Supplementary Information

Intramolecular alkene hydroamination and degradation of amidines: divergent behavior of rare earth metal amidinate intermediates

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Table of Contents

1. General information .................................................................................................................. S2
2. Synthesis of substrates............................................................................................................. S2
3. Screening of reaction conditions for the synthesis of 2a...................................................... S4
4. General procedures for synthesis of products................................................................. S4
5. General procedures for degradation of amidines.............................................................. S5
6. General procedures for Yttrium-Catalyzed tandem amino-exchange/cyclization of amidines
   with aminoalkenes.............................................................................................................. S5
7. Mechanistic Studies ............................................................................................................. S5
8. Characterization of substrates .......................................................................................... S7
9. Characterization of the products ....................................................................................... S17
10. References.......................................................................................................................... S24
11. Copies of $^1$H NMR and $^{13}$C NMR Spectra ............................................................... S25
Experimental Section

1. General information: All manipulations involving air- and moisture-sensitive compounds were performed under dinitrogen atmosphere, using Schlenk techniques or an Mbraun glovebox (Unilab Mbraun; < 0.1 ppm O₂, < 0.1 ppm H₂O). Toluene, hexane and tetrahydrofuran (THF) were taken from a solvent purification system (PS-400-5, Unilab Mbraun, Inc.). Nitriles, N-methyl allylamine and diallylamines were obtained commercially from Sigma Aldrich or J&K. Other allylamines were prepared according to literature procedures.¹ CDCl₃, THF-d₈ and benzene-d₆ were obtained from Cambridge Isotope.¹ H NMR and ¹³C NMR spectra of organic compounds were recorded on a JEOL ECA-400 NMR spectrometer (FT, 400 MHz for ¹H; 100 MHz for ¹³C) in CDCl₃ at room temperature. All chemical shift values are quoted in ppm referenced to an internal tetramethylsilane standard for ¹H NMR. Chemical shifts of ¹³C NMR are reported relative to CDCl₃ (δ 77.16). The following abbreviations were used to describe peak splitting patterns when appropriate: br = broad, s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet. Coupling constants (J) was reported in Hz unit. GC-MS analyses were measured on a Focus GC-ISQ MS instrument. High resolution mass spectrometry (HRMS) spectra were obtained on a micro TOF II Instrument using ESI ionization sources.

2. Synthesis of Substrates

2.1 General Procedure for Preparation of N-Allyl Amidines 1a-l, 1n-p, and 1al-ar.²

\[
\text{Ar-CN} + \text{R'}\text{N} + \text{AlCl}_3 \xrightarrow{\text{neat, 120 °C, 12 h}} \text{NH} \text{R'}\text{N} + \text{Ar-CN}
\]

In a glovebox, nitrile (5.0 mmol), AlCl₃ (5.0 mmol, 0.665 g) and N-substituted allylamine (6.0 mmol, 1.2 equiv) were charged into a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. The sealed tube was taken out from the glovebox, and heated at 120 °C for 12 h. After cooling down to room temperature, to the reaction mixture 25 mL of ice-water was added under vigorous stirring. Then, an aqueous NaOH solution was added until a pH of 14 was reached. The aqueous layer was extracted with ethyl acetate (3 × 30 mL). The combined organic extracts were dried over Na₂SO₄ and then the solvent was evaporated under reduced pressure. The residue was purified by flash column chromatography on silica gel with ethyl acetate/hexane as eluent silica gel chromatography to give the desired product.

2.2 General Procedure for Preparation of N-Allyl Amidines 1m, 1q-t, 1u and 1aa-ab.³

\[
\text{R-CN} + \text{R'}\text{N} + \text{AlCl}_3 \xrightarrow{n-\text{BuLi, 0 °C, 15 min}} \text{NH} \text{R'}\text{N} + \text{Ar-CN}
\]

A 2.5 M solution of n-BuLi in hexane (6.0 mmol) was added to a solution of amine (5.0 mmol) in anhydrous THF (10 mL) at 0 °C. After stirring for 15 min at 0 °C, the mixture was cooled down to -78 °C, to which a solution of nitrile (5.0 mmol) in anhydrous THF (10 mL) was added dropwise. The brown reaction mixture was slowly warmed to room temperature and continued to stir for 2 h, and then quenched by adding aq NH₄Cl. The two phases were separated and the aqueous layer was extracted three times with ethyl acetate. The combined organic extracts were dried over anhydrous Na₂SO₄ and then the solvent was evaporated under reduced pressure. The residue was purified by silica gel chromatography to give the desired product.
2.3 General Procedure for Preparation of Amidines 1ac-1ak.

A solution of AlMe$_3$ (2.0 M in toluene, 5.6 mmol) was added dropwise to a mixture of aromatic amine (2.2 mmol) and primary carboxamide (2.0 mmol) in anhydrous toluene (3 mL) at 0 °C. After stirring for 30 min at room temperature, the resulting mixture was then heated at 120 °C (oil bath temperature) for the given time. The mixture was cooled to 0 °C and diluted with CH$_2$Cl$_2$ (20 mL) and then slowly poured into ice-cold saturated NH$_4$Cl solution (4 mL). THF (15 mL) was added and the mixture was stirred for 30 min at room temp, then filtered through a Celite bed, dried with sodium sulphate, and concentrated to give the crude product. The residue was purified by silica gel chromatography to give the desired product.

2.4 General Procedure for Preparation of Amidines 1as.

The amide (5.0 mmol) was activated with triflic anhydride (6.5 mmol) and pyridine (15.0 mmol) for two hours at 0°C, after which time a solution of n-butylamine in dichloromethane (15.0 mmol) was added and allowed to react overnight at room temperature. The reaction was then diluted with CH$_2$Cl$_2$ and saturated aqueous NaHCO$_3$. The aqueous layer was diluted with CH$_2$Cl$_2$ (20 mL), and the combined organic layers were dried over MgSO$_4$ and concentrated under reduced pressure. Flash chromatography using MeOH : EtOAc (0 to 10% MeOH) afforded the desired product.
3. Screening of Reaction Conditions for the Synthesis of 2a

**Table S1. Optimization of Reaction Conditions**

<table>
<thead>
<tr>
<th>Entry</th>
<th>Catalyst</th>
<th>Amount (mol%)</th>
<th>Time/h</th>
<th>Yield(%) b</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>La[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>18</td>
</tr>
<tr>
<td>2</td>
<td>Sm[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>89</td>
</tr>
<tr>
<td>3</td>
<td>Gd[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>92</td>
</tr>
<tr>
<td>4</td>
<td>Yb[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>92</td>
</tr>
<tr>
<td>5</td>
<td>Y[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>&gt;99(99) c</td>
</tr>
<tr>
<td>6</td>
<td>Lu[N(SiMe3)2]3</td>
<td>5</td>
<td>12</td>
<td>93</td>
</tr>
<tr>
<td>7</td>
<td>Y[N(SiMe3)2]3</td>
<td>3</td>
<td>12</td>
<td>71</td>
</tr>
<tr>
<td>8</td>
<td>Y[N(SiMe3)2]3</td>
<td>1</td>
<td>12</td>
<td>16</td>
</tr>
<tr>
<td>9</td>
<td>Y[N(SiMe3)2]3</td>
<td>5</td>
<td>8</td>
<td>91</td>
</tr>
<tr>
<td>10</td>
<td>Y[N(SiMe3)2]3</td>
<td>5</td>
<td>4</td>
<td>63</td>
</tr>
<tr>
<td>11</td>
<td>KN(SiMe3)2</td>
<td>5</td>
<td>12</td>
<td>0</td>
</tr>
<tr>
<td>12</td>
<td>Yb(OTf)3</td>
<td>10</td>
<td>12</td>
<td>0</td>
</tr>
<tr>
<td>13</td>
<td>YbCp3</td>
<td>10</td>
<td>12</td>
<td>0</td>
</tr>
</tbody>
</table>

Conditions: a Reactions were carried out with 1a (0.50 mmol), toluene (2 mL) at 25 °C under N2. b Determined by 1H NMR using xylene as the internal standard. c Isolated yield in parentheses.

4. General Procedures for Synthesis of 2 and 3

In a glovebox, Y[N(SiMe3)2]3 (14.3 mg, 0.025 mmol, 5 mol%), toluene (2 mL), and amidine (0.50 mmol) were added to a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. After stirring for 12 h at room temperature, the reaction was quenched with water (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na2SO4. The pure 2-imidazoline and tetrahydropyrimidines was obtained by flash column chromatography on silica gel with ethyl acetate/TEA (20:1) as eluent.
5. General Procedures for Degradation of Amidines

In a glovebox, $Y[N(SiMe_3)_2]_3$ (14.3 mg, 0.025 mmol, 5 mol%), and amidine (0.50 mmol) were added to a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. After stirring for 12 h at room temperature (25 °C), the reaction mixture was quenched with 1 M HCl (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na$_2$SO$_4$. The pure nitrile and amine were obtained by flash column chromatography on silica gel with PE/EA (20:1) as eluent.

6. General Procedures for Yttrium-Catalyzed Tandem Reconstruction /Cyclization of Amidines with Aminoalkenes

$Y[N(SiMe_3)_2]_3$ (14.3 mg, 0.025 mmol), $N$-substituted allylamine (1 mL), and amidines (0.50 mmol) were charged into a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. After stirring for 12 h at room temperature, the reaction mixture was quenched with water (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na$_2$SO$_4$. The pure 2-imidazoline was obtained by flash column chromatography on silica gel with EA/TEA (20:1) as eluent.

7. Mechanistic Studies

7.1 Confirmation of Yttrium Amidinate Intermediates Involved in Tandem Y-Catalyzed Amino-Exchange/Cyclization of Amidines with Aminoalkenes.

7.1.1 Stoichiometric Reaction of Precatalyst $Y[N(SiMe_3)_2]_3$, with 1ao

$Y[N(TMS)_2]_3$ (Y-1) (570.1 mg, 1.0 mmol) and N-(2,6-dimethylphenyl)benzimidamide (672.9 mg, 3.0 mmol) were dissolved in 5 mL toluene. After stirring at room temperature for 24 hours, the white precipitate was filtered and washed by cold toluene (2 × 3 mL) and hexane (3 × 10 mL), and dried under reduced pressure, affording a white powder. Diffusion of toluene into a THF solution of the white powder afforded Y-2 as colorless crystals (1.1434g, 87%). $^1$H NMR (400 MHz, THF-$d_8$): $\delta$ 7.18-7.11 (m, 3H), 7.09-7.05 (m, 2H), 6.67-6.65 (m, 2H), 6.56-6.52 (m, 1H), 5.67 (d, $J = 1.8$ Hz, 1H), 2.08 (s, 6H). $^{13}$C NMR
(100 MHz, THF-\textit{d}_8): \delta 177.54, 149.29, 140.31, 132.63, 129.20, 128.03, 128.00, 127.45, 121.97, 19.76.

### 7.1.2 Determination of Molecular Structure of Y-2

X-ray diffraction data for Y-2 (CCDC 1831321) were collected on a SMART APEX CCD diffractometer (graphite-monochromated MoK\textalpha radiation, \phi-\omega scan technique, \( \lambda = 0.71073 \) Å). The intensity data were integrated by means of the SAINT program. SADABS was used to perform area-detector scaling and absorption corrections. The structure was solved by direct methods and was refined against \( F^2 \) using all reflections with the aid of the SHELXTL package. All non-hydrogen atoms were found from the difference Fourier syntheses and refined anisotropically. The H atoms were included in calculated positions with isotropic thermal parameters related to those of the supporting carbon atoms but were not included in the refinement. All calculations were performed using the Bruker Smart program.

CCDC 1831321 (complex Y-2) contains the supplementary crystallographic data for this paper. These data can be obtained free of charge via www.ccdc.cam.ac.uk/conts/retrieving.html (or from The Cambridge Crystallographic Data Centre, 12, Union Road, Cambridge CB2 1EZ, UK; fax: (+44)-1223-336033; or deposit@ccdc.cam.ac.uk).

### 7.1.3 Thermal Decomposition of Y-2

Y-2 (83.1 mg, 0.1 mmol) and toluene (3 mL) were added to a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. After heating at 100 °C for 5 days, the reaction mixture was quenched with water (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na\textsubscript{2}SO\textsubscript{4}. The pure nitrile and amine were obtained by flash column chromatography on silica gel with ethyl PE/EA (20:1) as eluent.

### 7.1.4 Reaction of Yttrium Amidinate Intermediate (Y-2) with Aminoalkene (6a)

A mixture of Y-2 (83.1 mg, 0.1 mmol) and 6a (1 mL) was charged into a Schlenk tube equipped with a magnetic stirring bar and a Teflon cap. After stirring for 5 days at 100 °C, the reaction mixture was quenched with water (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na\textsubscript{2}SO\textsubscript{4}. 2a (29.8 mg, 57%) was obtained by flash column chromatography on silica gel with ethyl EA/TEA (20:1) as eluent.

### 7.1.5 Examination of Catalytic Activation of Y-2 on the Tandem Amino-Exchange/ Cyclization of Amidine with Aminoalkene

Y-2 (41.5 mg, 0.05 mmol, 20 mol%), 1ao (56.1 mg, 0.25 mmol) and 6a (1.0 mL) were added to a
Schlenk tube equipped with a magnetic stirring bar and a Teflon cap in sequence. After heating for 5 days at 100 °C, the reaction mixture was quenched with water (2 mL), and extracted with ethyl acetate (3 × 5 mL). The organic layers were combined, and dried over anhydrous Na₂SO₄ and concentrated. 2a (37.0 mg, 85%) was obtained by flash column chromatography on silica gel with ethyl EA/TEA (20:1) as eluent.

7.2 ¹H NMR-Monitoring the Reaction of 1ap with 6a

In a glovebox, Y-1 (20.5 mg, 0.036 mmol) was weighed into a NMR tube equipped with a Teflon valve (J-Young). Then, a mixture of 1ap (0.40 mmol) and 6a (0.60 mmol) in benzene-d₆ (0.5 mL) was added via syringe. The ensuing catalytic reaction was monitored by ¹H NMR spectroscopy. After 96 h at 100 °C the catalytic reaction reaches > 92% completion.

8. Characterization of substrates

_N-Allyl-N-methylbenzimidamide (1a).⁶_

![Image of 1a]

Light yellow oil, yield: 84%. ¹H NMR (400 MHz, CDCl₃): δ 7.36-7.34 (m, 5H), 5.82-5.72 (m, 1H), 5.20-5.15 (m, 2H), 3.78 (d, J = 4.8 Hz, 2H), 2.94 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ 169.76, 138.69, 133.92, 128.90, 128.46, 126.56, 116.71, 53.60, 35.44. Spectroscopic data for the title compound were consistent with those reported in the literature.

_N-Allyl-N-methyl-4-methylbenzimidamide (1b).⁶_

![Image of 1b]

Light yellow oil, yield: 86%. ¹H NMR (400 MHz, CDCl₃): δ 7.24-7.22 (m, 2H), 7.18-7.16 (m, 2H), 5.81-5.73 (m, 1H), 5.20-5.15 (m, 2H), 3.78 (d, J = 4.8 Hz, 2H), 2.93 (s, 3H), 2.36 (s, 3H); ¹³C NMR (100 MHz, CDCl₃): δ 170.06, 138.95, 135.93, 134.11, 129.15, 126.62, 116.73, 53.76, 35.54, 21.36. Spectroscopic data for the title compound were consistent with those reported in the literature.

_N-allyl-4-fluoro-N-methylbenzimidamide (1c)._

![Image of 1c]

Light yellow oil, Yield: 73%. ¹H NMR (400 MHz, CDCl₃): δ 7.36-7.32 (m, 2H), 7.08-7.03 (m, 2H), 5.80-5.73 (m, 1H), 5.20-5.15 (m, 2H), 3.78 (d, J = 4.0 Hz, 2H), 2.93 (s, 3H). ¹³C NMR (100 MHz, CDCl₃): δ 168.69, 164.07, 162.84 (d, J = 248.5 Hz), 134.66, 128.51 (d, J = 8.1 Hz), 116.74, 115.43 (d, J = 21.6 Hz), 53.61, 35.49. ¹⁹F NMR (376 MHz, CDCl₃): δ -111.88 (m). HRMS (ESI) (m/z): [M+H]+ Calcd for C₁₁H₁₄FN₂ 193.1141, found 193.1137.
**N-Allyl-N-methyl-4-chlorobenzimidamide (1d).**

![Chemical Structure](image)

Light yellow oil, yield: 53%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.36-7.34 (m, 2H), 7.30-7.28 (m, 2H), 5.80-5.71 (m, 1H), 5.20-5.15 (m, 2H), 3.77 (d, $J = 3.8$ Hz, 2H), 2.93 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.61, 137.03, 134.94, 133.71, 128.78, 128.06, 116.90, 53.64, 35.54. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{11}$H$_{14}$ClN$_2$ 209.0846, found 209.0841.

**N-Allyl-N-methyl-4-iodobenzimidamide (1e).**

![Chemical Structure](image)

Light yellow oil, yield: 66%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.72-7.70 (m, 2H), 7.10-7.08 (m, 2H), 5.78-5.70 (m, 1H), 5.20-5.15 (m, 2H), 3.77 (d, $J = 3.8$ Hz, 2H), 2.92 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.65, 137.99, 137.65, 133.66, 128.38, 116.90, 94.89, 53.60, 35.50. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{11}$H$_{14}$IN$_2$ 301.0202, found 301.0192.

**N-Allyl-N-methyl-4-(trifluoromethyl)benzimidamide (1f).**

![Chemical Structure](image)

Light yellow oil, yield: 88%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.66-7.64 (m, 2H), 7.48-7.46 (m, 2H), 5.80-5.73 (m, 1H), 5.22-5.17 (m, 2H), 3.78 (s, 2H), 2.95 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): 168.24, 142.12, 133.56, 131.13 (q, $J = 65.0$, 32.5 Hz), 127.10, 125.66 (q, $J = 3.5$ Hz). 123.88 (q, $J = 272.6$ Hz), 117.06, 53.59, 35.54. $^{19}$F NMR (376 MHz, CDCl$_3$): $\delta$ -62.82 (s). HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{12}$H$_{14}$F$_3$N$_2$ 243.1109, found 243.1107.

**N-Allyl-N-methyl-thiophene-2-carboximidamide (1g).**

![Chemical Structure](image)

Light yellow oil, yield: 46%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.33-7.29 (m, $J = 8$ Hz, 1H), 7.16-7.15 (m, 1H), 7.03-6.96 (m, 1H), 5.85-5.77 (m, 1H), 5.23-5.19 (m, 2H), 3.87 (d, $J = 5.3$ Hz, 2H), 2.95 (d, $J = 5.8$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): 162.85, 138.96, 133.91, 126.81, 126.72, 126.43, 116.94, 54.11, 35.82. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_9$H$_{13}$N$_2$S 181.0799, found 181.0797.
**N-Allyl-N-methylpicolinimidamide (1h).**

Light yellow oil, Yield: 58%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.64-8.62 (m, 2H), 7.69-7.67 (m, 1H), 7.35-7.32 (m, 1H), 5.81-5.74 (m, 1H), 5.23-5.17 (m, 2H), 3.81-3.80 (m, 2H), 2.97 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 166.28, 149.94, 147.36, 134.05, 133.85, 133.20, 123.10, 116.82, 53.36, 35.37. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{10}$H$_{14}$N$_3$ 176.1188, found 176.1175.

**N-Allyl-N-methyl-isonicotinimidamide (1i).**

Light yellow oil, yield: 50%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.65-8.64 (m, 2H), 7.26-7.25 (m, 2H), 5.81-5.71 (m, 1H), 5.23-5.17 (m, 2H), 3.78 (m, 2H), 2.94 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 166.79, 150.13, 145.74, 133.19, 121.03, 116.96, 53.22, 35.25. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{10}$H$_{14}$N$_3$ 176.1188, found 176.1182.

**N,N-Diallylbenzimidamide (1j).**

Yellow sticky oil, yield: 59%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.37-7.36 (m, 5H), 5.84-5.76 (m, 2H) 5.19-5.15(m, 4H), 3.90 (d, $J$ = 3.8 Hz, 4H); $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 169.28, 138.74, 133.91, 128.97, 128.53, 126.55, 116.83, 49.73. Spectroscopic data for the title compound were consistent with those reported in the literature.

**N-Allyl-N-benzylbenzimidamide (1k).**

Light yellow sticky oil, yield: 79%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.39-7.35 (m, 5H), 7.33-7.32 (m, 2H), 7.28-25 (m, 3H), 5.84-5.74 (m, 1H), 5.19-5.10 (m, 2H), 4.57 (s, 2H), 3.84 (s, 2H). $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 169.73, 138.79, 138.30, 133.76, 129.05, 128.68, 128.64, 127.58, 127.16, 126.66, 117.16, 50.30, 49.87. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{17}$H$_{19}$N$_2$ 251.1548, found 251.1548. Spectroscopic data for the title compound were consistent with those reported in the literature.
N-Allyl-N-butylbenzimidamide (1l).

\[
\begin{align*}
\text{Light yellow sticky oil, yield: 79\%.} \\
^1\text{H NMR (400 MHz, CDCl}_3\text{): } & \delta 7.37-7.35 (m, 3H), 7.32-7.27 (m, 2H), 5.85-5.78 (m, 1H), 5.20-5.14 (m, 2H), 3.90 (d, } J = 3.4 \text{ Hz, 2H), 3.27-3.23 (m, 2H), 1.57-1.51 (m, 2H), 1.32-1.20 (m, 2H), 0.86 (t, } J = 7.3 \text{ Hz, 3H).} \\
^1\text{C NMR (100 MHz, CDCl}_3\text{): } & \delta 169.35, 139.28, 134.55, 128.76, 128.53, 126.62, 116.51, 50.44, 47.40, 29.90, 20.20, 13.98. \\
\text{HRMS (ESI) (m/z): } & [\text{M+H}]^+ \text{ Calcd for C}_{14}\text{H}_{21}\text{N}_2 217.1705, \text{ found 217.1697.}
\end{align*}
\]

N-Allyl-N-cyclohexylbenzimidamide (1m).

\[
\begin{align*}
\text{Light yellow sticky oil, yield: 69\%.} \\
^1\text{H NMR (400 MHz, CDCl}_3\text{): } & \delta 7.38-7.36 (m, 3H), 7.30-7.27 (m, 2H), 5.94-5.87 (m, 1H), 5.18-5.07 (m, 2H), 3.98 (d, } J = 4.3 \text{ Hz, 2H), 1.76-1.71 (m, 4H), 1.56-1.26 (m, 4H), 1.15-0.97 (m, 3H).} \\
^1\text{C NMR (100 MHz, CDCl}_3\text{): } & \delta 168.78, 139.19, 136.27, 128.61, 128.48, 126.27, 115.03, 57.98, 45.24, 31.49, 25.84, 25.46. \\
\text{HRMS (ESI) (m/z): } & [\text{M+H}]^+ \text{ Calcd for C}_{16}\text{H}_{23}\text{N}_2 243.1861, \text{ found 243.1862.}
\end{align*}
\]

N-Allylbenzimidamide (1n).

\[
\begin{align*}
\text{Light yellow oil, yield: 90\%.} \\
^1\text{H NMR (400 MHz, CDCl}_3\text{): } & \delta 7.59-7.57 (m, 2H), 7.42-7.37 (m, 3H), 6.04-5.95 (m, 1H), 5.31-5.26 (m, } J = 17.2, 1.5 \text{ Hz, 1H), 5.18 (dd, } J = 10.2, 1.3 \text{ Hz, 1H), 3.99 (d, } J = 5.4 \text{ Hz, 2H).} \\
^1\text{C NMR (100 MHz, CDCl}_3\text{): } & \delta 163.10, 137.89, 134.88, 130.11, 128.72, 126.10, 116.10, 45.30. \\
\text{Spectroscopic data for the title compound were consistent with those reported in the literature.}
\end{align*}
\]

N,N-diallylthiophene-2-carboximidamide (1o).

\[
\begin{align*}
\text{S10}
\end{align*}
\]
Brown sticky oil, yield: 69%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.38-7.37 (m, 1H), 7.31-7.29 (m, 1H), 7.11-7.10 (m, 1H), 5.86-5.79 (m, 2H), 5.20-5.15 (m, 4H), 3.92 (d, $J = 5.0$ Hz, 4H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 164.42, 139.06, 133.94, 126.55, 125.91, 123.69, 116.75, 49.83. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{11}$H$_{15}$N$_2$S 207.0956, found 207.0946.

$N,N$-Diallyl-$4$-chlorobenzimidamide (1p).

\[
\begin{array}{c}
\text{Cl} \\
\text{N} \\
\text{N} \\
\text{H} \\
\end{array}
\]

Light yellow sticky oil, yield: 73%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.34-7.32 (m, 2H), 7.29-7.27 (m, 2H), 5.82-5.76 (m, 2H), 5.19-5.13 (m, 4H), 3.88 (s, 4H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 167.91, 136.95, 134.80, 133.58, 128.65, 127.86, 116.84, 49.64. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{13}$H$_{16}$ClN$_2$ 235.1002, found 235.0991.

$N$-Benzy$N$-(2-methylallyl)benzimidamide (1q).

\[
\begin{array}{c}
\text{H} \\
\text{N} \\
\text{N} \\
\text{1q} \\
\end{array}
\]

Light yellow sticky oil, yield: 63%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.39-7.33 (m, 5H), 7.32-7.30 (m, 2H), 7.27-7.22 (m, 3H), 4.92 (s, 1H), 4.85 (s, 1H), 4.57 (s, 2H), 3.77 (s, 2H), 1.64 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 169.78, 141.11, 138.69, 138.23, 128.87, 128.54, 128.50, 127.45, 126.52, 111.82, 52.60, 49.85, 20.21. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{18}$H$_{21}$N$_2$ 265.1705, found 265.1700.

$N$-(2-Methylallyl)$N$-(4-methylbenzyl)benzimidamide (1r).

\[
\begin{array}{c}
\text{H} \\
\text{N} \\
\text{N} \\
\text{1r} \\
\end{array}
\]

Light yellow sticky oil, yield: 70%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.33-7.29 (m, 3H), 7.27-7.25 (m, 4H), 7.18-7.16 (m, 2H), 4.93 (s, 1H), 4.85 (s, 1H), 4.58 (s, 2H), 3.79 (s, 2H), 2.36 (s, 3H), 1.65 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 169.85, 141.04, 139.20, 138.12, 135.33, 129.31, 128.61, 127.56, 127.16, 126.67, 111.96, 52.88, 50.24, 21.41, 20.32. HRMS(ESI)(m/z): [M+H]$^+$ Calcd for C$_{19}$H$_{23}$N$_2$ 279.1861, found 265.1873.

(E)$-N$-Benzy$N$-(but-2-en-1-yl)benzimidamide (1t).
N-Allyl-N-phenylbenzimidamide (1u).

Light yellow sticky oil, yield: 74%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.38-7.34 (m, 5H), 7.33-7.31 (m, 2H), 7.28-7.22 (m, 3H), 5.51-5.45 (m, 2H), 4.55 (s, 2H), 3.75 (s, 2H), 1.68 (d, $J = 4.9$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 169.66, 138.91, 138.43, 128.90, 128.68, 128.58, 128.53, 127.53, 127.00, 126.65, 126.35, 49.86, 49.21, 17.81. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{18}$H$_{21}$N$_2$ 265.1705, found 265.1691.

N-Benzyl-N-(but-3-en-1-yl)benzimidamide (1aa).

Light yellow sticky oil, yield: 78%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.33-7.26 (m, 2H), 7.22-7.15 (m, 3H), 7.15-7.09 (m, 2H), 7.02-6.94 (m, 3H), 6.11-6.01 (m, 1H), 5.25 (d, $J = 17.2$ Hz, 1H), 5.17 (d, $J = 10.4$ Hz, 1H), 4.58 (d, $J = 5.2$ Hz, 2H). Spectroscopic data for the title compound were consistent with those reported in the literature.

N-benzyl-N-(but-3-en-1-yl-4-methylbenzimidamide (1ab).

Light yellow sticky oil, Yield: 78%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.37-7.33 (m, 2H), 7.28-7.26 (m, 5H), 7.19-7.17 (m, 2H), 5.78-5.68 (m, 1H), 5.06-5.00 (m, 2H), 4.56 (s, 2H), 3.36-3.34 (m, 2H), 2.37-2.32 (m,
5H. $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 169.84, 138.83, 138.54, 136.16, 135.67, 129.24, 128.59, 127.33, 127.10, 126.64, 116.58, 51.50, 46.70, 31.94, 21.36. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{19}$H$_{23}$N$_2$ 279.1861, found 279.1858.

*N-Methyl-N-phenylbenzimidamide (1ac).*

![N-Methyl-N-phenylbenzimidamide (1ac)](image)

Yellow solid, yield: 53%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.28-7.24 (m, 2H), 7.19-7.11 (m, 5H), 7.00-6.95 (m, 3H), 3.50 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.73, 146.83, 138.57, 128.92, 128.85, 128.15, 127.83, 126.57, 124.97, 40.05. Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-Methyl-N-phenyl-(2-methyl)benzimidamide (1ad).*

![N-Methyl-N-phenyl-(2-methyl)benzimidamide (1ad)](image)

Yellow oil, yield: 43%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.22-7.01 (m, 9H), 3.48 (s, 3H), 2.27 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.06, 145.66, 138.64, 134.07, 130.23, 128.71, 128.50, 128.11, 126.49, 125.62, 125.45, 76.84, 39.05, 19.56. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{15}$H$_{17}$N$_2$ 225.1392, found 225.1386. Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-Methyl-N-phenyl(4-methyl)benzimidamide (1ae).*

![N-Methyl-N-phenyl(4-methyl)benzimidamide (1ae)](image)

Yellow oil, yield: 69%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.17 (t, $J$ = 8.0 Hz, 4H), 6.99 (dd, $J$ = 9.7, 5.9 Hz, 5H), 3.52 (s, 3H), 2.27 (d, $J$ = 2.9 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.83, 157.42, 147.06, 138.97, 135.66, 128.85, 127.82, 126.51, 124.83, 40.18, 21.32. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{15}$H$_{17}$N$_2$ 225.1392, found 225.1386. Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-Methyl-N-phenyl-(4-chloro)benzimidamide (1af).*
Yellow solid, yield: 51%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.22-7.14 (m, 6H), 7.04-6.93 (m, 3H), 3.48 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 167.38, 146.52, 136.90, 134.80, 129.19, 129.00, 128.38, 126.50, 125.19, 40.05. HRMS (ESI) (m/z): [M+H]$^+$ Caled for C$_{14}$H$_{14}$ClN$_2$ 245.0846, found 245.0861.

**N-Methyl-N-phenyl-(4-iodo)benzimidamide (1ag).**

Brown solid, yield: 47%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.53-7.51 (m, 2H), 7.19-7.15 (t, $J$ = 7.7 Hz, 2H), 7.05-7.00 (m, 3H), 6.96-6.94 (m, 2H), 3.48 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 167.66, 146.54, 138.01, 137.39, 129.62, 129.13, 126.59, 125.34, 95.14, 40.15. HRMS (ESI) (m/z): [M+H]$^+$ Caled for C$_{14}$H$_{14}$IN$_2$ 337.0202, found 337.0176.

**N-methyl-N-phenylthiophene-2-carboximidamide (1ah).**

Brown solid, yield: 53%. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.29-7.19 (m, 3H), 7.10-7.06 (m, 3H), 6.98-6.97 (m, 1H), 6.83-6.81 (m, 1H), 3.48 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 161.55, 147.00, 140.11, 129.05, 128.70, 127.53, 126.62, 126.45, 125.48, 40.47. HRMS (ESI) (m/z): [M+H]$^+$ Caled for C$_{12}$H$_{13}$N$_2$S 217.0799, found 217.0785.

**N-Methyl-N-phenylnicotinimidamide (1ai).**

Brown oil, yield: 43%. $^1$H NMR (400 MHz, CDCl$_3$): δ 8.55 (m, 1H), 8.43-8.42 (m, 1H), 7.55-7.53 (m, 1H), 7.18-7.13 (m, 2H), 7.12-7.07 (m, 1H), 7.05-7.01 (m, 1H), 6.80-6.95 (m, 2H), 3.49 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 165.51, 149.78, 148.78, 146.04, 135.00, 134.20, 129.13, 126.77, 125.56, 122.86, 39.87. HRMS (ESI) (m/z): [M+H]$^+$ Caled for C$_{13}$H$_{14}$N$_3$ 212.1188, found 212.1173.
**N-Methyl-N-phenylisonicotinimidamide (1aj).**

![Structure of 1aj](image)

Brown solid, yield: 45%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.45-8.43 (m, 2H), 7.18-7.15 (m, 4H), 7.06-7.02 (m, 1H), 6.97-6.95 (m, 2H), 3.48 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.91, 149.76, 145.80, 145.67, 129.10, 126.52, 125.72, 122.10, 39.73. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{13}$H$_{14}$N$_2$ 212.1188, found 212.1190.

**N-Methyl-N-phenyl-2-naphthimidamide (1ak).**

![Structure of 1ak](image)

Yellow solid, yield: 67%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.89 (m, 1H), 7.78-7.77 (m, 2H), 7.64-7.62 (m, 1H), 7.48-7.46 (m, 2H), 7.35-7.29 (m, 1H), 7.15-7.11 (m, 2H), 7.06-7.05 (m, 2H), 6.99-6.96 (m, 1H), 3.59 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.67, 146.80, 136.12, 133.24, 132.80, 128.93, 128.38, 127.75, 127.71, 127.59, 126.82, 126.53, 126.48, 125.20, 124.96, 40.22. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{18}$H$_{17}$N$_2$ 261.1392, found 261.1379.

**N,N-Diethylbenzimidamide (1al).**

![Structure of 1al](image)

Yellow oil, yield: 77%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.36-7.34 (m, 3H), 7.31-7.29 (m, 2H), 3.31 (q, $J$ = 6.9 Hz, 4H), 1.13 (t, $J$ = 7.1 Hz, 6H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 168.59, 139.39, 128.34, 128.28, 126.22, 41.75, 13.14. Spectroscopic data for the title compound were consistent with those reported in the literature.

**N-Phenylbenzimidamide (1an).**

![Structure of 1an](image)

Yellow solid, yield: 68%. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.87-7.86 (m, 2H), 7.50-7.43 (m, 3H), 7.40-7.36 (m, 2H), 7.11-7.07 (m, 1H), 7.01-7.00 (m, 2H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 155.11, 149.60, 135.81,
Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-(2,6-Dimethylphenyl)benzimidamide* (1ao).\(^{12}\)

![1ao](image)

Yellow solid, yield: 45%. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.87-7.85 (m, 2H), 7.45-7.38 (m, 3H), 7.05-7.03 (m, 2H), 6.90-6.87 (m, 1H), 4.60 (s, 2H), 2.13 (s, 6H). \(^13\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 153.16, 146.45, 135.65, 130.50, 128.84, 128.56, 128.19, 126.76, 122.84, 17.84. Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-(2,6-Dimethylphenyl)-4-methylbenzimidamide* (1ap).

![1ap](image)

Yellow solid, yield: 49%. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.75-7.73 (m, 2H), 7.19-7.17 (m, 2H), 7.03-7.01 (m, 2H), 6.89-6.85 (m, 1H), 4.57 (s, 2H), 2.37 (s, 3H), 2.11 (s, 6H). \(^13\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 153.13, 146.46, 140.59, 132.73, 129.14, 128.91, 128.11, 126.65, 122.72, 21.39, 17.81. HRMS (ESI) (m/z): [M+H]+ Caled for C\(_{16}\)H\(_{19}\)N\(_2\): 239.1548, found 239.1546.

*N-(2,6-Diisopropylphenyl)benzimidamide* (1aq).\(^{12}\)

![1aq](image)

Yellow solid, yield: 40%. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.90-7.88 (m, 2H), 7.49-7.42 (m, 3H), 7.17-7.15 (m, 2H), 7.09-7.05 (m, 1H), 4.61 (s, 2H), 3.07-3.01 (m, 2H), 1.18 (dd, \(J = 6.8, 4.2\) Hz, 12H). \(^13\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 153.50, 144.23, 139.36, 135.85, 130.52, 128.67, 126.83, 123.52, 123.46, 28.30, 23.79, 23.70. Spectroscopic data for the title compound were consistent with those reported in the literature.

*N-propylbenzimidamide* (1ar).\(^{13}\)

![1ar](image)

Yellow oil, yield: 83%. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.33-7.29 (m, 3H), 7.16-7.12 (m, 2H), 5.49 (s, 1H), 3.01 (t, \(J = 7.2\) Hz, 2H), 1.48-1.39 (m, 2H), 0.77 (t, \(J = 7.4\) Hz, 3H). \(^13\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\)
Spectroscopic data for the title compound were consistent with those reported in the literature.

**(Z)-N,N-diethyl-N'-propylbenzimidamide (1as).**

Yellow oil, yield: 63%. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.39-7.29 (m, 3H), 7.12-7.10 (m, 2H), 3.19-3.18 (m, 4H), 2.83 (t, \(J = 7.1\) Hz, 2H), 1.41 (dd, \(J = 14.4, 7.2\) Hz, 2H), 1.03 (t, \(J = 6.9\) Hz, 6H), 0.75 (t, \(J = 7.4\) Hz, 3H). \(^{13}\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 160.59, 135.54, 128.33, 127.90, 127.66, 52.66, 41.31, 25.69, 13.50, 11.89. HRMS (ESI) (m/z): [M+H]+ Calcd for C\(_{14}\)H\(_{23}\)N\(_2\) 219.1861, found 219.1859.

9. Characterization of the products

**1,4-Dimethyl-2-phenyl-4,5-dihydro-1H-imidazole (2a).**

Light yellow oil. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.54-7.53 (m, 2H), 7.38-7.37 (m, 3H), 4.18-4.09 (m, 1H), 3.57 (t, \(J = 8.0\) Hz, 1H), 2.98 (t, \(J = 8.0\) Hz, 1H), 2.76 (s, 3H), 1.32 (d, \(J = 6.6\) Hz, 3H). \(^{13}\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 166.50, 131.25, 129.54, 128.33, 128.14, 60.88, 59.61, 36.23, 22.09. Spectroscopic data for the title compound were consistent with those reported in the literature.

**1,4-Dimethyl-2-(p-tolyl)-4,5-dihydro-1H-imidazole (2b).**

Brown oil. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.45-7.43 (m, 2H), 7.20-7.18 (m, 2H), 4.17-4.08 (m, 1H), 3.57 (t, \(J = 9.5\) Hz, 1H), 2.97 (t, \(J = 8.7\) Hz, 1H), 2.77 (s, 3H), 2.36 (s, 3H), 1.32 (d, \(J = 6.6\) Hz, 3H). \(^{13}\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 166.67, 139.68, 128.92, 128.31, 128.20, 61.02, 59.53, 36.43, 22.20, 21.39. Spectroscopic data for the title compound were consistent with those reported in the literature.

**2-(4-Fluorophenyl)-1,4-dimethyl-4,5-dihydro-1H-imidazole (2c).**

Brown oil. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.57-7.53 (m, 2H), 7.10-7.06 (m, 2H), 4.16-4.08 (m, 1H), 3.59 (t, \(J = 9.5\) Hz, 1H), 2.99 (t, \(J = 8.8\) Hz, 1H), 2.77 (s, 3H), 1.32 (d, \(J = 6.6\) Hz, 3H). \(^{13}\)C NMR (100 MHz,
CDCl$_3$): $\delta$ 165.68, 163.54 (d, $J = 249.3$ Hz), 130.31 (d, $J = 8.4$ Hz), 127.35, 115.36 (d, $J = 21.7$ Hz), 61.01, 59.61, 36.34, 22.16. $^{19}$F NMR (376 MHz, CDCl$_3$): $\delta$ -108.82 (m), -110.90 (m). HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{11}$H$_{14}$FN$_2$ 193.1141, found 193.1138.

2-(4-Chlorophenyl)-1,4-dimethyl-4,5-dihydro-1H-imidazole (2d).$^1$

![Image of 2d]

Brown oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.50-7.48 (m, 2H), 7.38-7.36 (m, 2H), 4.18-4.08 (m, 1H), 3.58 (t, $J = 9.5$ Hz, 1H), 2.98 (t, $J = 8.8$ Hz, 1H), 2.76 (s, 3H), 1.32 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.53, 135.59, 129.81, 129.63, 128.49, 60.97, 59.79, 36.32, 22.11.

2-(4-Iodophenyl)-1,4-dimethyl-4,5-dihydro-1H-imidazole (2e).$^1$

![Image of 2e]

Yellow solid. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.74-7.72 (m, 2H), 7.30-7.27 (m, 2H), 4.17-4.07 (m, 1H), 3.57 (t, $J = 9.5$ Hz, 1H), 2.97 (t, $J = 8.8$ Hz, 1H), 2.75 (s, 3H), 1.31 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.62, 137.34, 130.74, 129.89, 95.85, 60.97, 59.74, 36.28, 22.06. Spectroscopic data for the title compound were consistent with those reported in the literature.

1,4-Dimethyl-2-(trifluoromethyl)phenyl-4,5-dihydro-1H-imidazole (2f).$^1$

![Image of 2f]

Brown oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.70-7.65 (m, 4H), 4.21- 4.12 (m, 1H), 3.61 (t, $J = 9.8$ Hz, 1H), 3.01 (t, $J = 8.8$ Hz, 1H), 2.77 (s, 3H), 1.34 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$: 165.38, 135.07, 131.61 (q, $J = 32.7$ Hz), 128.72, 127.03, 125.64, 125.26 (q, $J = 3.6$ Hz), 121.14 (d, $J = 277.6$ Hz), 60.93, 60.02, 36.17, 22.02. $^{19}$F NMR (376 MHz, CDCl$_3$): $\delta$ -62.89 (s). Spectroscopic data for the title compound were consistent with those reported in the literature.

1,4-dimethyl-2-(thiophen-2-yl)-4,5-dihydro-1H-imidazole (2g) $^1$.

![Image of 2g]

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.37-7.34 (m, 2H), 7.06-7.04 (m, 1H), 4.15-4.08 (m, 1H), 3.60 (t, $J = 9.5$ Hz, 1H), 3.02 (t, $J = 8.7$ Hz, 1H), 2.97 (s, 3H), 1.31 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100
MHz, CDCl$_3$): $\delta$ 159.89, 132.93, 127.99, 127.59, 126.96, 61.45, 59.41, 36.11, 21.96. Spectroscopic data for the title compound were consistent with those reported in the literature.

3-(1,4-Dimethyl-4,5-dihydro-1H-imidazol-2-yl)pyridine (2h).

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.79-8.78 (m, 1H), 8.66-8.64 (m, 1H), 7.91-7.88 (m, 1H), 7.37-7.33 (m, 1H), 4.22-4.12 (m, 1H), 3.63 (t, $J = 9.6$ Hz, 1H), 3.03 (t, $J = 8.8$ Hz, 1H), 2.80 (s, 3H), 1.34 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 163.89, 150.70, 149.03, 135.99, 127.28, 123.27, 60.80, 59.83, 36.10, 22.06. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{10}$H$_{14}$N$_3$ 176.1188, found 176.1188.

4-(1,4-Dimethyl-4,5-dihydro-1H-imidazol-2-yl)pyridine (2i).

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.68-8.67 (m, 2H), 7.46-7.45 (m, 2H), 4.21-4.14 (m, 1H), 3.62 (t, $J = 9.6$ Hz, 1H), 3.01 (t, $J = 8.9$ Hz, 1H), 2.78 (s, 3H), 1.34 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 164.6, 150.2, 139.3, 122.8, 60.9, 60.2, 36.1, 22.0. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{10}$H$_{14}$N$_3$ 176.1188, found 176.1188.

1-Allyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2j).

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.55-7.53 (m, 2H), 7.39-7.36 (m, 3H), 5.79-5.70 (m, 1H), 5.24-5.15 (m, 2H), 4.22-4.13 (m, 1H), 3.73-3.55 (m, 3H), 2.99 (dd, $J = 9.1$, 8.3 Hz, 1H), 1.33 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.75, 134.16, 131.28, 129.67, 128.22, 128.01, 116.55, 59.89, 57.86, 51.46, 22.36. Spectroscopic data for the title compound were consistent with those reported in the literature.

1-Benzyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2k).

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.61-7.59 (m, 2H), 7.37-7.31 (m, 5H), 7.27-7.22 (m, 3H), 4.33-4.13 (m, 3H), 3.51 (t, $J = 8.0$ Hz, 1H), 2.92 (t, $J = 8.0$ Hz, 1H), 1.31 (d, $J = 4.0$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.73, 137.98, 131.27, 129.74, 128.56, 128.35, 128.14, 127.19, 127.01, 59.89, 57.78, 52.77, 22.41. Spectroscopic data for the title compound were consistent with those reported in the literature.
**1-Butyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2l).**

![Image of 1-Butyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2l)](image)

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.52-7.49 (m, 2H), 7.38-7.34 (m, 3H), 4.18-4.10 (m, 1H), 3.58 (t, $J$ = 9.6 Hz, 1H), 3.03-2.95 (m, 3H), 1.51-1.44 (m, 2H), 1.32 (d, $J$ = 4.0 Hz, 3H), 1.30-1.25 (m, 2H), 0.86 (t, $J$ = 7.3 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 166.24, 131.77, 129.39, 128.11, 128.08, 59.63, 57.83, 48.71, 30.99, 22.37, 19.77, 13.75. HRMS (ESI) (m/z): [M+H]$^+$ Calcd for C$_{14}$H$_{21}$N$_2$ 217.1705, found 217.1703.

**1-Cyclohexyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2m).**

![Image of 1-Cyclohexyl-4-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2m)](image)

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.48-7.46 (m, 2H), 7.39-7.36 (m, 3H), 4.16-4.07 (m, 1H), 3.61-3.57 (m, 1H), 3.31-3.23 (m, 1H), 3.02 (t, $J$ = 8.6 Hz, 1H), 1.72-1.64 (m, 3H), 1.59-1.56 (m, 2H), 1.45-1.34 (m, 2H), 1.29 (d, $J$ = 6.6 Hz, 3H), 1.15-0.98 (m, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.38, 132.13, 129.40, 128.25, 128.08, 59.22, 54.83, 51.38, 31.15, 30.55, 25.63, 25.52, 25.42, 22.48. Spectroscopic data for the title compound were consistent with those reported in the literature.

**1-allyl-4-methyl-2-(thiophen-2-yl)-4,5-dihydro-1H-imidazole (2o).**

![Image of 1-allyl-4-methyl-2-(thiophen-2-yl)-4,5-dihydro-1H-imidazole (2o)](image)

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.60 (m, 1H), 7.32-7.28 (m, 2H), 5.83-5.74 (m, 1H), 5.22 (dd, $J$ = 22.9, 13.7 Hz, 2H), 4.20-4.11 (m, 1H), 3.82-3.71 (m, 2H), 3.56 (t, $J$ = 9.7 Hz, 1H), 2.99 (t, $J$ = 8.7 Hz, 1H), 1.31 (d, $J$ = 6.5 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 161.00, 134.16, 131.95, 127.36, 126.09, 125.66, 116.41, 59.47, 57.91, 51.21, 22.30. Spectroscopic data for the title compound were consistent with those reported in the literature.

**1-Allyl-2-(4-chlorophenyl)-4-methyl-4,5-dihydro-1H-imidazole (2p).**

![Image of 1-Allyl-2-(4-chlorophenyl)-4-methyl-4,5-dihydro-1H-imidazole (2p)](image)

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.50-7.48 (m, 2H), 7.37-7.35 (m, 2H), 5.78-5.72 (m, 1H), 5.24-5.17 (m, 2H), 4.19-4.14 (m, 1H), 3.71-3.57 (m, 3H), 3.03-2.97 (m, 1H), 1.32 (d, $J$ = 6.6 Hz, 3H). $^{13}$C
NMR (100 MHz, CDCl₃): δ 164.86, 135.81, 134.00, 129.82, 129.52, 128.64, 116.82, 59.97, 58.04, 51.54, 22.42. Spectroscopic data for the title compound were consistent with those reported in the literature.

1-Benzyl-4,4-dimethyl-2-phenyl-4,5-dihydro-1H-imidazole (2q).¹

Brown oil. ¹H NMR (400 MHz, CDCl₃): δ 7.61-7.58 (m, 2H), 7.37-7.32, (m, 5H), 7.27-7.22 (m, 3H), 4.28 (s, 2H), 3.13 (s, 2H), 1.31 (s, 6H). ¹³C NMR (100 MHz, CDCl₃): δ 163.93, 138.11, 131.41, 129.78, 128.66, 128.44, 128.33, 127.29, 127.08, 64.72, 63.14, 52.61, 29.42. Spectroscopic data for the title compound were consistent with those reported in the literature.

1-Benzyl-4,4-dimethyl-2-(p-tolyl)-4,5-dihydro-1H-imidazole (2r).¹

Brown oil. ¹H NMR (400 MHz, CDCl₃): δ 7.50-7.48 (m, 2H), 7.35-7.31 (m, 2H), 7.26-7.21 (m, 3H), 7.17-7.15 (m, 2H), 4.28 (s, 2H), 3.11 (s, 2H), 2.33 (s, 3H), 1.31 (s, 6H). ¹³C NMR (100 MHz, CDCl₃): δ 163.85, 139.63, 138.14, 128.97, 128.52, 128.35, 128.15, 127.12, 126.96, 64.48, 63.07, 52.59, 29.35, 21.28. Spectroscopic data for the title compound were consistent with those reported in the literature.

1,4-Dibenzyl-2-phenyl-4,5-dihydro-1H-imidazole (2s).¹

Light yellow oil. ¹H NMR (400 MHz, CDCl₃): δ 7.57-7.55 (m, 2H), 7.40-7.35 (m, 3H), 7.31-7.19 (m, 8H), 7.06-7.05 (m, 2H), 4.47-4.06 (m, 1H), 4.13 (m, 2H), 3.35 (t, J = 10.0 Hz, 1H), 3.14-3.08 (m, 2H), 2.79 (m, 1H). ¹³C NMR (100 MHz, CDCl₃): δ 166.44, 138.49, 137.94, 131.21, 129.99, 129.67, 128.67, 128.55, 128.28, 128.23, 127.26, 126.96, 126.24, 65.60, 54.76, 52.63, 42.29. Spectroscopic data for the title compound were consistent with those reported in the literature.

4-Methyl-1,2-diphenyl-4,5-dihydro-1H-imidazole (2u).¹
Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.50-7.48 (m, 2H), 7.34-7.32 (m, 1H), 7.28-7.25 (m, 2H), 7.16-7.12 (m, 2H), 6.98-6.94 (m, 1H), 6.77-6.75 (m, 2H), 4.37-4.28 (m, 1H), 4.14 (t, $J$ = 9.5 Hz, 1H), 3.63 (dd, $J$ = 8.9, 8.3 Hz, 1H), 1.41 (d, $J$ = 6.6 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 161.20, 143.00, 131.09, 130.02, 128.82, 128.74, 128.19, 123.23, 122.31, 60.79, 59.53, 22.30. Spectroscopic data for the title compound were consistent with those reported in the literature.

$1,4$-$\text{Dimethyl-2-(o-tolyl)-4,5-dihydro-1H-imidazole (2v).}$$^1$

Yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 7.28-7.24 (m, 2H), 7.20-7.16 (m, 2H), 4.22-4.11 (m, 1H), 3.55 (t, $J$ = 6.6 Hz, 1H), 2.97 (t, $J$ = 6.6 Hz, 1H), 2.57 (s, 3H), 2.32 (s, 3H), 1.33 (d, $J$ = 4.0 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.80, 136.11, 131.35, 129.96, 128.87, 128.36, 125.48, 59.82, 59.73, 34.44, 22.14, 19.14. Spectroscopic data for the title compound were consistent with those reported in the literature.

$1,4$-$\text{Dimethyl-2-(naphthalen-2-yl)-4,5-dihydro-1H-imidazole (2w).}$$^1$

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.06 (m, 1H), 7.86-7.79 (m, 3H), 7.62-7.60 (m, 1H), 7.47-7.45 (m, 2H), 4.20-4.14 (m, 1H), 3.57 (dd, $J$ = 9.8, 9.3 Hz, 1H), 2.97 (t, $J$ = 8.8 Hz, 1H), 2.76 (s, 3H), 1.35 (d, $J$ = 6.6 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 166.69, 133.80, 132.80, 128.66, 128.37, 128.10, 127.97, 127.70, 126.83, 126.41, 125.44, 61.03, 59.81, 36.48, 22.24. Spectroscopic data for the title compound were consistent with those reported in the literature.

$1$-$\text{Allyl-4-methyl-2-(naphthalen-2-yl)-4,5-dihydro-1H-imidazole (2x).}$

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 8.08 (m, 1H), 7.85-7.79 (m, 3H), 7.64-7.61 (m, 1H), 7.47-7.45 (m, 2H), 5.80-5.70 (m, 1H), 5.24-5.15 (m, 2H), 4.27-4.18 (m, 1H), 3.75-3.57 (m, 3H), 3.03-2.98 (m, 1H), 1.37 (d, $J$ = 6.6 Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 165.78, 134.10, 133.75, 132.71,
128.58, 128.28, 127.91, 127.84, 127.57, 126.69, 126.27, 125.14, 116.57, 59.91, 57.93, 51.60, 22.37. HRMS (ESI) (m/z): [M+H]+ Calcd for C_{17}H_{15}N_{2} 251.1548, found 251.1546.

4-methyl-1-(naphthalen-2-ylmethyl)-4,5-dihydro-1H-imidazole (2y).

![2y]

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.84-7.82 (m, 3H), 7.71 (m, 1H), 7.67-7.64 (m, 2H), 7.52-7.47 (m, 2H), 7.42-7.38 (m, 3H), 7.36-7.33 (m, 1H), 4.44 (m, 2H), 4.26-4.17 (m, 1H), 3.58 (t, $J = 9.7$ Hz, 1H), 2.98 (dd, $J = 9.0$, 8.5 Hz, 1H), 1.33 (d, $J = 6.6$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 166.01, 135.73, 133.52, 132.87, 131.42, 130.04, 128.62, 128.39, 127.83, 126.42, 125.98, 125.82, 125.35, 60.11, 58.12, 53.20, 22.61. HRMS (ESI) (m/z): [M+H]+ Calcd for C$_{21}$H$_{21}$N$_{2}$ 301.1705, found 301.1703.

4-benzyl-1-methyl-2-phenyl-4,5-dihydro-1H-imidazole (2z).

![2z]

Light yellow oil. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.53-7.51 (m, 2H), 7.38-7.36 (m, 3H), 7.28-7.21 (m, 5H), 4.38-4.32 (m, 1H), 3.39 (t, $J = 9.8$ Hz, 1H), 3.20-3.19 (m, 2H), 2.74 (m, 1H), 2.65 (s, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 167.03, 138.83, 131.21, 129.73, 129.41, 128.27, 126.15, 65.57, 58.05, 42.27, 36.06. HRMS (ESI) (m/z): [M+H]+ Calcd for C$_{21}$H$_{21}$N$_{2}$ 251.1548, found 251.1545.

1-Benzyl-4-methyl-2-phenyl-1,4,5,6-tetrahydropyrimidine (3a).

![3a]

Brown oil. $^1$H NMR (400 MHz, CDCl$_3$): δ 7.47-7.44 (m, 2H), 7.32-7.29 (m, 5H), 7.27-7.21 (m, 1H), 7.16-7.14 (m, 2H), 4.30-4.16 (m, 2H), 3.59-3.51 (m, 1H), 3.25-3.13 (m, 2H), 1.96-1.90 (m, 1H), 1.65-1.56 (m, 1H), 1.28 (d, $J = 6.7$ Hz, 3H). $^{13}$C NMR (100 MHz, CDCl$_3$): δ 158.15, 137.91, 137.62, 128.59, 128.55, 128.29, 127.95, 126.91, 55.51, 49.00, 44.49, 28.85, 23.38. Spectroscopic data for the title compound were consistent with those reported in the literature.
1-benzyl-4-methyl-2-(p-tolyl)-1,4,5,6-tetrahydropyrimidine (3b)\(^1\)

\[
\begin{array}{c}
\text{N} \\
\text{N} \\
\text{3b}
\end{array}
\]

Brown oil. \(^1\)H NMR (400 MHz, CDCl\(_3\)): \(\delta\) 7.27-7.25 (m, 2H), 7.23-7.18 (m, 2H), 7.16-7.14 (m, 1H), 7.06-7.05 (m, 2H), 7.03-7.01 (m, 2H), 4.15 (m, 2H), 3.45 (d, \(J = 2.8\) Hz, 1H), 3.13-3.04 (m, 2H), 2.21 (s, 3H), 1.85-1.81 (m, 1H), 1.53-1.48 (m, 1H), 1.19 (d, \(J = 6.6\) Hz, 3H). \(^{13}\)C NMR (100 MHz, CDCl\(_3\)): \(\delta\) 158.21, 138.37, 138.00, 134.69, 128.90, 128.51, 127.87, 127.13, 126.90, 55.55, 48.98, 44.46, 28.86, 23.39, 21.20. Spectroscopic data for the title compound were consistent with those reported in the literature.

**10. References**

11. Copies of $^1\text{H}$ and $^{13}\text{C}$ NMR spectra

The $^1\text{H}$ NMR spectrum of 1c in CDCl$_3$ of 1c in CDCl$_3$.

The $^{13}\text{C}$ NMR spectrum of 1c in CDCl$_3$. 
The $^{19}$F NMR spectrum of 1c in CDCl$_3$

The $^1$H NMR spectrum of 1d in CDCl$_3$
The $^{13}$C NMR spectrum of 1d in CDCl$_3$

![13C NMR spectrum of 1d in CDCl$_3$]

The $^1$H NMR spectrum of 1e in CDCl$_3$

![1H NMR spectrum of 1e in CDCl$_3$]
The $^{13}$C NMR spectrum of 1e in CDCl$_3$

The $^1$H NMR spectrum of 1f in CDCl$_3$
The $^{13}$C NMR spectrum of 1f in CDCl$_3$

The $^{19}$F NMR spectrum of 1f in CDCl$_3$
The $^1$H NMR spectrum of 1g in CDCl$_3$

The $^{13}$C NMR spectrum of 1g in CDCl$_3$
The $^1$H NMR spectrum of 1h in CDCl$_3$
The $^1$H NMR spectrum of 1i in CDCl$_3$

![H NMR spectrum](image)

The $^{13}$C NMR spectrum of 1i in CDCl$_3$

![C NMR spectrum](image)
The $^1$H NMR spectrum of $1l$ in CDCl$_3$.

The $^{13}$C NMR spectrum of $1l$ in CDCl$_3$. 
The $^1$H NMR spectrum of 1m in CDCl$_3$

![1H NMR spectrum of 1m in CDCl$_3$]

The $^{13}$C NMR spectrum of 1m in CDCl$_3$

![13C NMR spectrum of 1m in CDCl$_3$]
The $^1$H NMR spectrum of 1o in CDCl$_3$

The $^{13}$C NMR spectrum of 1o in CDCl$_3$
The $^1$H NMR spectrum of 1p in CDCl$_3$

The $^{13}$C NMR spectrum of 1p in CDCl$_3$
The $^1$H NMR spectrum of 1q in CDCl$_3$

![1H NMR Spectrum]

The $^{13}$C NMR spectrum of 1q in CDCl$_3$

![13C NMR Spectrum]
The $^1$H NMR spectrum of 1r in CDCl$_3$

The $^{13}$C NMR spectrum of 1r in CDCl$_3$
The $^1$H NMR spectrum of 1t in CDCl$_3$

The $^{13}$C NMR spectrum of 1t in CDCl$_3$
The $^1$H NMR spectrum of 1aa in CDCl$_3$

The $^{13}$C NMR spectrum of 1aa in CDCl$_3$
The $^1$H NMR spectrum of 1ab in CDCl$_3$

The $^{13}$C NMR spectrum of 1ab in CDCl$_3$
The $^1$H NMR spectrum of 1af in CDCl$_3$

The $^{13}$C NMR spectrum of 1af in CDCl$_3$
The $^1$H NMR spectrum of 1ag in CDCl$_3$

The $^{13}$C NMR spectrum of 1ag in CDCl$_3$
The $^1$H NMR spectrum of 1ah in CDCl$_3$

The $^{13}$C NMR spectrum of 1ah in CDCl$_3$
The $^1$H NMR spectrum of 1ai in CDCl$_3$

![1H NMR spectrum](image)

The $^{13}$C NMR spectrum of 1ai in CDCl$_3$

![13C NMR spectrum](image)
The $^1$H NMR spectrum of 1aj in CDCl$_3$

The $^{13}$C NMR spectrum of 1aj in CDCl$_3$
The $^1$H NMR spectrum of 1ak in CDCl$_3$

The $^{13}$C NMR spectrum of 1ak in CDCl$_3$
The $^1$H NMR spectrum of 1ap in CDCl$_3$

The $^{13}$C NMR spectrum of 1ap in CDCl$_3$
The $^1$H NMR spectrum of 1as in CDCl$_3$

The $^{13}$C NMR spectrum of 1as in CDCl$_3$
The $^1$H NMR spectrum of $2a$ in CDCl$_3$

The $^{13}$C NMR spectrum of $2a$ in CDCl$_3$
The $^1$H NMR spectrum of $2b$ in CDCl$_3$.

The $^{13}$C NMR spectrum of $2b$ in CDCl$_3$. 
The $^1$H NMR spectrum of 2c in CDCl$_3$

The $^{13}$C NMR spectrum of 2c in CDCl$_3$
The $^{19}$F NMR spectrum of $2c$ in CDCl$_3$

The $^1$H NMR spectrum of $2d$ in CDCl$_3$
The $^{13}$C NMR spectrum of 2d in CDCl$_3$

The $^1$H NMR spectrum of 2e in CDCl$_3$
The $^{13}$C NMR spectrum of 2e in CDCl$_3$
The $^{13}$C NMR spectrum of 2f in CDCl$_3$

The $^{19}$F NMR spectrum of 2f in CDCl$_3$
The $^1$H NMR spectrum of 2g in CDCl$_3$

The $^{13}$C NMR spectrum of 2g in CDCl$_3$
The $^1$H NMR spectrum of 2h in CDCl$_3$

The $^{13}$C NMR spectrum of 2h in CDCl$_3$
The $^1$H NMR spectrum of 2i in CDCl$_3$

![H NMR spectrum of 2i](image)

The $^{13}$C NMR spectrum of 2i in CDCl$_3$

![C NMR spectrum of 2i](image)
The $^1$H NMR spectrum of 2j in CDCl$_3$

The $^{13}$C NMR spectrum of 2j in CDCl$_3$
The $^1$H NMR spectrum of 2k in CDCl$_3$

The $^{13}$C NMR spectrum of 2k in CDCl$_3$
The $^1$H NMR spectrum of 2l in CDCl$_3$

![NMR spectrum]

The $^{13}$C NMR spectrum of 2l in CDCl$_3$

![NMR spectrum]
The $^1$H NMR spectrum of 2m in CDCl$_3$

The $^{13}$C NMR spectrum of 2m in CDCl$_3$
The $^1$H NMR spectrum of 2o in CDCl$_3$

The $^{13}$C NMR spectrum of 2o in CDCl$_3$
The $^1$H NMR spectrum of 2p in CDCl$_3$

The $^{13}$C NMR spectrum of 2p in CDCl$_3$
The $^1$H NMR spectrum of 2q in CDCl$_3$

The $^{13}$C NMR spectrum of 2q in CDCl$_3$
The $^1$H NMR spectrum of 2r in CDCl$_3$

The $^{13}$C NMR spectrum of 2r in CDCl$_3$
The $^1$H NMR spectrum of 2s in CDCl$_3$

The $^{13}$C NMR spectrum of 2s in CDCl$_3$
The $^1$H NMR spectrum of $2u$ in CDCl$_3$

The $^{13}$C NMR spectrum of $2u$ in CDCl$_3$
The $^1$H NMR spectrum of 2v in CDCl$_3$

![H NMR spectrum](image)

The $^{13}$C NMR spectrum of 2v in CDCl$_3$

![C NMR spectrum](image)
The $^1$H NMR spectrum of 2w in CDCl$_3$

The $^{13}$C NMR spectrum of 2w in CDCl$_3$
The $^1$H NMR spectrum of 2x in CDCl$_3$

The $^{13}$C NMR spectrum of 2x in CDCl$_3$
The $^1$H NMR spectrum of 2y in CDCl$_3$

The $^{13}$C NMR spectrum of 2y in CDCl$_3$
The $^1$H NMR spectrum of $2z$ in CDCl$_3$

The $^{13}$C NMR spectrum of $2z$ in CDCl$_3$
The $^1$H NMR spectrum of 3a in CDCl$_3$

The $^{13}$C NMR spectrum of 3a in CDCl$_3$
The $^1$H NMR spectrum of $3b$ in CDCl$_3$

The $^{13}$C NMR spectrum of $3b$ in CDCl$_3$
The $^1$H NMR spectrum of Y-2 in THF-$d_8$

The $^{13}$C NMR spectrum of Y-2 in THF-$d_8$