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# A Multimetallic Piano-Stool Ir–$Sn_3$ Catalyst for Nucleophilic Substitution Reaction of γ-Hydroxy Lactams through *N*-Acyliminium Ions†

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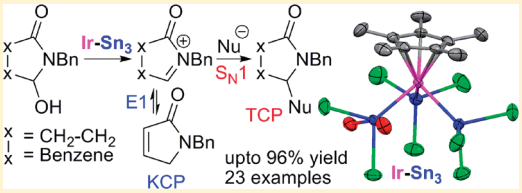
Supporting Information

**ABSTRACT:** A multimetallic piano-stool complex [Cp*Ir-$(SnCl_3)_2\{SnCl_2(H_2O)_2\}$] (**1**) having Ir–$Sn_3$ motif has been synthesized from $[Cp^*IrCl_2]_2$ and $SnCl_2$. The multimetallic complex catalytically promotes the nucleophilic substitution reaction (here after α-amidoalkylation reaction) of γ-hydroxylactams generated from phthalimidals to obtain decorated isoindolinones in excellent yields. Succinamidals, however, lead to the substituted pyrrolidinones (thermodynamic control product) via $S_N1$-type path as well as eliminated pyrrolinones (kinetic control product) via an E1-type path, depending on the reaction parameters. A straightforward application of this methodology is to synthesize benzo-fused indolizidine alkaloid mimics.




The development of multimetallic catalysis has drawn a significant attention in modern organic synthesis due to its superiority in terms of enhanced reactivity and selectivity over monometal catalysis.[1] Like enzymes, in a cooperative multimetallic catalyst two or more metal centers can simultaneously activate nucleophiles and electrophiles leading to higher level of efficiency. This multifunctional synergism has been well exploited in a number of organic transformations.[2] Within the above broad domain, a new class of catalysts can be envisaged where multiple centers coordinate to bring a stronger activation of the substrate and thus offer superior activity. Such multisite interactions between adjacent metal centers are the essence of cluster catalysis; for example, a metal–metal bond is viewed as the edge of a cluster, while three metals constitute a face.[3] Our continuing success in illustrating the unique reactivity of heterobimetallic catalysts[4] bearing a transition metal (Tm) and a main-group metal (Mgm) partner in carbon–carbon bond forming reactions attracted us in designing a substrate activation model across Tm–Mgm for the intermolecular α-amidoalkylation reaction. Related recent developments of note include the trapping of *N*-acyliminium ions by stoichiometric Lewis and Brønsted acids and acidic additives like TMSCl.[5] Therefore, *N*-acyliminium ion chemistry justifies development of catalytic version of the α-amidoalkylation reaction. Indeed recent excitement in the field testifies this view with the emergence of thiourea,[6] Brønsted acids,[7] as well as Lewis acidic metal triflates and triflimidates as catalysts.[8] It is pertinent that in case of catalysis involving a metal complex, the ligand motif is tunable to achieve desired selectivity (chemo-, regio-, stereo-, enantio-). Such an opportunity does not exist in case of "*catalysis via simple metal-salts*". To our knowledge, in the chemistry involving the generation and utilization of *N*-acyliminium ion $Au(PPh_3)OTf$ is the only complex utilized to date.[9] In keeping with our program on multimetallic catalysis, we report here the first multimetallic piano-stool Ir–$Sn_3$ complex **1** which catalytically activates γ-hydroxylactams to produce *N*-acyliminium ions as intermediates for efficient α-amidoalkylation reaction. The catalyst also shows unique differential reactivity for phthalimidals versus succinamidals.

The multimetallic Ir–$Sn_3$ complex **1** was synthesized via insertion of $SnCl_2$ across the iridium–chlorine bonds of $[Cp^*IrCl_2]_2$ in refluxing 1,2-dichloroethane.[10] The initially brick-red solution slowly turned greenish yellow on completion of the reaction. Upon slow diffusion of *n*-hexane to this solution, the product $Cp^*Ir(SnCl_3)_2\{SnCl_2(H_2O)_2\}$ (**1**) crystallized out as greenish yellow blocks in good yield. As shown in Figure 1A, **1** adopts a perfect three-legged piano-stool geometry, as is common to the $Cp^*Ir^{III}$ complexes.[11] In solution, the three stannyl ligands in complex **1** are equivalent in NMR time scale due to plausible fluxional behavior.[12] The satellite peaks in $^1H$ and $^{119}Sn$ NMR spectra of **1** also conclude the presence of direct Ir–Sn bonds. The $^1H$–$^{119}Sn$ coupling constant value ($^4J_{H-Sn}$ = 29 Hz) lies in the expected range of other known complexes.[13] On the other hand, the $^{119}Sn$ chemical shift and the $^{117}Sn$–$^{119}Sn$ coupling constant value (−311 ppm and $^2J_{Sn-Sn}$ = 8 MHz) is well within the expected region of Tm-$SnCl_3$ complexes.[10b] We believe that the piano-stool geometry having three Ir–Sn bonds holds the key to superior catalytic activity of **1** since the three tin centers provide an interactive face to bind the alkylating agent R–Y, (Figure 1B; Y = OH in the instant case).

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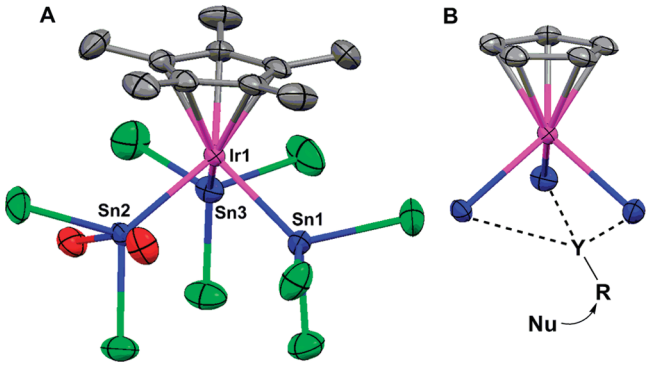



**Figure 1.** (A) Molecular structure of **1**. H-atoms are not shown for clarity. (B) Proposed electrophilic activation model involving **1**. R−Y = alkylating agent, Nu = nucleophile.

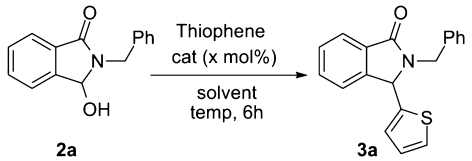
Initially a model study on the $\alpha$-amidoalkylation reaction of thiophene with hydroxylactam **2a** was scrutinized under a broad set of conditions using a variety of Lewis acid catalysts (Table 1). Quantitative conversion was achieved with 1 mol % of **1**, and at 80 °C; the catalytic activity rapidly dropped down at lower temperature (entries 1−3). We also noted the following: (i) in terms of catalytic efficiency, **1** is superior over Lewis acids like $Sc(OTf)_3$, $La(OTf)_3$ or $SnCl_4$ (entries 3, 8, 9, and 11), (ii) individually $[Cp^*IrCl_2]_2$, $SnCl_4$, or $SnCl_2$ are poorly active (entries 12−14), and (iii) the $Ir^I$−$SnCl_3$ unit constructed on a simple ligand-frame showed negligible catalytic activity (entry 7). All of the above observations possibly highlight a multimetallic synergism in an Ir−$Sn_3$ core that promotes the alkylation reaction. However, further work is needed to comprehend the nature of such a synergy.

The scope of the present multimetallic Ir−$Sn_3$-catalyzed alkylation reaction for phthalimidals is illustrated in Table 2. Under optimal conditions, quantitative conversions and excellent yields were achieved in the $\alpha$-amidoalkylation reaction with various electron-rich aromatics (anisole, 1-methoxy-3,5-dimethylbenzene, and 1,3,5-trimethoxybenzene) and heteroaromatics (thiophene, 2-methylthiophene, 2,5-dimethylfuran, and indole). Less electron-rich arenes like benzene or toluene remained unreactive. Organosilicon nucleophiles (allyltrimethylsilane) and $\beta$-dicarbonyl nucleophiles (4-hydroxycoumarin) also afforded the alkylated products **3d**, **3q**, **3t**, and **3g** almost quantitatively. Delightfully, in addition to the $\alpha$-amidoalkylation of carbon nucleophiles; oxygen (2-propanol and 3-phenylpropanol), sulfur (thiophenol and 4-methoxythiophenol), or nitrogen (*N*-tosylamine) nucleophiles were also found to provide the alkylated products **3h−l** in excellent yields. All the new compounds were characterized by $^1$H NMR, $^{13}$C NMR, HRMS, or elemental analysis data, and the structure of **3r** was confirmed by X-ray crystallographic analysis (see the Supporting Information).

In contrast to phthalimidals, a first set of reactions for succinamidals with the model hydroxylactam **2d** and 2-methylthiophene in presence of catalyst **1** led to the expected alkylated product **3u** along with an eliminated product **4** (Table 3). The extent to which each product is formed varied with reaction time and catalyst loading. For example, at 1 mol % loading of **1**, **3u** was not observed even after 15 h (Table 3, entry 2). The result was just reversed at 5 mol % catalyst loading (entry 6). Further, at initial stage of reaction, **4** was formed; but with the progress of time, **3u** became the major product (entries 3 versus 4 and 5 versus 6). It is apparent from the above facts that under our reaction conditions pyrrolidinone **3u** is a thermodynamically controlled product while *N*-benzyl-3-pyrroline-2-one **4** is a kinetically controlled product.

Formation of **4** from **2d** can be explained by a prior E1-type elimination reaction followed by a double bond isomerization (Scheme 1).[14] We conjecture that the formation of **4** from *N*-acyliminium ion is reversible and also much faster compared to the substitution step ($S_N1$-type) which leads to the alkylated product **3**. Gratifyingly, in a separate set of reaction when **4** was reacted with 2-methylthiophene in presence of catalyst **1** (5 mol %) under similar conditions, **3u** was the major product

**Table 1. Screening and Optimization of $\alpha$-Amidoalkylation Reaction**[a]




**2a** **3a**

| no. | catalyst | *x* | solvent | temp, °C | conv,[b] % | yield,[c] % |
|---|---|---|---|---|---|---|
| 1 | **1** | 1 | DCE | 60 | 12 | 5 |
| 2 | **1** | 1 | DCE | 70 | 43 | 38 |
| 3 | **1** | 1 | DCE | 80 | 100 | 96 |
| 4 | **1** | 1 | MeCN | 80 | 17 | 10 |
| 5 | **1** | 1 | $MeNO_2$ | 80 | 58 | 50 |
| 6 | **1** | 1 | toluene | 80 | 37 | 30 |
| 7 | $(COD)_2IrSnCl_3$ | 1 | DCE | 80 | 5 | 1 |
| 8 | $Sc(OTf)_3$ | 5 | DCE | 80 | 50 | 22 |
| 9 | $La(OTf)_3$ | 5 | DCE | 80 | 5 | 0 |
| 10 | PTSA | 5 | DCE | 80 | 10 | 0 |
| 11 | $SnCl_4$ | 5 | DCE | 80 | 10 | 5 |
| 12 | $[Cp^*IrCl_2]_2$ | 5 | DCE | 80 | 0 | 0 |
| 13 | $SnCl_2$ | 5 | DCE | 80 | 0 | 0 |
| 14 | $SnCl_2.2H_2O$ | 5 | DCE | 80 | 0 | 0 |

[a]General conditions: **2a** (0.5 mmol), thiophene (1 mmol), catalyst (0.005 mmol for 1 mol % and 0.025 mmol for 5 mol %), solvent (2 mL). [b]Determined by NMR. [c]Isolated yield.


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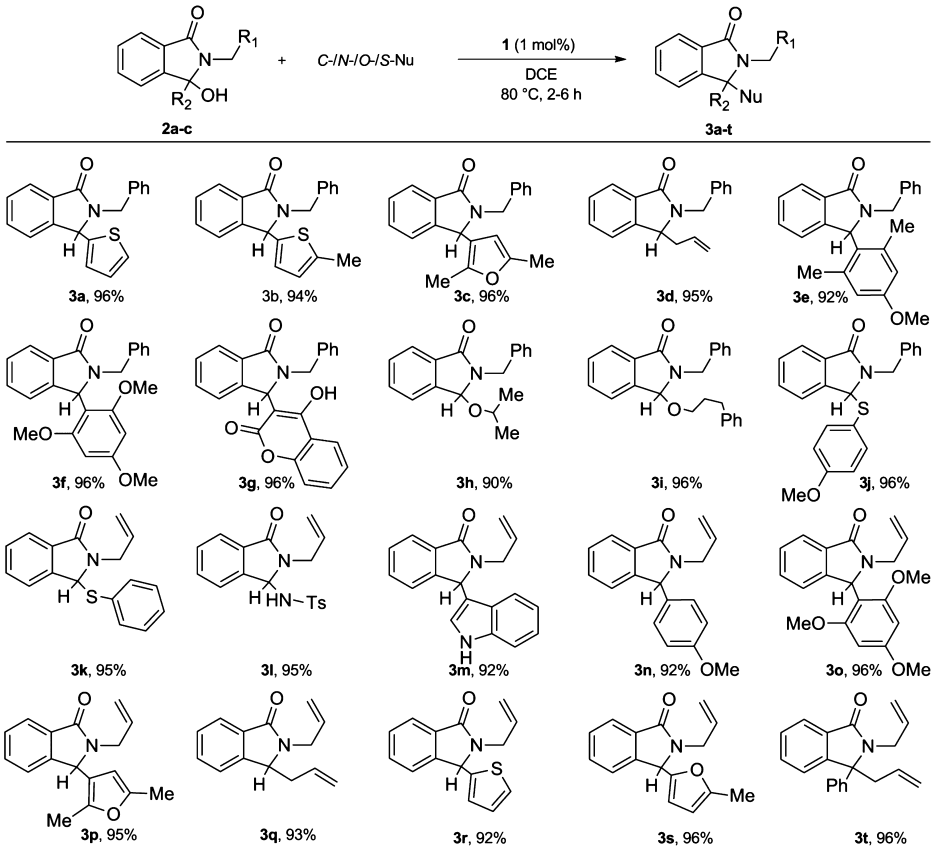
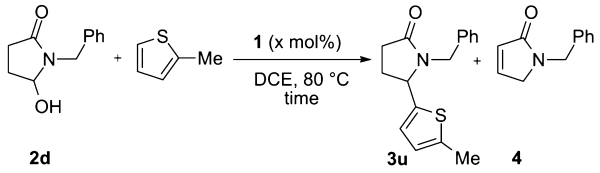
**Table 2. Scope of $\alpha$-Amidoalkylation Reaction with Phthalimidals Catalyzed by 1[a]**




[a]Reactions were performed on a 0.25 mmol scale.

**Table 3. Elimination vs Substitution Reaction for Succinamidal: Dependency on Catalyst Concentration and Reaction Time[a]**




| entry | $x$ | time, h | **3u**, % | **4**, % |
|---|---|---|---|---|
| 1 | 1 | 2 | 0 | 16 |
| 2 | 1 | 15 | 0 | 39 |
| 3 | 3 | 2 | 3 | 35 |
| 4 | 3 | 15 | 17 | 24 |
| 5 | 5 | 2 | 39 | 13 |
| 6 | 5 | 10 | 50 | 0 |

[a]General conditions: **2d** (0.25 mmol), 2-methylthiophene (0.50 mmol), DCE (2 mL).

formed.[15] One may also note the influence of nucleophilicity of arene in the reaction of succinamidals. While highly electron rich 1,3-dimethoxybenzene and 1,3,5-trimethoxybenzene gave the corresponding alkylated products **3w** and **3x** in excellent yields, anisole did not promote alkylation even after 20 h; the elimination product **4** remained the only product (Scheme 2).[16] However, it may be noted that hydroxylactams derived from phthalimide were directly transformed to substituted isoindolinones **3a**–**t** via $S_N1$-type pathway as elimination reaction is not an option here.

We propose that the catalyst **1** acts as a mild and controlled Lewis acid to activate and remove the −OH group of the substrate. Further, from the NMR spectroscopic study (see the Supporting Information), Lewis acidic nature of the hard tin centers to interact with −OH group is well established.[4,8b] Although at this stage the exact role of iridium is unclear, whether it directly activates the substrate or it helps the tin centers to do it and overall maintains an electrophilic environment around the Ir−$Sn_3$ motif for sustainable catalysis.[17] Although Sn−Cl bonds (as in $SnCl_4$) are susceptible for hydrolysis, those in "Ir−$Sn_3$" are very inert; in fact, **1** is stable enough to crystallize out even after catalysis (as confirmed by crystal structure analysis). As water is producing as a byproduct in our case, somehow, the multimetallic Ir−$Sn_3$ system manages to overpower the hydrolytic decomposition and thus as little as 1% of the catalyst **1** is sufficient for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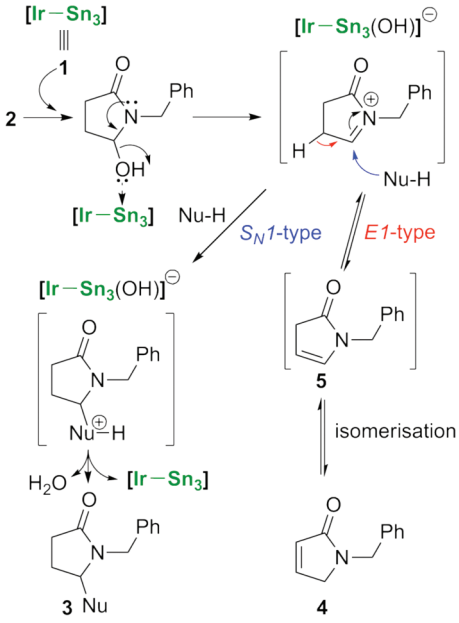
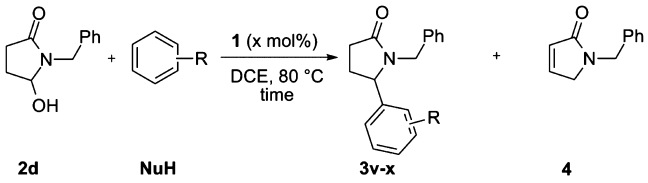
**Scheme 1. Proposed Reaction Mechanism**




**Scheme 2. Elimination vs Substitution Reaction for Succinamidal: Dependency on Nucleophilicity of Arenes**[a]




**NuH:** R = OMe x = 5 time = 20 h **3v:** 0% **4:** 57%
**NuH:** R = 1,3-$(OMe)_2$ x = 5 time = 10 h **3w:** R = 2,4-$(OMe)_2$; 83% **4:** 0%
**NuH:** R = 1,3,5-$(OMe)_3$ x = 1 time = 6 h **3x:** R = 2,4,6-$(OMe)_3$; 85% **4:** 0%

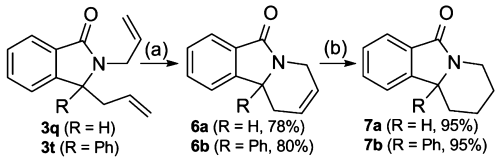
[a]General conditions: **2d** (0.25 mmol), **NuH** (0.50 mmol), DCE (2 mL)

quantitative organic transformation while the traditional Lewis acids like $SnCl_4$ fails (see Table 1).[18] One worth noticing is that the combination of high valent iridium and stannyl ligand as in Ir−$Sn_3$ motif is necessary for superior catalytic activity. Detailed studies on the exact roles of these two metal partners in Ir−$Sn_3$ motif are ongoing and will be reported in due course.

Alkaloids incorporating the indolizidine skeletons comprise a large group of natural compounds having an interesting range of biological activity.[19] In this context, we perceive that there remains a great applicability of the present route toward the synthesis of benzo-fused indolizidine alkaloid mimics. As a proof-of-principle, we constructed the desired indolizidine skeletons from the alkylated products (**3q** or **3t**) by ring closing metathesis followed by olefin hydrogenation (Scheme 3). Significantly, compound **7b** is a non-nucleoside HIV-1 reverse transcriptase inhibitor.[19a]

In summary, we have synthesized a new multimetallic Ir−$Sn_3$ catalyst, $Cp^*Ir(SnCl_3)_2\{SnCl_2(H_2O)_2\}$, having piano-stool geometry for amidoalkylation reaction. The differential reactivity of succinamidals (over phthalimidals) toward elimination and substitution reaction is yet another discrete outcome of this study. Asymmetric induction and further synthetic elaboration are underway.

**Scheme 3. Synthesis of Indolizidine Skeletons**[a]




[a]Reagents and conditions: (a) Grubbs-I (1 mol %)/DCM/50 °C/12 h; (b) $H_2$/Pd−C/MeOH/rt/12 h.

## EXPERIMENTAL SECTION

**General Procedures.** All reactions were carried out under an argon atmosphere in flame-dried glassware using standard Schlenk techniques. Solvents are dried prior to use. Column chromatography was performed on 100−200 mesh silica gel.

**Starting Materials.** $[Cp^*IrCl_2]_2$ and $[(COD)_2Ir(SnCl_3)]$ were synthesized according to the literature procedures.[20] Organic starting materials **2a**, **2b**, and **2d** were synthesized by $NaBH_4$ reduction,[8a] and **2c** was synthesized by Grignard reagent PhMgBr addition to imides according to the literature procedures.[21]

**2-Benzyl-3-hydroxyisoindolin-1-one (2a).** White solid.[22]

**2-Allyl-3-hydroxyisoindolin-1-one (2b).** White solid.[21]

**2-Allyl-3-hydroxy-3-phenylisoindolin-1-one (2c).** White solid.[21]

**1-Benzyl-5-hydroxypyrrolidin-2-one (2d).** White solid.[23]

**Synthesis of $Cp^*Ir(SnCl_3)_2\{SnCl_2(H_2O)_2\}\cdot 3H_2O$ (1).** To $[Cp^*IrCl_2]_2$ (40 mg, 0.05 mmol) in 2 mL of dichloroethane was added anhydrous $SnCl_2$ (60 mg, 0.3 mmol). The mixture was heated at 80 °C for 3 h. The initially brick red solution slowly turned a greenish yellow color after completion of the reaction. After the mixture was cooled to room temperature, the greenish yellow solution was taken out from the reaction flask and slow diffusion of *n*-hexane to this solution causes greenish yellow block like crystals: yield 90 mg (80%); mp >300 °C; solubility: highly soluble in DMSO, MeCN, acetone; moderately soluble in methanol, DCM, DCE, $CHCl_3$; insoluble in benzene, toluene and other hydrocarbons; $^1$H NMR (200 MHz, acetone-$d_6$) $\delta$ 2.29 (s, 15H, $C_5(CH_3)_5$) with tin satellites ($J_{H-Sn}$ = 29 Hz); $^{13}$C NMR (54.6 MHz, acetone-$d_6$) $\delta$ 10.4 ($C_5(CH_3)_5$), 98.0 ($C_5(CH_3)_5$); $^{119}$Sn NMR (149 MHz, acetone-$d_6$) $\delta$ −311 with tin satellites ($J_{Sn-Sn}$ = 8000 Hz). Anal. Calcd for $C_{10}H_{25}Cl_8IrO_5Sn_3$ (1057.59): C, 11.36; H, 2.38. Found: C, 11.53; H, 2.56.

**General Procedure for the Nucleophilic Substitution Reaction of γ-Hydrxylactams 2a−d Catalyzed by Complex 1.** To a solution of γ-hydroxylactam **2a** (60 mg, 0.25 mmol) in 2 mL of DCE were added under an argon atmosphere thiophene (40 μL, 0.5 mmol) and **1** (2.6 mg, 0.0025 mmol). The reaction mixture was stirred at 80 °C. When the reaction was over (monitored by TLC using ethyl acetate/petroleum ether 60−80 °C 1:3 v/v), a saturated aqueous solution of sodium hydrogencarbonate was added. The aqueous layer was then extracted two times with $CH_2Cl_2$ (2 × 5 mL). The combined organic layers were dried over $Na_2SO_4$; the solvent was removed under reduced pressure. The resulting product was purified by column chromatography on silica gel to give the expected coupled product **3**.

**2-Benzyl-3-(thiophene-2-yl)isoindolin-1-one (3a):** crystalline white solid (73 mg, 96%); mp 125−127 °C; $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 3.89 (d, 1H, $J$ = 14.8 Hz), 5.40 (d, 1H, $J$ = 14.8 Hz), 5.57 (s, 1H), 7.00−7.04 (m, 2H), 7.23−7.33 (m, 7H), 7.46−7.51 (m, 2H), 7.92−7.96 (m, 1H); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 43.6, 58.7, 123.2, 123.8, 126.7, 126.9, 127.6, 127.7, 128.4, 128.6, 128.7, 131.1, 131.9, 137.0, 140.0, 145.6, 167.8; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 43.7 ($CH_2$), 58.7 (CH), 123.3 (CH), 123.8 (CH), 126.8 (CH), 127.0 (CH), 127.7 (CH), 127.8 (CH), 128.5 (CH), 128.76 (CH), 128.79 (CH), 132.0 (CH). Anal. Calcd for $C_{19}H_{15}NOS$ (305.09): C, 74.72; H, 4.95; N, 4.59. Found: C, 74.91; H, 5.13; N, 4.41.

**2-Benzyl-3-(5-methylthiophene-2-yl)isoindolin-1-one (3b):** white crystalline solid (75 mg, 94%); mp 110−112 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 2.43 (s, 3H), 3.91 (d, 1H, $J$ = 14.8 Hz), 5.40 (d, 1H, $J$ = 15 Hz), 5.48 (s, 1H), 6.66 (d, 1H, $J$ = 2.4 Hz), 6.85 (d, 1H, $J$ =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3.2 Hz), 7.23−7.31 (m, 6H), 7.47−7.51 (m, 2H), 7.91−7.95 (m, 1H); $^{13}$C NMR (54.6 Hz, $CDCl_3$) $\delta$ 16.5, 44.6, 60.0, 124.2, 124.7, 125.8, 128.5, 128.9, 129.5, 129.6, 129.7, 132.2, 132.9, 138.1, 138.4, 142.6, 146.7, 168.7; HRMS (ESI) calcd for $C_{20}H_{17}NOS + H^+$ 320.1109, found 320.1103.

**2-Benzyl-3-(2,5-dimethylfuran-3-yl)isoindolin-1-one (3c):** colorless oil (76 mg, 96%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 2.07 (s, 3H), 2.16 (s, 3H), 3.83 (d, 1H, $J$ = 14.8 Hz), 5.15 (s, 1H), 5.35 (s, 1H), 5.37 (d, 1H, $J$ = 14.8 Hz), 7.15−7.31 (m, 6H), 7.45−7.49 (m, 2H), 7.90−7.95 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 11.5, 13.5, 43.7, 55.4, 104.8, 115.5, 123.0, 123.7, 127.5, 128.3, 128.7, 131.7, 137.3, 145.8, 148.8, 150.9, 168.1; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 11.5 ($CH_3$), 13.5 ($CH_3$), 44.2 ($CH_2$), 55.4 (CH), 104.7 (CH), 123.0 (CH), 123.7 (CH), 127.5 (CH), 128.2 (CH), 128.3 (CH), 128.6 (CH), 131.7 (CH). Anal. Calcd for $C_{21}H_{19}NO_2$ (317.14): C, 79.47; H, 6.03; N, 4.41. Found: C, 79.33; H, 6.27; N, 4.25.

**3-Allyl-2-benzylisoindolin-1-one (3d):** colorless oil (63 mg, 95%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 2.56−2.80 (m, 2H), 4.17 (d, 1H, $J$ = 15.2 Hz), 4.42 (t, 1H, $J$ = 5.2 Hz), 4.96−5.07 (m, 2H), 5.24−5.41 (m, 1H), 5.42 (d, 1H, $J$ = 15.2 Hz), 7.22−7.55 (m, 8H), 7.87−7.91 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 35.2, 43.9, 58.0, 119.3, 122.4, 123.8, 127.6, 128.1, 128.8, 131.2, 131.4, 132.3, 137.1, 144.9, 168.5. Anal. Calcd for $C_{18}H_{17}NO$ (263.13): C, 82.10; H, 6.51; N, 5.32. Found: C, 82.31; H, 6.37; N, 5.44.

**2-Benzyl-3-(4-methoxy-2,6-dimethylphenyl)isoindolin-1-one (3e).** Mixture of ortho and para regioisomers and isolated as colorless oil (82 mg, 92%): $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 1.52 (s, 3H), 1.58 (s, 3H), 2.03 (s, 3H), 2.33 (s, 3H), 3.62 (d, 1H, $J$ = 14.6 Hz), 3.70 (s, 3H), 3.71 (d, 1H, $J$ = 14.6 Hz), 3.79 (s, 3H), 5.27 (d, 1H, $J$ = 14.4 Hz), 5.43 (d, 1H, $J$ = 14.6 Hz), 5.72 (s, 1H), 6.30 (s, 1H), 6.48 (s, 2H), 6.61−6.66 (m, 2H), 7.07−7.30 (m, 11H), 7.40−7.48 (m, 5H), 7.87−7.97 (m, 2H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 18.3, 18.9, 21.0, 21.5, 43.8, 44.0, 55.1, 55.5, 55.7, 59.1, 109.6, 114.1, 115.8, 118.2, 122.3, 122.6, 123.3, 123.5, 123.7, 125.4, 127.3, 127.6, 127.8, 128.1, 128.3, 128.5, 128.7, 128.8, 131.6, 131.8, 132.5, 132.7, 137.2, 137.4, 139.4, 139.9, 145.4, 145.9, 158.9, 168.6. Anal. Calcd for $C_{24}H_{23}NO_2$ (357.17): C, 80.64; H, 6.49; N, 3.92. Found: C, 80.79; H, 6.66; N, 3.80.

**2-Benzyl-3-(2,4,6-trimethoxyphenyl)isoindolin-1-one (3f):** colorless crystalline solid (93 mg, 96%); 120−122 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 3.25 (s, 3H), 3.69 (s, 3H), 3.79 (s, 3H), 3.83 (d, 1H, $J$ = 14.6 Hz), 5.13 (d, 1H, $J$ = 14.6 Hz), 5.91 (d, 1H, $J$ = 2.0 Hz), 5.98 (s, 1H), 6.16 (d, 1H, $J$ = 2.0 Hz), 7.09−7.23 (m, 6H), 7.35−7.39 (m, 2H), 7.86−7.91 (m, 1H); $^{13}$C NMR (54.6 Hz, $CDCl_3$) $\delta$ 44.2, 54.8, 55.3, 55.8, 90.4, 91.2, 103.8, 121.8, 123.0, 127.0, 127.1, 128.1, 128.6, 130.9, 132.9, 137.8, 147.0, 160.1, 160.5, 161.7, 169.0; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 44.2 ($CH_2$), 54.8 ($CH_3$), 55.3 ($CH_3$), 55.4 ($CH_3$), 55.8 (CH), 90.4 (CH), 91.2 (CH), 121.8 (CH), 123.0 (CH), 127.0 (CH), 127.1 (CH), 128.1 (CH), 128.6 (CH), 130.9 (CH); HRMS (ESI) calcd for $C_{24}H_{23}NO_4 + H^+$ 390.1705, found 390.1701.

**2-Benzyl-3-(4-hydroxy-2-oxo-2*H*-chromen-3-yl)isoindolin-1-one (3g):** off-white solid (92 mg, 96%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 4.32 (d, 1H, $J$ = 14.8 Hz), 4.90 (d, 1H, $J$ = 14.8 Hz), 6.09 (s, 1H), 7.07−7.65 (m, 12H), 8.34 (d, 1H, $J$ = 7.8 Hz); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 45.1, 57.6, 99.1, 115.8, 116.9, 122.4, 122.9, 123.3, 124.2, 127.5, 128.0, 128.1, 128.2, 128.5, 131.5, 132.0, 132.8, 136.0, 145.3, 153.1, 164.0, 164.3, 170.5; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 45.2, 57.7, 117.0, 122.5, 123.0, 123.5, 124.4, 127.7, 128.2, 128.3, 128.6, 132.1, 132.9; HRMS (ESI) calcd for $C_{24}H_{17}NO_4 + Na^+$ 406.1055, found 406.1055.

**2-Benzyl-3-isopropoxyisoindolin-1-one (3h):** colorless oil (65 mg, 90%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 1.07 (d, 3H, $J$ = 6.2 Hz), 1.21 (d, 3H, $J$ = 6.0 Hz), 3.64 (septet, 1H, $J$ = 6.0 Hz), 4.25 (d, 1H, $J$ = 15.0 Hz), 5.27 (d, 1H, $J$ = 15.0 Hz), 5.64 (s, 1H), 7.26−7.31 (m, 5H), 7.48−7.57 (m, 3H), 7.85 (d, 1H, $J$ = 7.4 Hz); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 23.4, 23.7, 43.1, 68.7, 85.2, 123.5, 127.5, 128.2, 128.4, 128.7, 129.7, 131.9, 132.3, 137.0, 142.2, 167.3; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 23.5 ($CH_3$), 23.8 ($CH_3$), 43.2 ($CH_2$), 68.7 (CH), 85.2 (CH), 123.5 (CH), 127.6 (CH), 128.2 (CH), 128.7 (CH), 129.7 (CH), 132.0 (CH). Anal. Calcd for $C_{18}H_{19}NO_2$ (281.14): C, 76.84; H, 6.81; N, 4.98. Found: C, 76.62; H, 6.98; N, 4.81.

**2-Benzyl-3-(3-phenylpropoxy)isoindolin-1-one (3i):** colorless oil (85 mg, 96%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 1.75−1.85 (m, 2H), 2.62 (t, 2H, $J$ = 7.6 Hz), 2.97 (dt, 1H, $J$ = 6.4 Hz, 15.6 Hz), 3.14 (dt, 1H, $J$ = 6.4 Hz, 15.2 Hz), 4.25 (d, 1H, $J$ = 14.8 Hz), 5.15 (d, 1H, $J$ = 14.4 Hz), 5.75 (s, 1H), 7.14−7.21 (m, 3H), 7.26−7.38 (m, 7H), 7.47−7.58 (m, 3H), 7.87 (d, 1H, $J$ = 6.8 Hz); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 31.1, 32.3, 43.2, 61.6, 85.4, 123.4, 123.6, 125.9, 127.6, 128.2, 128.3, 128.5, 128.7, 129.8, 132.0, 132.6, 136.9, 141.0, 141.5, 167.4; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 31.2 ($CH_2$), 32.4 ($CH_2$), 43.3 ($CH_2$), 61.7 ($CH_2$), 85.5 (CH), 123.5 (CH), 123.7 (CH), 125.9 (CH), 127.6 (CH), 128.3 (CH), 128.4 (CH), 128.6 (CH), 128.7 (CH), 129.9 (CH), 132.1 (CH). Anal. Calcd for $C_{24}H_{23}NO_2$ (357.17): C, 80.64; H, 6.49; N, 3.92. Found: C, 80.81; H, 6.65; N, 3.77.

**2-Benzyl-3-(4-methoxyphenylthio)isoindolin-1-one (3j):** white solid (86 mg, 96%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 3.68 (s, 3H), 4.55 (d, 1H, $J$ = 14.4 Hz), 5.40 (s, 1H), 5.44 (d, 1H, $J$ = 14.8 Hz), 6.57 (d, 2H, $J$ = 8.4 Hz), 6.94 (d, 2H, $J$ = 8.4 Hz), 7.26−7.36 (m, 6H), 7.52 (t, 1H, $J$ = 7.6 Hz), 7.58−7.59 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 43.0, 55.1, 65.7, 114.1, 117.7, 123.3, 123.7, 127.6, 128.4, 128.5, 128.7, 131.5, 131.5, 136.7, 137.2, 143.0, 160.4, 167.3; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 43.1 ($CH_2$), 55.2 ($CH_3$), 65.8 (CH), 114.1 (CH), 123.3 (CH), 123.7 (CH), 127.7 (CH), 128.5 (CH), 128.6 (CH), 128.8 (CH), 131.6 (CH), 137.3 (CH). Anal. Calcd for $C_{22}H_{19}NO_2S$ (361.11): C, 73.10; H, 5.30; N, 3.88. Found: C, 73.31; H, 5.12; N, 4.13.

**2-Allyl-3-(phenylthio)isoindolin-1-one (3k):** colorless oil (67 mg, 95%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 4.07 (dd, 1H, $J$ = 7.8 and 15.4 Hz), 4.77 (dd, 1H, $J$ = 4.2 and 15.4 Hz), 5.18−5.25 (m, 2H), 5.72 (s, 1H), 5.83 (dddd, 1H, $J$ = 1.4 Hz, 7.0 Hz, 11.2 and 18.8 Hz), 7.04−7.19 (m, 5H), 7.34 (t, 1H, $J$ = 7.4 Hz), 7.50−7.66 (m, 3H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 41.9, 66.0, 118.4, 123.2, 123.8, 128.1, 128.6, 129.0, 131.6, 132.5, 135.2, 142.9, 167.1; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 41.9 ($CH_2$), 66.0 (CH), 118.4 ($CH_2$), 123.2 (CH), 123.8 (CH), 128.6 (CH), 128.7 (CH), 129.0 (CH), 131.6 (CH), 132.5 (CH), 135.2 (CH). Anal. Calcd for $C_{17}H_{15}NOS$ (281.09): C, 72.57; H, 5.37; N, 4.98. Found: C, 72.43; H, 5.18; N, 5.21.

***N*-(2-Allyl-3-oxoisoindolin-1-yl)-4-methylbenzenesulfonamide (3l):** white solid (80 mg, 95%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 2.48 (s, 3H), 3.49 (dd, 1H, $J$ = 6.8 and 16.0 Hz), 3.87 (d, 1H, $J$ = 14.4 Hz), 4.95 (d, 1H, $J$ = 17.2 Hz), 5.06 (d, 1H, $J$ = 10.4 Hz), 5.60 (ddd, 1H, $J$ = 3.0 Hz, 9.0 and 13.4 Hz), 5.71 (d, 1H, $J$ = 10.0 Hz), 6.38 (d, 1H, $J$ = 10.0 Hz), 6.88 (s, 1H), 7.37−7.41 (m, 4H), 7.62 (s, 1H), 7.87 (d, 2H, $J$ = 8.0 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 21.5, 41.0, 67.4, 117.6, 123.0, 123.4, 126.9, 129.6, 129.9, 130.9, 131.8, 132.4, 138.5, 142.2, 143.9, 167.0; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 21.6 ($CH_3$), 41.1 ($CH_2$), 67.5 (CH), 117.7 ($CH_2$), 123.1 (CH), 123.5 (CH), 126.9 (CH), 129.7 (CH), 130.0 (CH), 131.9 (CH), 132.5 (CH). Anal. Calcd for $C_{18}H_{18}N_2O_3S$ (342.10): C, 63.14; H, 5.30; N, 8.18. Found: C, 62.90; H, 5.14; N, 8.01.

**2-Allyl-3-(1*H*-indol-3-yl)isoindolin-1-one (3m):** colorless crystalline solid (66 mg, 92%); mp 138−140 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 3.47 (dd, 1H, $J$ = 7.2 and 15.6 Hz), 4.65 (dd, 1H, $J$ = 4.0 and 15.6 Hz), 5.05−5.18 (m, 2H), 5.82 (dddd, 1H, $J$ = 2.1 Hz, 4.9 Hz, 12.1 and 19.3 Hz), 5.86 (s, 1H), 6.88−6.94 (m, 2H), 7.09−7.18 (m, 1H), 7.26−7.53 (m, 5H), 7.98−8.02 (m, 1H), 9.45 (s, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 42.5, 57.9, 110.1, 111.8, 117.4, 118.9, 119.9, 122.4, 123.4, 125.2, 125.3, 128.3, 131.9, 133.2, 137.1, 146.3, 168.4; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 42.5 ($CH_2$), 57.8 (CH), 111.7 (CH), 117.4 ($CH_2$), 118.9 (CH), 119.9 (CH), 122.4 (CH), 123.4 (CH), 123.4 (CH), 125.1 (CH), 128.3 (CH), 131.8 (CH), 133.2 (CH); HRMS (ESI) calcd for $C_{19}H_{16}N_2O + H^+$ 289.1341, found 289.1334.

**2-Allyl-3-(4-methoxyphenyl)isoindolin-1-one (3n):** colorless oil (64 mg, 92%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 3.31 (dd, 1H, $J$ = 7.6 and 15.4 Hz), 3.78 (s, 3H), 4.64 (dd, 1H, $J$ = 4.2 and 15.4 Hz), 5.01−5.17 (m, 2H), 5.41 (s, 1H), 5.77 (dddd, 1H, $J$ = 2.1 Hz, 7.7 Hz, 11.9 and 19.7 Hz), 6.85 (d, 2H, $J$ = 8.8 Hz), 7.01 (d, 2H, $J$ = 8.6 Hz), 7.13−7.17 (m, 1H) 7.39−7.49 (m, 2H), 7.84−7.91 (m, 1H); $^{13}$C

NMR (54.6 MHz, $CDCl_3$) $\delta$ 42.5, 55.3, 63.4, 114.4, 117.9, 123.1, 123.5, 128.2, 128.6, 129.0, 131.5, 131.8, 133.0, 146.6, 159.8, 168.2; HRMS (ESI) calcd for $C_{18}H_{17}NO_2 + H^+$, 280.1338; found, 280.1331.

**2-Allyl-3-(2,4,6-trimethoxyphenyl)isoindolin-1-one (3o):** colorless crystalline solid (80 mg, 96%); mp 128−130 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 3.28 (s, 3H), 3.38 (dd, 1H, $J$ = 7.0 and 15.2 Hz), 3.81 (s, 3H), 3.89 (s, 3H), 4.48 (dd, 1H, $J$ = 5.0 and 15.4 Hz), 4.96−5.08 (m, 2H), 5.78 (dddd, 1H, $J$ = 3.2 Hz, 8 Hz, 13.2 and 20.2 Hz), 5.96 (d, 1H, $J$ = 2.0 Hz), 6.15 (s, 1H), 6.20 (d, 1H, $J$ = 2.0 Hz), 7.15−7.19 (m, 1H), 7.38−7.42 (m, 2H), 7.88−7.92 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 43.0, 55.1, 55.5, 55.6, 56.2, 90.7, 91.5, 104.4, 117.1, 122.0, 123.1, 127.3, 131.0, 133.2, 133.7, 147.2, 160.4, 160.7, 161.8, 168.9; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 42.8 ($CH_2$), 55.0 ($CH_3$), 55.3 ($CH_3$), 55.4 ($CH_3$), 56.0 (CH), 90.5 (CH), 91.3 (CH), 116.9 ($CH_2$), 121.8 (CH), 123.0 (CH), 127.1 (CH), 130.8 (CH), 133.4 (CH); HRMS (ESI) calcd for $C_{20}H_{21}NO_4 + H^+$ 340.1549, found 340.1544.

**2-Allyl-3-(2,5-dimethylfuran-3-yl)isoindolin-1-one (3p):** colorless viscous oil (63 mg, 95%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 2.11 (s, 3H), 2.25 (s, 3H), 3.36 (dd, 1H, $J$ = 7.8 and 15.6 Hz), 4.61 (dd, 1H, $J$ = 4.0 and 15.4 Hz), 5.03−5.16 (m, 2H), 5.29 (s, 1H), 5.33 (s, 1H), 5.76 (dddd, 1H, $J$ = 1.9 Hz, 7.7 Hz, 11.7 and 19.5 Hz), 7.18−7.22 (m, 1H), 7.38−7.50 (m, 2H), 7.82−7.86 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 11.6, 13.4, 42.3, 55.6, 104.7, 115.6, 117.5, 123.0, 123.5, 128.2, 131.6, 131.8, 133.3, 145.7, 148.7, 150.8, 167.8; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 11.6 ($CH_3$), 13.4 ($CH_3$), 42.3 ($CH_2$), 55.6 (CH), 104.7 (CH), 117.5 ($CH_2$), 123.0 (CH), 123.4 (CH), 128.2 (CH), 131.6 (CH), 133.3 (CH). Anal. Calcd for $C_{17}H_{17}NO_2$ (267.13): C, 76.38; H, 6.41; N, 5.24. Found: C, 76.53; H, 6.59; N, 5.11.

**2,3-Diallylisoindolin-1-one (3q).** Colorless oil (50 mg, 93%) and identified by spectral comparison with literature data.[8a]

**2-Allyl-3-(thiophene-2-yl)isoindolin-1-one (3r):** white crystalline solid (58 mg, 92%); mp 95−97 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 3.47 (dd, 1H, $J$ = 7.8 and 15.6 Hz), 4.66 (dd, 1H, $J$ = 4.4 and 15.4 Hz), 5.07−5.20 (m, 2H), 5.69−5.89 (m, 1H), 5.78 (s, 1H), 6.97−7.14 (m, 2H), 7.29−7.35 (m, 2H), 7.44−7.56 (m, 2H), 7.86−7.93 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 42.5, 59.0, 118.1, 123.2, 123.6, 126.6, 126.9, 127.6, 128.7, 131.2, 131.9, 132.8, 140.2, 145.6, 167.6; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 42.5 ($CH_2$), 59.0 (CH), 118.1 ($CH_2$), 123.2 (CH), 123.7 (CH), 126.6 (CH), 126.9 (CH), 127.5 (CH), 128.7 (CH), 131.9 (CH), 132.8 (CH); HRMS (ESI) calcd for $C_{15}H_{13}NOS + Na^+$ 278.0616, found, 278.0616.

**2-Allyl-3-(5-methylfuran-2-yl)isoindolin-1-one (3s):** white crystalline solid (60 mg, 96%); mp 115−117 °C; $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 2.18 (s, 3H), 3.58 (dd, 1H, $J$ = 7.2 and 15.4 Hz), 4.57 (dd, 1H, $J$ = 4.6 Hz, 15.4 Hz), 5.07−5.16 (m, 2H), 5.51 (s, 1H), 5.76 (dddd, 1H, $J$ = 3.1 Hz, 7.7 Hz, 12.1 and 19.5 Hz), 5.91 (d, 1H, $J$ = 2.2 Hz), 6.19 (d, 1H, $J$ = 3.0 Hz), 7.30−7.34 (m, 1H), 7.43−7.54 (m, 2H), 7.85−7.89 (m, 1H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 13.6, 43.0, 57.6, 106.3, 110.7, 117.7, 123.0, 123.6, 128.6, 131.6, 132.0, 132.9, 143.2, 147.2, 153.3, 167.8; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 13.5 ($CH_3$), 43.0 ($CH_2$), 57.6 (CH), 106.3 (CH), 110.7 (CH), 117.7 ($CH_2$), 123.0 (CH), 123.6 (CH), 128.5 (CH), 131.6 (CH), 132.9 (CH); HRMS (ESI) calcd for $C_{16}H_{15}NO_2 + H^+$ 254.1181, found 254.1175.

**2,3-Diallyl-3-phenylisoindolin-1-one (3t):** colorless oil (70 mg, 96%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 3.23 (d, 2H, $J$ = 6.8 Hz), 3.70 (dd, 1H, $J$ = 7.2 and 15.6 Hz), 4.07 (dd, 1H, $J$ = 5.6 and 15.6 Hz), 4.87−5.07 (m, 4H), 5.13−5.24 (m, 1H), 5.69−5.79 (m, 1H), 7.08 (d, 1H, $J$ = 7.2 Hz), 7.16 (d, 2H, $J$ = 6.4 Hz), 7.25−7.31 (m, 3H), 7.38−7.46 (m, 2H), 7.86 (d, 1H, $J$ = 7.2 Hz); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 38.3, 43.2, 70.4, 117.3, 119.6, 122.1, 123.3, 126.7, 128.0, 128.6, 131.0, 131.5, 131.9, 133.5, 139.7, 149.8, 168.7; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 40.9 ($CH_2$), 45.8 ($CH_2$), 119.9 ($CH_2$), 122.2 ($CH_2$), 124.7 (CH), 126.0 (CH), 129.3 (CH), 130.6 (CH), 131.2 (CH), 133.6 (CH), 134.5 (CH), 136.1 (CH). Anal. Calcd for $C_{20}H_{19}NO$ (289.15): C, 83.01; H, 6.62; N, 4.84. Found: C, 83.20; H, 6.50; N, 4.98.

**1-Benzyl-5-(5-methylthiophene-2-yl)pyrrolidin-2-one (3u):** colorless oil (35 mg, 50%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 2.01−2.08 (m, 1H), 2.38−2.53 (m, 5H), 2.64−2.72 (m, 1H), 3.64 (d, 1H, $J$ = 14.8 Hz), 4.62−4.65 (m, 1H), 5.09 (d, 1H, $J$ = 14.8 Hz), 6.61−6.62 (m, 1H), 6.66 (d, 1H, $J$ = 3.6 Hz), 7.17−7.19 (m, 2H), 7.26−7.34 (m, 3H); $^{13}$C NMR (54.6 MHz, $CDCl_3$) $\delta$ 15.4, 28.6, 30.1, 44.1, 56.9, 124.6, 126.0, 127.4, 128.4, 128.5, 136.5, 140.3, 142.0, 174.4; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 15.5 ($CH_3$), 28.6 ($CH_2$), 30.2 ($CH_2$), 44.1 ($CH_2$), 57.0 (CH), 124.6 (CH), 126.1 (CH), 127.5 (CH), 128.5 (CH), 128.6 (CH). Anal. Calcd for $C_{16}H_{17}NOS$ (271.10): C, 70.81; H, 6.31; N, 5.16. Found: C, 70.95; H, 6.12; N, 5.02.

**1-Benzyl-5-(2,4-dimethoxyphenyl)pyrrolidin-2-one (3w):** colorless oil (65 mg, 83%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 1.84−1.92 (m, 1H), 2.26−2.36 (m, 1H), 2.41−2.49 (m, 1H), 2.55−2.64 (m, 1H), 3.53 (d, 1H, $J$ = 14.4 Hz), 3.70 (s, 3H), 3.81 (s, 3H), 4.72 (dd, 1H, $J$ = 4.0 and 8.4 Hz), 5.03 (d, 1H, $J$ = 14.4 Hz), 6.45−6.47 (m, 2H), 6.92 (d, 1H, $J$ = 8.0 Hz), 7.09 (d, 2H, $J$ = 6.0 Hz), 7.22−7.28 (m, 3H); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 26.2, 30.2, 44.3, 55.2, 55.3, 56.3, 98.8, 104.0, 120.5, 127.2, 128.0, 128.3, 136.6, 158.2, 160.5, 175.7; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 26.3 ($CH_2$), 30.3 ($CH_2$), 44.4 ($CH_2$), 55.3 ($CH_3$), 55.4 ($CH_3$), 56.3 (CH), 98.9 (CH), 104.1 (CH), 127.3 (CH), 128.1 (CH), 128.4 (CH). Anal. Calcd for $C_{19}H_{21}NO_3$ (311.15): C, 73.29; H, 6.80; N, 4.50. Found: C, 73.11; H, 6.93; N, 4.34.

**1-Benzyl-5-(2,4,6-trimethoxyphenyl)pyrrolidin-2-one (3x):** colorless oil (72 mg, 85%); $^1$H NMR (200 MHz, $CDCl_3$) $\delta$ 1.85−2.75 (m, 4H), 3.49 (d, 1H, $J$ = 14.4 Hz), 3.63−3.70 (m, 6H), 3.81 (s, 3H), 4.79 (d, 1H, $J$ = 14.6 Hz), 5.14 (dd, 1H, $J$ = 4.8 and 9.6 Hz), 6.06 (s, 2H), 7.04−7.08 (m, 2H), 7.19−7.23 (m, 3H); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 23.4, 31.1, 44.3, 51.5, 55.2, 55.4, 90.0, 90.7, 108.0, 126.8, 127.9, 128.5, 137.1, 160.9, 175.5. $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 23.5 ($CH_2$), 31.9 ($CH_2$), 44.4 ($CH_2$), 51.6 ($CH_3$), 55.3 ($CH_3$), 55.5 ($CH_3$), 90.1 (CH), 90.8 (CH), 126.9 (CH), 127.9 (CH), 128.0 (CH), 128.6 (CH); HRMS (ESI) calcd for $C_{20}H_{23}NO_4 + H^+$, 342.1705; found, 342.1700.

**1-Benzyl-1*H*-pyrrol-2(5*H*)-one (4).** Colorless oil (25 mg, 57%) and identified by spectral comparison with literature data.[24]

**1,10b-Dihydropyrido[2,1-*a*]isoindol-6(4*H*)-one (6a).** Compound **6a** was synthesized as an off-white solid (72 mg, 78%) according to the literature procedure.[25a]

**10b-Phenyl-1,10b-dihydropyrido[2,1-*a*]isoindol-6(4*H*)-one (6b).** Compound **6b** was prepared similarly as above: off-white solid (105 mg, 80%); $^1$H NMR (400 MHz, $CDCl_3$) $\delta$ 2.34−2.41 (m, 1H), 3.43−3.53 (m, 2H), 4.72−4.79 (m, 1H), 5.70−5.75 (m, 1H), 5.91−5.96 (m, 1H), 7.21−7.35 (m, 6H), 7.43−7.49 (m, 2H), 7.92−7.94 (m, 1H); $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ 33.0, 38.2, 64.6, 121.6, 122.4, 123.9, 124.4, 126.0, 127.8, 128.1, 128.8, 130.5, 131.9, 138.6, 151.9, 167.1; $^{13}$C DEPT-135 NMR (100 MHz, $CDCl_3$) $\delta$ 33.0 ($CH_2$), 38.2 ($CH_2$), 121.6 (CH), 122.4 (CH), 123.9 (CH), 124.4 (CH), 126.0 (CH), 127.8 (CH), 128.1 (CH), 128.8 (CH), 131.9 (CH). Anal. Calcd for $C_{18}H_{15}NO$ (261.12): C, 82.73; H, 5.79; N, 5.36. Found: C, 82.87; H, 5.99; N, 5.21.

**1,2,3,4-Tetrahydropyrido[2,1-*a*]isoindol-6(10b*H*)-one (7a).** Compound **7a** was synthesized as a colorless oil (90 mg, 95%) according to the literature procedure[25a] and identified by spectral comparison with literature data.[25b]

**10b-Phenyl-1,2,3,4-tetrahydropyrido[2,1-*a*]isoindol-6-(10b*H*)-one (7b).** Prepared similarly as above. Off-white solid (125 mg, 96%). The product was and identified by spectral comparison with literature data.[19a]

**Experimental Procedure To Study the Elimination vs Substitution Reaction for Succinamidal.** To a solution of succinamidal **2d** (24 mg, 0.125 mmol) in 2 mL of DCE were added under an argon atmosphere 2-methylthiophene (25 $\mu$L, 0.25 mmol), triphenylmethane (7.3 mg, 0.03 mmol) as internal standard, and **1** (1.3 mg for 1 mol % loading). The reaction mixture was stirred at 80 °C. At different time interval 100 $\mu$L of aliquot was taken out, solvent was removed under reduced pressure, and $^1$H NMR was recorded using $CDCl_3$ as NMR solvent.

**X-ray Crystallographic Analysis of 1 and 3r.** The X-ray diffraction intensity data for all compounds were collected at 293K

using a CCD diffractometer (graphite-monochromated Mo K$\alpha$ radiation, $\lambda$ = 0.71073 Å). The structures were solved by direct methods with SHELXS-86 or SHELXS-97 and refined with SHELXL-97. The non-hydrogen atoms were refined anisotropically. Hydrogen atoms were refined isotropically.

**1** (CCDC 805407): $C_{10}H_{15}Cl_8IrO_5Sn_3$, Fw = 1047.17, monoclinic, $P2_{1/a}$ (No. 14), $a$ = 9.5026(4) Å, $b$ = 34.1358(13) Å, $c$ = 9.7418(4) Å, $\beta$ = 116.9220(9)°, $U$ = 2817.6(2) Å$^3$, $Z$ = 4, $D_c$ = 2.468 mg/m$^3$, $\mu$ = 8.112 mm$^{-1}$, $F$(000) =1912, GOF = 1.299.

**3r** (CCDC 805408): $C_{15}H_{13}NOS$, Fw = 255.33, orthorhombic, $Pc2_1n$ (No. 33), $a$ = 5.985(5) Å, $b$ = 7.913(5) Å, $c$ = 27.626(5) Å, $U$ = 1308.3(14) Å$^3$, $Z$ = 4, $D_c$ = 1.296 mg/m$^3$, $\mu$ = 0.234 mm$^{-1}$, $F$(000) = 536, GOF = 1.06.

## ■ ASSOCIATED CONTENT

### Supporting Information

Spectra for all new compounds; NMR experiments; X-ray crystallographic data for **1** and **3r** (CIF). This material is available free of charge via the Internet at http://pubs.acs.org.

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### Notes

The authors declare no competing financial interest.

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## ■ DEDICATION

†Dedicated to the fond memory of Prof. B. D. Gupta (1949−2011), Chemistry Department, IIT Kanpur.

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