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A highly efficient catalyst, Cu-doped poly (N-[3-(dimethylamino)propyl]-methacrylamide, Acrylic Acid, N-vinyl Imidazole) (PDAI) magnetic carbon aerogel (CPIMCA), was successfully employed for the synthesis of Isatin nitrone derivatives. The 3-D porous CPIMCA catalyst demonstrated outstanding performance through the application of a coupling reaction between Isatin oxime and phenylboronic acid-derivatives, providing up to 98% of the required nitrone derivatives. Remarkably, the incorporation of copper within the polymeric structure of the magnetic carbon aerogel exhibited a significant influence on catalytic activity, even at a low overall copper content of approximately 2%. This was confirmed through EDX elemental mapping analysis, further establishing the competence of the catalyst for catalytic reactions. Comparative studies with Cu(OAc)$_2$ catalyst revealed that CPIMCA outperformed it, providing a notable 10-15% increase in product yield. This superior performance can be attributed to the unique synergistic effect of copper, iron, and carbon aerogel as the polymeric matrix, highlighting the exceptional capabilities of CPIMCA as a catalyst.

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Abstract A highly efficient catalyst, Cu-doped poly (N-[3-(dimethylamino)propyl]-methacrylamide, Acrylic Acid, N-vinyl Imidazole) (PDAI) magnetic carbon aerogel (CPIMCA), was successfully employed for the synthesis of isatin nitrones derivatives. The 3-D porous CPIMCA catalyst demonstrated outstanding performance through the application of a coupling reaction between isatin oxime and phenylboronic acid-derivatives, providing up to 98% of the required nitrone derivatives. Remarkably, the incorporation of copper within the polymeric structure of the magnetic carbon aerogel exhibited a significant influence on catalytic activity, even at a low overall copper content of approximately 2%. This was confirmed through EDX elemental mapping analysis, further establishing the competence of the catalyst for catalytic reactions. Comparative studies with Cu(OAc)2, catalyst revealed that CPIMCA outperformed it, providing a notable 10-15% increase in product yield. This superior performance can be attributed to the unique synergetic effect of copper, iron, and carbon aerogel as the polymeric matrix, highlighting the exceptional capabilities of CPIMCA as a catalyst.

Keywords Catalytic coupling reaction, Recyclable magnetic catalyst, Carbon Aerogel, Iron, Copper-doping.

Selective and efficient synthesis of heteroatomic compounds plays a crucial role in the preparation of various pharmaceuticals, agrochemicals, and functional materials. The synthesis of many different organic molecules depends on catalytic coupling processes, which have attracted a lot of interest in the field of organic chemistry.1-4 These processes, which result in the formation of new carbon-carbon or carbon-heteroatom bonds, provide an adaptable and successful method for developing complex compounds. Among the different approaches, catalytic methods have gained considerable attention due to their environmentally friendly nature and mild reaction conditions. Coupling and cross-coupling reactions use metal-based catalysts such as palladium (Pd), platinum (Pt), and rhodium (Rh), and some reactions specifically use heavy metals like mercury (Hg), lead (Pb), or cadmium (Cd), which can pose environmental and health risks and are highly expensive and non-recoverable.5-14 In coupling reactions, the metal catalysts can be prone to poisoning or deactivation by various species present in the reaction mixture.14 The release of these metals or their derivatives into the environment during catalytic processes can be harmful to ecosystems and human health. Making the reaction cost-prohibitive and not favorable to the environment is another factor. The most adverse disadvantage of using heavy catalysts is that they are not recoverable and can be used only a single time.15,16 Some nano techniques are used to deal with the disposed metals, such as nanoparticles, nanotubes, and carbon-based materials.17-19 To enhance the efficiency and selectivity of these reactions, studies have focused on acquiring advanced catalysts that exhibit remarkable catalytic properties and will also be more eco-friendly.20,21

Carbon aerogels are highly porous materials with a three-dimensional network structure, providing a large surface area and excellent stability. In recent years, carbon-based catalysts have emerged as more promising candidates for organic transformations owing to their unique properties and tunable reactivity. Vinyl-based hydrophilic co-monomers N-[3-(dimethylamino)propyl]-methacrylamide (DMAPMA), Acrylic Acid (AAc), and N-vinyl Imidazole (NVI) make a promising polymer (referred as-PDAl). The presence of polar functional groups in vinyl-based monomers, such as amine or carboxyl groups, imparts hydrophilic properties to the resulting aerogel.22 Notably, DMAPMA can be polymerized readily in the aqueous medium with other monomers, while acrylic monomers are making polymers more flexible and controlling hardness, and in N-vinyl imidazole, the imidazole moiety affords hydrophilicity to the resulting hydrogel; also, imidazole groups have the ability to create coordination bonds with metal ions, resulting in the formation of metal-polymer complexes.23-26

Moreover, allowing the incorporation of copper and iron into carbon aerogels adds the advantages of facile separation and recyclability, minimizing the environmental impact and improving the overall sustainability of the process. Interestingly, copper activates C-H bonds, and iron makes the carbon aerogel magnetic, enabling multiple cycles of catalytic reactions by recovering it after reaction completion. Copper itself is a proficient catalyst, so incorporation of copper is a greener strategy because it is economical and one of the most abundant elements on earth. Compared to expensive metal catalysts, copper catalysts often work in less adverse conditions. It is potentially selective towards C-N, C-C, and C-O bonds by varying the reaction conditions in different coupling reactions.27-31 A variety of schemes are already known for the synthesis of isatin nitrones using different reaction conditions. Chen et al. and Mo et al.32,33 had synthesized nitrene product
using Cu(OAc)$_2$ as catalyst in different solvents to select the best solvent to get the better yield. To get more better results, the primary goal of this research is to create a magnetic carbon aerogel catalyst with copper doping and evaluate the catalytic performance for the selective N-arylation reaction of Isatin oxime with phenylboronic acid derivatives. The integration of copper and iron into the carbon aerogel matrix provides a synergistic effect, combining the unique properties of carbon aerogels with the catalytic activity of copper. To achieve this goal, catalyst synthesis was carried out via the copolymerization procedure and the adsorption of Fe and Cu inside the polymeric structure was done via the in-situ adsorption method. This synthetic method produces a high-quality hydrophilic copper- and iron-adsorbed hydrogel, which was then lyophilized and further carbonized to produce the Cu-doped PDAI magnetic carbon aerogel (CPIMCA). In addition, morphological and chemical characterization of synthesized catalyst was done by various analytical techniques, such as FTIR, vibration sample magnetometry (VSM), X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive X-ray (EDX), transition electron microscopy (TEM), and X-ray photoelectron spectroscopy (XPS). Overall, this research presents an innovative approach to developing a highly efficient and recyclable catalyst for catalytic coupling reactions. The unique combination of copper and magnetic carbon aerogels offers significant potential for the advancement of catalysis, enabling the synthesis of complex organic molecules with improved efficiency and selectivity.

**Chemicals**

$N$-[3-(dimethylamino)propyl]-methacrylamide (DMAPMA), Acrylic Acid (AAc), $N$-vinyl Imidazole (NVI), ammonium persulphate (APS), N,N,N',N'-tetramethyl ethylene diamine (TEMED), Ferric(III) chloride ($FeCl_3\cdot 6H_2O$), ferrous(II) sulphate ($FeSO_4\cdot 7H_2O$), Copper(II) chloride ($CuCl_2$), Isatin, boronic acid derivatives, anhydrous Na$_2$SO$_4$, pyridine, hydroxyl amine hydrochloride ($NH_2OH\cdot HCl$), nitrogen-gas, sodium hydride (NaH), ethanol, methanol, dimethylformamide (DMF) and distilled water is used throughout the reaction.

**Synthesis of Cu-doped PDAI Magnetic Carbon Aerogel (CPIMCA)**

PDAI hydrogel was created by the free radical copolymerization method, in which the monomers were polymerized through vinyl bonds, leading to the formation of a polymeric matrix. In the synthetic procedure of PDAI hydrogel, firstly the monomers- DMAPMA, AAc, and $N$-vinyl imidazole were mixed sequentially in the concentration 1.44 ml, 1.12 ml, and 0.24 ml, respectively. All three vinyl monomers were dissolved in 1.56 ml of distilled water in an ice-bath. Shortly after thoroughly mixing the monomers with the distilled water, 100 µl of APS was added as initiator which initiates the cleavage of vinyl bonds in monomers, making the free radicals available for chain initiation, after homogenizing the monomeric mixture to begin the polymerization process, 2.5 µl of accelerator TEMED was blended consistently that accelerates the reaction for chain propagation maintaining the temperature < 10 °C. After 14 h of heating in an oil bath, the PDAI hydrogel was finally produced. The formed hydrogel was washed with water three times to pull out the remaining unreacted monomers that might still be present. Which was then followed by drying at 60 °C in the oven. Once the hydrogel has dried, it is magnetized by being submerged in a solution containing both FeSO$_4\cdot 7H_2O$ and FeCl$_3\cdot 6H_2O$ for adsorption of Fe ions. Following alkali treatment, the magnetized hydrogel was kept in an aqueous ammonia solution overnight for the reduction of Fe$^{3+}$ and Fe$^{2+}$. After the adsorption of the Fe-ion onto the surface of the hydrogel, it is next dipped in a solution of CuCl$_2\cdot 2H_2O$ for the adsorption of copper. All three ions, FeSO$_4\cdot 7H_2O$, FeCl$_3\cdot 6H_2O$, and CuCl$_2\cdot 2H_2O$, are taken in the ratio 1:2:1. Once the Cu and Fe adsorption was complete, the PDAI Hydrogel was freeze-dried or lyophilized at -71 °C for 8 h in order to evaporate all the hydro inside it. This is the most important step in the process of aerogel formation in which all the water from the polymeric matrix is removed while maintaining the internal structure of...
the aerogel. Subsequently, to form a carbon magnetic aerogel, the prepared aerogel was treated at 800 °C for 8 h in an inert atmosphere with heating rate of 4 °C/min in a tube furnace for carbonization. During carbonization, the adsorbed ions of Cu and Fe were incorporated inside the carbon structure of the polymer. Hence, carbonization also facilitates Cu and Fe inside the polymeric chain, this, in turn, leads to the electrostatic binding of copper and iron to the carbon structure, which, consequently, makes it possible for copper to be doped onto PDAl carbon aerogel.

**Instrumental Techniques for Characterization**

The morphology of the synthesized CPIMCA was analyzed using X-Ray Diffraction spectroscopy by utilizing a Rigaku SmartlabSE X-Ray diffractometer having voltage of 40 kV and a current of 30 mA, with a Cu-Kα radiation in 2θ range 10-60°. For surface morphology Transition Electron Microscopy (TEM) images were captured by a JEOL JEM-2100F electron microscope supplying a 200 kV accelerating voltage. The CPIMCA sample was drop-cast onto a copper-grid after dispersion and allowed to dry at room temperature. Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (EDX) was performed by ZEISS, Merlin Compact which has tungsten electron and it worked at a voltage of 20 kV, with Magnification K × 5 to K × 10. Prior to proceeding the sample was coated with a thin layer of gold in a gold sputtering machine. For chemical morphology sample was determined by Fourier transform Infrared spectroscopy FTIR using Perkin Elmer-Spectrum two FTIR analyser in the range 400-4000 cm⁻¹ range. First the sample was mixed with dry KBr, before keeping in the analyser. For the objective of obtaining magnetic measurements, vibrating sample magnetometry (VSM) was carried out using the Microsense-ADE-EV9 magnetometer with a magnetic field strength of up to 2.2 tesla. The magnetic measurement range was -10000 Oe to +10000 Oe. Furthermore, X-ray photoelectron spectroscopy (XPS; ULVAC-PHI, INC., PHI 5000 Versa Probe III Scanning XPS Microprobe) has been utilized to demonstrate the elemental composition, oxidation state, and binding energy. The NMR spectra of the products formed by the CPIMCA catalyzed reactions was recorded by Bruker 400 MHz instrument in DMSO-d₆, or MeOD with TMS as internal standard.

**Catalytic activity of as-synthesized CPIMCA for N-arylation of Isatin**

This organic transformation was a three-step mechanistic process, at the beginning Isatin was converted to its methyl Isatin derivative summarized in Scheme 2. In the very first step of the mechanism, deprotonation of Isatin through alkylation was implemented by the treatment of Isatin (1) with chloromethane in the presence of NaH (1.5 equivalent) in DMF for 6 h at 0 °C, resulting to the formation of 1-methylisatin (2). In the proceeding step the generated 2 reacted with NH₂OH.HCl (1.2 equivalent) in ethanol to produce 1-methylisatin oxime (3) at 80 °C for 6 h. In order to obtain the final product Isatin Nitrone (5a-g), the previously synthesized 3 underwent a reaction with phenylboronic acid derivatives 4(a-g) (1.2 equivalent) in methanol along with pyridine at 25 °C for 16 h, utilizing (0.1 equivalent) Cu(OAc)₂ and (0.1 equivalent) CPIMCA as catalyst detailed in Scheme 2.
Scheme 2 Arylation of isatin oxime via catalytic coupling reaction with phenylboronic acid and comparative yield of nitrone products.

**Result and discussion**

**Characterizations**

The functional group and chemical bonds of the CPIMCA were investigated by FTIR in the range 4000-500 cm⁻¹. The results of the FTIR reveal the peaks at 3443 cm⁻¹, 2920 cm⁻¹, 1623 cm⁻¹, 1265 cm⁻¹ and 1094 cm⁻¹ were observed in overall spectra, engraved in Fig. 1a. The detected peak at 3443 cm⁻¹ corresponds to aliphatic amine (NH₂). The peak at 2920 cm⁻¹ represents the C–H component. Adsorption at 1623 cm⁻¹ is attributed to C=C. The peak at 1265 cm⁻¹ corresponds to C–N from the aromatic amine. While peak at 1094 cm⁻¹ is attributed to C=C.  It is noticeable that carbonaceous components C–C and C=C are highly present in the structure, and after carbonization some compounds like hydroxyl, carboxyl will be eliminated partially or completely. The magnetic property of CPIMCA was analyzed with the help of VSM technique. The sample was kept in the field range from -10000 Oe to +10000 Oe at room temperature. From the hysteresis curve of the magnetization measurement of CPIMCA sample, it was observed that the sample is supermagnetic in nature and displaying an S-like curve, depicted in Fig. 1c. The saturation magnetization value (Ms) value of the sample is 40.94 emu/g. Carbonization increases the magnetic saturation value and decreases the coercivity of the sample resulting in increasing the magnetization of the sample. 

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The magnetic behavior can also be identified physically, as on applying external magnetic field the sample is strongly attracted towards the magnet Fig.1c.; i.e. admirably useful for the catalytic applications of the magnetic sample. XRD was performed for crystalline and amorphous nature of the sample. As demonstrated in Fig.1b, the peaks at 2θ = 22.16°, 38.80°, and 43.70° corresponds to the lattice planes [101], [200], and [111] respectively. The peak at 22.16° shows that the carbon skeleton of carbon aerogel is semi-crystalline in nature. While the peak at 38.80° corresponds to Fe ions and peak at 43.70° is consistent with Cu ions. From the XRD observations it can be concluded that copper and iron are completely encapsulated in the semi-crystalline Cu-doped magnetic carbon aerogel framework.44,45

SEM, TEM, and EDX were utilized to investigate the shape, morphology, particle size, elemental analysis, and particle length of the as-synthesized catalyst CPIMCA. In accordance with Fig. 2a and Fig. 2b. SEM images at 10 µm and 2 µm demonstrated that the CPIMCA has an interconnected 3-D network structure, which reflects the proper binding of vinyl monomers via covalent interactions, which is further confirmed by TEM images. Alongside this, it is admirably porous in nature and has the incorporation of metal ions inside the carbon framework of the aerogel.46,47 It can also be confirmed that copper and iron particles are uniformly distributed throughout the polymeric structure. It is noticeable that after carbonization in an inert atmosphere, the 3-D polymeric structure remains porous. Furthermore, the elemental composition was confirmed by EDX-mapping, and the presence of Cu and Fe can also be confirmed by the EDX image as depicted in Fig. 2f. The SEM -EDX mapping images explains about that the presence of the carbon component covers the maximum area as the sample was carbonized shown in Fig. 2g., along with this copper and iron are embedded consistently in the structure exhibited in Fig. 2j and Fig.2k, respectively. It revealed that copper is present in 1.68% and Fe is present in the structure in 7.3%. It also displayed that carbon is present in the highest concentration, i.e., 49.94%, which indicates that the sample is carbonized correctly. All other elements O and N are also spread homogeneously as depicted in Fig. 2h, and Fig. 2i. Interestingly, the encapsulation of the metals effectively increases the surface area and overall porosity of the material.

Moreover, the typical TEM image discloses more about the surface morphology of the CPIMCA can be seen in Fig.2c. At 20 nm directs that 3-D packed aerogel have Cu and Fe incorporated dense network structure.48,49 It is detected that the mesoporous particles are elliptical and spherical in shape having a diameter range 12-30 nm. Additionally, the material possesses lattice fringes in a single direction with a D-spacing of 0.46 nm as illustrated in Fig.2d. Regarding Fig.2f. SAED pattern interprets the diffraction rings corresponding to the plane [101], [200], and [111] on the lattice planes.

XPS analysis is used to predict the surface chemical composition and oxidation state of the present elements inside the material. The XPS survey exhibits five leading peaks Cu1s at 932 eV, N1s at 400 eV, O1s at 532 eV, Fe2p at 724 eV, and Cu2p at 974 eV as shown in Fig.3a. Further Cu1s spectrum has been deconvoluted in three peaks, first is C-metal peak that appears at 284 eV, the peak at 284.63 eV that corresponds to (sp2), and 285.55 eV is for C-C (sp3), represented in Fig.3b. The N1s data is depicted in Fig.3d. Typically shows the deconvoluted peaks at 397.39 eV and 400.18 eV correlate to N-metal and C-N, respectively. The Data for O1s is elucidated in Fig.3c. matching with the C=O at 531.2 eV, and C-O=C at 532.9 eV. The Cu2p spectrum could fit three peaks and one satellite peak, that are corresponding to Cu2p3/2 at 936 eV, Cu2p1/2 at 950.6 eV, CuO 535 at 972.7 eV, and satellite peak at 997 eV due to extra energy loss, illustrated in Fig.3e. This data confirms that copper is present in two oxidation states.50,51 The Fe2p data has been split into two peaks 712.17 eV which is assigned to Fe2+ and 725.23 eV which belongs to Fe2+ supporting the fact that iron is present in two oxidation states pictured in Fig.3f. Hereafter, carbonization the copper is completely encapsulated inside the structure.52,53

Catalyst investigation in coupling reaction

With the objective of examining the catalytic activity of the CPIMCA as catalyst, the organic transformation has been performed in which coupling reaction of Isatin Oxime with boronic acid derivatives was conducted to generate Isatin nitrone derivatives. To the best of our knowledge, this is the first time that metal doped magnetic carbon aerogel is used as a catalyst for the formation of N-aryl nitrones product by N-arylation. In the preparative procedure of Isatin nitrone, initially Isatin was converted to 1-methylisatin, for that Isatin, NaH and chloromethane was taken in the mole ratio 1:2:1, Isatin was mixed in DMF, followed by addition of heterogeneous basic deprotonating agent NaH the reaction mixture left for 6 h at 0 °C, the reaction was monitored by TLC and LC/MS. Once the reaction is completed, the mixture is diluted with EtOAc and washed with dil. HI, finally providing 1-methylisatin giving 91% yield. Furthermore, to facilitate the Isatin Oxime, a slurry of 31.77 mmol (5.28 g) of 1-methylisatin in 50 ml ethanol, mixed with 47.66 mmol (3.31 g) of NH4OH.
HCl in 6 ml H$_2$O left for stirring at 80 °C for 6 h. The resulting mixture was a greenish yellow paste, triturated with H$_2$O, filtered, and washed with H$_2$O, subsequent product left for drying overnight in vacuum oven. In the proceeding final step for N-arylation 1 equivalent (0.3 mmol) of 1-methylisatin oxime along with phenyl boronic acid derivatives (1.2 equivalent) was taken. The reaction commenced with the addition of 0.1 equivalent catalyst CPIMCA (0.002 equivalent Cu as 2% in CPIMCA catalyst) in the presence of anhydrous Na$_2$SO$_4$, pyridine, and polar aprotic

![Figure 2 SEM](image)

**Figure 2** SEM (a) 10 µm (b) 2µm, TEM (c) 20 nm, (d) 1 nm, SAED (e) 1Å (f) EDX image, elemental mapping images; (g) Carbon, (h) Oxygen, (i) Nitrogen, (j) Iron (k) Copper of CPIMCA.
solvent methanol, the reaction mixture was stirred for 16 h at 25 °C. Methanol was used as solvent as methanol is green solvent and it can be renewable, according to an article by Byrne et al., methanol is the solvent that is recommended and can be renewable while, solvents like DCM, DCE, toluene etc. are not safe to use as these cannot be renewable and reused. Therefore, using a green solvent is a greener approach for this reaction procedure. The monitoring of reaction was done by using TLC and HRMS, additionally the product formation was confirmed by 1H-NMR. The compound was simply removed by flash column chromatography. The catalytic investigation was conducted by comparing the catalytic activities of Cu(OAc)₂ and CPIMCA. The same set of reactions was done by using Cu(OAc)₂ as catalyst (Scheme 2). Remarkably, it is noticeable that produced nitrone derivatives gave up to 98% yield by using CPIMCA catalyst. As can be seen in table in scheme-2, CPIMCA had significantly higher levels of catalytic activity than Cu(OAc)₂. The yield of the products that had been produced using the CPIMCA catalyst improved by 10-15%.

<table>
<thead>
<tr>
<th>Sr No.</th>
<th>R Value for Phenyl boronic acid</th>
<th>Product</th>
<th>yield % using Cu(OAc)₂</th>
<th>Yield % using CPIMCA catalyst (our catalyst)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4a: R=H</td>
<td></td>
<td>60</td>
<td>80</td>
</tr>
<tr>
<td>2</td>
<td>4b: R=3-F</td>
<td></td>
<td>46</td>
<td>62</td>
</tr>
<tr>
<td>3</td>
<td>4c: R=2-F</td>
<td></td>
<td>50</td>
<td>65</td>
</tr>
<tr>
<td>4</td>
<td>4d: R=4-Me</td>
<td></td>
<td>40</td>
<td>59</td>
</tr>
<tr>
<td>5</td>
<td>4e: R=2-Me, 4-F</td>
<td></td>
<td>80</td>
<td>95</td>
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<tr>
<td>6</td>
<td>4f: R=4-F</td>
<td></td>
<td>82</td>
<td>98</td>
</tr>
<tr>
<td>7</td>
<td>4g: R=4-SO₂O₂NH-tBu</td>
<td></td>
<td>44</td>
<td>60</td>
</tr>
</tbody>
</table>

Table 1 Isatin Nitrone derivatives using different catalysts.

Figure 3 XPS of CPIMCA (a) Survey, (b) Cls, (c) O1s, (d) N1s, (e) Cu2p, (f) Fe2p

Figure 4 Coupling reaction for synthesis of isatin nitrone catalytic activity of CPIMCA.
Our catalyst showed good catalytic activity as the three-dimensional porous structure of carbon aerogel provides a large surface area and an interconnected porous network; the carbon framework allows the proper uniform agglomeration in the structure consequently have more active catalytic sites. Moreover, incorporation of Cu and Fe ions in the carbon framework shows synergistic effects along with carbon, increases its catalytic performance, as it fastens the movement of electrons, also which allows the better dispersion and accessibility of copper ions, while copper ions in Cu(OAc)₂ are not easily accessible, which can obstruct the catalytic activity. Fe makes the aerogel magnetic which enables easy separation from the reaction mixture by an external magnetic field. This makes it a sustainable catalyst and reduces compound waste.

**1-methyl-2-oxo-1′-phenylindolin-3-imine oxide** (5a): The compound was brownish yellow solid, 37 mg, 62% yield. 1H NMR (400 MHz, DMSO-d₆): δ 8.304-8.286 (d, J = 7.2 Hz, 1H), 7.503-7.436 (m, 3H), 7.313-7.293 (d, J = 8.0 Hz, 2H), 7.145-7.090 (q, 2H), 7.21 (d, J = 5.2 Hz, 1H). 1H NMR (100 MHz, DMSO-d₆): δ 25.95, 109.11, 116.26, 116.73, 122.72, 124.07, 125.24, 126.10, 126.17, 132.97, 134.12, 135.35, 134.41, 142.38, 147.29, 158.74, 160.24, 162.68. 13C NMR (400 MHz, DMSO-d₆): δ 112.009; HRMS (ESI) m/z calcd for C₁₉H₁₉FN₂O₂: found 271.0874.

**N-(4-fluorophenyl)-1-methyl-2-oxindolin-3-imine oxide** (5f): The compound is orangish yellow solid, 52 mg, 98% yield. 1H NMR (400 MHz, DMSO-d₆): δ 8.287-8.307 (d, J = 8.0, 1H), 7.937-7.958 (d, J = 8.2, 2H), 7.770-7.783 (d, J = 5.2 Hz, 2H), 7.496-7.534 (t, J = 8.0 Hz, 1H), 7.16-7.20 (m, 2H), 3.13 (s, 3H), 1.13 (s, 9H). 1C NMR (100 MHz, DMSO-d₆): δ 26.001, 29.793, 53.541, 108.91, 117.35, 122.57, 124.055, 124.947, 127.09, 132.696, 134.665, 142.476, 145.712, 148.166, 158.782; HRMS (ESI) m/z calcd for C₁₉H₁₉FN₂O₂: found 288.1319.

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Supporting Information

YES (this text will be updated with links prior to publication)

CRediT author statement

DM: Writing - Original Draft, Methodology, Formal analysis; NY and RP: Investigation, Formal analysis; and VM: Conceptualization, Data Curation, Formal analysis, Methodology, Supervision

Conflict of Interest

The authors declare no conflict of interest.

References


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Table-S1 Mole percentage feed ratio of monomers and other materials with respect to monomers.

<table>
<thead>
<tr>
<th>Distilled water</th>
<th>DMAPMA</th>
<th>AAc</th>
<th>NVI</th>
<th>APS</th>
<th>TEMED</th>
</tr>
</thead>
<tbody>
<tr>
<td>320 mole %</td>
<td>30 mole %</td>
<td>60 mole %</td>
<td>10 mole %</td>
<td>3.2 mole %</td>
<td>7.1 mole %</td>
</tr>
</tbody>
</table>

Figure-S1 EDX- Mapping survey image of CPIMCA catalyst
$^1$H-NMR and LCMS data of Isatin Nitrone derivatives

5a: 1-methyl-2-oxo-N-phenylindolin-3-imine oxide

Solvent - DMSO-$d_6$

Figure- S2 $^1$H NMR of compound 5a.

Figure- S3 $^{13}$C NMR of compound 5a.
Figure-S4 HRMS of compound 5a.

5b: N-(3-fluorophenyl)-1-methyl-2-oxoindolin-3-imine oxide

Solvent- DMSO-\textit{d}_6

Figure-S5 \textit{\textsuperscript{1}H NMR of compound 5b.}
Figure S6  $^{13}$C NMR of compound 5b.

Figure S7  $^{19}$F NMR of compound 5b.
5c: N-(2-fluorophenyl)-1-methyl-2-oxoindolin-3-imine oxide

Solvent- DMSO-\textit{d}_6

Figure-S8 HRMS of compound 5b.

Figure- S9 $^1$H NMR of compound 5c.
Figure S10 $^{13}$C NMR of compound 5c.

Figure S11 $^{19}$F NMR of compound 5c.
**Figure S12** HRMS of compound 5c.

5d: 1-methyl-2-oxo-N-(p-tolyl) indolin-3-imine oxide

Solvent: DMSO-\(d_6\)

**Figure S13** \(^1\)H NMR of compound 5d.
Figure- S14 $^{13}$C NMR of compound 5d.

Figure- S15 HRMS of compound 5d.
5e: N-(4-fluoro-2-methylphenyl)-1-methyl-2-oxoindolin-3-imine oxide (Solvent- DMSO-d$_6$)

**Figure-S16** $^1$H NMR of compound 5e.

**Figure-S17** $^{13}$C NMR of compound 5e.
Figure S18 $^{19}$F NMR of compound 5e.

Figure S19 HRMS of compound 5e.
5f: N-(4-fluorophenyl)-1-methyl-2-oxoindolin-3-imine oxide (Solvent- DMSO-d6)

Figure- S20 $^1$H NMR of compound 5f.

Figure- S21 $^{13}$C NMR of compound 5f.
Figure- S22 $^{19}$F NMR of compound 5f.

Figure- S23 HRMS of compound 5f.
Figure S24 COSY spectra of compound 5f.

Figure S25 HSQC spectra of compound 5f.
5g: \(N\)-(4-(N-(tert-butyl)sulfamoyl)phenyl)-1-methyl-2-oxoindolin-3-imine oxide

Solvent- DMSO-\textit{d}_6

Figure- S26 \(^1\)H NMR of compound 5g.

Figure- S27 \(^{13}\)C NMR of compound 5g.
Figure- S28 HRMS of compound 5g.