the same reaction using different palladium catalyst such as $PdCl_2(PPh_3)_2$,[19] $PdCl_2(PPh_3)_2$/ $PdCl_2$(dppf),[20] $Pd(OAc)_2$-imidazolium salts,[21] $Pd(OAc)_2$/ N,N-bis(2,6-diisopropylphenyl)dihydroimidazolium chloride,[22] an MCM-41-supported bidentate phosphane palladium complex,[23] $Pd(OAc)_2$/di-1-adamantyl-n-butylphosphane[24] and Pd/thiourea.[25] In spite of their huge potentiallity, those methods suffered from several disadvantages, like use of air/moisture sensitive and expensive phosphane donating ligands, longer reaction time and requirement of high pressure equipment. Very recently, Bhanage and Han *et al*. reported the Suzuki carbonylation of aryl and heterocyclic halides using palladacyle complex, KCC-1 supported palladium nanoparticles,[26] iron and nickel nano particle[27] where carbon monoxide gas used as carbonyl source. In these protocols, high pressure of CO gas is essentially required not only to get the good yield of desired ketones but also to reduce the formation of byproducts. A search in the literature revealed that several compounds like formic acid,[28] formates,[29] chloroform,[30] aldehydes[31] and other CO surrogates[32] have been used as CO source. In addition, few metal carbonyls were also used as CO source.[33] Above all, the incorporation of carbon monoxide *via* metal carbonyl appears much appealing and effective than any other methods due to easy handling and *in-situ* generation of CO upon heating.

Previously, Jafarpour *et al*. reported the carbonylative Suzuki coupling of aryl halides and arylboronic acid in presence of $Mo(CO)_6$ and Palladium acetate.[34] However, the method requires high loading of Pd catalyst (10 mol%), longer reaction time and high temperature. As a part of our ongoing effort towards develop the simple protocol for the formation of C-C bond, herein we report a convenient methodology for carbonylative Suzuki reaction (Scheme 1) between aryl halides and arylboronic acids in presence of our previously synthesized Pd-NHC catalyst (Figure 2).[35]

Further, we apply this protocol for the synthesis of biologically active 2-substituted-3-Aroylquinolin-4(1*H*)-one and acridone scaffolds (Scheme 1).

2 | RESULTS AND DISCUSSION

To investigate the feasibility of the reaction, we commenced our journey with the carbonylative cross coupling reaction between phenylboronic acid and iodobenzene using Pd-NHC (1 mol%) as catalyst, K_2CO_3 as base in DMF. After 24 h of reaction, we obtained only 15% yield of the desired diaryl ketone (Table I; entry 1). Then, we screened solvents and found anisole as a media, served the better yields of diaryl ketone compared to other solvents like DMF, Toluene, 1,4-dioxane and THF (Table 1; entries 1-5). Combination of K_2CO_3 and anisole proved to be best suited for this carbonylative Suzuki coupling (Table 1; entry 4). Most surprisingly, the reaction completed within 1 h only and resulted in 85% of the desired product. We next turned our attention to choose



SCHEME 1 Our approach for the synthesis of diaryl ketone scaffolds

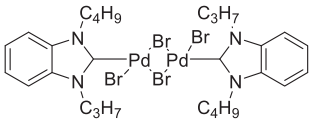


FIGURE 2 Structure of Pd-NHC catalyst

the better base other than K_2CO_3. Inferior results of desired product was obtained upon using common bases like DBU, *t*-BuOK, Cs_2CO_3 and Et_3N (Table 1; entries 6-9).

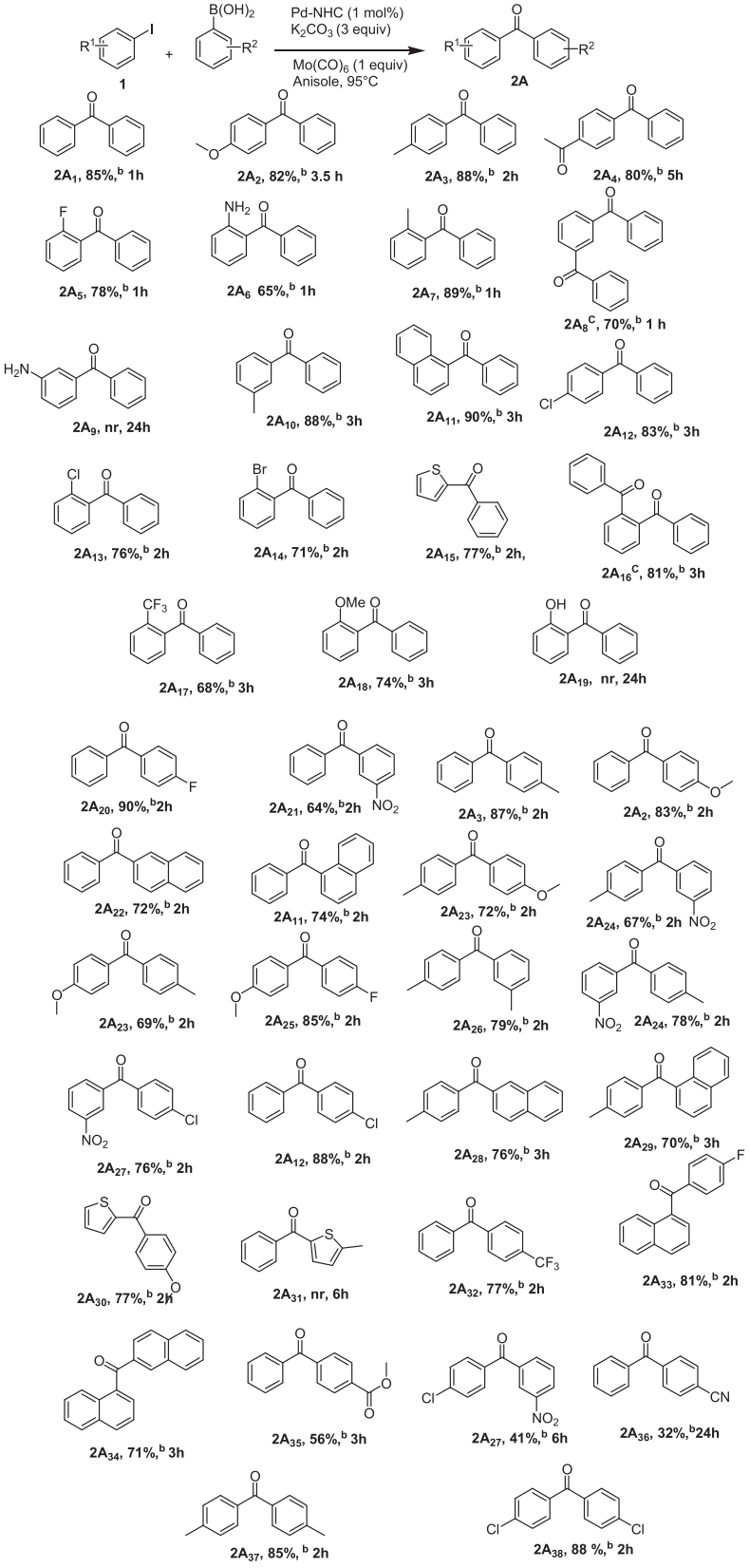
Then, we screened out the catalytic activity of different palladium catalyst (Table 1; entries 10-12) as well as reaction temperature (Table 1; entries 4, 13-15). The product obtained in moderate to good yield upon using commonly available Pd-catalysts. It was also found that 95 °C is the optimal reaction temperature (Table 1; entry 4) for this coupling reaction. At low temperature, yield of the diaryl ketone decreases might be due to the unavailability of sufficient carbon monoxide (Table 1; entry 13, 14). The same reaction at 120 °C resulted in 84% yield of product **2A$_1$** within 1 h (Table 1; entry 15). Combination of Pd-NHC (1 mol %) and K_2CO_3 (3 equiv.), in anisole at 95 °C, was found to be optimal for the carbonylative cross coupling reaction and led the product **2A$_1$** (benzophenone) in 85% yield after only 1 h.

TABLE 1 Optimization of the reaction conditions[a]

Entry	Catalyst (mol %)	Solvent	Base	Temp (° c)	Time (h)	Yield (%)[b]
1	Pd-NHC (1)	DMF	K_2CO_3	95	24	15
2	Pd-NHC (1)	THF	K_2CO_3	95	24	20
3	Pd-NHC (1)	1,4-dioxane	K_2CO_3	95	24	NR
4	**Pd-NHC (1)**	**Anisole**	**K_2CO_3**	**95**	**1**	**85**
5	Pd-NHC (1)	Toluene	K_2CO_{3s}	95	24	30
6	Pd-NHC (1)	Anisole	*t*-BuOK	95	24	10
7	Pd-NHC (1)	Anisole	DBU	95	24	NR
8	Pd-NHC (1)	Anisole	Cs_2CO_3	95	24	10
9	Pd-NHC (1)	Anisole	Et_3N	95	24	25
10	$Pd(OAc)_2$ (5)	Anisole	K_2CO_3	95	7	64
11	$PdCl_2$ (5)	Anisole	K_2CO_3	95	7	68
12	$Pd_2(dba)_3$ (5)	Anisole	K_2CO_3	95	7	73
13	Pd-NHC (1)	Anisole	K_2CO_3	80	4	65
14	Pd-NHC (1)	Anisole	K_2CO_3	60	12	52
15	Pd-NHC (1)	Anisole	K_2CO_3	120	1	84

[a]Reaction conditions: 0.25 mmol of iodobenzene, 0.375 mmol of phenylboronic acid, base (3 equiv.), $Mo(CO)_6$ (1 equiv., 66 mg), were heated under N_2 atm.

[b]Yield = isolated yields after column chromatography.



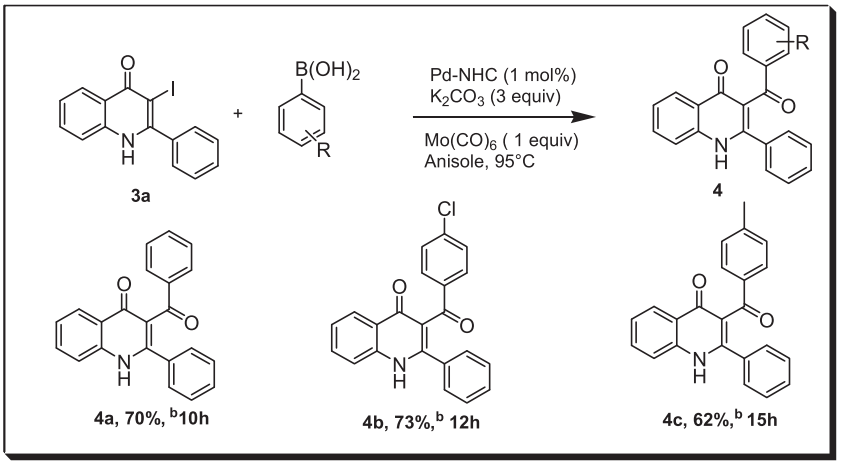
SCHEME 2 Substrate scope of various aryl iodides and (het)arylboronic acids in the carbonylative Suzuki coupling*

*Reaction condition: 0.25 mmol of iodoarenes, 0.375 mmol of boronic acids, base (0.75 mmol, 104 mg), Pd-NHC catalyst (2.5 mg, 1 mol%) and $Mo(CO)_6$ (1 equiv., 66 mg) were heated 95°C under N_2- atmosphere in anisole. [b]Isolated yield after column chromatography.

The representative results of this carbonylative Suzuki reaction between different aryliodides, phenylboronic acid and $Mo(CO)_6$ are summarized in Scheme 2. It is clear from the results that iodoarenes having electron withdrawing and electron releasing group were equally participated in the carbonylative Suzuki reaction with phenylboronic acid without any difficulties (Scheme 2; entries **$2A_1$-$2A_{19}$**). Aryl iodides containing electron donating groups like –Me, -OMe, $-NH_2$ reacted efficiently with phenylboronic acid and gave corresponding unsymmetrical ketones in high yield (Scheme 2, entries **$2A_3$**, **$2A_7$**, **$2A_{10}$**, **$2A_2$**, **$2A_{18}$**, and **$2A_6$**). On the other hand, 4-iodoacetophenone took longer time (5 h) and resulted in 80% yield of the desired product (Scheme 2, entry **$2A_4$**). This might be due to the presence of electron withdrawing group which slow down the reaction. Aspect of steric hindrance in making the diaryl ketones *via* this present protocol was also studied. Ortho substituted (-F, $-NH_2$, -Me, -Cl, -Br, $-CF_3$ and -OMe) iodoarenes afforded the corresponding diaryl ketones in moderate to good yields (Scheme 2, entries **$2A_5$**, **$2A_6$**, **$2A_7$**, **$2A_{13}$**, **$2A_{14}$**, **$2A_{17}$**, **$2A_{18}$**).

Whereas 1-naphthyl iodoarenes responded very well and gave 90% of the desired product in 1 h (Scheme 2, entry **$2A_{11}$**). We then moved to explore the possibility for making diketo compounds using our protocol. Reaction of 1,3- and 1,2-diiodo benzene with phenylboronic acid and $Mo(CO)_6$ afforded the bis couple product in 70% and 81% yield, respectively (Scheme 2; entry **$2A_8$** and **$2A_{16}$**). Isolation of the product **$2A_{16}$** in 81% yield further justified that the steric hindrance did not affect the reaction. Surprisingly, 3-iodo aniline and 2-iodophenol did not afford the corresponding diaryl ketone even after 24 h stirring (Scheme 2, entries **$2A_9$** and **$2A_{19}$**) at 95 °C. We also found the smooth participation of heterocyclic iodoarene (2-iodothiophene) in this reaction and resulted in the good yield of the corresponding product (Scheme 2, entry **$2A_{15}$**).

To study the scope of this protocol, a broad array of arylboronic acid possessing both electron donating and electron withdrawing groups were allowed to react with various iodoarenes under the optimized conditions (Scheme 2). Our protocol proved to be quite compatible with various functional groups like methoxy, chloro, fluoro, ester and cyano. 1-naphthyl and 2-naphthylboronic acid were individually coupled with iodobenzene and 4-iodotoluene and afforded the desired products with high yields (Scheme 2; entries **$2A_{22}$**, **$2A_{11}$**, **$2A_{28}$** and **$2A_{29}$**). We also observed that a heterocyclic iodoarenes (2-iodo thiophene) coupled with 4-methoxyphenyl boronic acid effectively (Scheme 2; entry **$2A_{30}$**). Importantly, 1-iodonaphthalene quite successfully underwent the carbonylative Suzuki coupling with 4-fluoro phenylboronic acid and 2-naphthylboronic acid (Scheme 2; entries **$2A_{33}$** and **$2A_{34}$**). Unfortunately, 4-cyano phenylboronic acid afforded only 32% yield of the desired diaryl ketone when it coupled with normal iodobenzene (Scheme 2; entry **$2A_{36}$**). To our delight, iodoarene possessing nitro group at meta position did not hamper the process and coupled with both electron deficient and electron rich arylboronic acid (Scheme 2; entries **$2A_{24}$**, **$2A_{27}$**). More interestingly, 4-iodotoluene coupled with *p*-tolylboronic acid excellently and furnished the



SCHEME 3 Scope of carbonylative Suzuki coupling in 4-quinolone scaffold†

†Reaction conditions: 0.25 mmol of 3-iodo substituted 4-quinolone (86 mg), 0.375 mmol of substituted arylboronic acid, base (3 equiv.), $Mo(CO)_6$ (66 mg, 1 equiv.) and Pd-NHC (2.5 mg, 1 mol%) were heated at 95°C under N_2 atmosphere in anisole. [b] Isolated yield after column chromatography.

corresponding product **2A$_{37}$** (85%) in a very short span. Similarly, we also prepared the symmetrical diaryl carbonyls of 4-chloro-iodobenzene in 88% yield (Scheme 2; entry **2A$_{38}$**).

3 | APPLICATIONS

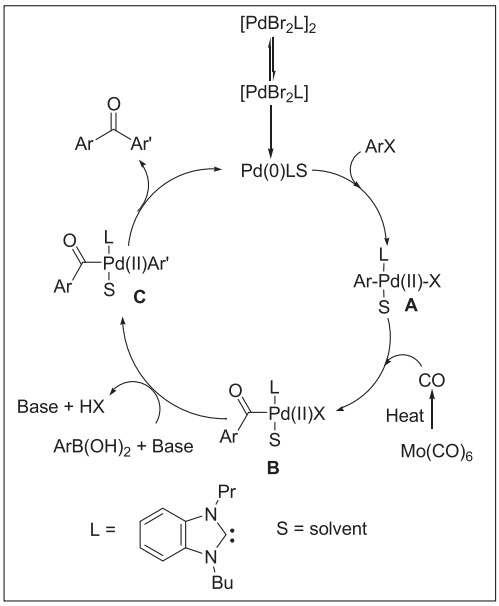
As a part of our ongoing interest towards the synthesis of functionalized 4-quinolones.[35c, 36] we further explored the scope of this coupling reaction for the preparation of biologically active 4-quinolone scaffolds. Very recently Alfonsi *et al*. reported a palladium catalysed sequential synthesis of 3-aroylquinolin-4(1*H*) one derivatives from 2-iodoaniline.[7] But the method requires the use of expensive phosphine ligand, high pressure (20 atm.) of toxic CO gas, high temperature and longer reaction time (72 h) which limit the broad application scope of the method. We took the challenge and decided to apply our protocol in synthesis of 3-aroylquinolin-4(1*H*) one derivatives (Scheme 3).

Reaction between 3-iodo-2-aryl substituted 4-quinolone and arylboronic acid at our optimized condition resulted in the good yield of desired products within 10-15 h. It has been found that both electron withdrawing and electron donating arylboronic acid effectively underwent the reaction and resulted in the desired products in 72% and 63%, respectively (Scheme 3; entries **4b** and **4c**).

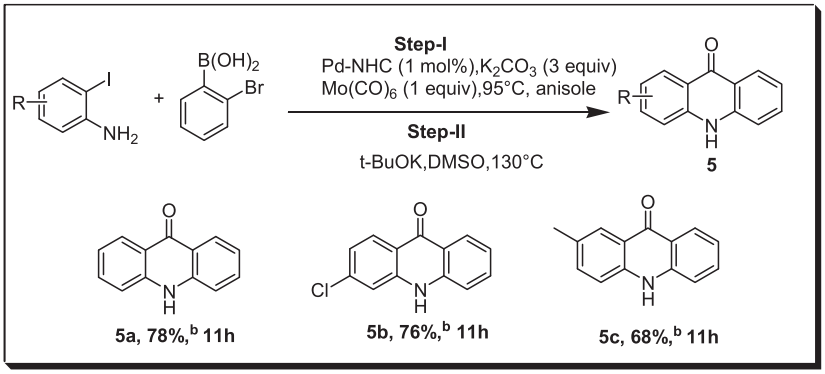
Next, we again applied our protocol in the modular synthesis of pharmaceutically important acridone scaffolds in one pot as shown in scheme 4. Acridones are widely existed in antifungal, antileishmanial, DNA-intercalating anticancer drugs[6b] and also as fluorescent labels.[37]

Substituted 2-iodo aniline and 2-bromo phenylboronic acid are selected as coupling partners in this case. Without purifying the intermediate diaryl ketones, we turned our attention to cyclize them under refluxing conditions in presence of *t*-BuOK and getting isolated the biologically active acridone moieties in excellent yields (Scheme 4, entries **5a-5c**).

A plausible reaction pathway for Carbonylative Suzuki cross coupling reactions has been shown in Scheme 5. The reaction initiated with generation of active Pd(0) species. It underwent an oxidative addition with aryl iodides to



SCHEME 5 Plausible reaction pathway for the catalytic cycles of Carbonylative Suzuki coupling reaction



SCHEME 4 Synthesis of pharmaceutically important acridone scaffolds‡

‡Reaction conditions: Step-I – 0.25 mmol of substituted 2-iodo aniline, 0.375 mmol of 2-bromo phenylboronic acid (76 mg), base (3 equiv.), $Mo(CO)_6$ (66 mg, 1 equiv.) and Pd-NHC were heated at 95°C under N_2 atmosphere in anisole for 3 h; Step-II – ^{t}BuOK (3 equiv.) reflux at 130°C in DMSO for next 8 h.
[b] Isolated yield after column chromatography.

generate an aryl palladium intermediate (A). Consequently, the aryl palladium intermediate get converted into the acylpalladium species (B) in the presence of CO which was generated *in situ* upon decomposition of $Mo(CO)_6$ under the reaction condition. Later, the complex **B** participated in transmetallation reaction with arylboronic to form new complex **C**. Finally, the desired diarylketone product is obtained after the reductive elimination with the concomitant generation of Pd (0) species, which proceeds to participate in the next catalytic cycle.

4 | CONCLUSIONS

In summary, we have developed a mild and efficient protocol for the synthesis of diaryl ketones *via* Pd-NHC catalysed carbonylative Suzuki coupling reaction between the commercially available iodoarenes and arylboronic acids in the presence of $Mo(CO)_6$. This new strategy offers several advantages such as, non-requirement of toxic CO gas, shorter reaction time, good to excellent yield of the desired products and broad substrate scope. Most importantly, this protocol requires only 1 mol% of palladium catalyst which is much lower than the previously reported methods. Noteworthy, in this protocol various sensitive functional groups are found well tolerated (-COMe, -COOMe, $-NH_2$, -CN, -Br, -F, -Cl). The present methodology could be applied for the synthesis of biologically active 2- phenyl substituted-3-Aroylquinolin-4(1*H*)-ones and acridone motifs.

5 | EXPERIMENTAL SECTION

5.1 | General considerations

Unless stated otherwise, all reagents such as various Iodoarenes, aryl(Het)boronic acid, K_2CO_3, Anisole, $Mo(CO)_6$ and solvents were used as received from commercial suppliers. NMR spectra were recorded on 300 MHz spectrometer at 298 K with calibration done on the basis of solvent residual peak. Products were purified using column chromatography on silica gel (60-120 mesh). Ethyl acetate and petroleum ether (60-80 °C) were used as eluents. Progress of reaction was monitored using silica gel TLC.

5.2 | Preparation of Biaryl ketones from the Carbonylative Suzuki coupling of various iodoarenes and aryl(het)boronic acids ($2A_1$-$2A_{38}$)

Initially, various iodoarenes (0.25 mmol), aryl(Het) boronic acid (0.375 mmol), K_2CO_3 (0.75 mmol, 104 mg), $Mo(CO)_6$ (0.25 mmol, 66 mg), Pd-NHC (1 mol%, 2.5 mg) and anisole (2 ml) were taken in a sealed tube under N_2 atmosphere and heated at 95 °C. The reaction was continued for 1 h to several hours for completion of the reaction. After monitoring the TLC analysis, the reaction mixture was diluted with 30 ml water and the organic layer was extracted with DCM (30 ml). Then, it was dried over anhydrous sodium sulphate and concentrated under reduced pressure. The crude residue was purified through column chromatography using petroleum ether and ethyl acetate as an eluent.

5.3 | Preparation of 3-iodo-2- phenyl substituted 4-quinolones (3a)

Initially, 2-aryl quinolin-4(1*H*)-one (0.25 mmol), iodine (2 equiv.) and sodium carbonate (1.5 equiv.) in THF (2 ml) was stirred at room temperature for 18 hours. Then, the reaction mixture was quenched with sodium thiosulphate and the precipitate was collected by filtration and washed with ice-cold water. Afterwards, the crude product was purified through column chromatography.

5.4 | Preparation of 2-phenyl substituted-3-aroylquinolin-4(1H)-ones from the carbonylative Suzuki coupling of 3a (4a-4c)

Initially, 3-iodo-4-quinolone derivatives (0.25 mmol, 86 mg), arylboronic acid (0.375 mmol), K_2CO_3 (0.75 mmol, 104 mg), $Mo(CO)_6$ (0.25 mmol, 66 mg), Pd-NHC (1 mol%, 2.5 mg) and anisole (2 ml) were taken in a sealed tube under N_2 atmosphere and heated at 95 °C. The reaction was continued for 10 h to 15 h for completion of the reaction. After monitoring the TLC analysis, the reaction mixture was diluted with 30 ml water and the organic layer was extracted with ethyl acetate (30 ml). Then, it was dried over anhydrous sodium sulphate and concentrated under reduced pressure. The crude residue was purified through column chromatography using petroleum ether and ethyl acetate as an eluents.

5.5 | Preparation of pharmaceutically important acridone scaffolds (5a-5c)

Primarily, substituted 2-iodo aniline (0.25 mmol), 2-bromophenylboronic acid (0.375 mmol, 76 mg), K_2CO_3 (0.75 mmol, 104 mg), $Mo(CO)_6$ (0.25 mmol, 66 mg), Pd-NHC (1 mol%, 2.5 mg) and anisole (2 ml) were taken in a sealed tube under N_2 atmosphere and heated at 95 °C. The reaction was continued for 3 h for completion of the reaction. After monitoring the TLC analysis, the reaction mixture was diluted with 30 ml water and the organic layer was extracted with DCM (30 ml). Then, it was dried

over anhydrous sodium sulphate and concentrated under reduced pressure. In the next step, the crude reaction mixture was refluxed in DMSO (5 ml) at 130 °C under basic condition (*t*-BuOK, 3 equiv.) for 8 h. After monitoring the TLC analysis, the reaction mixture was diluted with 30 ml water and the organic layer was extracted with ethyl acetate (30 ml). Then, it was dried over anhydrous sodium sulphate and concentrated under reduced pressure. The crude residue was purified through column chromatography using petroleum ether and ethyl acetate as an eluent.

5.6 | Physical characteristics and spectral data of compounds

Benzophenone (**2A$_1$**)[12e]: White solid, Yield = (85%, 39 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.47-7.53 (m, 4H), 7.58-7.64 (m, 2H), 7.80-7.84 (m, 4H); 13C NMR ($CDCl_3$, 75 MHz) δ 128.3, 130.1, 132.4, 137.6, 196.8.

(4-methoxy phenyl) phenyl methanone (**2A$_2$**)[12e]: White solid, Yield = (82%, 43.5 mg and 83%, 44 mg),^{1}H NMR ($CDCl_3$, 300 MHz) δ 3.89 (s, 3H), 6.95-6.98 (m, 2H), 7.44-7.49 (m, 2H), 7.53-7.59 (m, 1H), 7.73-7.76 (m, 2H), 7.77-7.84 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 55.5, 113.6, 128.2, 129.7, 130.2, 131.9, 132.5, 138.3, 163.2, 195.6.

Phenyl (p-tolyl)methanone (**2A$_3$**)[12e]: White solid, Yield = (88%, 43 mg and 87%, 42.6 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 2.44 (s, 3H), 7.26-7.30 (m, 2H), 7.44-7.50 (m, 2H), 7.54-7.60 (m, 1H), 7.70-7.74 (m, 2H),7.76-7.80 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 21.6, 128.3, 129.0, 129.9, 130.3, 132.1, 134.9, 138.0, 143.2, 196.5.

1-(4-Benzoylphenyl)ethan-1-one (**2A$_4$**)[28]: Yellow oil, Yield = (80%, 44.8 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 2.69 (s, 3H), 7.50-7.55 (m, 2H), 7.62-7.65 (m, 1H), 7.81-7.84 (m, 2H), 7.87-7.90 (m, 2H), 8.07-8.09 (m, 2H),^{13}C NMR ($CDCl_3$, 75 MHz) δ 26.8, 128.1, 128.4, 130.0, 130.1, 132.9, 136.9, 139.6, 141.4, 195.9, 197.5.

(2-fluoro phenyl)(phenyl)methanone (**2A$_5$**)[38]: Colourless liquid, Yield = (78%, 39.0 mg), ^{1}H NMR (DMSO-d_6, 300 MHz) δ 7.36-7.42 (m, 2H), 7.55-7.60 (m, 3H), 7.69-7.71 (m, 2H), 7.75-7.78 (m, 2H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 116.6, 116.9, 125.3, 125.3, 126.8, 127.0, 129.3, 129.8, 130.8, 130.9, 134.0, 134.1, 134.3, 137.1, 158.0, 161.3, 193.2.

(2-aminophenyl)(phenyl)methanone (**2A$_6$**)[39]: Yellow solid, Yield = (65%, 32.0 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 6.00 (bs, 2H), 6.57-6.72 (m, 1H), 6.73-6.75 (m, 1H), 7.26-7.32 (m, 1H), 7.43-7.55 (m, 4H), 7.62-7.65 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 115.5, 117.0, 118.2, 128.1, 129.1, 131.1, 134.2, 134.6, 140.1, 150.9, 199.1.

Phenyl(o-tolyl)methanone (**2A$_7$**)[12e]: Colourless liquid, Yield = (89%, 43.6 mg), ^{1}H NMR (DMSO-d_6, 300 MHz) δ 2.23 (s, 3H), 7.30-7.39 (m, 3H), 7.55-7.57 (m, 3H), 7.69-7.72 (m, 3H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 19.9, 125.9, 128.5, 129.3, 130.1, 130.8, 131.4, 134.0, 136.2, 137.5, 138.8, 198.2.

1,3-Phenylenebis(phenylmethanone) (**2A$_8$**)[40a]: White solid, Yield = (70%, 50.0 mg), ^{1}H NMR (DMSO-d_6, 300 MHz) δ 7.57-7.62 (m, 1H), 7.68-7.70 (m, 3H), 7.79-7.82 (m, 3H), 7.90-8.00 (m, 4H), 8.00-8.04 (m, 1H), 8.05-8.08 (m, 2H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 129.1, 129.7, 130.2, 131.2, 133.5, 133.9, 136.9, 137.5, 195.5.

Phenyl (m-tolyl)methanone (**2A$_{10}$**)[12e]: Colourless liquid, Yield = (88%, 43.1 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 2.43 (s, 3H), 7.37-7.40 (m 2H), 7.49-7.51 (m, 2H), 7.57-7.64 (m, 3H), 7.79-7.82 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 20.8, 126.8, 127.6, 127.7, 129.5, 130.0, 131.8, 132.6, 137.2, 137.3, 137.6, 196.4.

(Naphthalen-2-yl)(phenyl)methanone (**2A$_{11}$**)[12e]: Colourless oil, Yield = (90%, 52.2 mg and 74%, 40.7 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.45-7.64 (m, 7H), 7.88-7.97 (m, 3H), 8.01-8.03 (m, 1H), 8.10-8.14 (m, 1H); ^{13}C NMR ($CDCl_3$, 75 MHz) δ 124.4, 125.7, 126.5, 127.3, 127.97, 128.4, 128.51, 130.5, 131.0, 131.4, 133.3, 133.7, 138.3, 198.1.

(4-chloro phenyl)(phenyl) methanone (**2A$_{12}$**)[12e]: White solid, Yield = (83%, 45.0 mg and 88%, 47.5 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.44-7.52 (m, 4H), 7.58-7.63 (m, 1H), 7.74-7.79 (m, 4H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 128.4, 128.6, 129.9, 131.5, 132.6, 135.9, 137.2, 138.9, 195.2.

(2-chlorophenyl)phenyl methanone (**2A$_{13}$**)[41]: Colourless oil, Yield = (76%, 41.2 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.38-7.44 (m, 2H), 7.46-7.51 (m, 4H), 7.60-7.65 (t, J = 7.5 Hz, 1H), 7.83-7.85 (d, J = 7.8 Hz, 2H),^{13}C NMR ($CDCl_3$, 75 MHz) δ 126.7, 128.6, 129.1, 130.1, 130.2, 131.1, 131.3, 133.7, 136.4, 138.5, 195.5.

(2-bromophenyl)(phenyl)methanone (**2A$_{14}$**)[41]: Yellow solid, Yield = (71%, 46.3 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.33-7.50 (m, 5H), 7.58-7.66 (m, 2H), 7.80-7.83 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 119.5, 127.2, 128.6, 129.0, 130.2, 131.2, 133.2, 133.8, 136.1, 140.7, 195.9.

Phenyl(thiophen-2-yl)methanone (**2A$_{15}$**)[38]: White solid, Yield = (77%, 36.2 mg), ^{1}H NMR (DMSO-d_6, 300 MHz) δ 7.28-7.31 (m, 1H), 7.55-7.60 (m, 2H), 7.66-7.73 (m, 2H), 7.82-7.85 (m, 2H), 8.13 (dd, J = 4.8 Hz, 0.9 Hz, 1H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 129.1, 129.3, 129.3, 133.0, 136.1, 136.2, 137.9, 143.2, 187.8.

1,2-Phenylenebis(phenylmethanone) (**2A$_{16}$**)[42]: White solid, Yield = (81%, 58.0 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.35-7.41 (m, 4H), 7.49-7.55 (m, 2H), 7.63-7.65 (m, 4H), 7.69-7.72 (m, 4H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 128.3, 129.7, 129.8, 130.4, 133.0, 137.2, 140.0, 196.6.

(2-(trifluoromethyl)phenyl)(phenyl) methanone (**2A$_{17}$**)[43]: Colourless liquid, Yield = (68%, 42.5 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.38-7.41 (m, 1H), 7.44-7.49 (m, 2H), 7.58-7.64 (m, 3H), 7.77-7.80 (m, 3H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 121.7, 125.4,

126.5, 126.6, 126.7, 126.8, 128.0, 128.1, 128.5, 129.8, 130.2, 131.3, 133.8, 136.4, 138.4, 195.5.

(2-methoxyphenyl)(phenyl) methanone (**2A$_{18}$**)[43]: Colourless liquid, Yield = (74%, 39.2 mg), ^{1}H NMR (DMSO-d$_6$, 300 MHz) δ 3.67 (s, 3H), 7.09 (dt, *J* = 7.5 Hz, 0.9 Hz, 1H), 7.17-7.20 (m, 1H), 7.32 (dd, *J* = 7.5 Hz, 0.9 Hz, 1H), 7.49-7.57 (m, 3H), 7.62-7.71 (m, 3H), ^{13}C NMR (DMSO-d$_6$, 75 MHz) δ 56.0, 112.4, 121.0, 128.8, 129.1, 129.2, 129.7, 132.5, 133.8, 137.5, 157.1, 196.2.

4-Fluoro phenyl) phenyl methanone (**2A$_{20}$**)[41]: Yellow oil, Yield = (90%, 45.0 mg),^{1}H NMR (CDCl$_3$, 300 MHz) δ 7.13-7.20 (m, 2H), 7.46-7.51 (m, 2H), 7.57-7.62 (m, 1H), 7.75-7.79 (m, 2H), 7.82-7.87 (m, 2H),^{13}C NMR (CDCl3, 75 MHz) δ 115.3, 115.6, 127.5, 128.3, 128.7, 129.2, 129.8, 132.4, 132.6, 132.7, 133.8, 137.5, 163.7, 167.1, 195.2.

(3-nitrophenyl)(phenyl)methanone (**2A$_{21}$**)[44]: Yellow solid, Yield = (64%, 36.3 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 7.53-7.57 (m, 2H), 7.64-7.74 (m, 2H), 7.79-7.83 (m, 2H), 8.15 (td, *J* = 3.0 Hz, 1.2 Hz, 1H), 8.45 (qd, *J* = 2.4 Hz, 1.2 Hz, 1H), 8.62 (t, *J* = 1.8 Hz, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 124.3, 126.3, 128.3, 129.2, 129.6, 133.0, 135.0, 135.9, 138.7, 147.7, 193.7.

(Naphthalen-3-yl)(phenyl) methanone (**2A$_{22}$**)[12e]: White solid, Yield = (72%, 41.8 mg),^{1}H NMR (CDCl$_3$, 300 MHz) δ 7.41-7.60 (m, 7H), 7.84-7.92 (m, 3H), 7.97-7.99 (m, 1H), 8.07-8.10 (m, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 124.4, 125.7, 126.5, 127.3, 127.8, 128.4, 128.5, 130.4, 131.0, 131.3, 133.3, 133.7, 136.4, 138.3, 198.1.

4-methoxy phenyl(*p*-tolyl) methanone (**2A$_{23}$**)[12e]: White solid, Yield = (72%, 40.7 mg and 69%, 39.0 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 2.43 (s, 3H), 3.88 (s, 3H), 6.95 (dt, *J* = 4.8 Hz, 2.7 Hz, 2H), 7.26-7.29 (m, 2H), 7.67 (dd, *J* = 8.1 Hz, 1.8 Hz, 2H), 7.78-7.82 (m, 2H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 21.6, 55.5, 113.5, 128.9, 130.0, 130.5, 132.5, 135.5, 142.6, 163.0, 195.4.

(3-nitrophenyl)(*p*-tolyl)methanone (**2A$_{24}$**)[43]: Yellow solid, Yield = (67%, 40.3 mg and 78%, 47.0 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 2.47 (s, 3H), 7.32-7.34 (m, 2H), 7.67-7.73 (m, 3H), 8.12 (td, *J* = 2.7 Hz, 1.5 Hz, 1H), 8.43 (qd, *J* = 3.6 Hz, 1.2 Hz, 1H), 8.60 (t, *J* = 1.8 Hz, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 21.7, 124.6, 126.4, 129.4, 129.5, 130.2, 133.6, 135.3, 139.5, 144.4, 148.1, 193.8.

(4-fluorophenyl)(4-methoxyphenyl)methanone (**2A$_{25}$**)[46]: White solid, Yield = (85%, 48.9 mg), ^{1}H NMR (DMSO-d$_6$, 300 MHz) δ 3.86 (s, 3H), 7.08-7.11 (m, 2H), 7.38 (dt, *J* = 6.9 Hz, 1.8 Hz, 2H), 7.72-7.79 (m, 2H); ^{13}C NMR (DMSO-d$_6$, 75 MHz) δ 56.0, 114.4, 115.8, 116.1, 129.7, 132.6, 132.7, 134.6, 134.7, 163.1, 163.4, 166.4, 193.5.

m-tolyl(p-tolyl) methanone (**2A$_{26}$**)[47]: White solid, Yield = (79%, 41.5 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 2.41 (s, 3H), 2.43 (s, 3H), 7.25-7.38 (m, 4H), 7.53-7.60 (m, 2H), 7.70-7.73 (m, 2H); ^{13}C NMR (CDCl$_3$, 75 MHz) δ 21.4, 21.6, 127.2, 128.0, 128.9, 130.3, 130.3, 132.9, 135.1, 138.0, 138.1, 143.1, 196.7.

(4-chloro phenyl)(m-nitro) methanone (**2A$_{27}$**)[48]: Yellow solid, Yield = (76%, 49.6 mg and 41%, 26.8 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 7.51 (td, *J* = 4.2 Hz, 2,1 Hz, 2H), 7.70-7.78 (m, 3H), 8.12 (td, *J* = 3.0 Hz, 1.5 Hz, 1H), 8.46 (qd, *J* = 2.4 Hz, 1.2 Hz, 1H), 8.59 (t, *J* = 1.8 Hz, 1H),^{13}C NMR (CDCl$_3$, 75 MHz) δ 124.6, 126.9, 129.1, 129.4, 129.8, 131.4, 134.5, 135.3, 138.7, 140.0, 148.1, 193.0.

(Naphthalen-2-yl)(p-tolyl)methanone (**2A$_{28}$**)[49]: White solid, Yield = (76%, 46.7 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 2.47 (s, 3H), 7.32 (d, *J* = 8.1 Hz, 2H), 7.54-7.63 (m, 2H), 7.78 (dd, *J* = 1.8 Hz, 2H), 7.90-7.93 (m, 4H), 8.25 (s, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 21.7, 125.9,126.7, 127.8, 128.1, 128.2, 129.1, 129.4, 130.4, 131.5, 132.4, 135.2, 135.3, 143.2, 196.5.

(Naphthalen-1-yl)(*p*-tolyl)methanone (**2A$_{29}$**)[12e, 40]: White solid, Yield = (70%, 43.0 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 2.41 (s, 3H), 7.22-7.25 (m, 2H), 7.44-7.56 (m, 4H), 7.77 (d, *J* = 8.1 Hz, 2H), 7.90 (dd, *J* = 7.5 Hz, 2.4 Hz, 1H), 7.96-7.98 (m, 1H), 8.02-8.06 (m, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 21.8, 124.4, 125.7, 126.4, 127.1, 127.4, 128.4, 129.2, 130.6, 130.9, 131.0, 133.7, 135.7, 136.8, 144.2, 197.8.

(4-methoxyphenyl)(thiophen-2-yl)methanone (**2A$_{30}$**)[27]: White solid, Yield = (77%, 41.9 mg),^{1}H NMR (CDCl$_3$, 300 MHz) δ 3.89 (s, 3H), 6.99 (dd, J = 4.8 Hz, 2.1 Hz, 2H), 7.14-7.17 (m, 1H), 7.64-7.70 (m, 2H), 7.90 (dd, J = 4.8 Hz, 2.1 Hz, 2H); ^{13}C NMR (CDCl$_3$, 75 MHz) δ 55.5, 113.7, 127.8, 130.7, 131.6, 133.4, 134.0, 143.8, 163.1, 186.9.

4-trifluoromethyl-benzophenone (**2A$_{32}$**)[41]: White solid, Yield = (77%, 48.1 mg), ^{1}H NMR (DMSO-d$_6$, 300 MHz) δ 7.56-7.62 (m, 2H), 7.70-7.80 (m, 3H), 7.93-7.96 (m, 4H), ^{13}C NMR (DMSO-d$_6$, 75 MHz) δ 125.4, 125.5, 128.7, 129.8, 130.1, 133.3, 136.2, 140.7, 194.9.

(4-fluorophenyl)(naphthalene-4-yl)methanone (**2A$_{33}$**)[26]: White solid, Yield = (81%, 53.9 mg), ^{1}H NMR (DMSO-d$_6$, 300 MHz) δ 7.35-7.41 (m, 2H), 7.55-7.64 (m, 4H), 7.83-7.92 (m, 3H), 8.06-8.09 (m, 1H), 8.16-8.19 (m, 1H), ^{13}C NMR (DMSO-d$_6$, 75 MHz) δ 116.3, 116.5, 125.2, 125.4, 126.2, 127.1, 127.9, 128.6, 129.1, 130.5, 131.7, 133.3, 133.4, 133.7, 134.7, 134.7, 136.0, 164.0, 167.3, 196.2.

(Napthalen-2-yl)(napthalen-5-yl)methanone (**2A$_{34}$**)[50]: White solid, Yield = (71%, 50.0 mg), ^{1}H NMR (CDCl$_3$, 300 MHz) δ 7.48-7.54 (m, 4H), 7.57-7.66 (m, 2H), 7.83 (d, *J* = 8.1 Hz, 1H), 7.89-7.97 (m, 3H), 8.03-8.13 (m, 3H), 8.24 (s, 1H), ^{13}C NMR (CDCl$_3$, 75 MHz) δ 124.4, 125.4, 125.7, 126.5, 126.8, 127.3, 127.7, 127.8, 128.4, 128.5, 128.7, 129.7, 131.0, 131.2, 132.4, 132.9, 133.8, 135.6, 135.7, 136.6, 198.0.

Methyl-4-benzoylbenzoate (**2A$_{35}$**)[51]: White solid, Yield = (56%, 33.6 mg),^{1}H NMR ($CDCl_3$, 300 MHz) δ 3.96 (s, 3H), 7.47-7.52 (m, 2H), 7.59-7.61 (m, 1H), 7.78-7.85 (m, 4H), 8.13-8.16 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 52.5, 128.5, 129.5, 129.8, 130.2, 133.0, 133.2, 136.9, 141.3, 166.3, 196.0.

(4-cyano phenyl) (phenyl) methanone (**2A$_{36}$**)[12e]: White solid, Yield = (32%, 17.0 mg), ^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.49-7.54 (m, 2H), 7.62-7.64 (m, 1H), 7.71-7.81(m, 4H), 7.86-7.89 (m, 2H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 115.7, 118.0, 128.6, 130.1, 130.2, 132.2, 133.3, 136.3, 141.2, 195.0.

di (*p*-tolyl)methanone (**2A$_{37}$**)[40b]: White solid, Yield = (85%, 44.6 mg), ^{1}H NMR (DMSO-d_6, 300 MHz) δ 2.41 (s, 6H), 7.36 (d, *J* = 7.8 Hz, 4H), 7.63(d, *J* = 8.1 Hz, 4H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 21.6, 129.5, 130.1, 135.0, 143.3, 195.6.

bis (4-chlorophenyl methanone) (**2A$_{38}$**)[52]: White solid, Yield = (88%, 55.2 mg),^{1}H NMR ($CDCl_3$, 300 MHz) δ 7.43-7.51 (m, 4H), 7.69-7.77 (m, 4H), ^{13}C NMR ($CDCl_3$, 75 MHz) δ 128.7, 131.2, 135.4, 139.1, 194.2.

3-iodo-2-phenylquinolin-4(1*H*)-one (**3a**)[53]: Light Yellow solid, Yield = (70%, 60.7 mg), ^{1}H NMR (300 MHz, DMSO-d_6) δ 7.36-7.42 (m, 1H), 7.53-7.58 (m, 5H), 7.63-7.69 (m, 2H), 8.13 (dd, *J* = 8.1 Hz, 0.9 Hz, 1H), 12.29 (s,1H); ^{13}C NMR (75 MHz, DMSO-d_6) δ 86.3, 118.8, 121.3, 124.7, 125.9, 128.8, 129.4, 130.3, 132.6, 138.3, 139.7, 153.6, 174.1.

3-benzoyl-2-phenyl-quinolin-4-(1*H*)-one (**4a**)[7]: White solid, Yield = (70%, 56.9 mg), ^{1}H NMR (DMSO-d6, 300 MHz) δ 7.38-7.48 (m, 8H), 7.56 (t, *J* = 7.5 Hz, 1H), 7.75-7.79 (m, 4H), 8.10 (d, *J* = 8.1 Hz, 1H), 12.20 (s, 1H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 119.3, 120.7, 124.5, 125.2, 125.3, 128.9, 129.0, 129.1, 129.4, 130.5, 132.9, 133.5, 134.0, 138.4, 140.3, 149.9, 175.5, 196.2.

3-benzoyl-(4-chlorophenyl)-quinolin-4-(1H)-one (**4b**)[7]: Pale yellow solid, Yield = (73%, 65.7 mg),^{1}H NMR (DMSO-d_6, 300 MHz) δ 7.41-7.51 (m, 8H), 7.75-7.81 (m, 4H), 8.10 (d, *J* = 8.1 Hz, 1H), 12.2 (s, 1H), ^{13}C NMR (DMSO-d_6, 75 MHz) δ 119.4, 120.2, 120.2, 124.5, 125.3, 129.0, 129.1, 129.2, 130.5, 131.3, 133.0, 134.0, 137.2, 138.3, 140.4, 150.5, 175.5, 195.1.

3-(4-Methylbenzoyl)-2-phenylquinolin-4(1H)-one (**4c**)[7]: Brown solid, Yield = (62%, 52.5 mg), ^{1}H NMR (300 MHz, DMSO-d_6) δ 2.33 (s, 3H),7.23 (d, *J* = 8.1 Hz, 2H), 7.42 – 7.44 (m, 6H),7.67 (d, *J* = 8.1 Hz, 2H), 7.74-7.76 (m, 2H), 8.10 (d, *J* = 8.0 Hz, 1H), 12.13 (s, 1H), ^{13}C NMR (75 MHz, DMSO-d_6) δ 21.5, 119.2, 120.9, 124.4, 125.1, 125.3, 128.9, 129.0, 129.5, 129.6, 130.4, 132.8, 134.0,135.9, 140.3, 143.9, 149.6, 175.5, 195.6.

Acridone-9(10*H*)-one (**5a**)[54]: Yellow solid, Yield = (78%, 38.0 mg), ^{1}H NMR (300 MHz, DMSO-d_6) δ 7.25 (t, *J* = 7.5 Hz, 2H), 7.53-7.59 (m, 2H), 7.70-7.75 (m, 2H), 8.23-8.26 (m, 2H), 11.76 (s, 1H), ^{13}C NMR (75 MHz, DMSO-d_6) δ 117.3, 120.4, 121.0, 126.0, 133.4, 140.8, 176.8.

3-chloroacridone-9(10*H*)-one (**5b**)[55]: Light yellow solid, Yield = (76%, 43.7 mg), ^{1}H NMR (300 MHz, DMSO-d_6) δ 7.24-7.31 (m, 2H), 7.51-7.56 (m, 2H), 7.72-7.77 (m, 1H), 8.21 (d, *J* = 8.4 Hz, 2H), ^{13}C NMR (75 MHz, DMSO-d_6) δ 116.8, 118.0, 119.6, 121.1, 121.8, 122.1, 126.5, 128.9, 134.4, 138.5, 141.4, 142.1, 176.7.

2-methylacridone-9(10*H*)-one (**5c**)[55]: Yellow solid, Yield = (68%, 35.5 mg), ^{1}H NMR (300 MHz, DMSO-d_6) δ 2.43 (s, 3H), 7.23 (t, *J* = 7.8 Hz, 1H), 7.45-7.59 (m, 3H), 7.68-7.73 (m, 1H), 8.03 (s, 1H), 8.22 (d, *J* = 8.1 Hz, 1H), 11.66 (s, 1H), ^{13}C NMR (75 MHz, DMSO-d_6) δ 21.0, 117.6, 117.7, 120.8, 121.2, 125.5, 126.4, 130.5, 133.7, 135.4, 139.4, 141.2, 177.0.

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