

Electronic Supporting Information

A greener approach towards the synthesis of *N*-heterocyclic thiones and selones using mechanochemical technique

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† Electronic supplementary information (ESI): Tables of structural data and spectroscopic data. CCDC 2189643-2189645. For ESI and crystallographic data in CIF or other electronic format see DOI:

‡ These authors contributed equally to this work.

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Crystal Structure Determination of Compounds 4-6.

Single crystals of all compounds were mounted on a Cryoloop with a drop of Paratone oil and positioned in the cold nitrogen stream on a Rigaku Saturn724+ (2x2 bin mode) diffractometer (for **5** and **6**) and Bruker D8 Venture (for **4**). The data were reduced using CrysAlisPro 1.171.41.93a (Rigaku Oxford Diffraction, 2020) software. The structures were solved using Olex2¹ with the ShelXT² structure solution program using intrinsic phasing and refined with the SHELXL³ refinement package using least-squares minimization. All non-hydrogen atoms were refined anisotropically. Hydrogen atoms were placed in calculated positions and included as riding contributions with isotropic displacement parameters tied to those of the attached non-hydrogen atoms. The given chemical formula and other crystal data do not take into account the unknown solvent molecule(s). The reflections with error/esd more than 10 were excluded in order to avoid problems related to better refinement of the data. The data completeness is more than 99.8% in most of the cases, which is enough to guarantee a very good refinement of data. The details of X-ray structural determinations are given in Tables S1. Crystallographic data for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication no. CCDC 2189643 (compound **4**), 2189644 (compound **5**) and 2189645 (compound **6**).

Table S1 Crystallography details.

| | Comp_4 | Comp_5 | Comp_6 |
|---|---|--|---|
| Empirical formula | C ₁₇ H ₁₇ N ₂ OPSe | C ₂₀ H ₂₃ N ₂ OPS | C ₂₀ H ₂₃ N ₂ OPSe |
| Formula weight | 375.25 | 370.43 | 417.33 |
| Temperature/K | 150.15 | 150 | 150.00 |
| Crystal system | orthorhombic | monoclinic | monoclinic |
| Space group | Pbca | P2 ₁ /c | P2 ₁ /c |
| a/Å | 10.9392(3) | 10.6482(15) | 10.6448(7) |
| b/Å | 8.0482(3) | 21.1068(17) | 21.0365(8) |
| c/Å | 37.8833(12) | 9.3551(10) | 9.5229(5) |
| α/° | 90 | 90 | 90 |
| β/° | 90 | 112.860(14) | 113.495(7) |
| γ/° | 90 | 90 | 90 |
| Volume/Å ³ | 3335.28(19) | 1937.4(4) | 1955.7(2) |
| Z | 8 | 4 | 4 |
| ρ _{calc} /cm ³ | 1.495 | 1.270 | 1.417 |
| μ/mm ⁻¹ | 2.350 | 0.260 | 2.012 |
| F(000) | 1520.0 | 784.0 | 856.0 |
| Crystal size/mm ³ | 0.102 × 0.068 × 0.056 | 0.089 × 0.067 × 0.058 | 0.098 × 0.068 × 0.056 |
| 2θ range for data collection/° | 4.3 to 65.32 | 4.152 to 49.994 | 3.872 to 62.346 |
| Reflections collected | 55566 | 12285 | 16625 |
| Independent reflections | 5557 [R _{int} = 0.0984] | 3409 [R _{int} = 0.1067] | 5747 [R _{int} = 0.0409] |
| Data/restraints/parameters | 5557/0/200 | 3409/55/264 | 5747/85/265 |
| Goodness-of-fit on F ² | 1.025 | 1.059 | 1.046 |
| R ₁ | 0.0528 | 0.0752 | 0.0476 |
| wR ₂ | 0.1419 | 0.1860 | 0.1314 |
| Largest diff. peak/hole /e Å ⁻³ | 0.30/-0.83 | 0.59/-0.40 | 1.57/-0.50 |

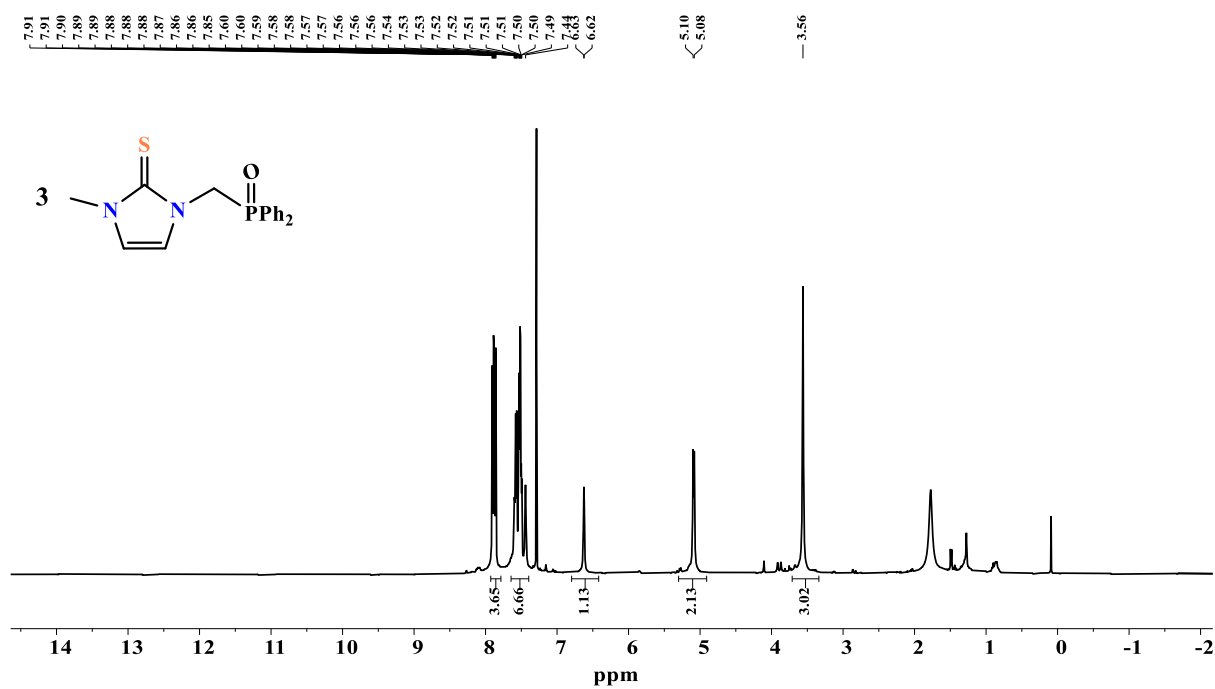


Fig. S1 ¹H NMR spectrum of **3**.

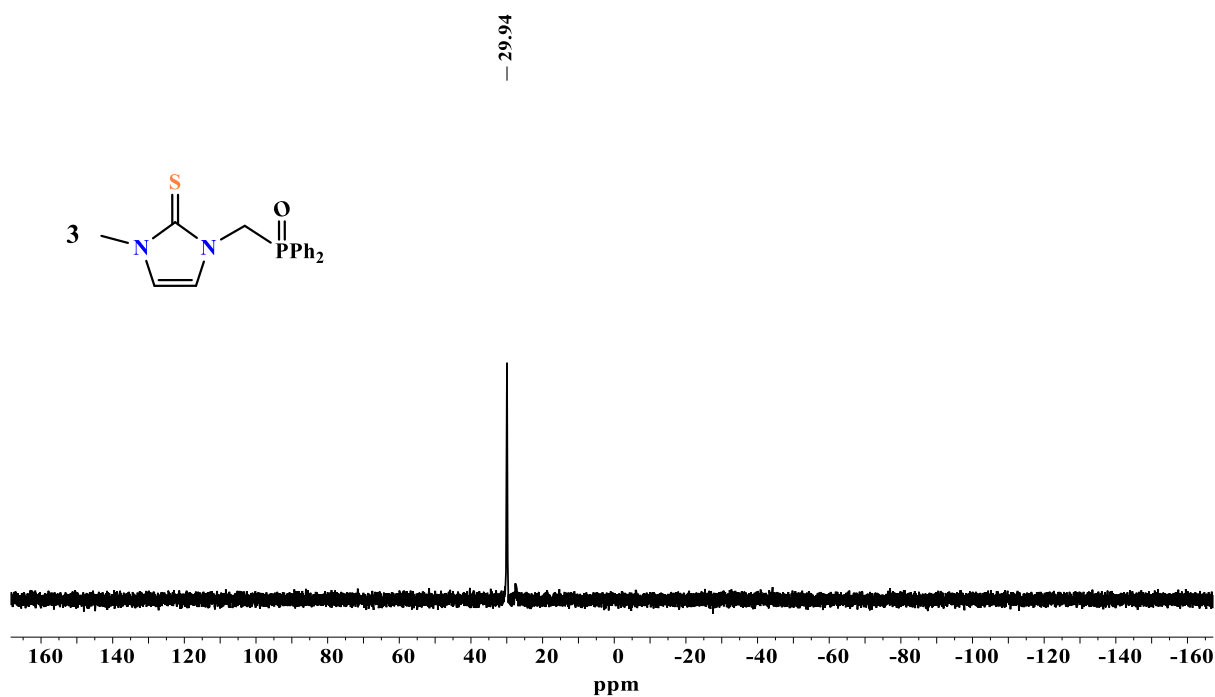


Fig. S2 ³¹P{¹H} NMR spectrum of **3**.

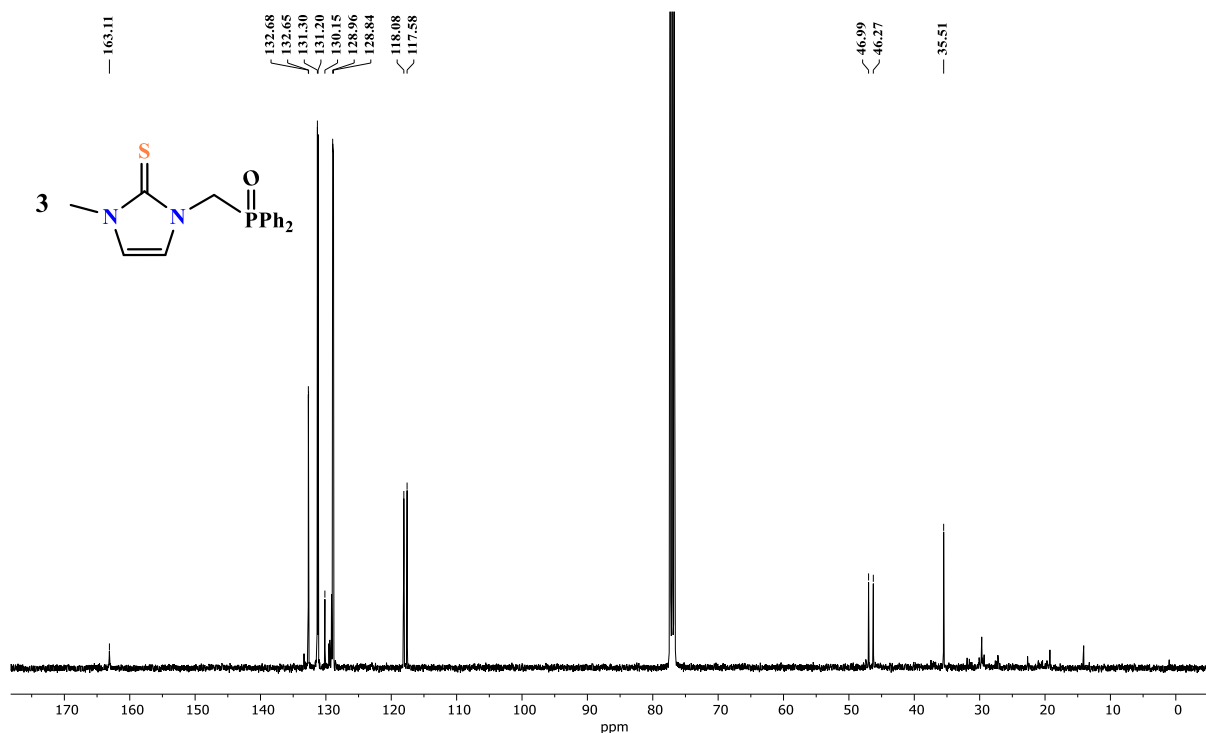


Fig. S3 ¹³C NMR spectrum of **3**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

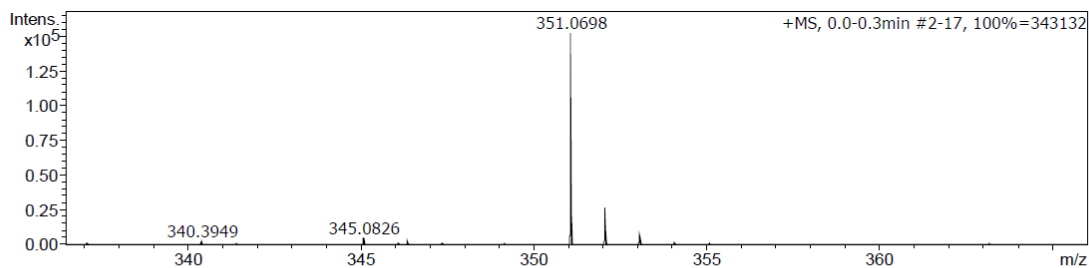
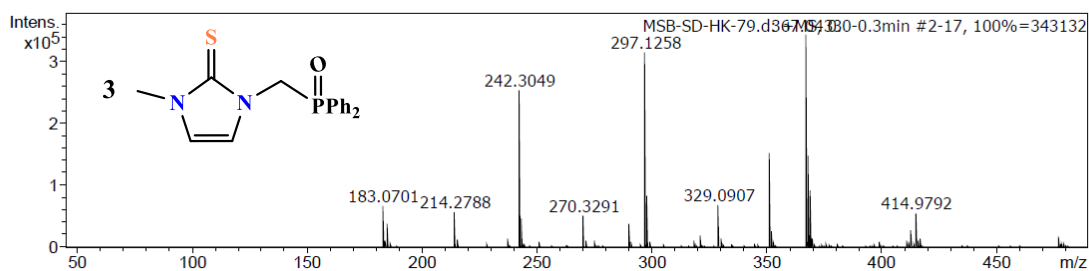
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 Sample Name MSB-SD-HK-79
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Acquisition Date 3/12/2020 11:40:07 AM

Operator SJG-out
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| Focus | Active | Set Capillary | 4000 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1200.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|---------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 351.0698 | 1 | C17H17N2NaOPS | 351.0691 | -1.9 | 16.3 | 1 | 100.00 | 10.5 | even | ok |

Fig. S4 HRMS spectrum of 3.

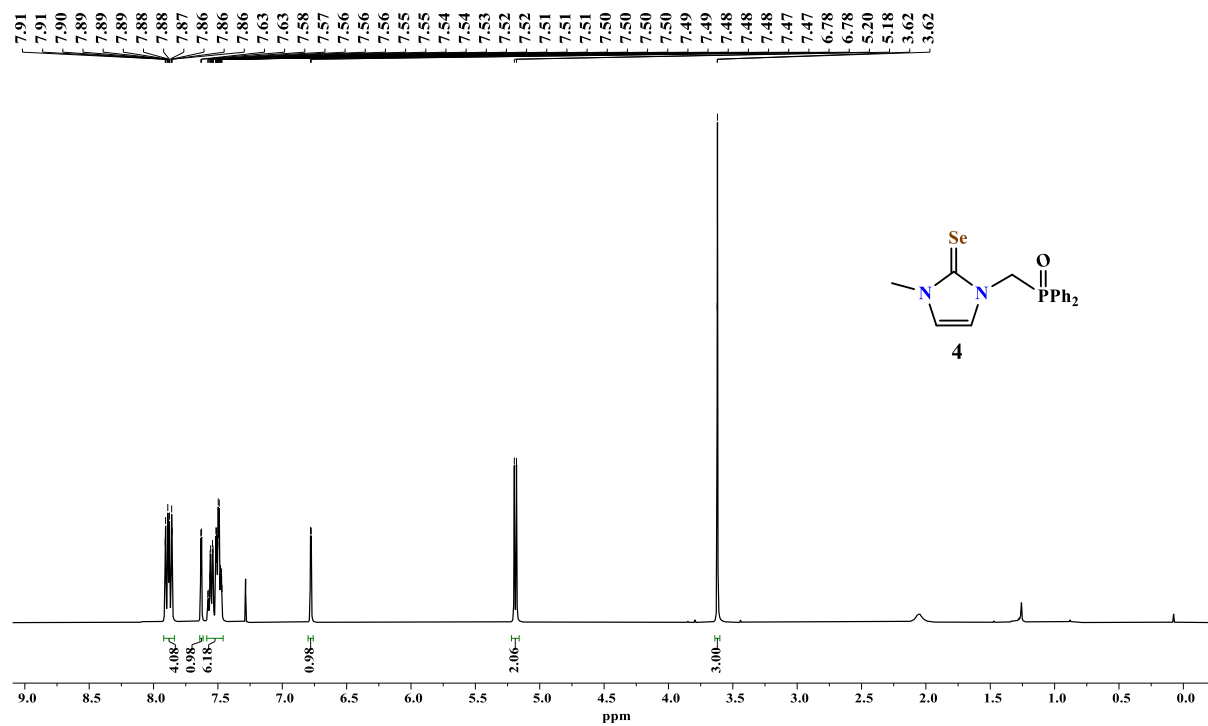


Fig. S5 ^1H NMR spectrum of 4.

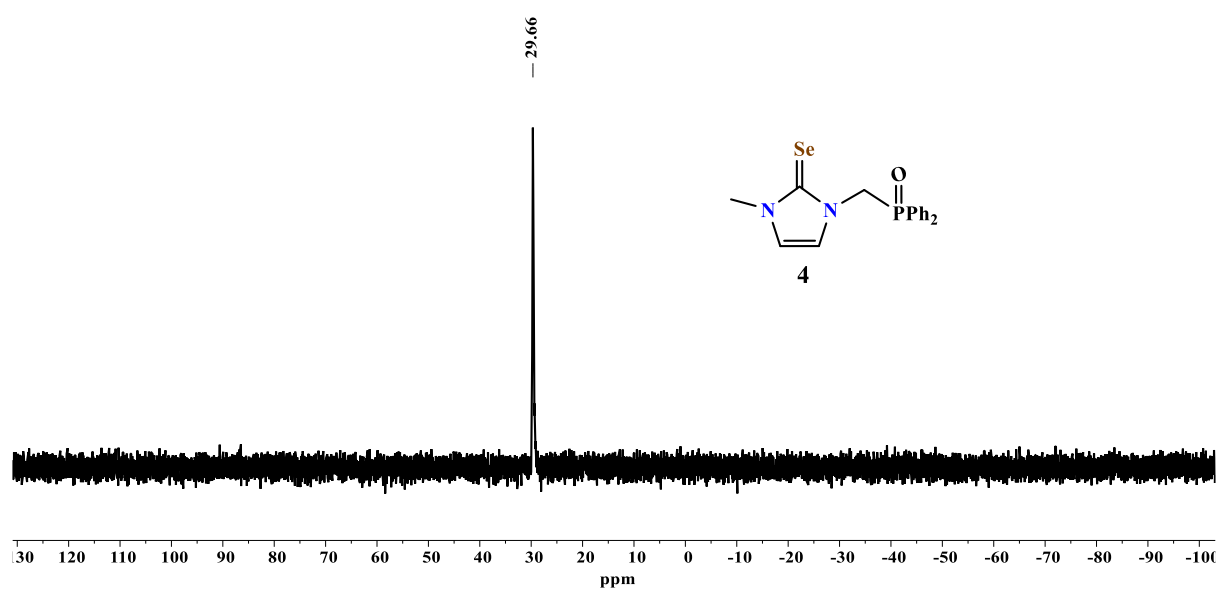


Fig. S6 $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of **4**.

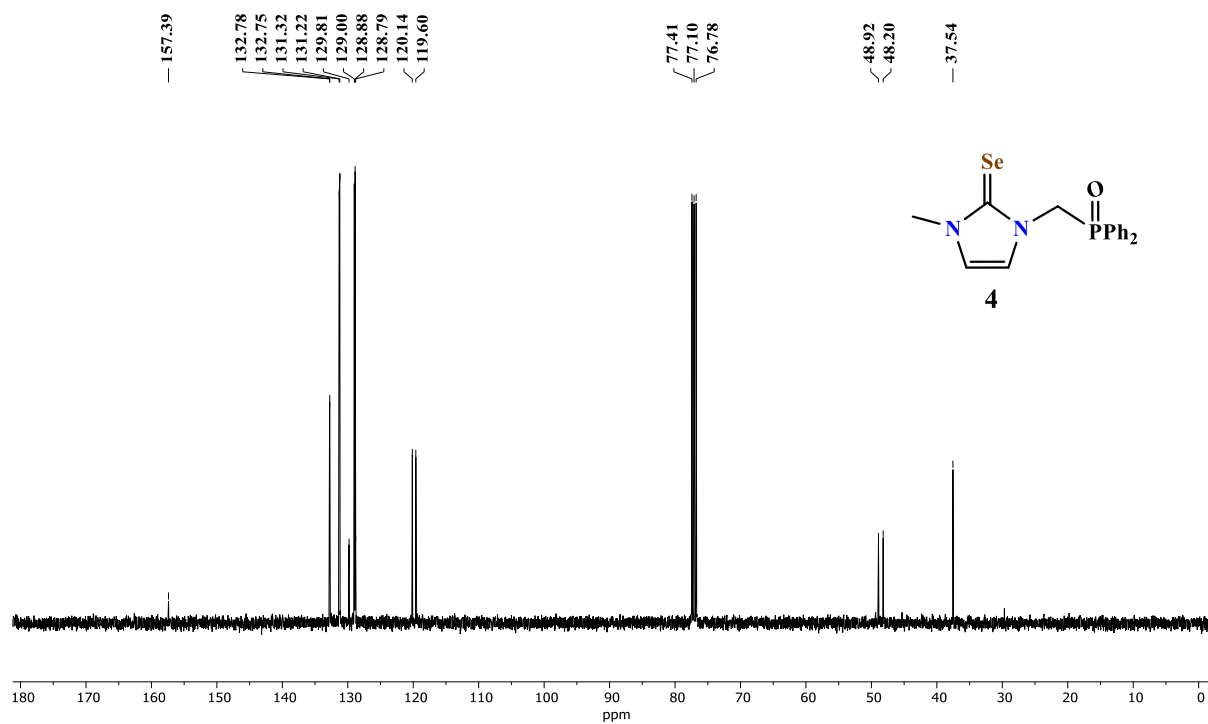


Fig. S7 ^{13}C NMR spectrum of **4**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

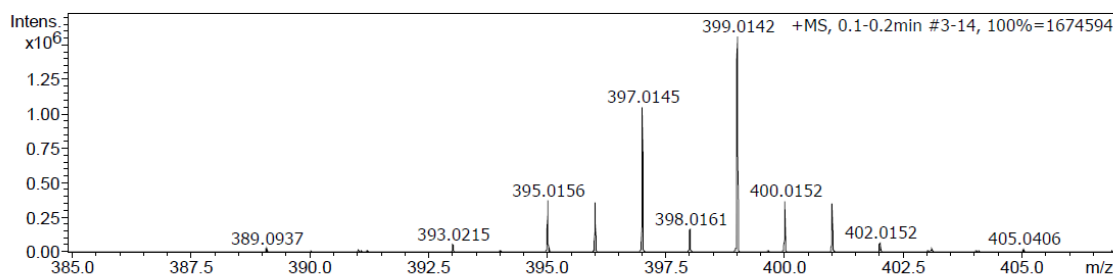
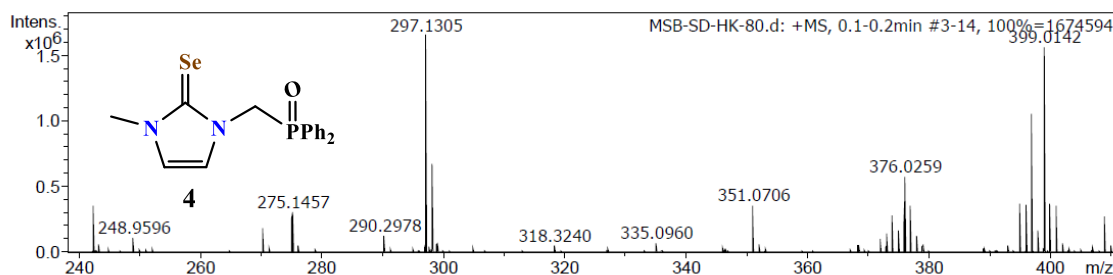
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Acquisition Date 3/12/2020 11:32:08 AM

Operator SJG-out
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1300.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|----------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 399.0142 | 1 | C17H17N2NaOPSe | 399.0137 | -1.4 | 59.3 | 1 | 100.00 | 10.5 | even | ok |

Fig. S8 HRMS spectrum of **4**.

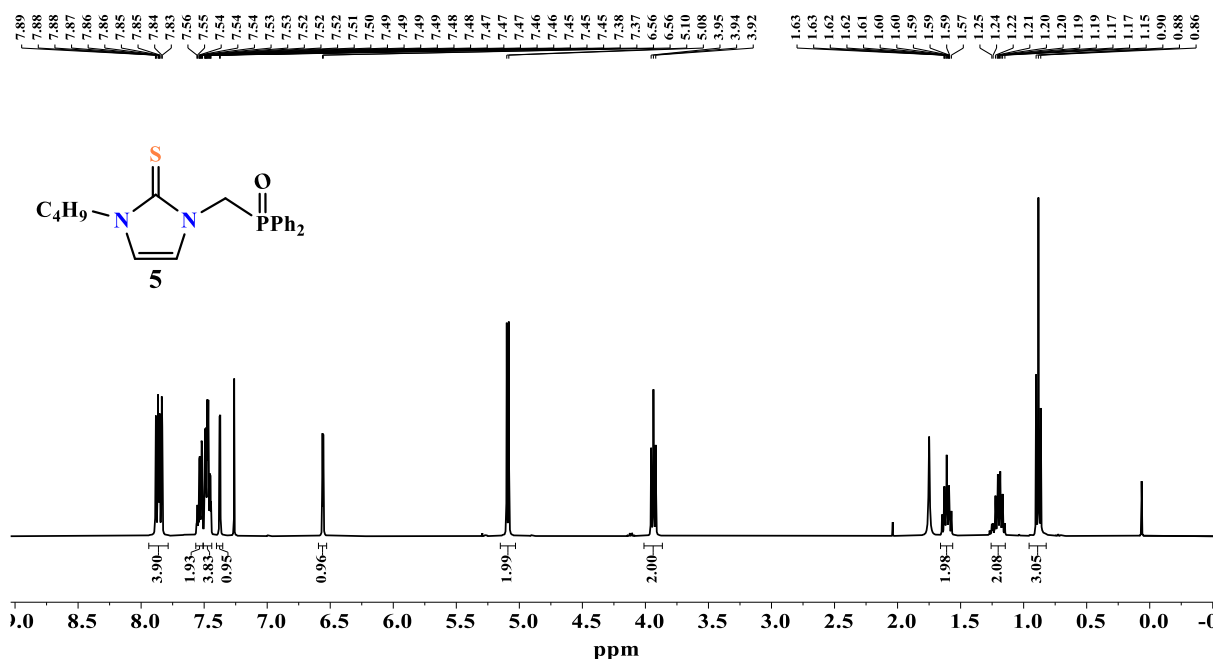


Fig. S9 ¹H NMR spectrum of **5**.

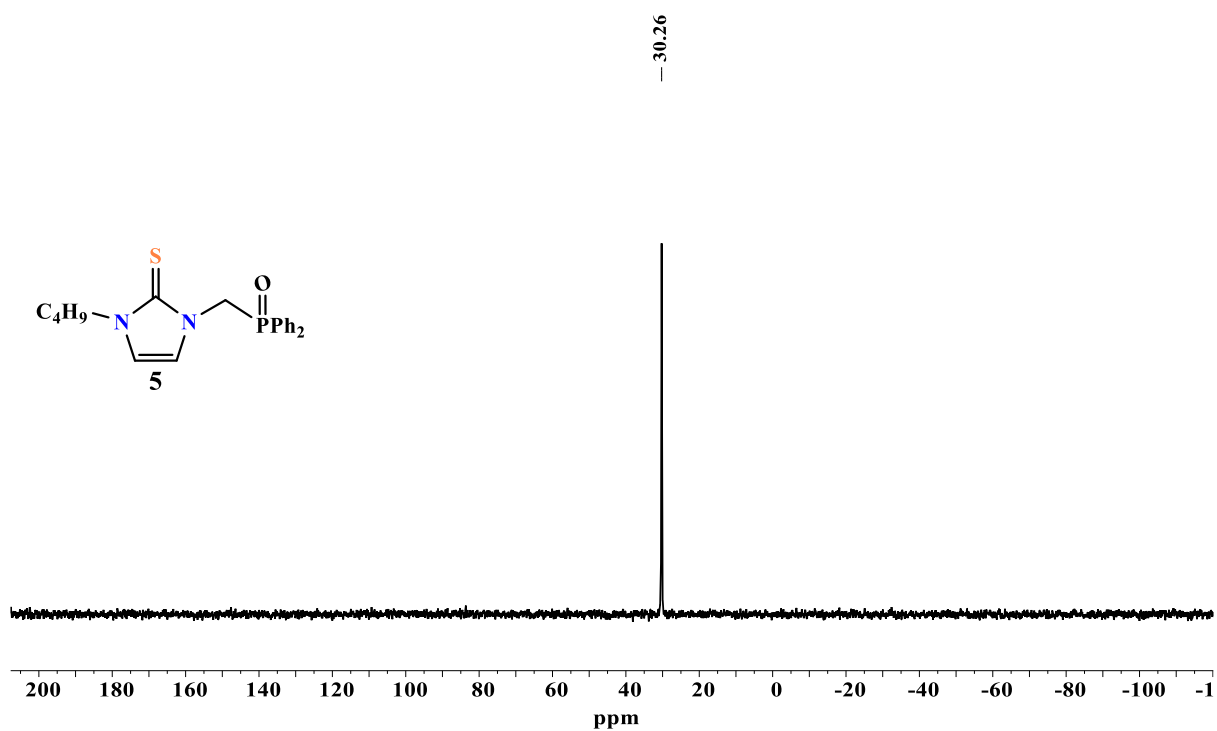


Fig. S10 $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of **5**.

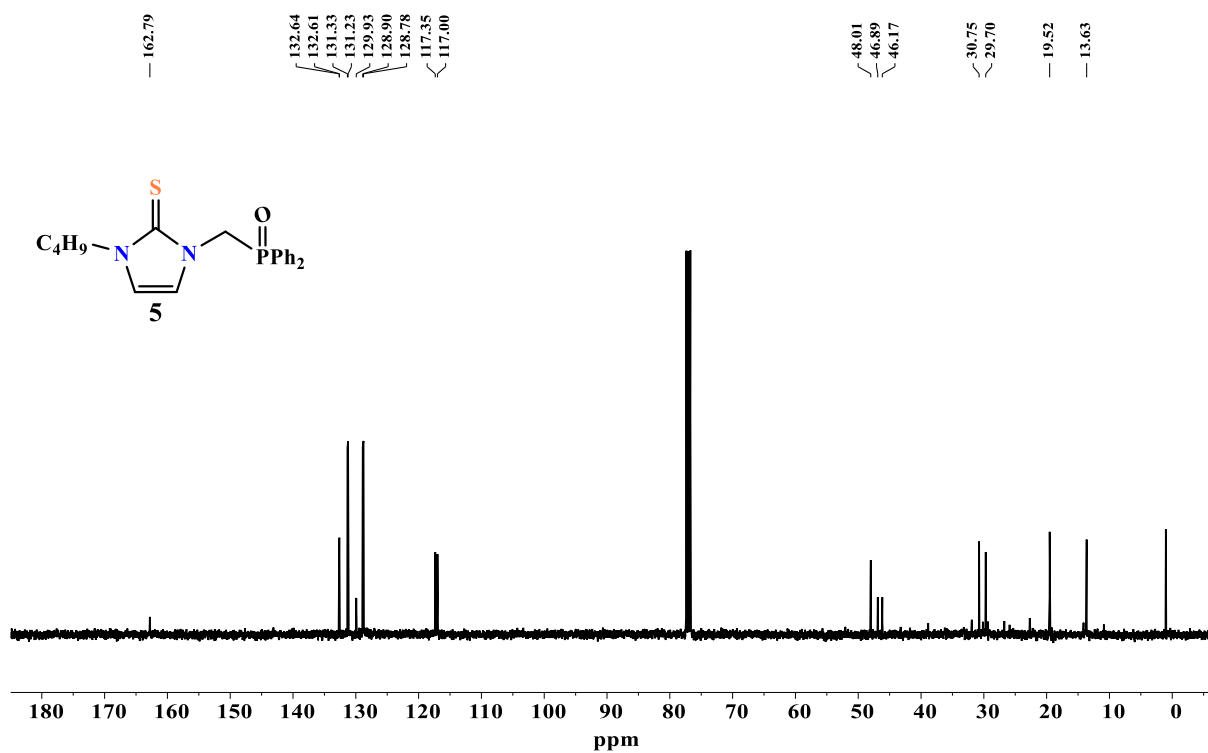


Fig. S11 ^{13}C NMR spectrum of **5**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

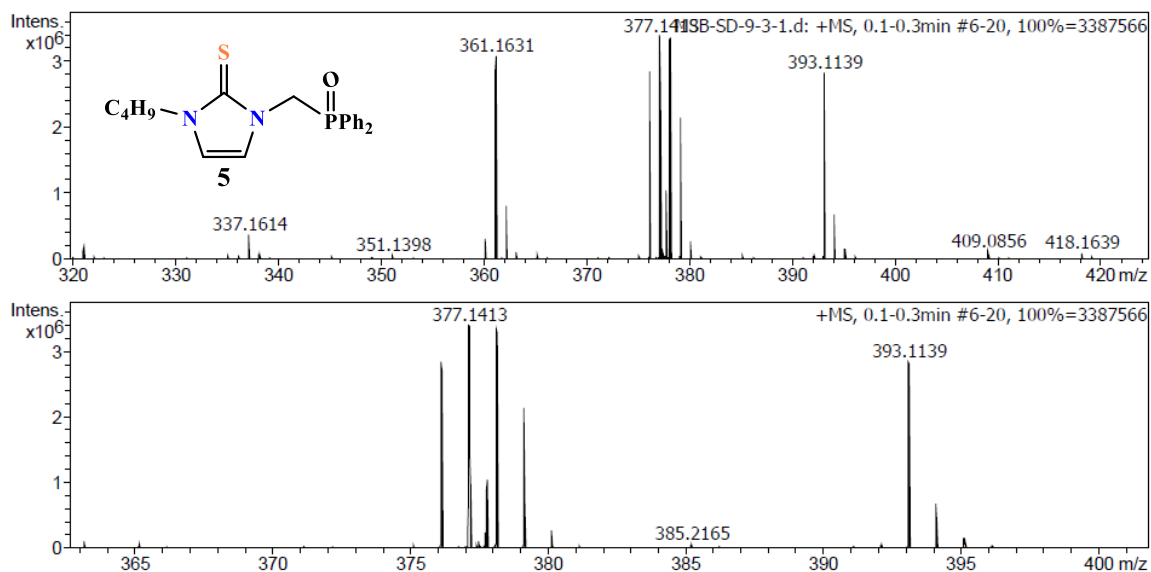
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 Sample Name MSB-SD-9-3-1
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Acquisition Date 11/26/2019 11:41:22 PM
 Operator INN-IN
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1200.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|---------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 377.1413 | 1 | C20H23LIN2OPS | 377.1424 | 2.8 | 538.9 | 1 | 100.00 | 10.5 | even | ok |

Fig. S12 HRMS spectrum of **5**.

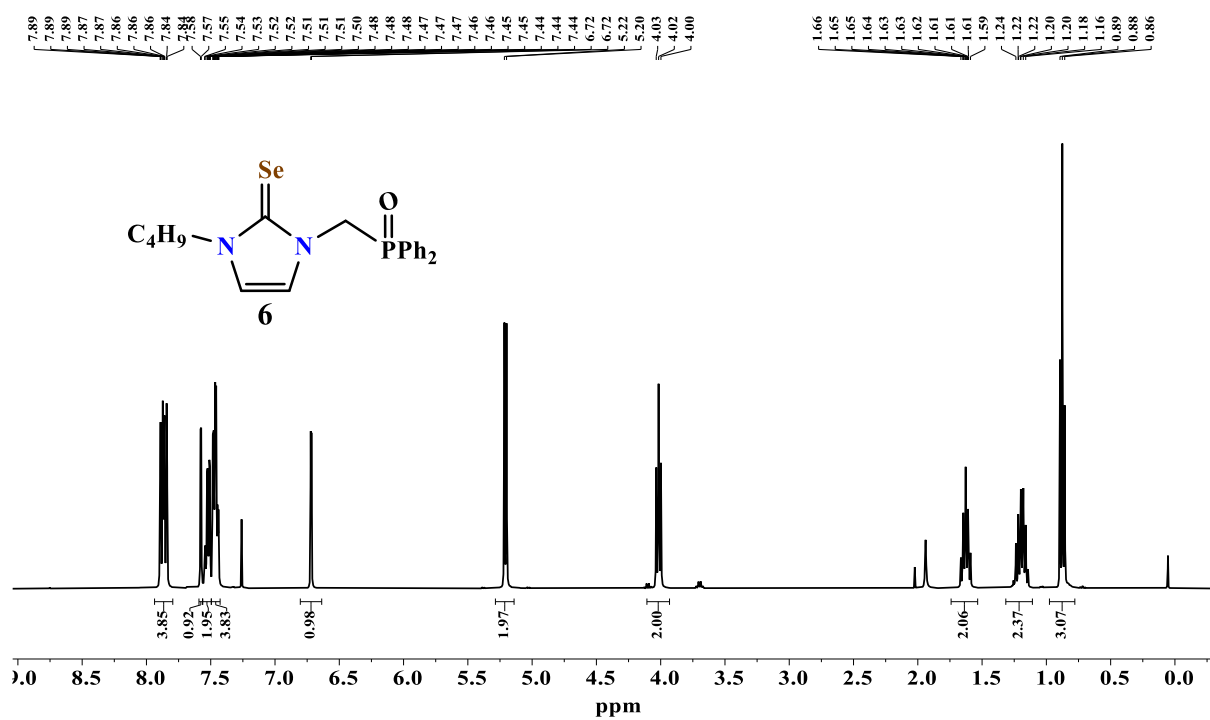


Fig. S13 ^1H NMR spectrum of **6**.

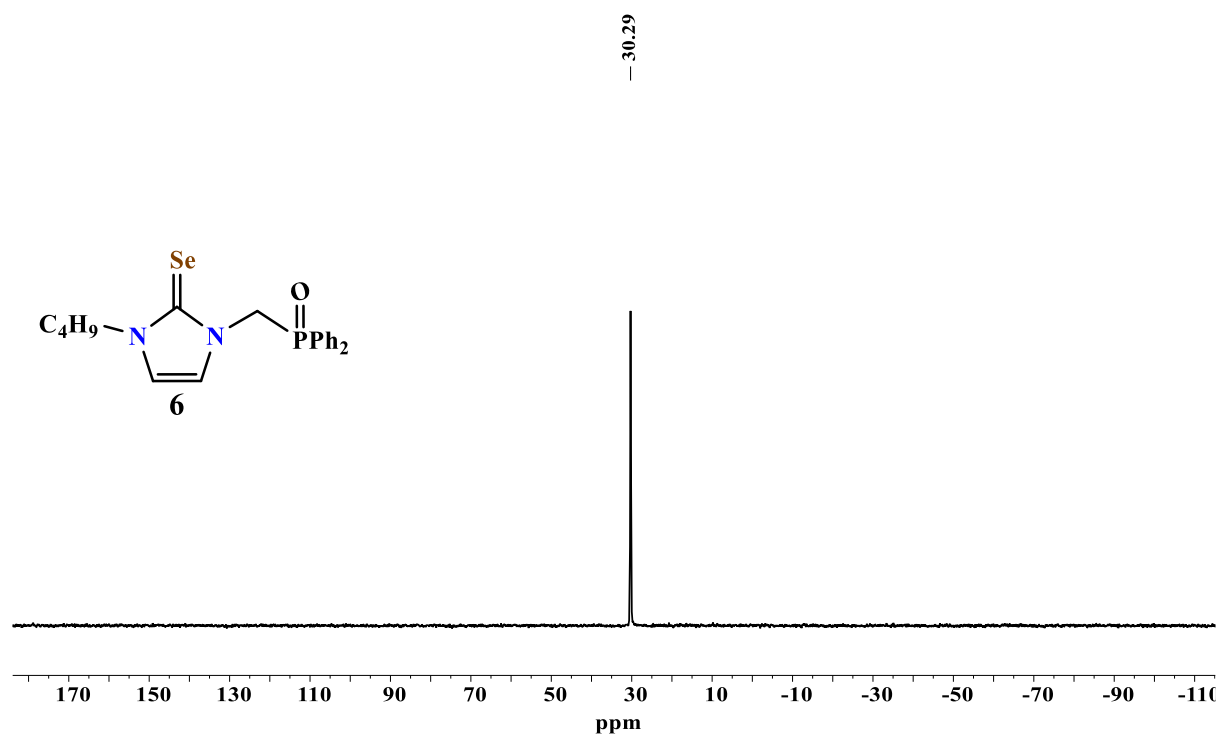


Fig. S14 $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of **6**.

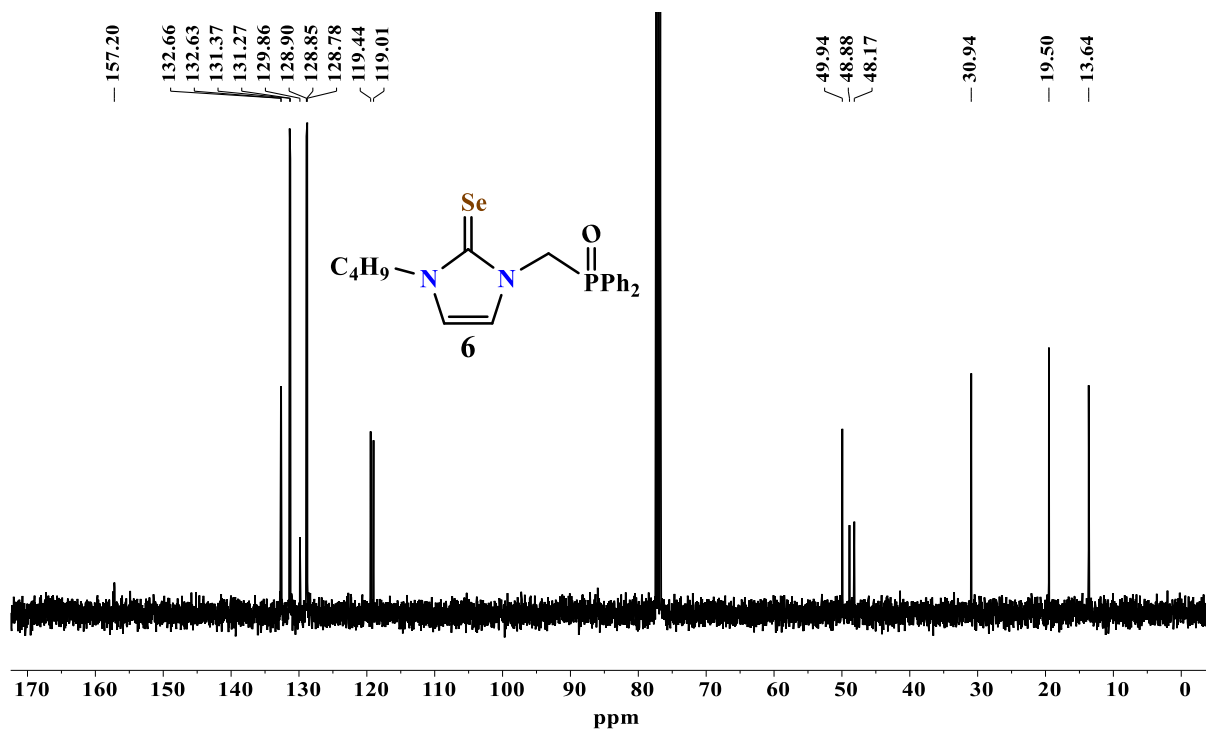


Fig. S15 ^{13}C NMR spectrum of **6**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

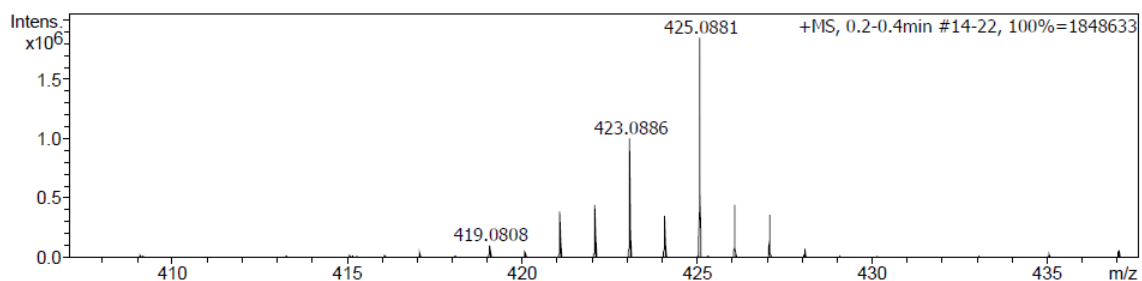
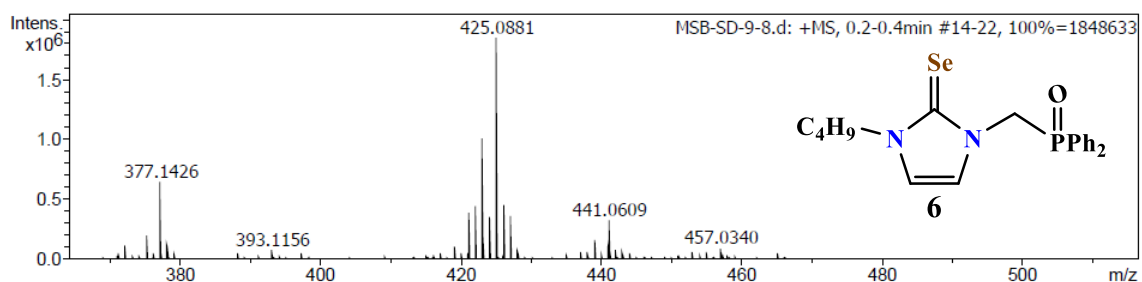
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Acquisition Date 11/26/2019 11:48:04 PM

Operator INN-IN
 Instrument maXis impact 282001.00081

Acquisition Parameter

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|-------------|----------|-----------------------|------------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1200.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|----------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 425.0881 | 1 | C20H23LiN2OPSe | 425.0869 | -2.8 | 11.6 | 1 | 100.00 | 10.5 | even | ok |

Fig. S16 HRMS spectrum of **6**.

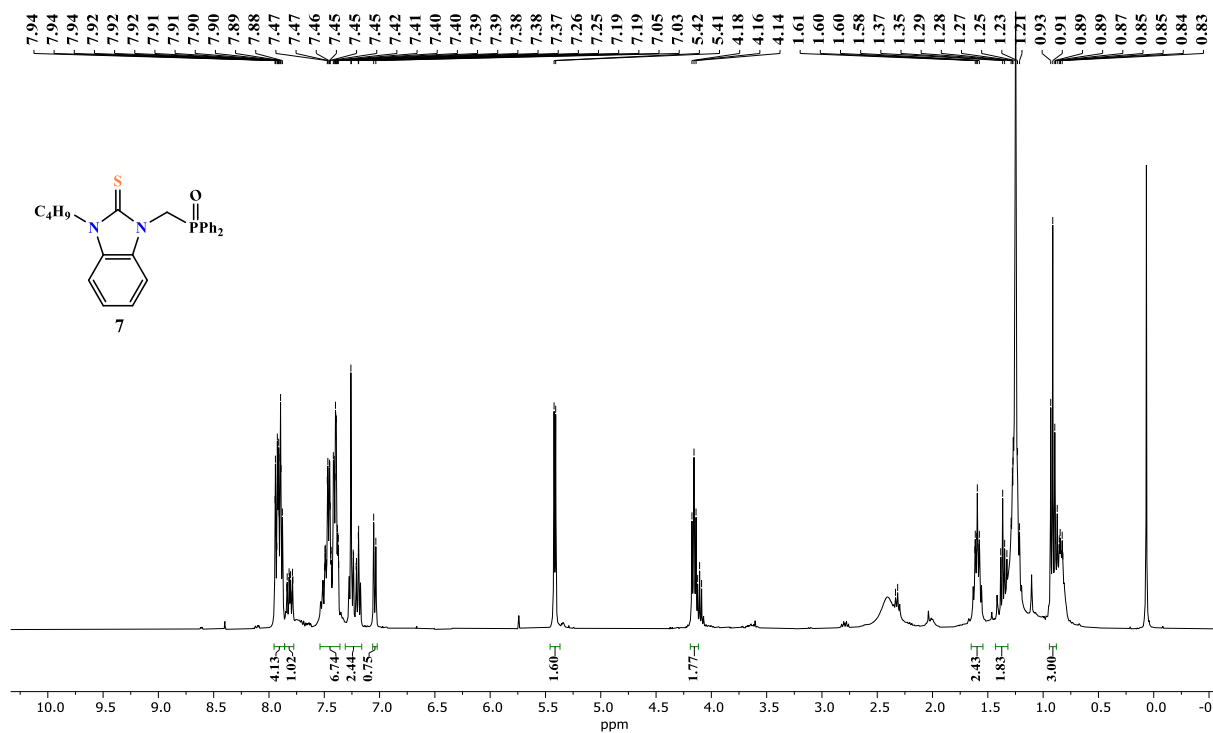


Fig. S17 ¹H NMR spectrum of 7.

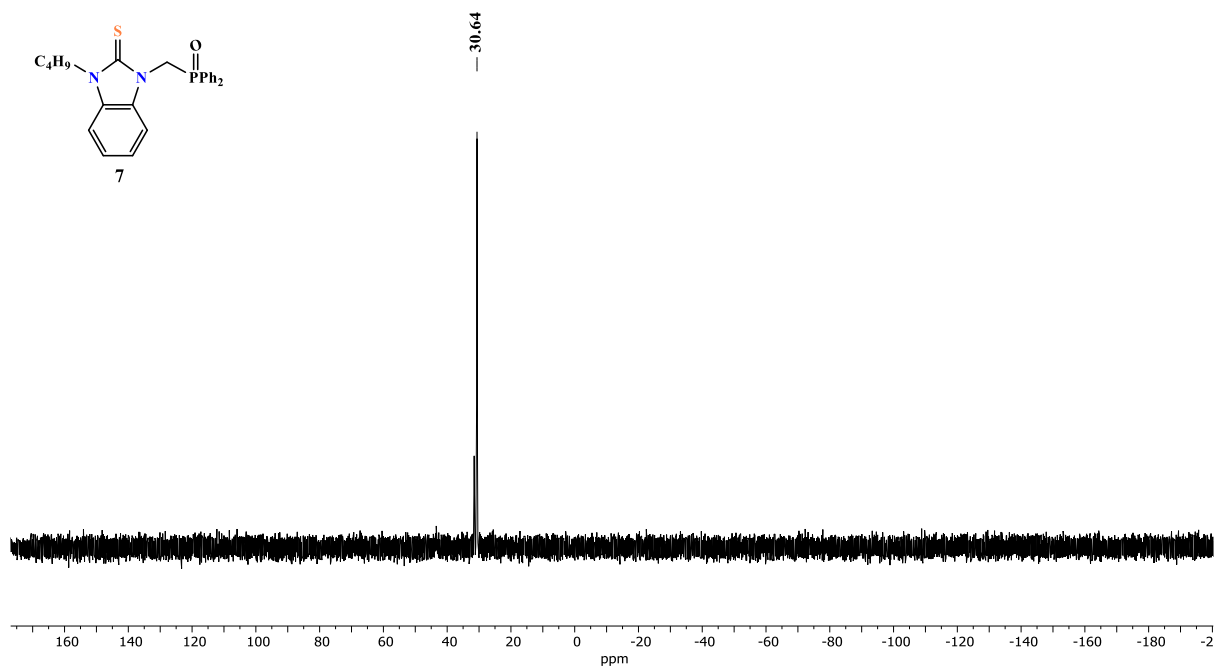


Fig. S18 ³¹P{¹H} NMR spectrum of 7.

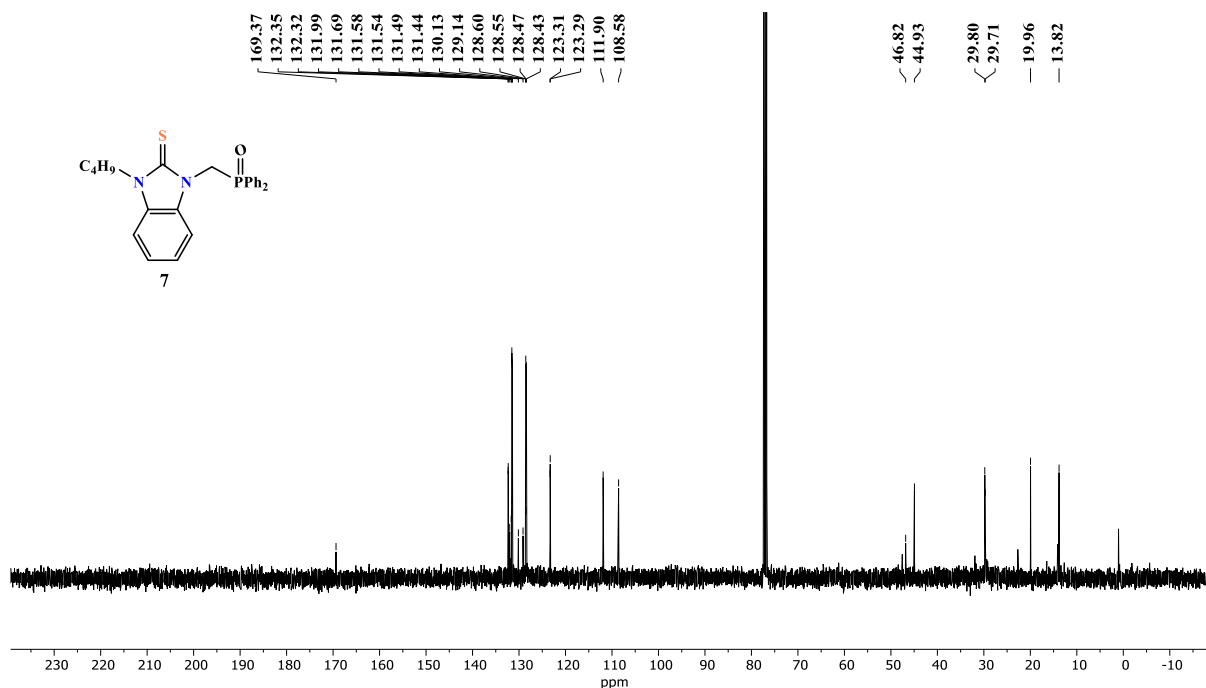


Fig. S19 ¹³C NMR spectrum of 7.

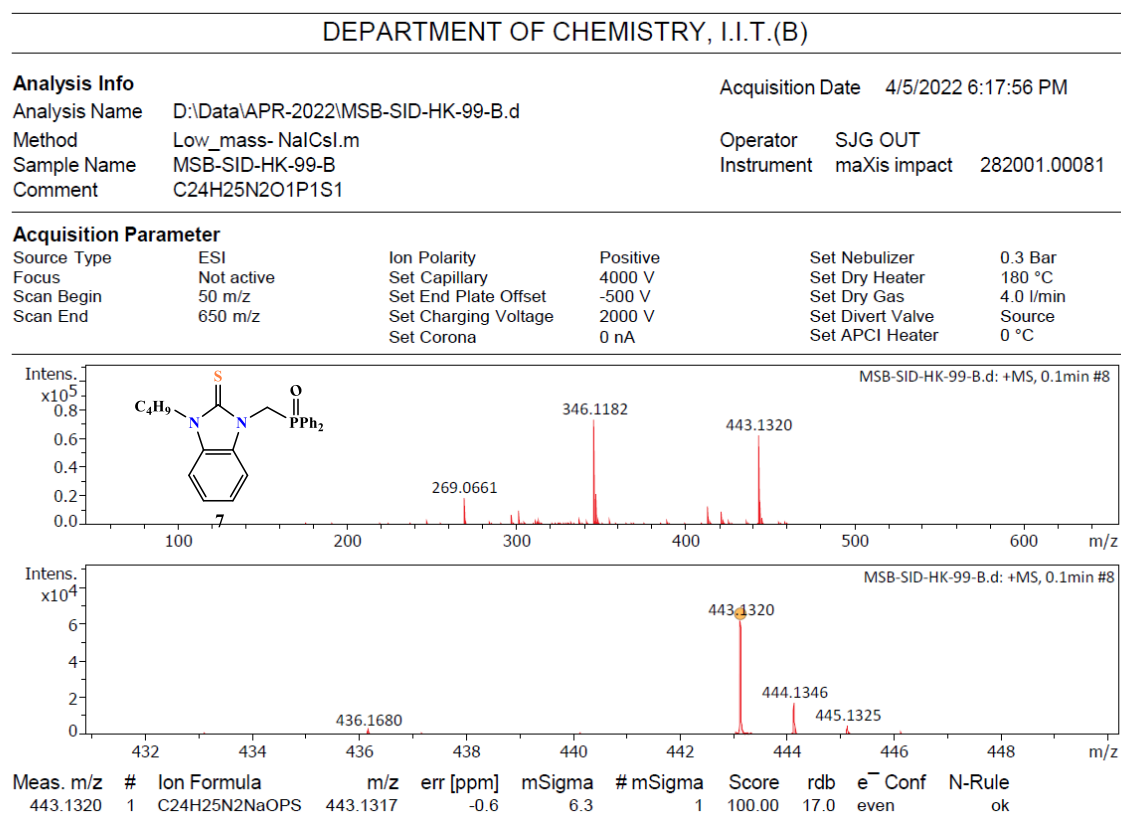


Fig. S20 HRMS spectrum of 7.

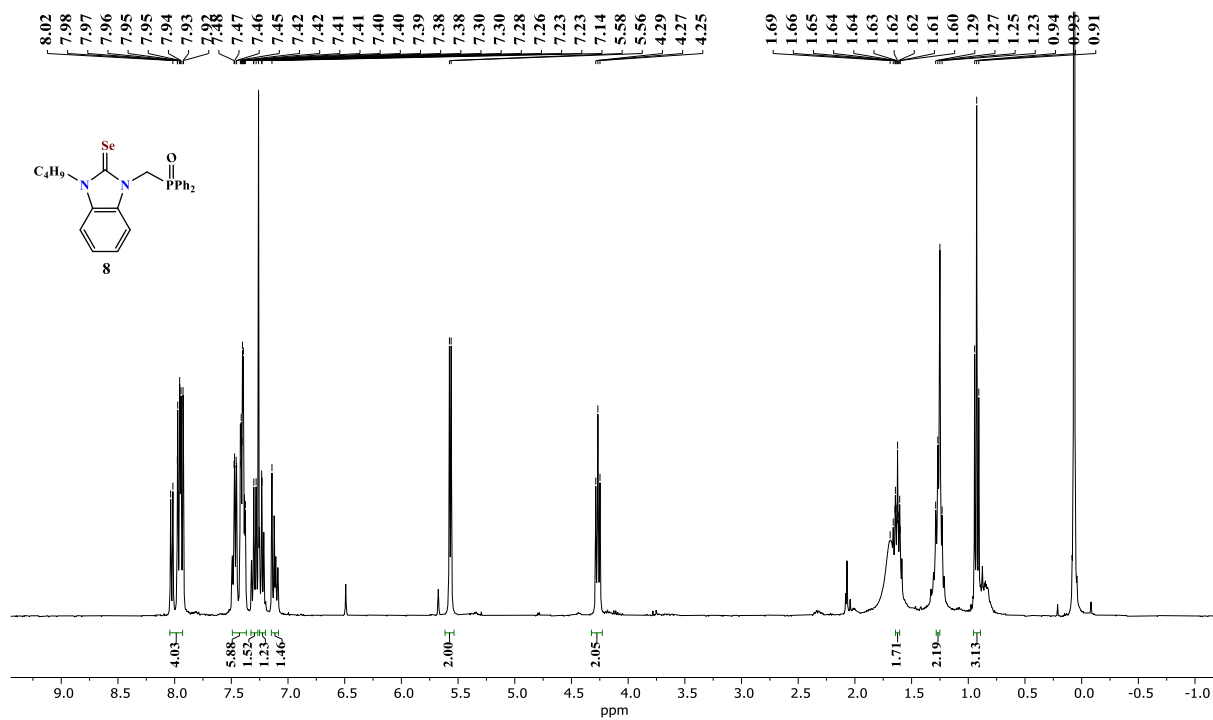


Fig. S21 ^1H NMR spectrum of **8**.

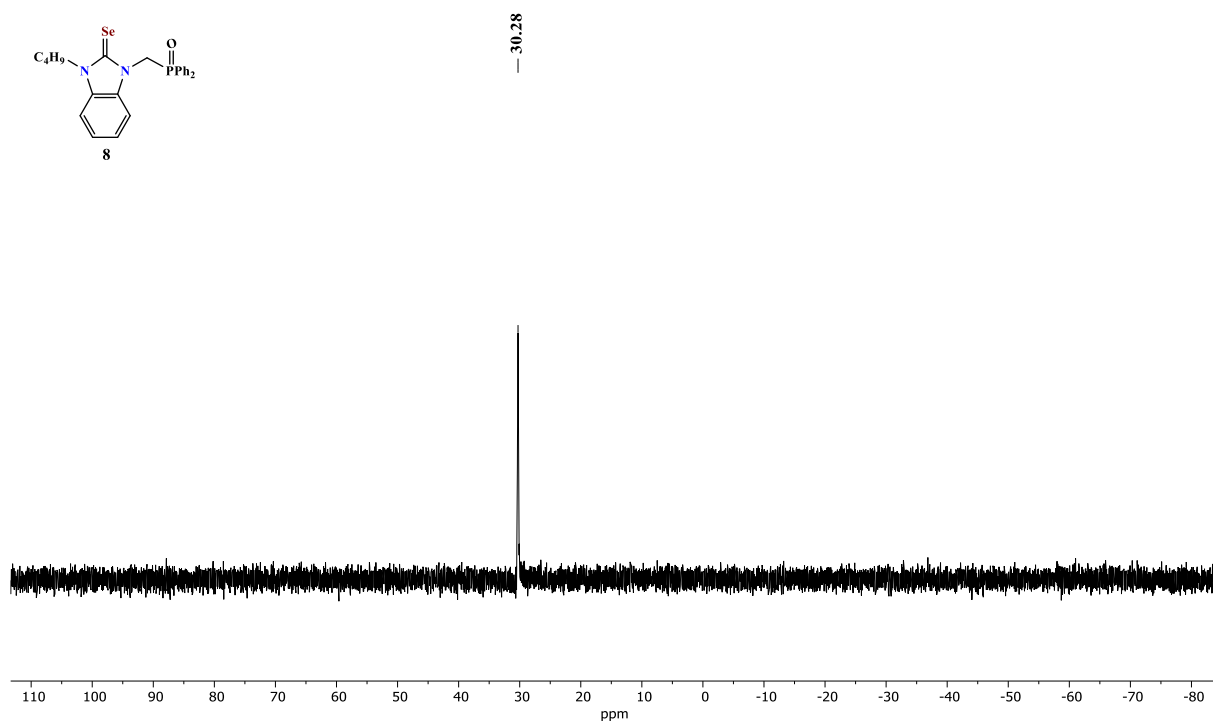


Fig. S22 $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of **8**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name D:\Data\APR-2022\MSB-SID-HK-100-B.d
 Method Naformat_pos_1000.m
 Sample Name MSB-SID-HK-100-B
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Acquisition Date 4/5/2022 6:28:23 PM

Operator SJG OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |

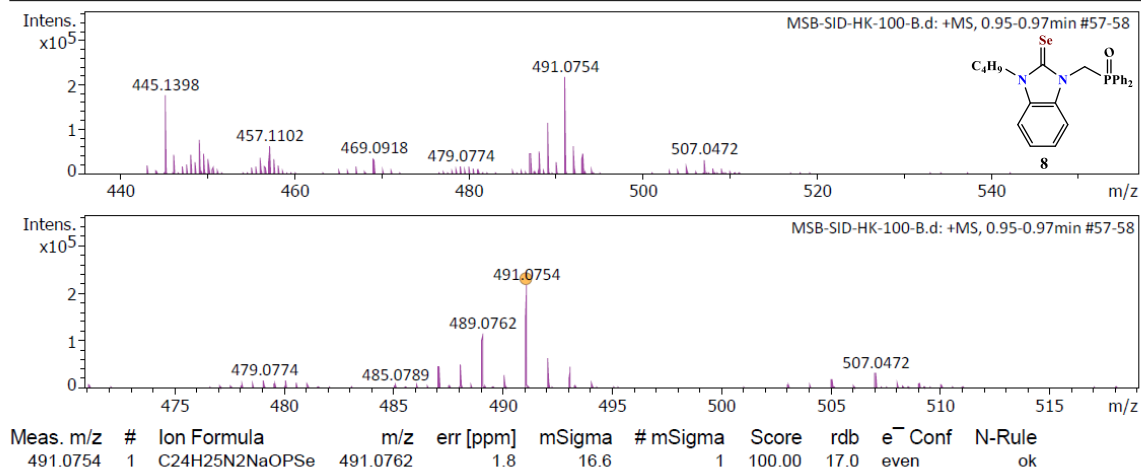


Fig. S24 HRMS spectrum of 8.

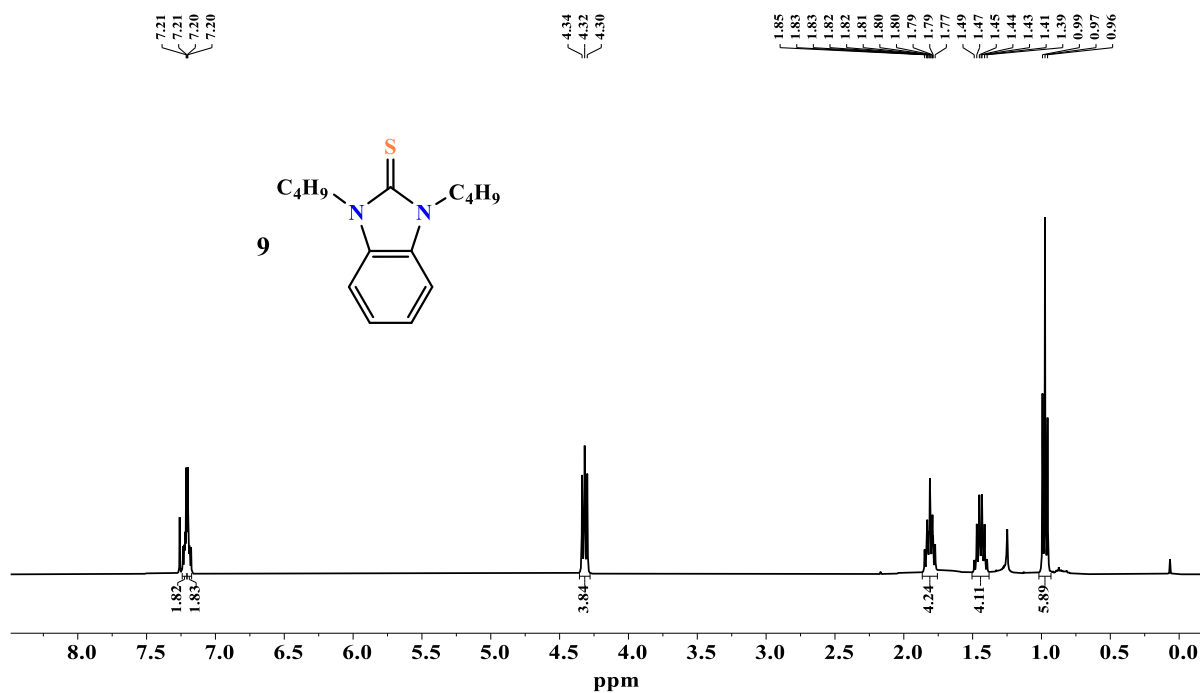


Fig. S25 ¹H NMR spectrum of 9.

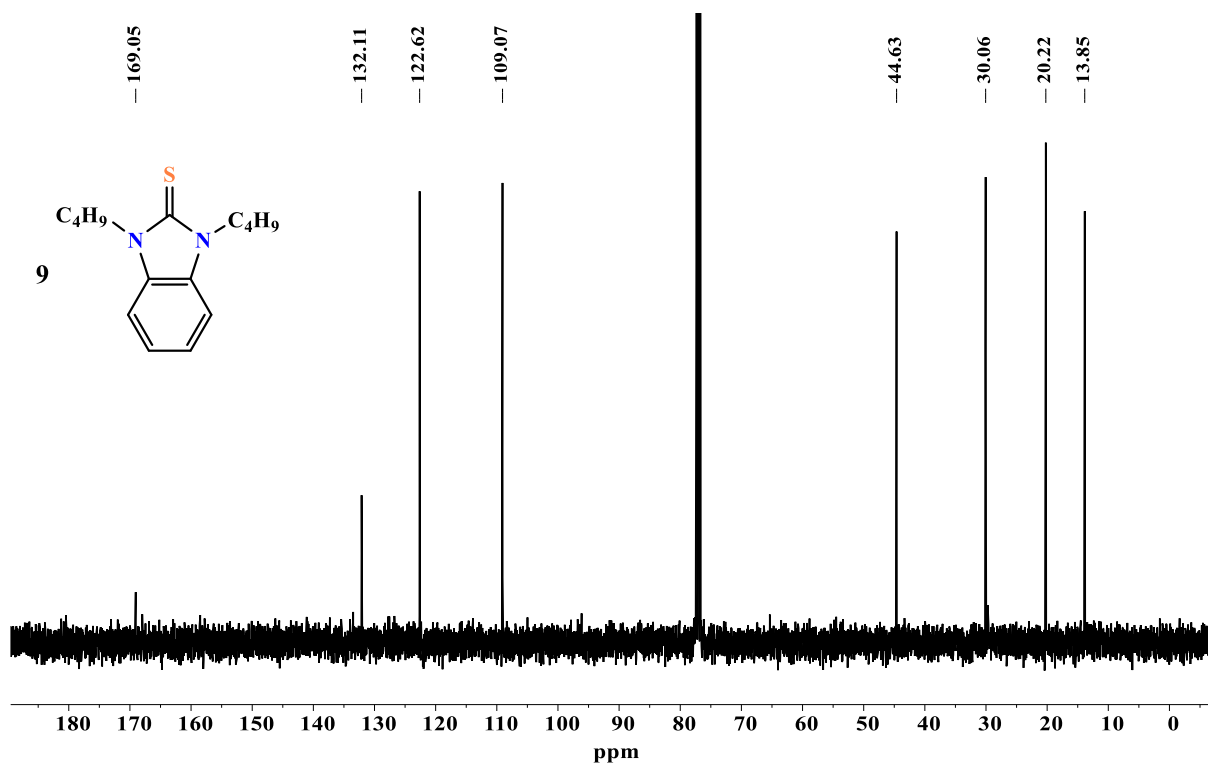


Fig. S26 ^{13}C NMR spectrum of **9**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

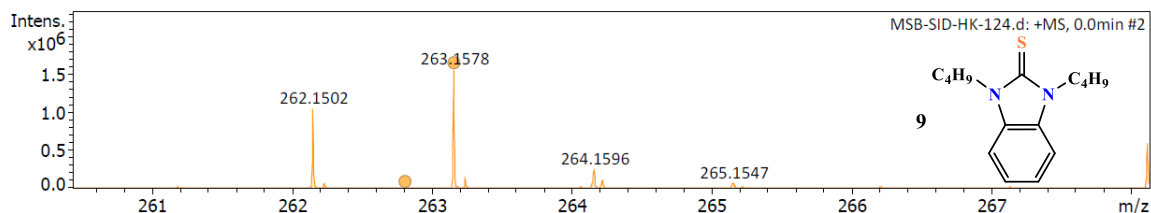
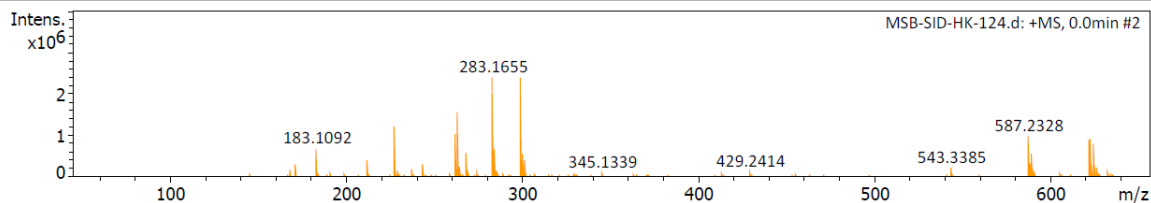
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 Comment C15H22N2Se

Acquisition Date 1/21/2022 8:17:48 PM
 Operator SJG-IN
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|----------|--------|-----|---------------------|--------|
| 263.1578 | 1 | C15H23N2S | 263.1576 | -0.7 | 15.7 | 1 | 100.00 | 8.0 | even | ok |

Fig. S27 HRMS spectrum of 9.

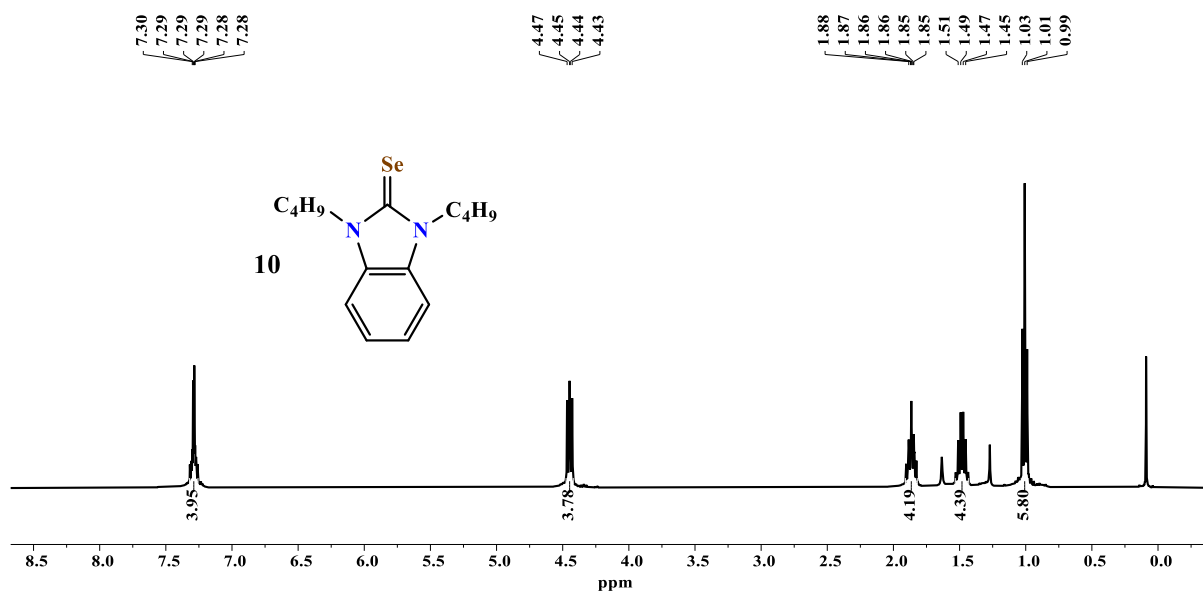


Fig. S28 ¹H NMR spectrum of 10.

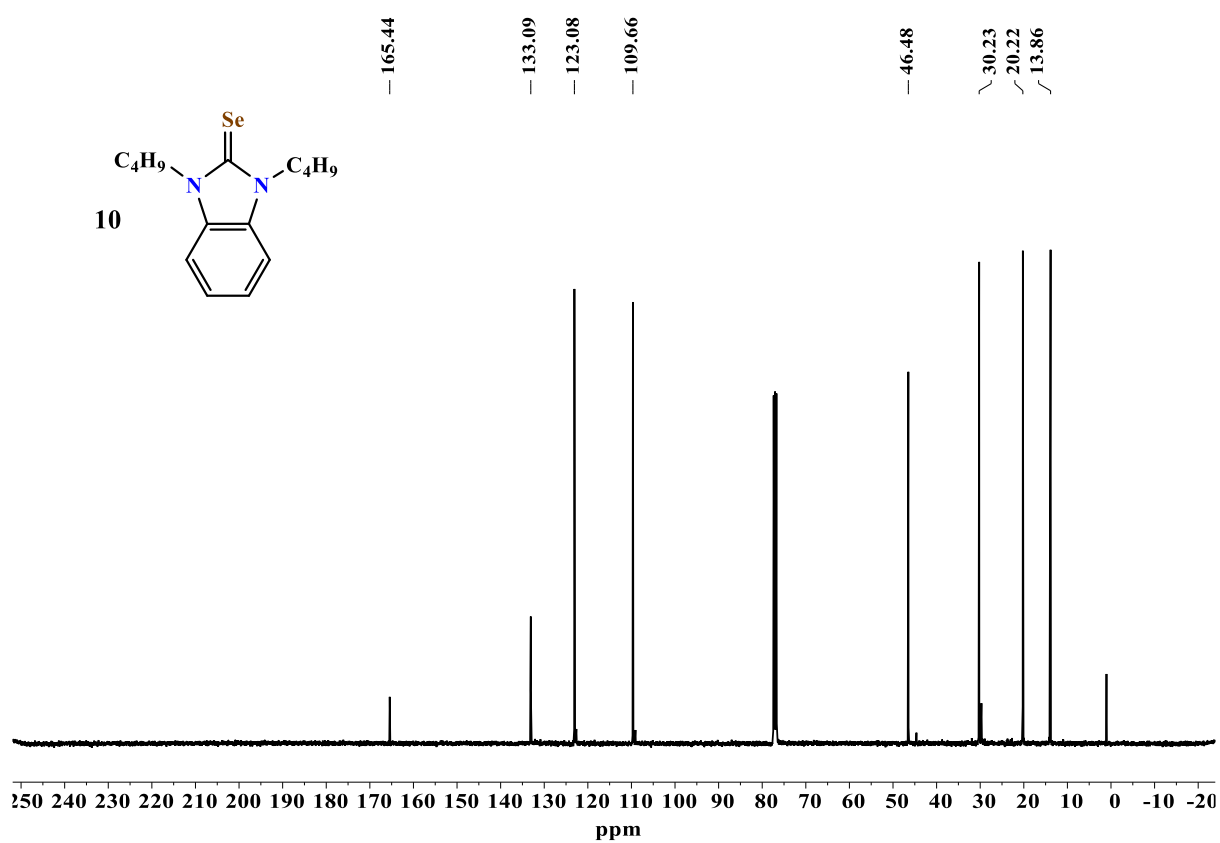


Fig. S29 ^{13}C NMR spectrum of **11**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name D:\Data\AUG 2021\MSB-SID-HK-125.d
 Method Low_mass- NAF.m
 Sample Name MSB-SID-HK-125
 Comment C19H30N2Se

Acquisition Date 8/12/2021 2:30:52 AM

Operator MSB-OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| Focus | Active | Set Capillary | 3200 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
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| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |

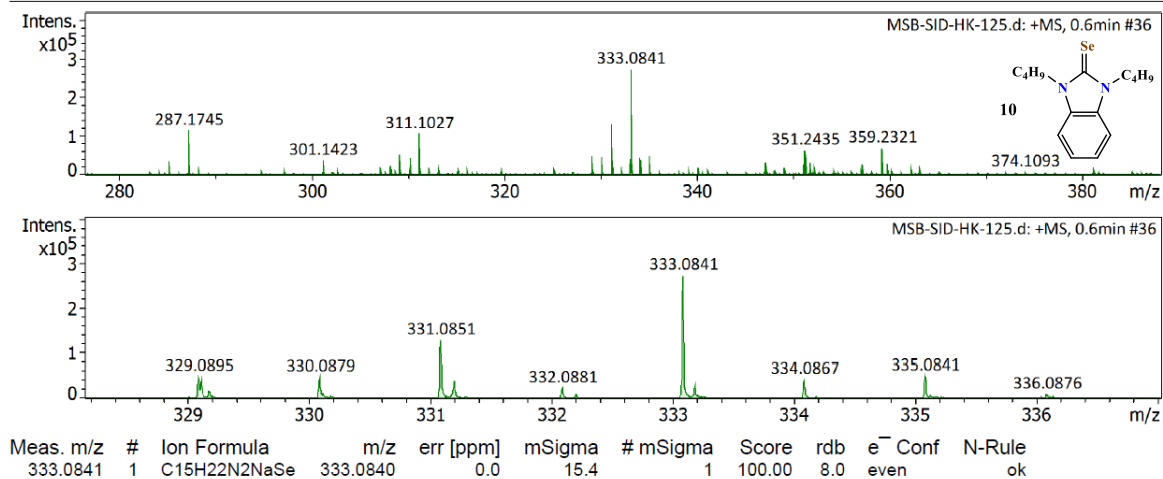


Fig. S30 HRMS spectrum of 10.

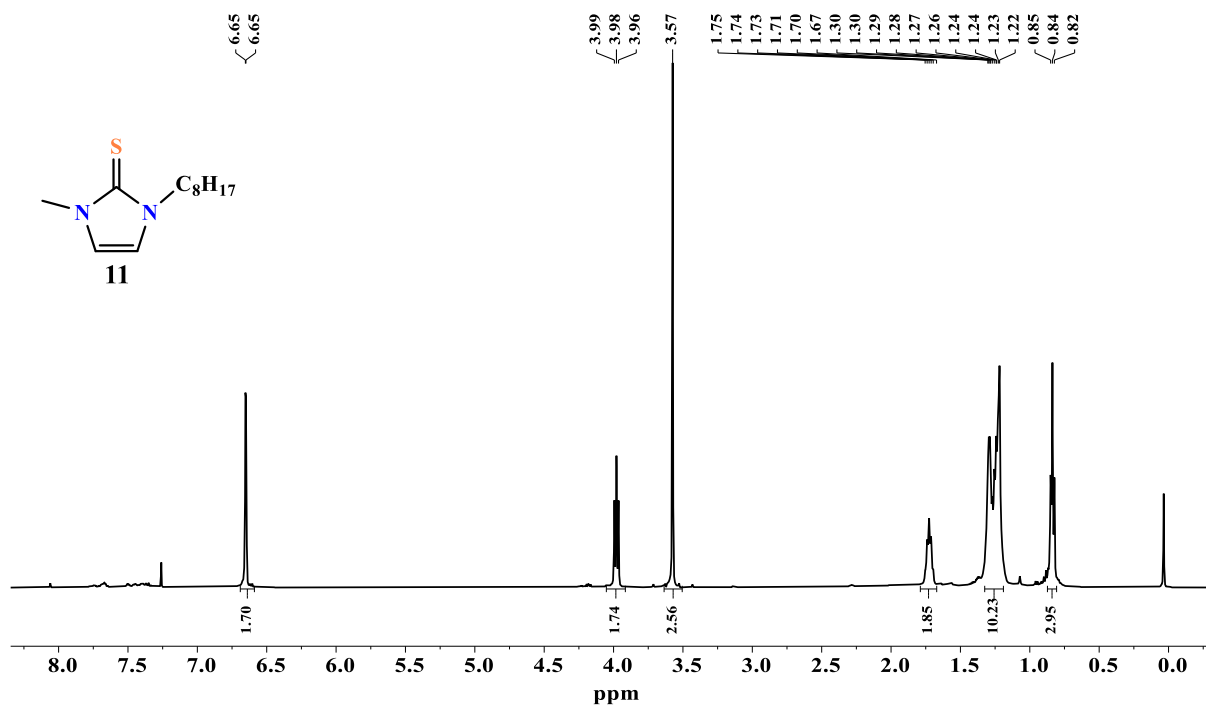


Fig. S31 ^1H NMR spectrum of **11**.

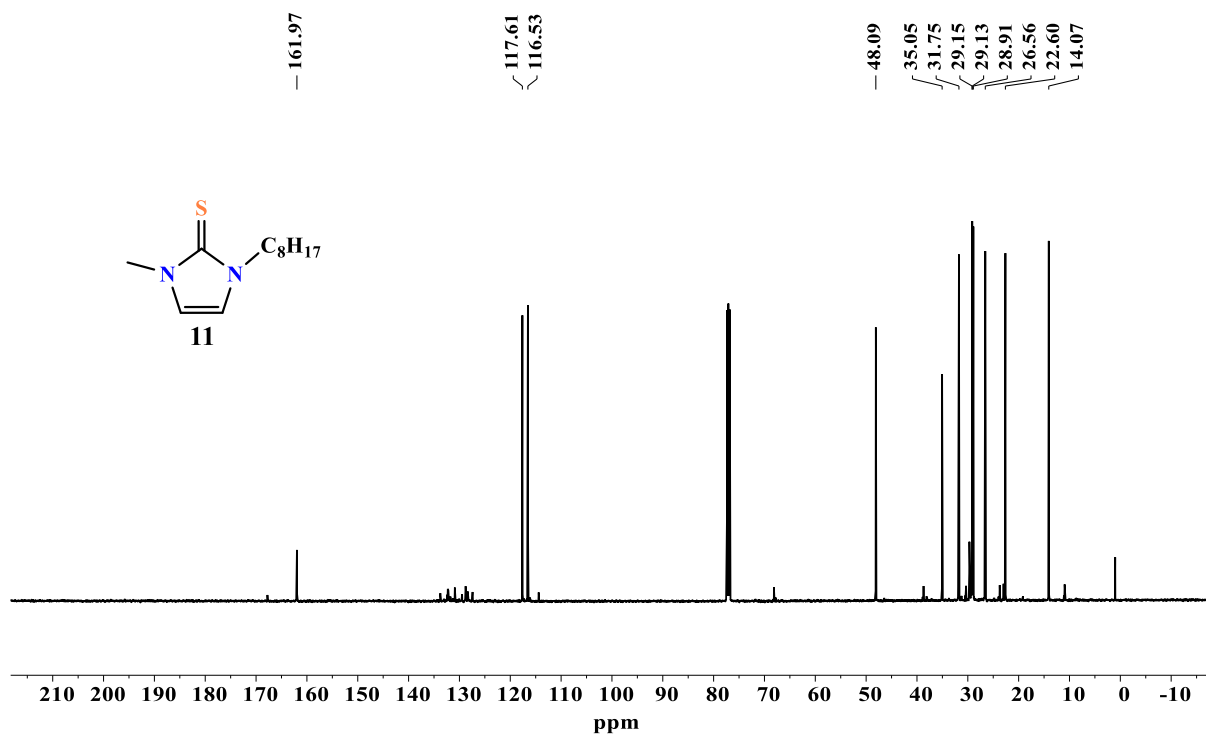


Fig. S32 ^{13}C NMR spectrum of **11**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

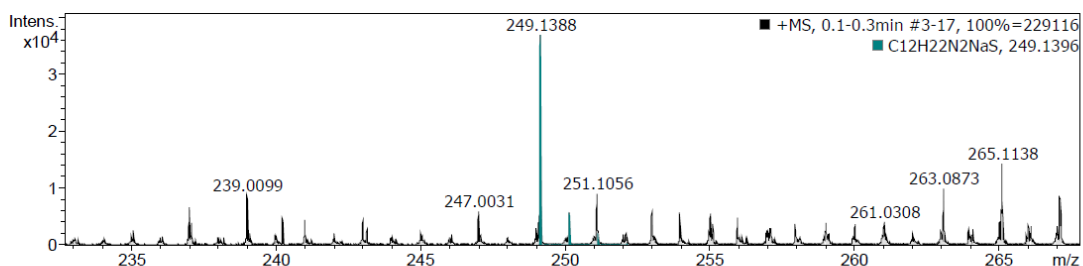
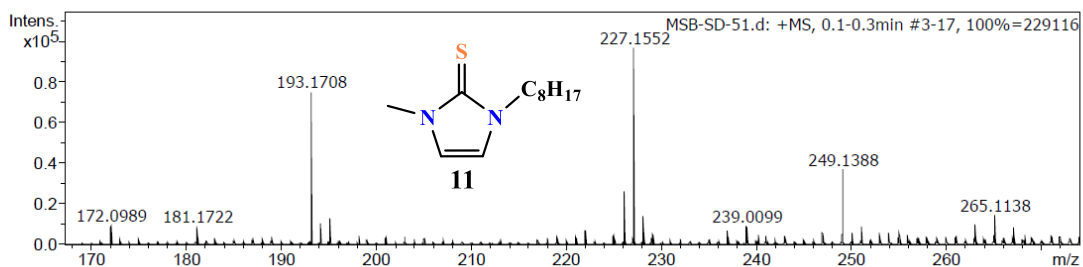
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Acquisition Date 12/10/2019 10:57:49 PM

Operator PG SRD OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

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| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 500 m/z | Set Collision Cell RF | 800.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
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Fig. S33 HRMS spectrum of 11.

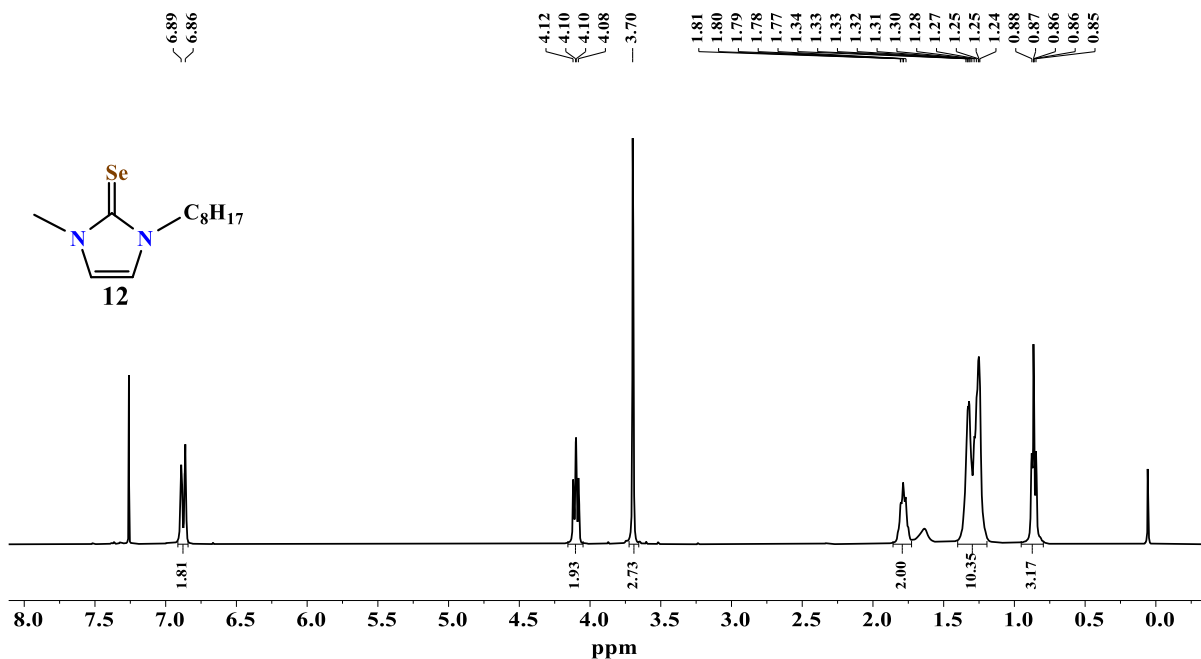


Fig. S34 ^1H NMR spectrum of **12**.

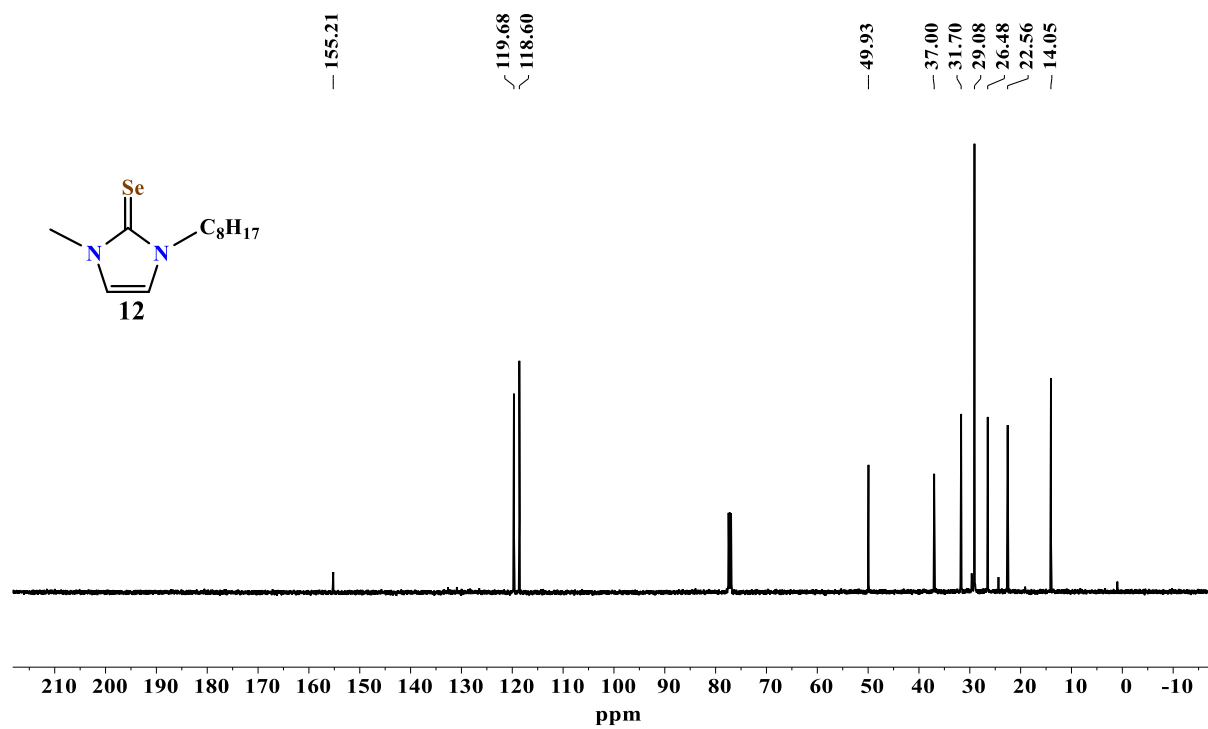


Fig. S35 ^{13}C NMR spectrum of **12**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

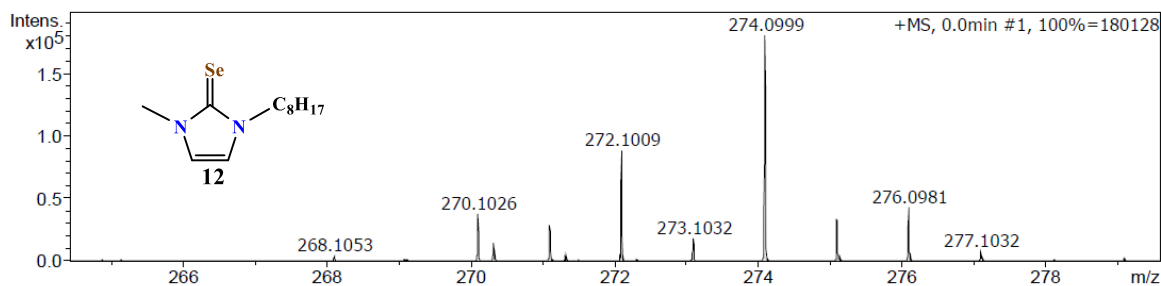
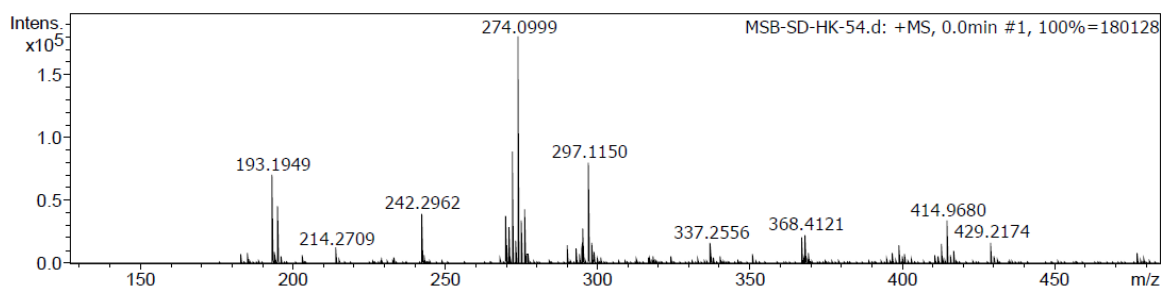
Analysis Name D:\Data\MAR-2020\MSB-SD-HK-54.d
 Method Tune_pos_NAICSI-1000.m
 Sample Name MSB-SD-HK-54
 Comment C12H22N2Se1

Acquisition Date 3/12/2020 11:48:10 AM

Operator SJG-out
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|----------|-----------------------|------------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.4 Bar |
| Focus | Active | Set Capillary | 4000 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1200.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 274.0999 | 1 | C12H22N2Se | 274.0943 | 20.4 | 28.2 | 1 | 100.00 | 3.0 | odd | ok |

Fig. S36 HRMS spectrum of **12**.

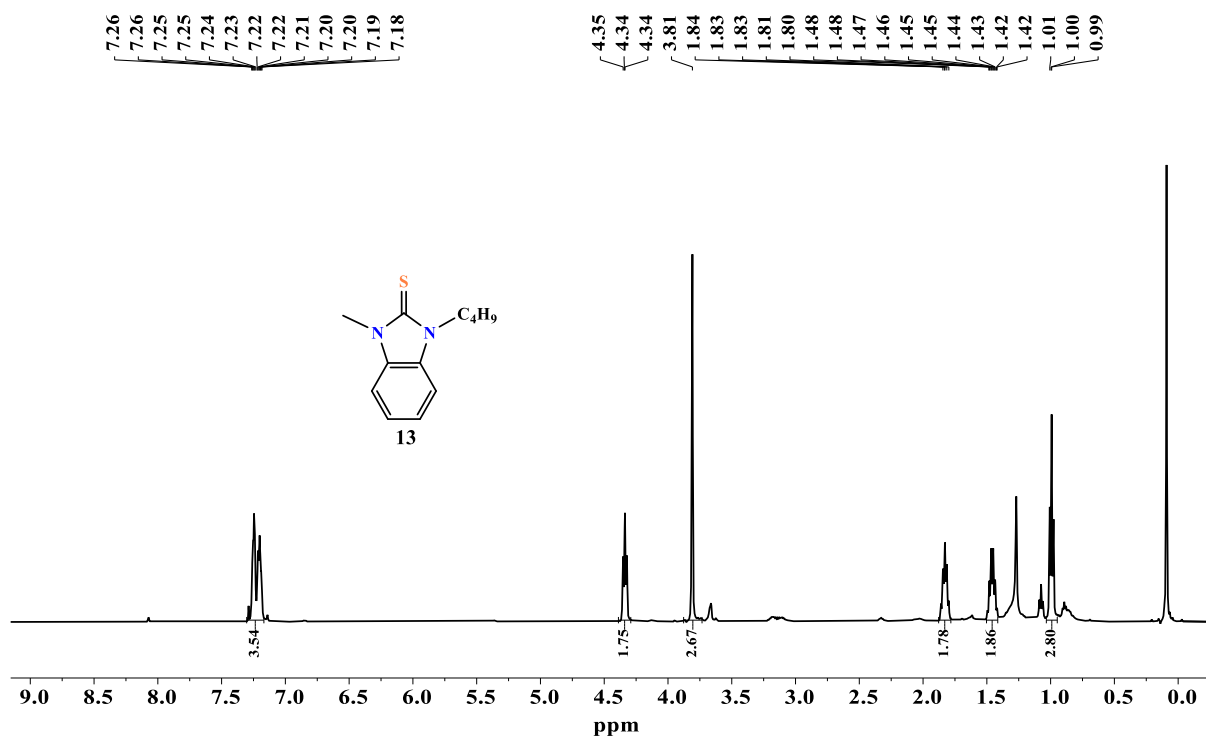


Fig. S37 ^1H NMR spectrum of **13**.

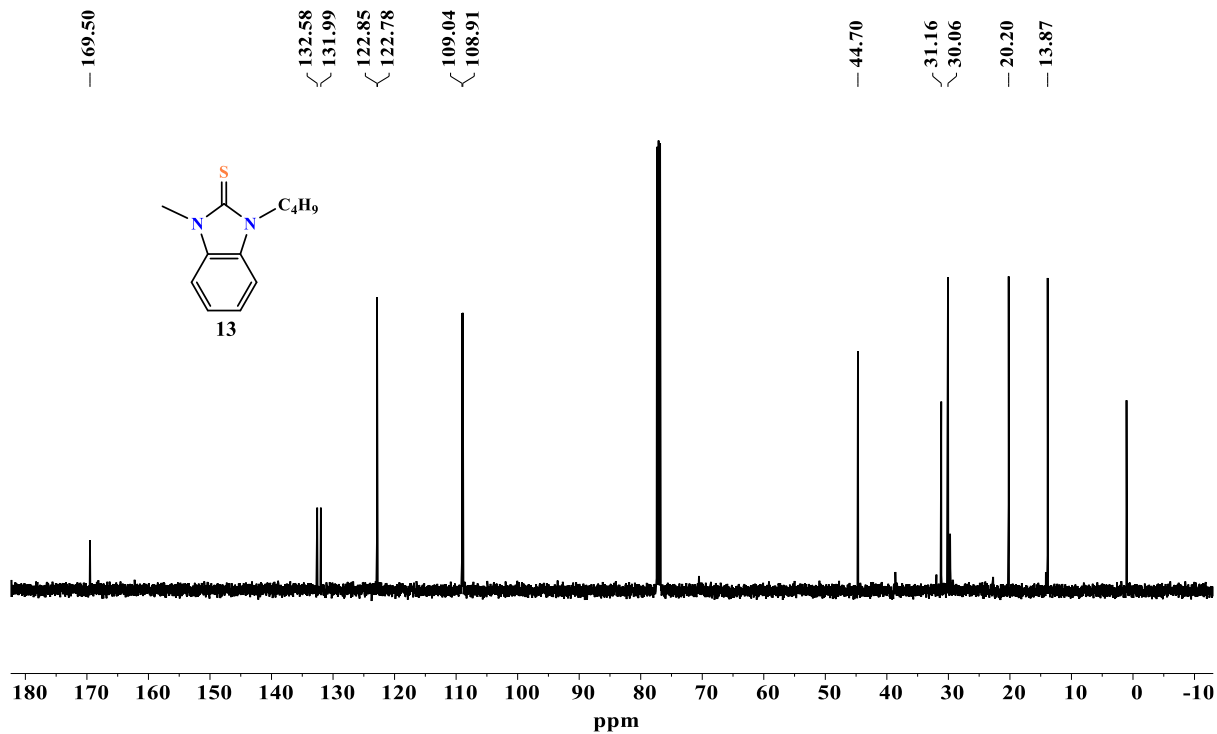


Fig. S38 ^{13}C NMR spectrum of **13**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name D:\Data\APR-2022\MSB-SID-HK-92.d
 Method Low_mass-NAF.m
 Sample Name MSB-SID-HK-92
 Comment C12H16N2S

Acquisition Date 4/5/2022 7:24:25 PM

Operator SJG OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 600 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |

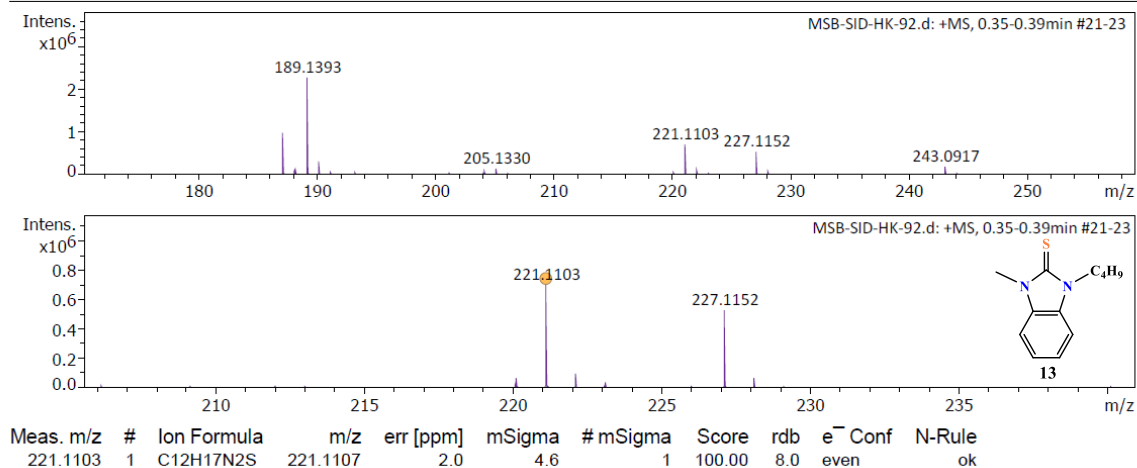


Fig. S39 HRMS spectrum of 13.

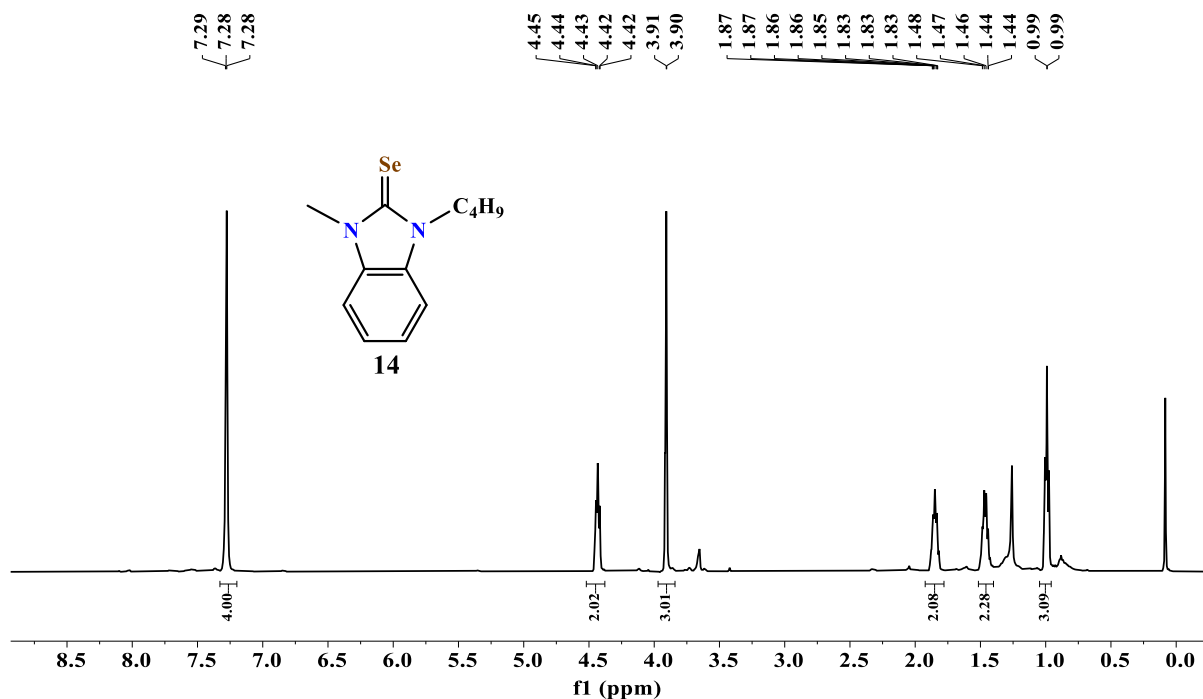


Fig. S40 ¹H NMR spectrum of 14.

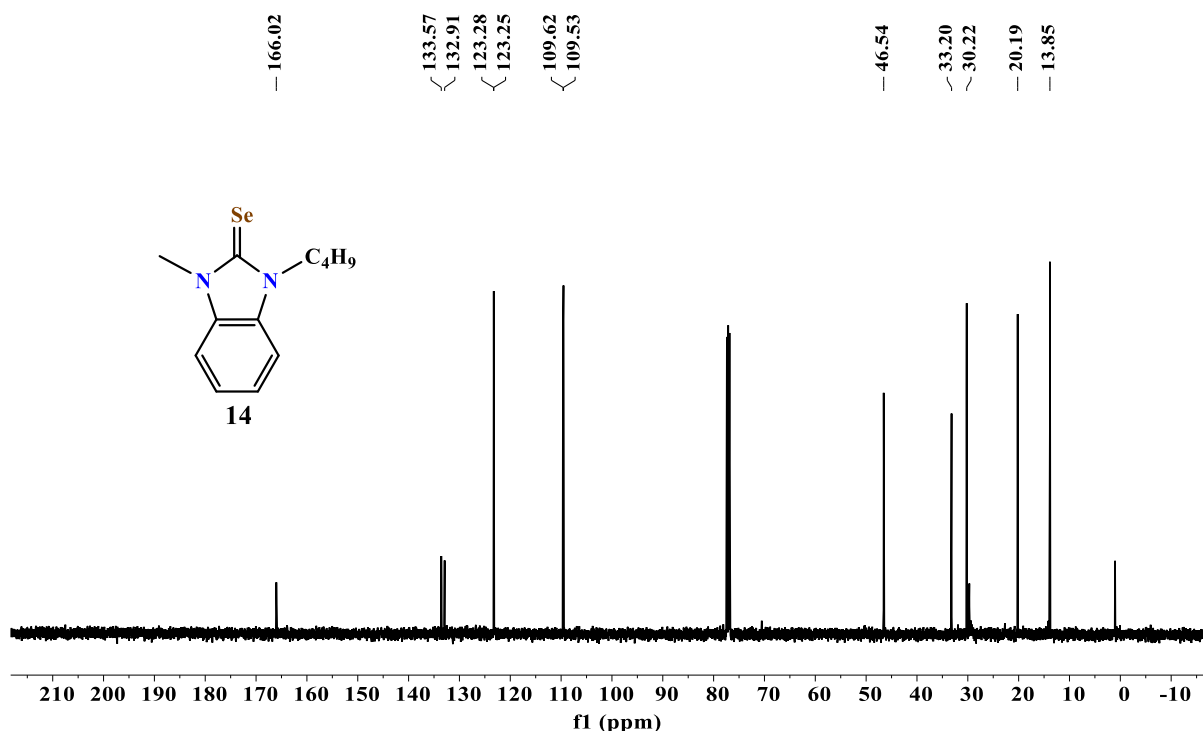


Fig. S41 ¹³C NMR spectrum of 14.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

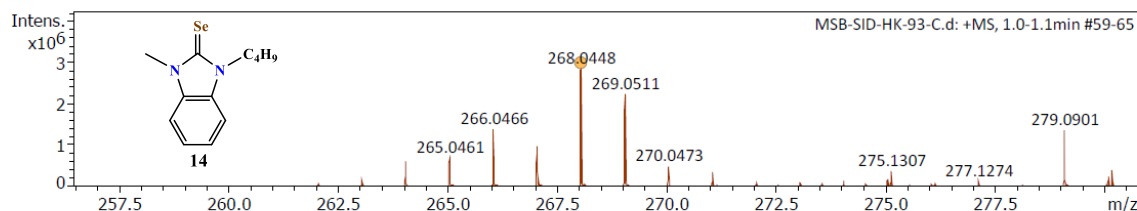
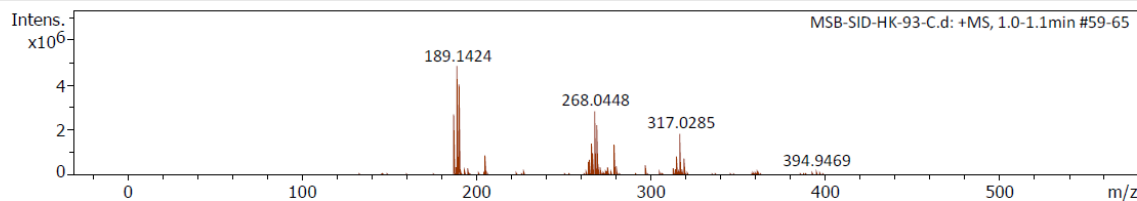
Analysis Name D:\Data\APR-2022\MSB-SID-HK-93-C.d
 Method Low_mass- NaICsI.m
 Sample Name MSB-SID-HK-93-C
 Comment C12H16N2Se1

Acquisition Date 4/5/2022 7:11:30 PM

Operator SJG OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 4000 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 650 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|----------|--------|-----|---------------------|--------|
| 268.0448 | 1 | C11H16N2OSe | 272.0422 | 0.5 | 515.5 | 1 | 100.00 | 7.5 | odd | ok |

Fig. S42 HRMS spectrum of 14.

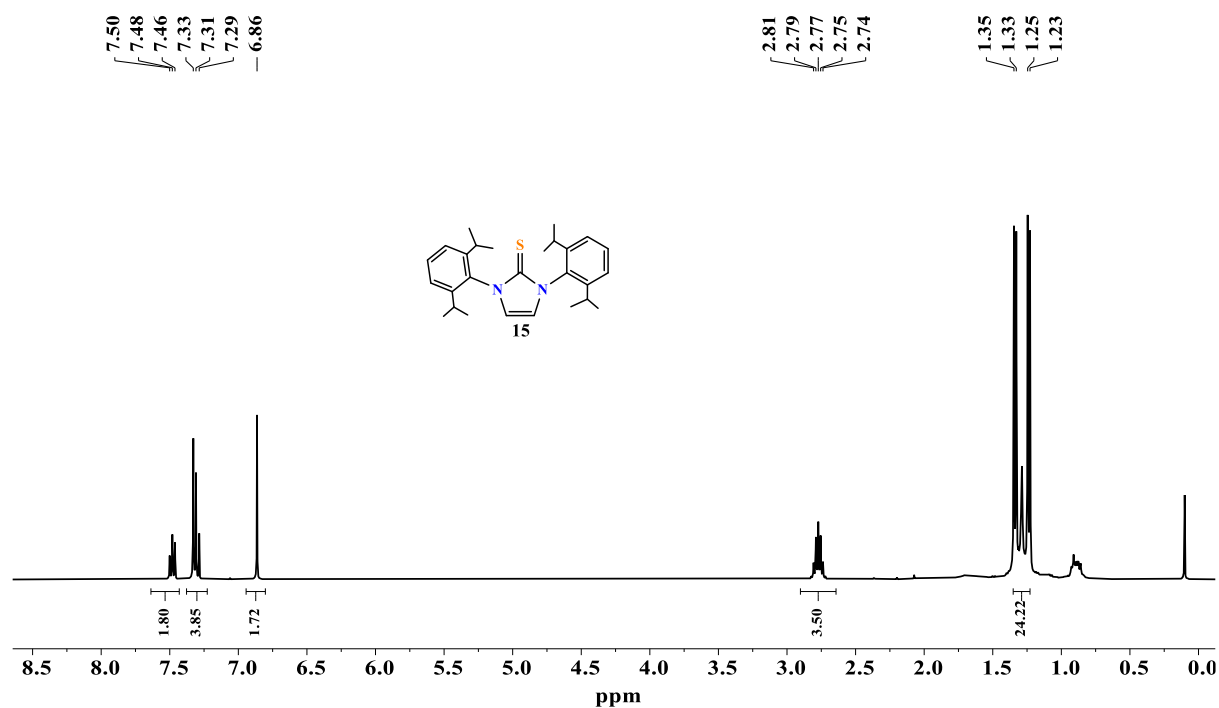


Fig. S43 ¹H NMR spectrum of 15.

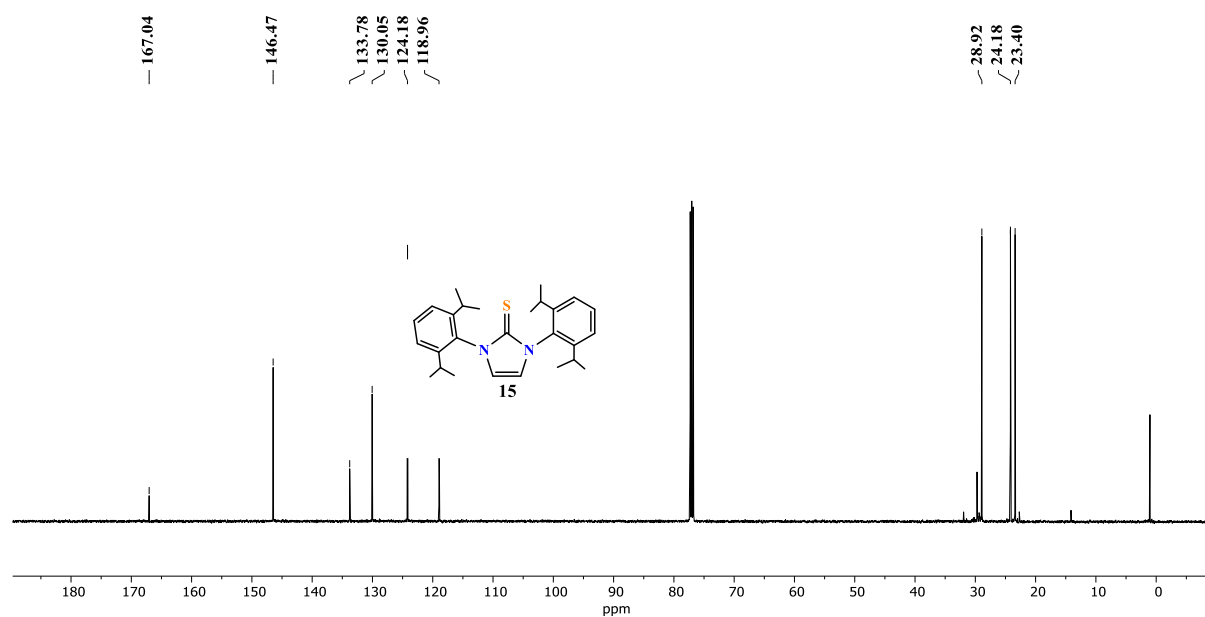


Fig. S44 ¹³C NMR spectrum of 15.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

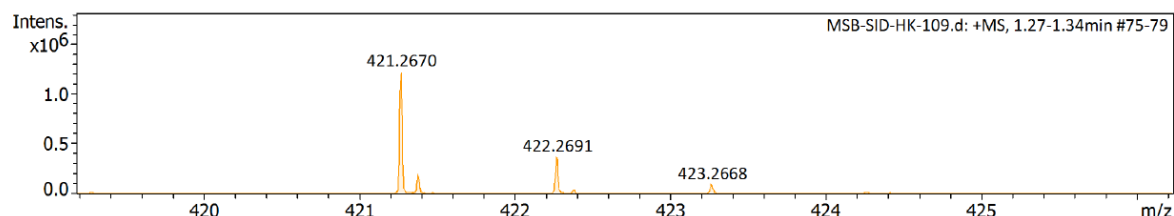
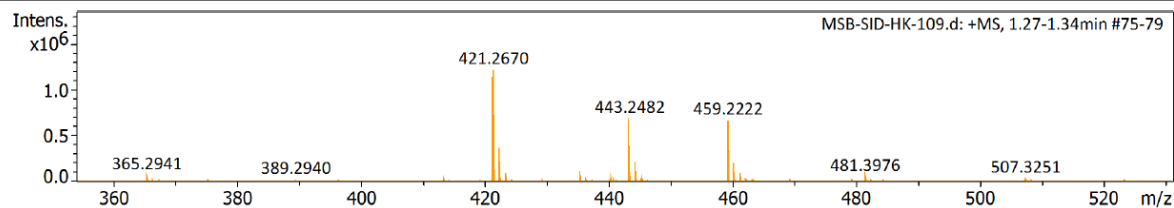
Analysis Name D:\Data\AUG 2021\MSB-SID-HK-109.d
 Method Low_mass- NAF.m
 Sample Name MSB-SID-HK-109
 Comment C27H36N2S

Acquisition Date 8/12/2021 1:20:05 AM

Operator MSB-OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|---------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 3200 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 600 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|----------|--------|------|---------------------|--------|
| 421.2670 | 1 | C27H37N2S | 421.2672 | 0.5 | 11.4 | 1 | 100.00 | 13.0 | even | ok |

Fig. S45 HRMS spectrum of 15.

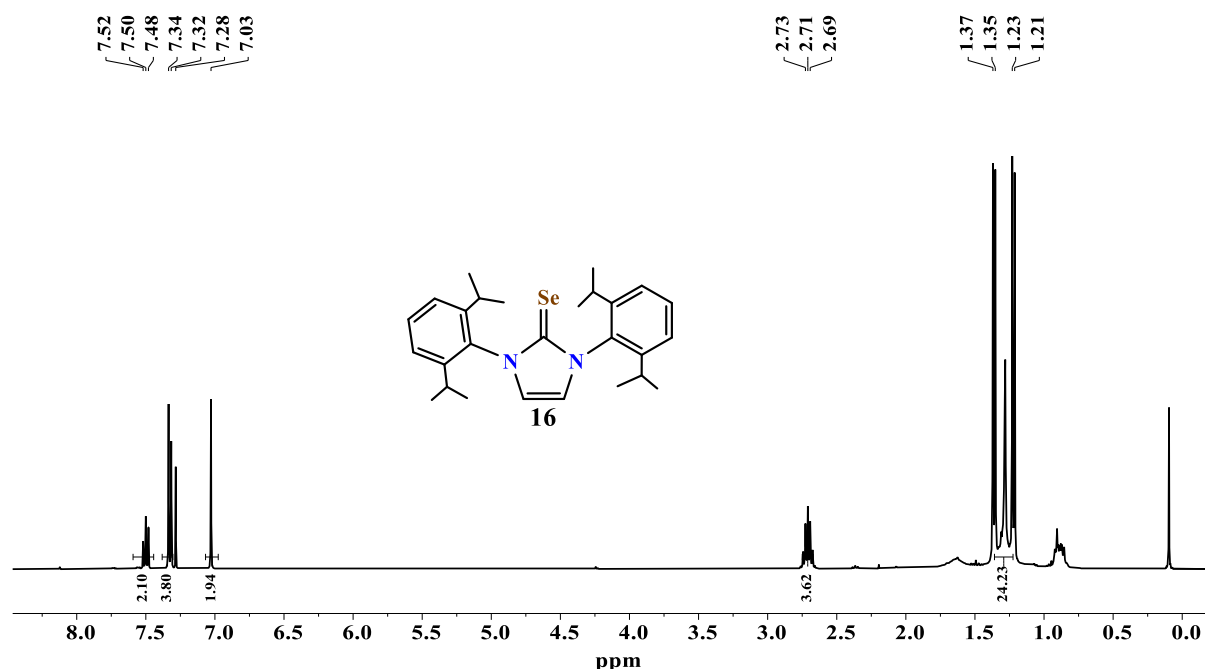


Fig. S46 ¹H NMR spectrum of 16.

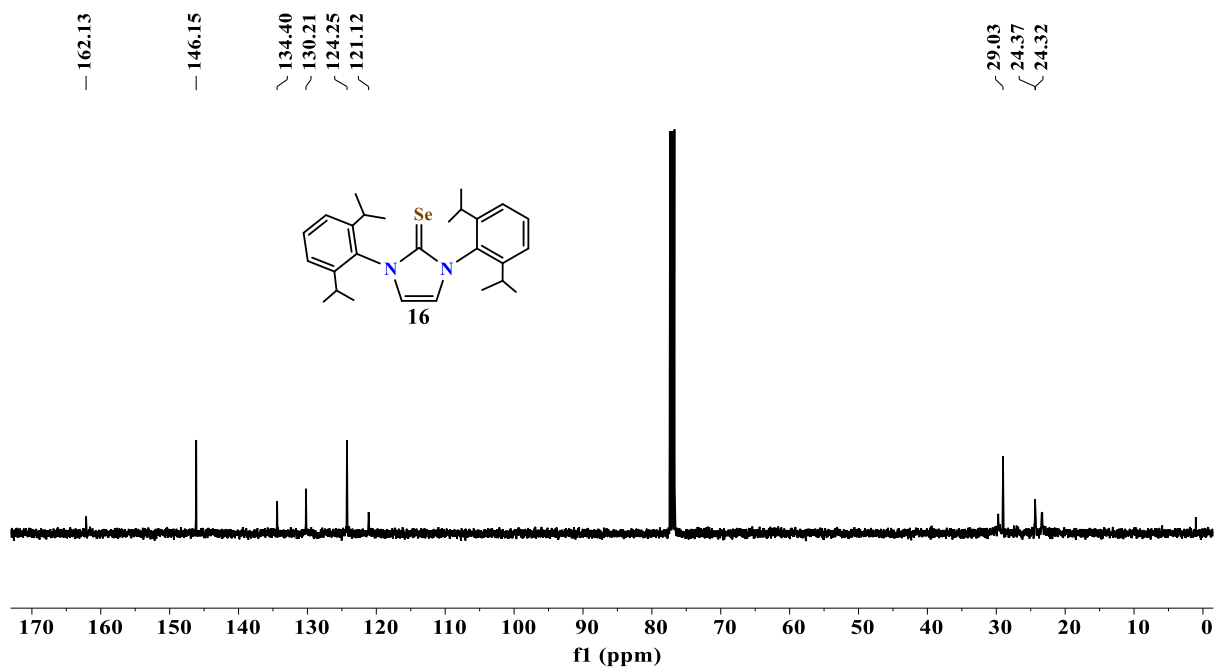


Fig. S47 ^{13}C NMR spectrum of 16.

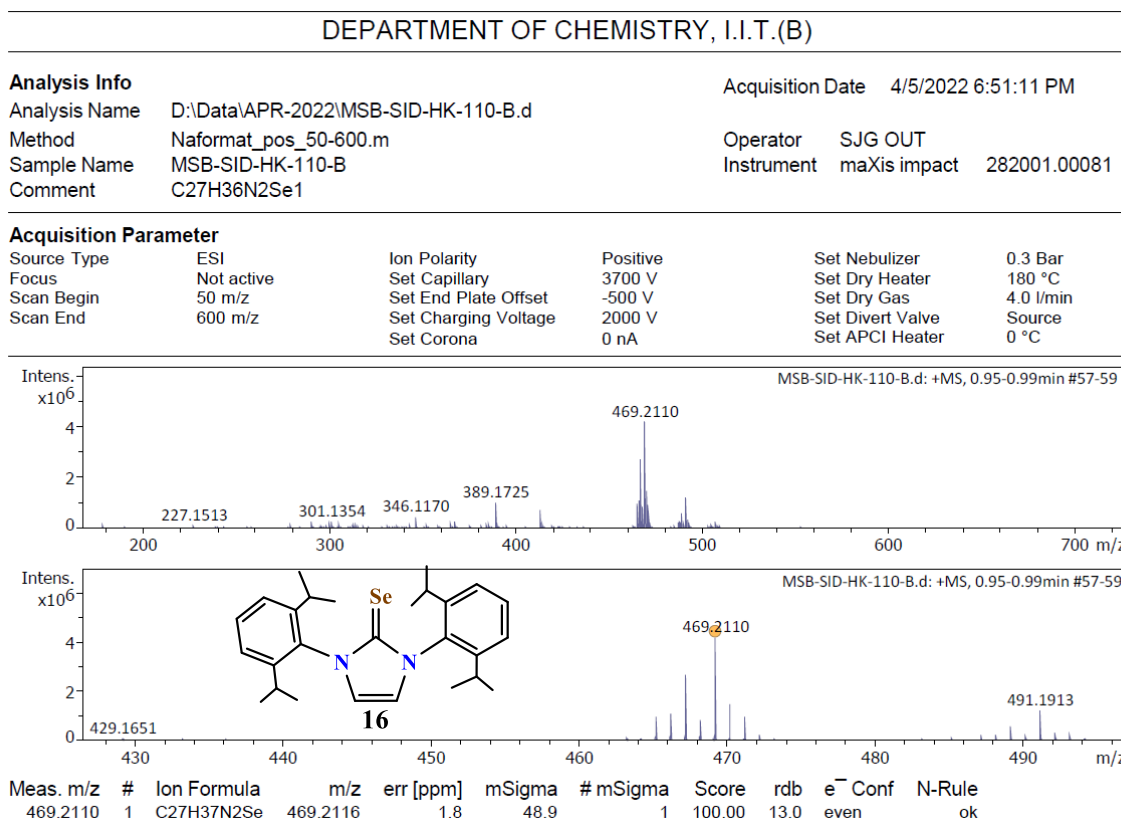


Fig. S48 HRMS spectrum of 16.

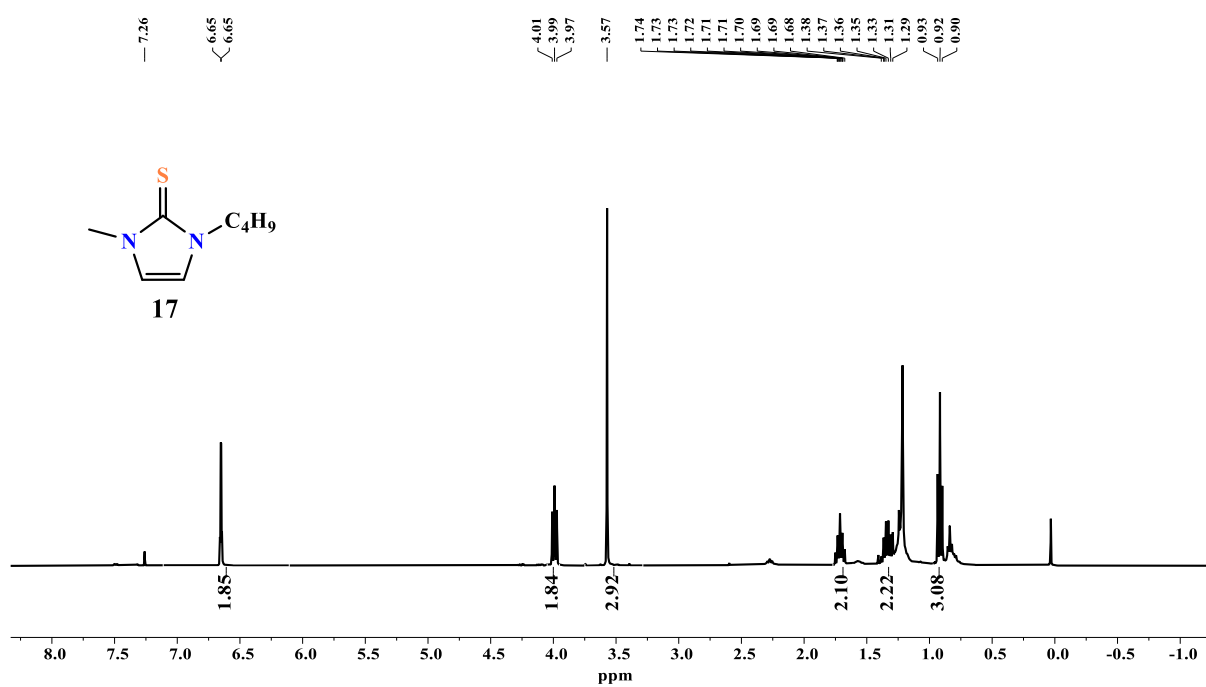


Fig. S49 ¹H NMR spectrum of 17.

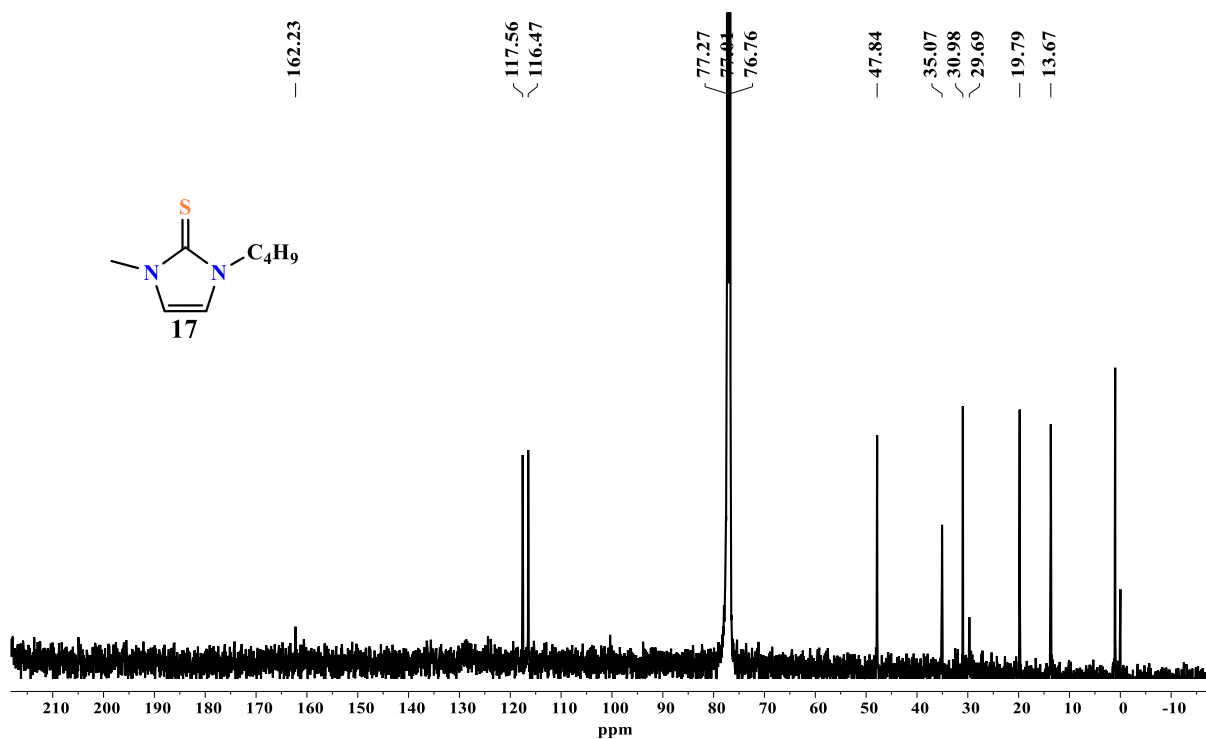


Fig. S50 ¹³C NMR spectrum of 17.

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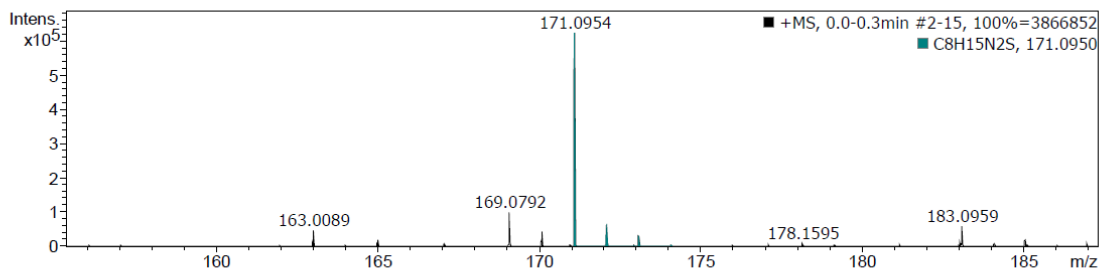
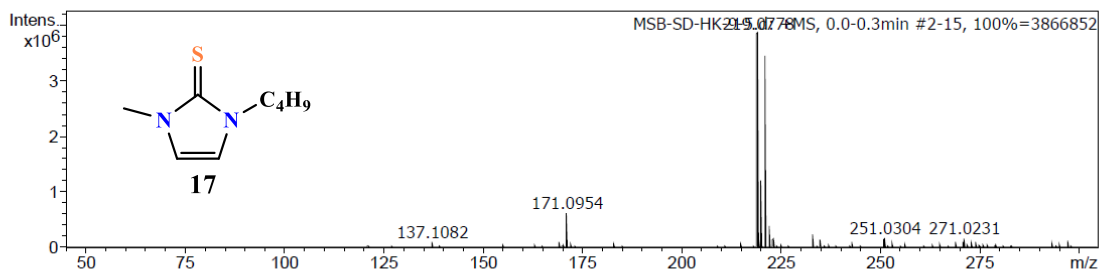
Analysis Info

Analysis Name D:\Data\DEC-2019\MSB-SD-HK-9-5.d
 Method Tune_pos_500_NAF.m
 Sample Name MSB-SD-HK-9-5
 Comment C8H14N2S1

Acquisition Date 12/10/2019 11:24:24 PM
 Operator PG SRD OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|---------|-----------------------|-----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 500 m/z | Set Collision Cell RF | 800.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 171.0954 | 1 | C8H15N2S | 171.0950 | -2.3 | 2.4 | 1 | 100.00 | 2.5 | even | ok |

Fig. S51 HRMS spectrum of **17**.

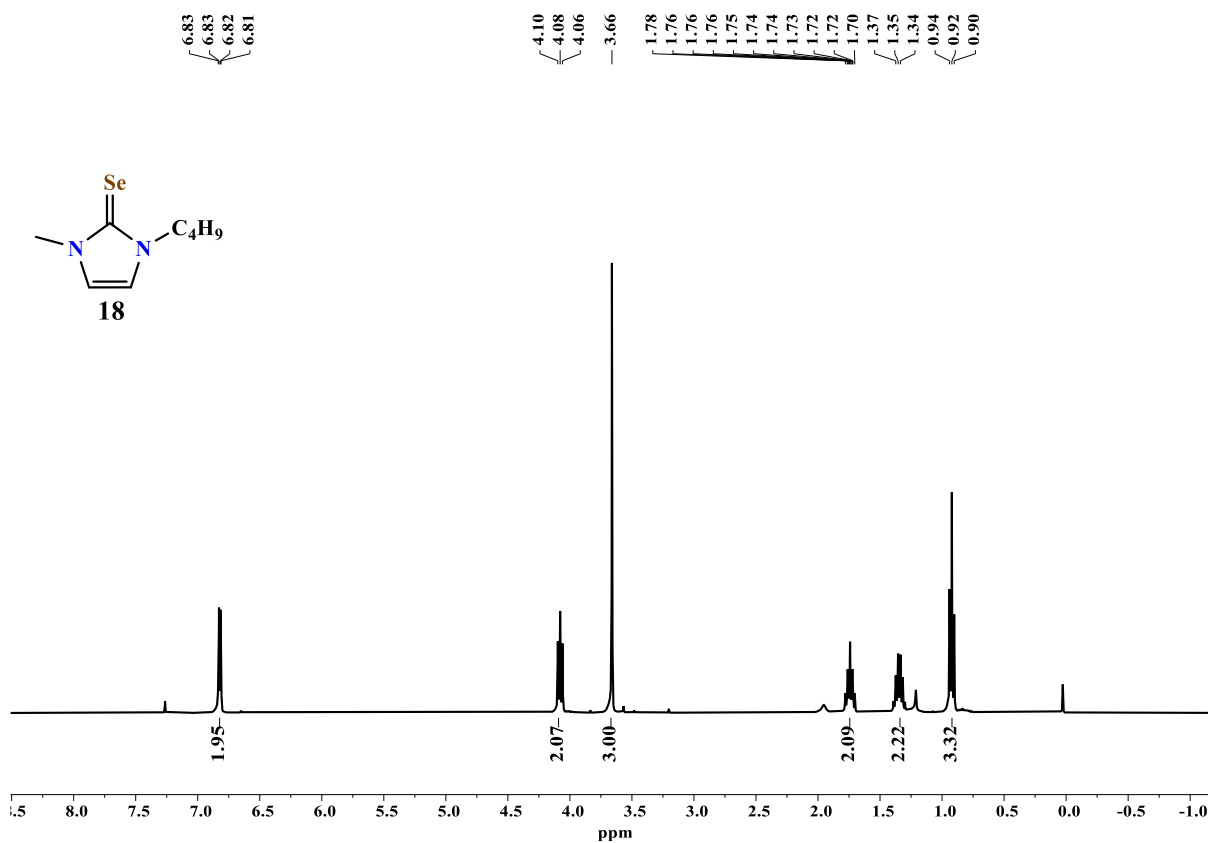


Fig. S52 ¹H NMR spectrum of **18**.

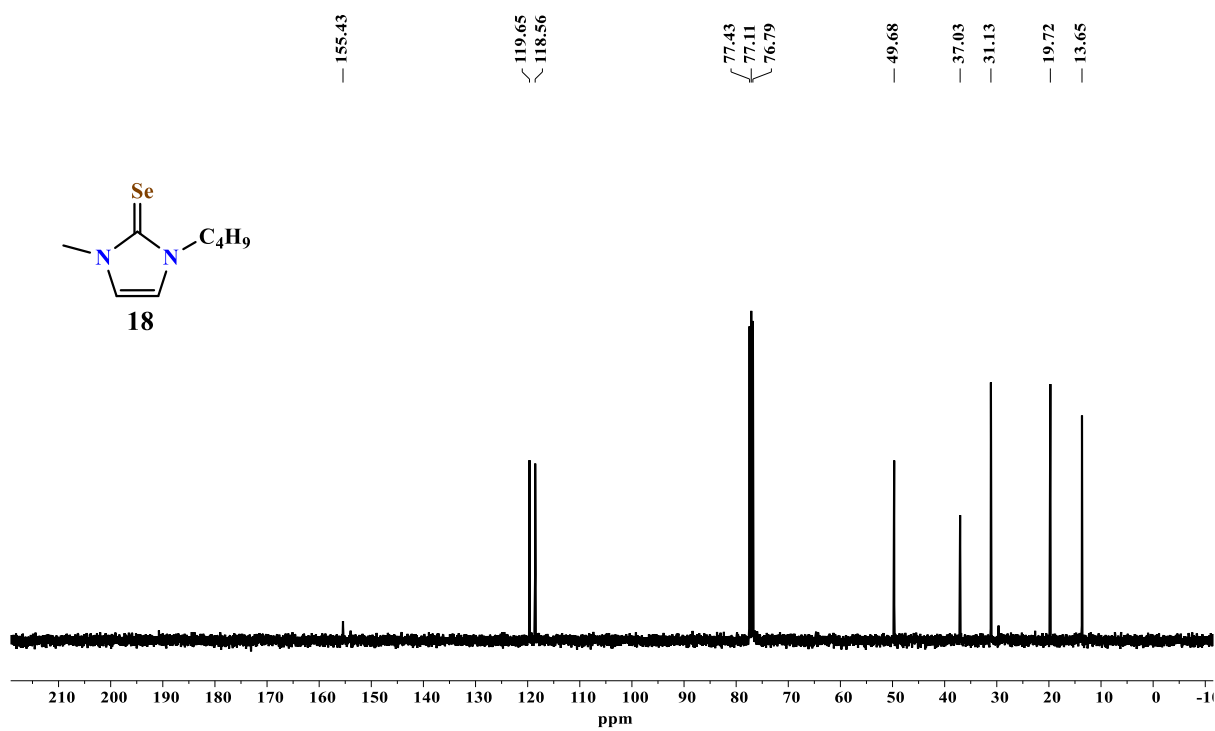


Fig. S53 ¹³C NMR spectrum of **18**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

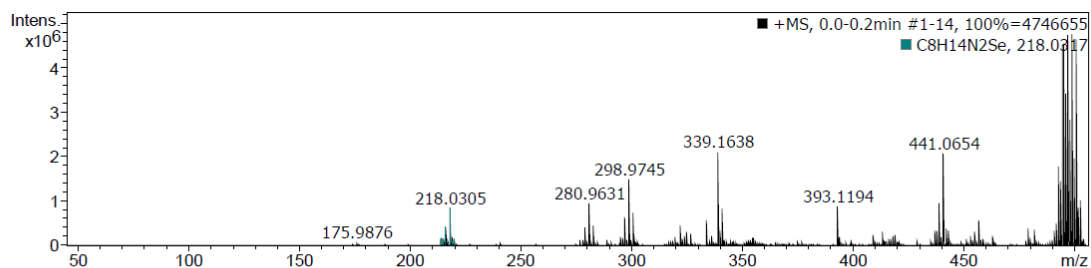
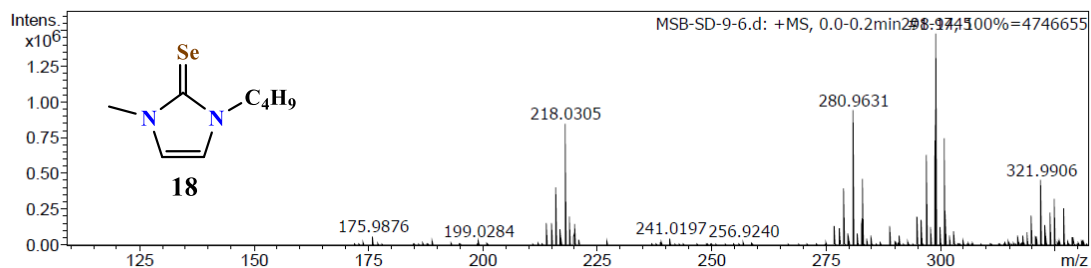
Analysis Name D:\Data\DEC-2019\MSB-SD-9-6.d
 Method Tune_pos_500_NAF.m
 Sample Name MSB-SD-9-6
 Comment C8H12N2Se

Acquisition Date 12/10/2019 11:06:22 PM

Operator PG SRD OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|---------|-----------------------|-----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 500 m/z | Set Collision Cell RF | 800.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 218.0305 | 1 | C8H14N2Se | 218.0317 | 5.4 | 56.0 | 1 | 100.00 | 3.0 | odd | ok |

Fig. S54 HRMS spectrum of **18**.

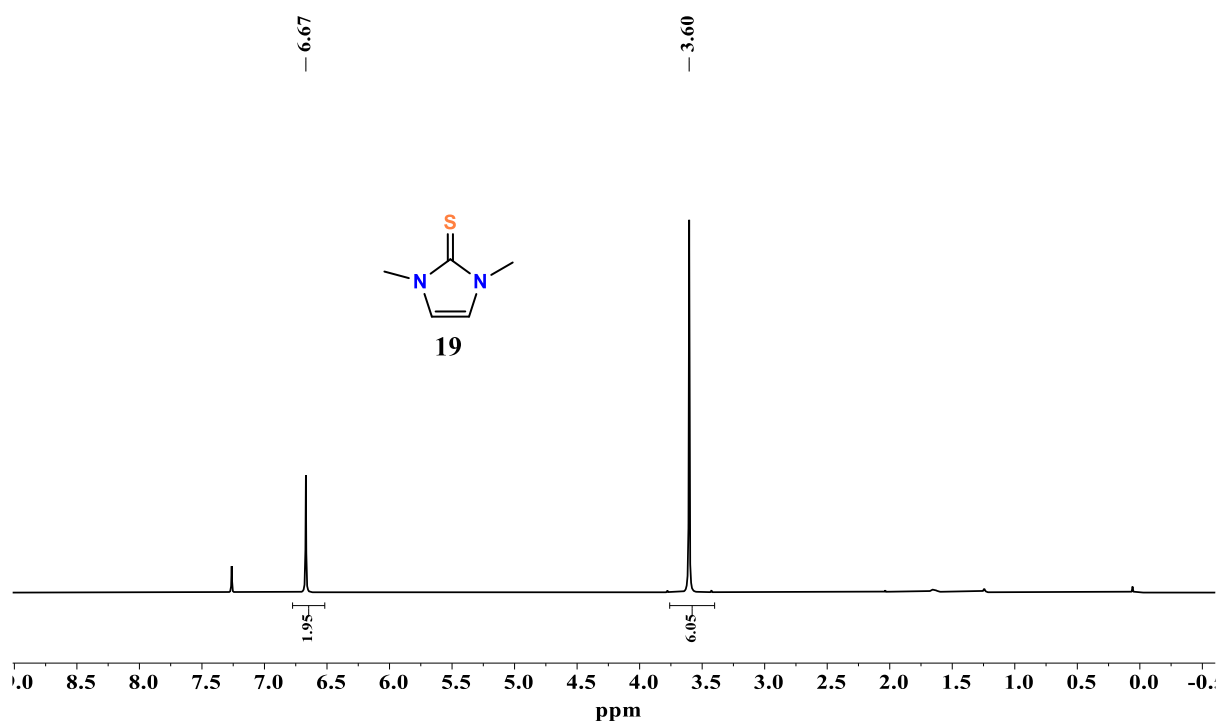


Fig. S55 ^1H NMR spectrum of 19.

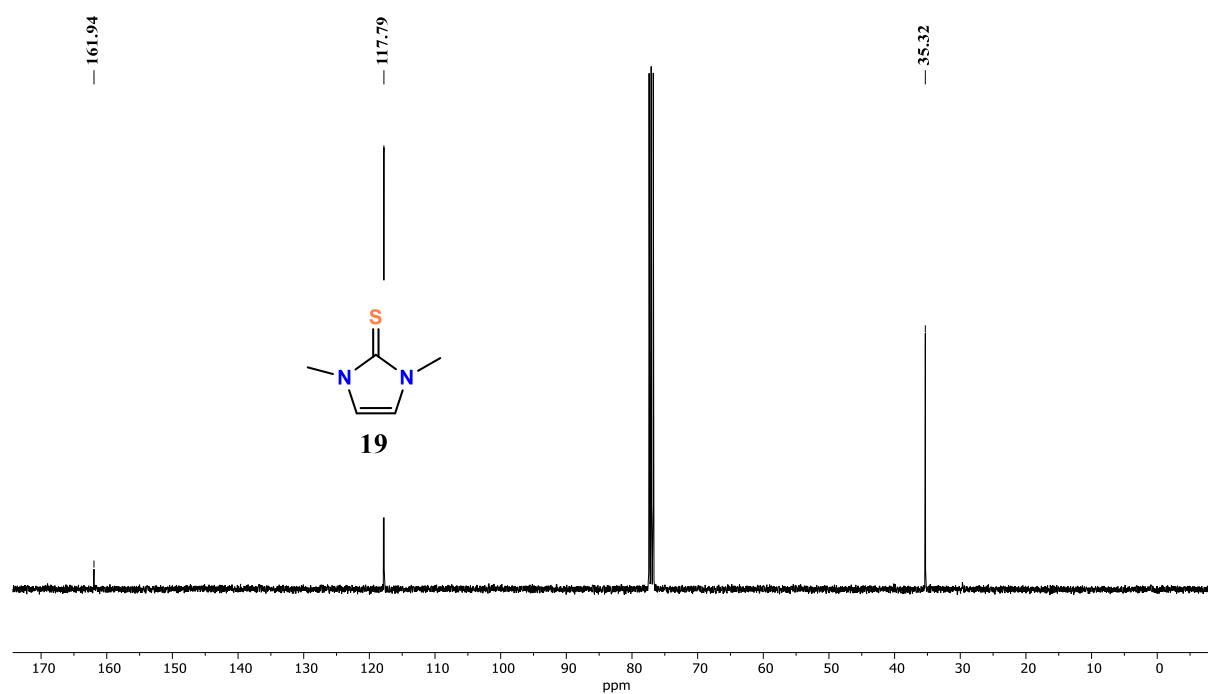


Fig. S56 ^{13}C NMR spectrum of 19.

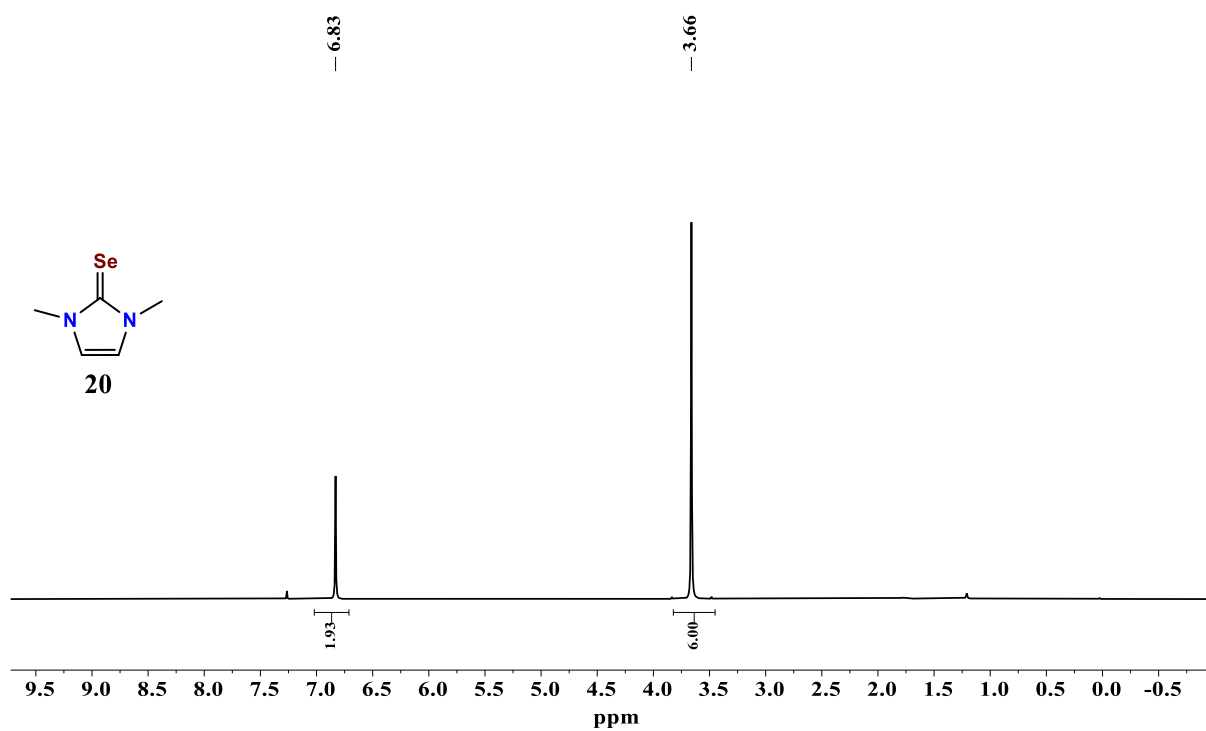


Fig. S57 ¹H NMR spectrum of **20**.

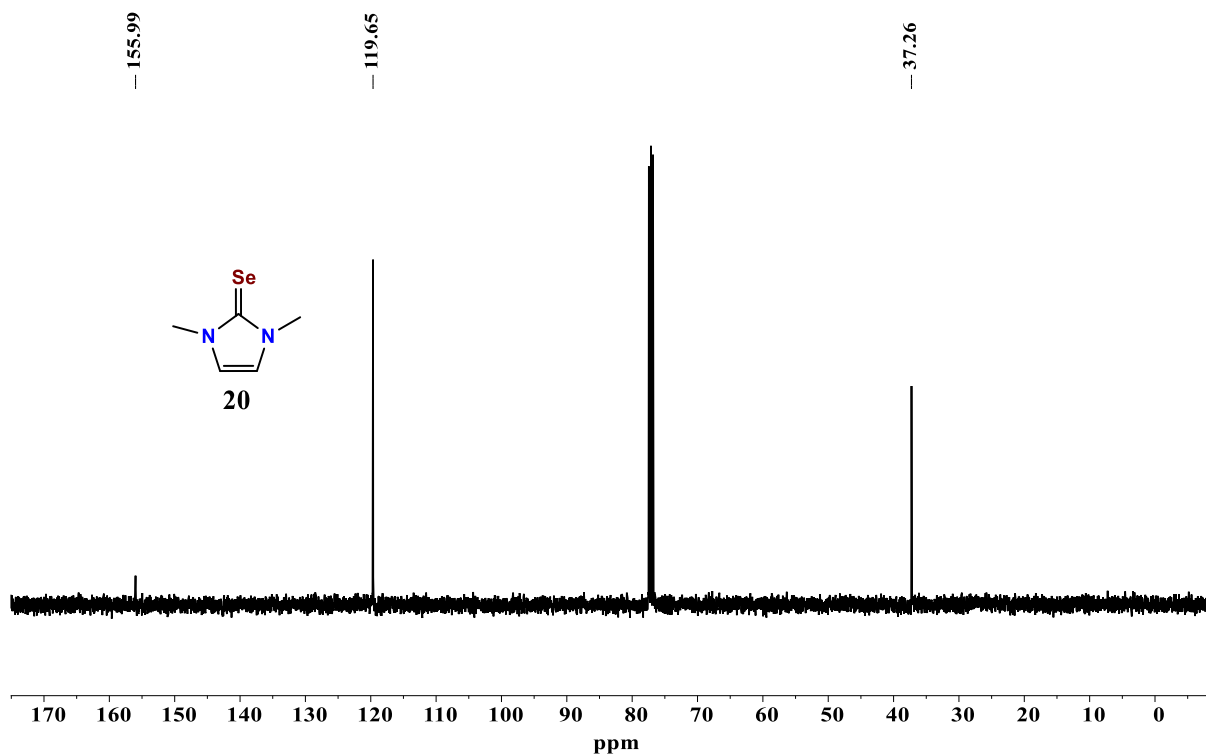


Fig. S58: ¹³C NMR spectrum of **20**.

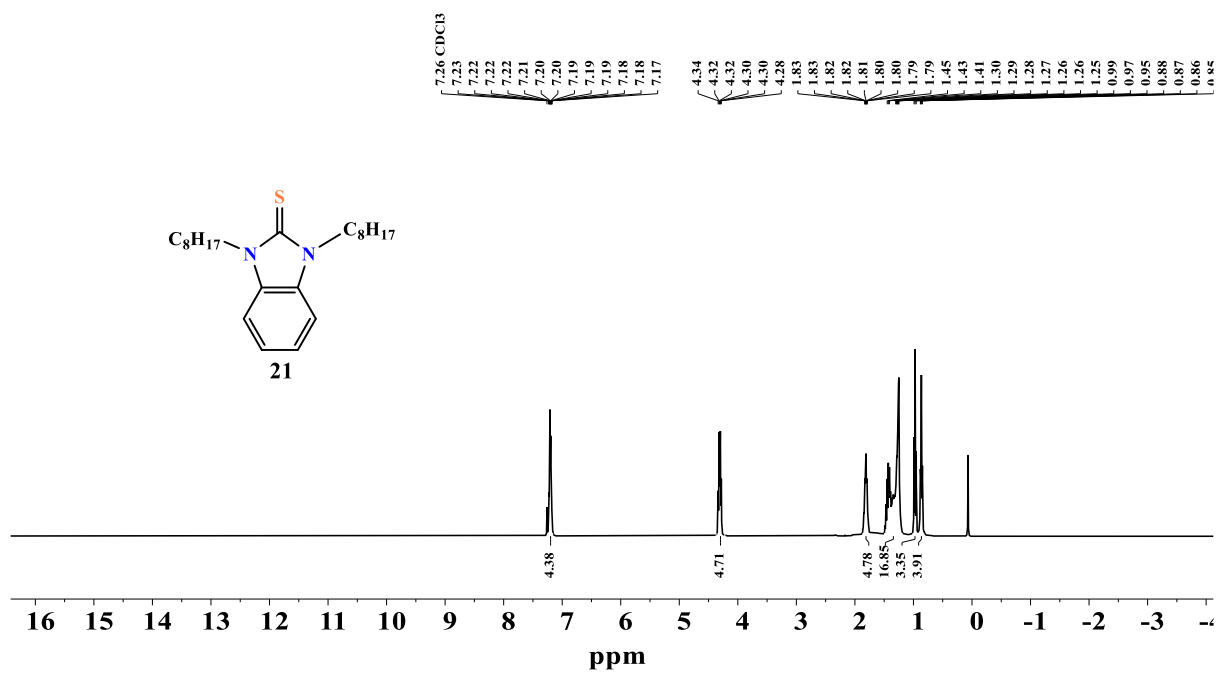


Fig. S59 ^1H NMR spectrum of **21**.

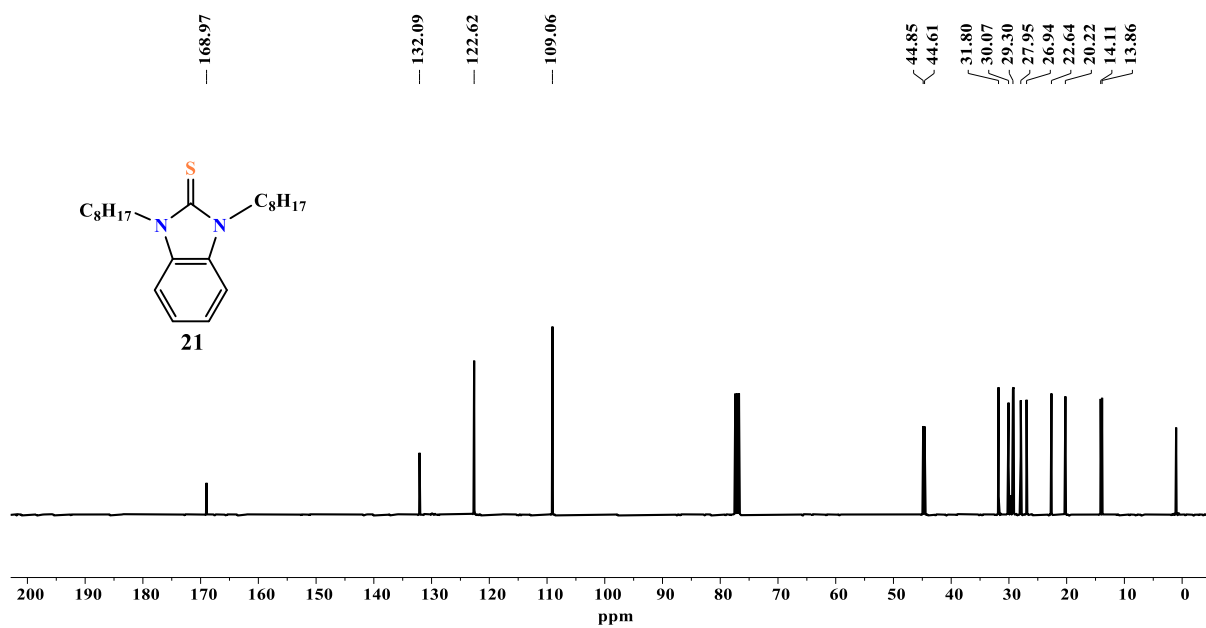


Fig. S60 ^{13}C NMR spectrum of **21**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

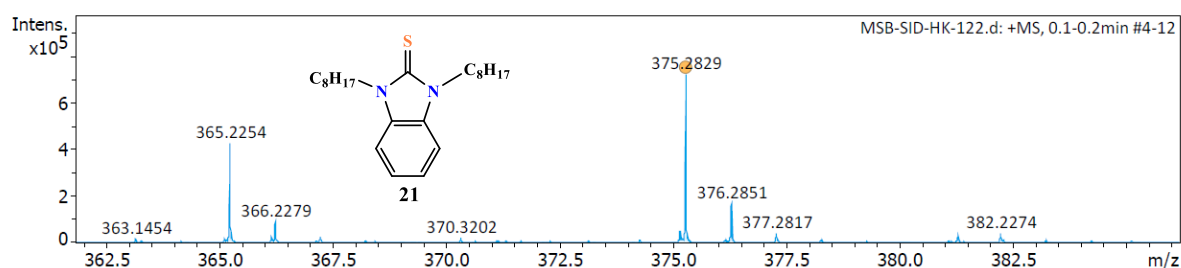
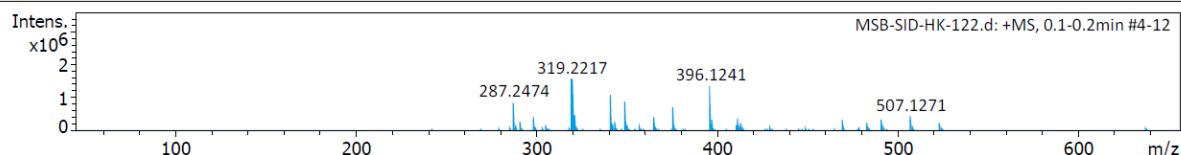
Analysis Name D:\Data\FEB-2022\MSB-SID-HK-122.d
 Method Low_mass- NaICsI.m
 Sample Name MSB-SID-HK-122
 Comment C23H38N2S

Acquisition Date 2/4/2022 7:48:26 PM

Operator SJGOUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 4000 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 650 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|----------|--------|-----|---------------------|--------|
| 375.2829 | 1 | C23H39N2S | 375.2828 | -0.0 | 21.7 | 1 | 100.00 | 8.0 | even | ok |

Fig. S61 HRMS Spectrum of 21.

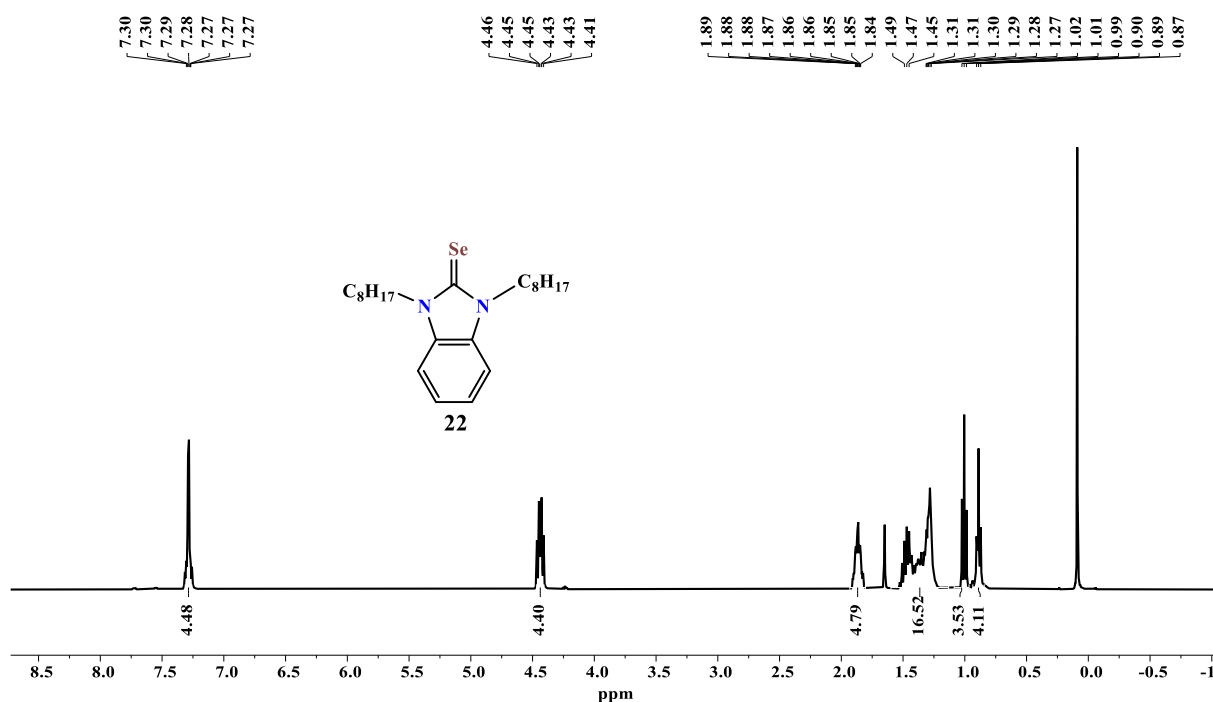


Fig. S62 ¹H NMR spectrum of 22.

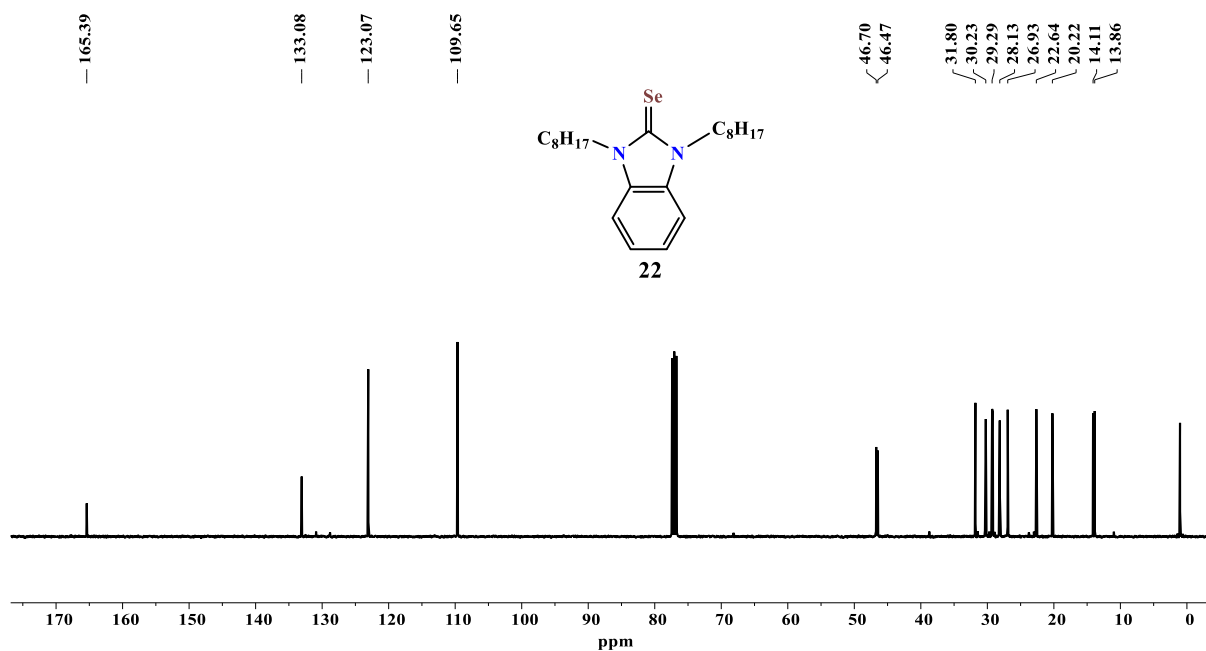


Fig. S63 ¹³C NMR spectrum of **22**.

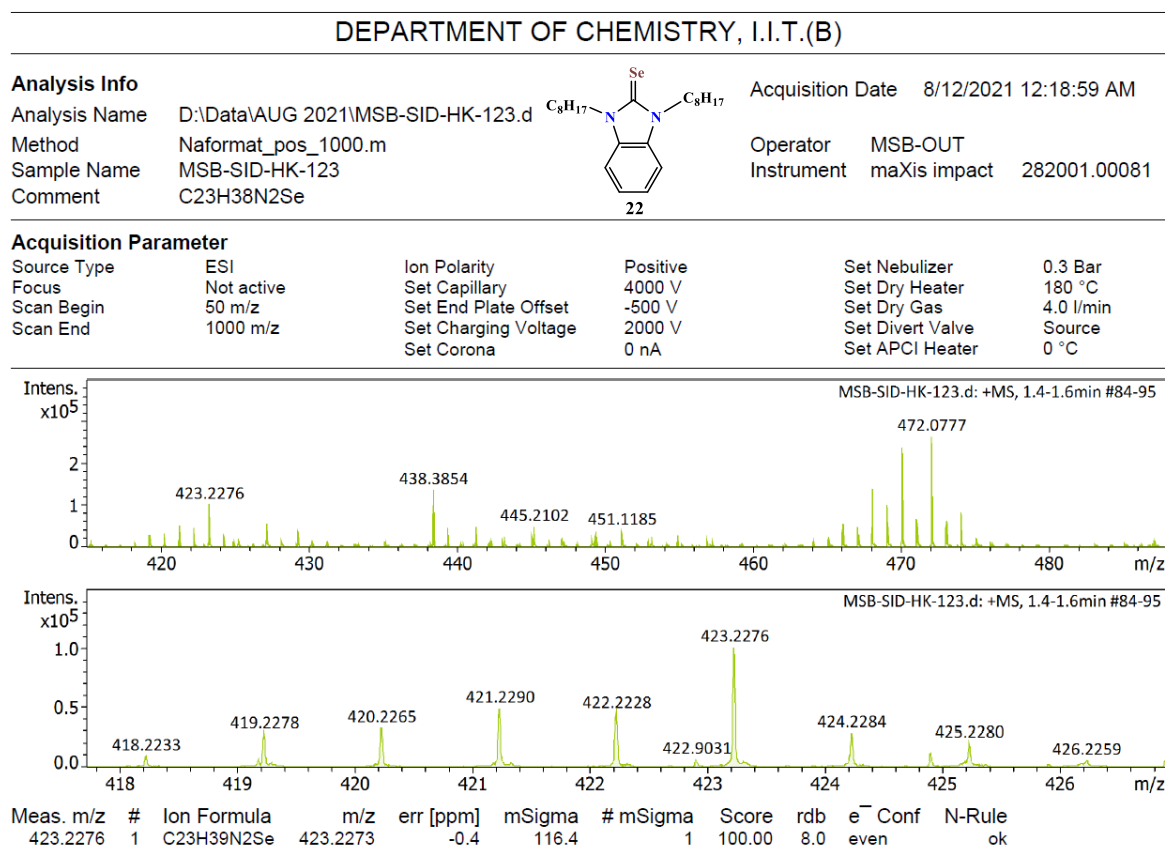


Fig. S64 HRMS spectrum of **22**.

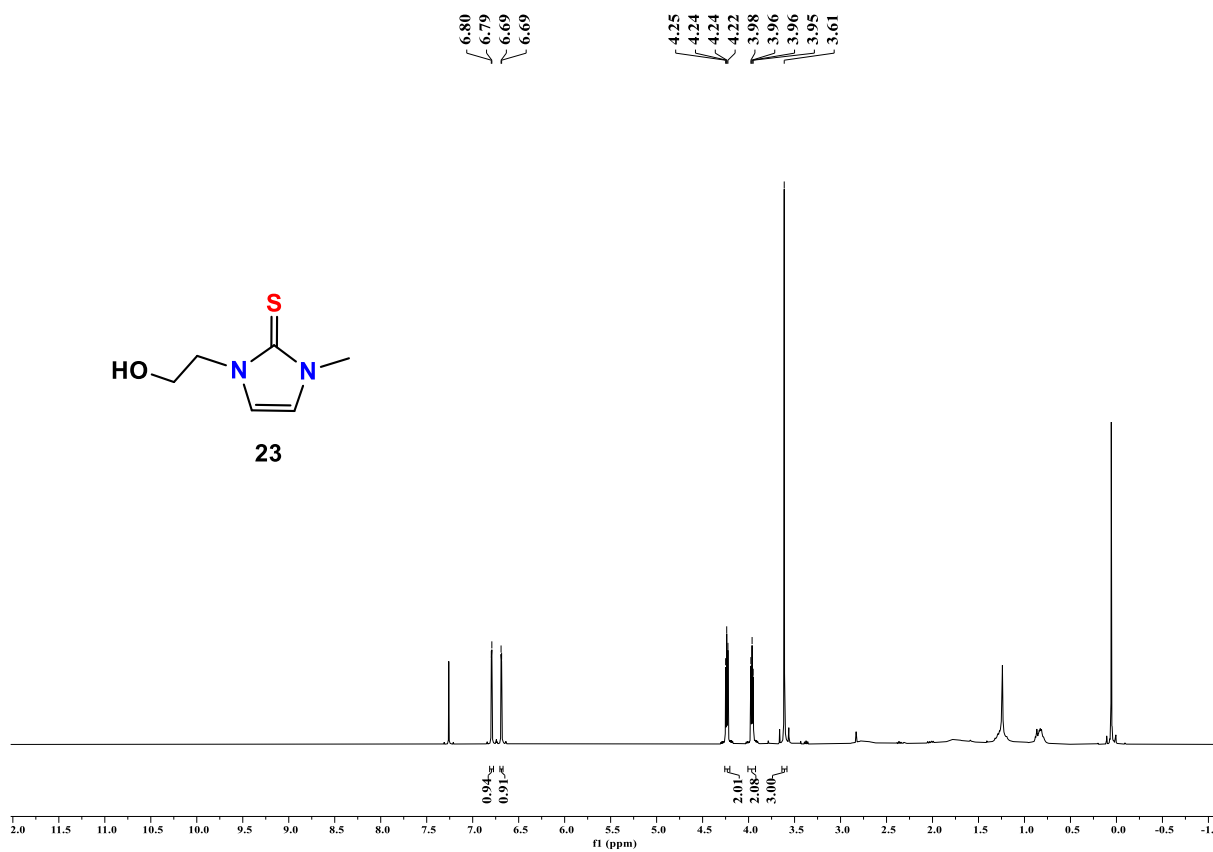


Fig. S65 ¹H NMR spectrum of **23**.

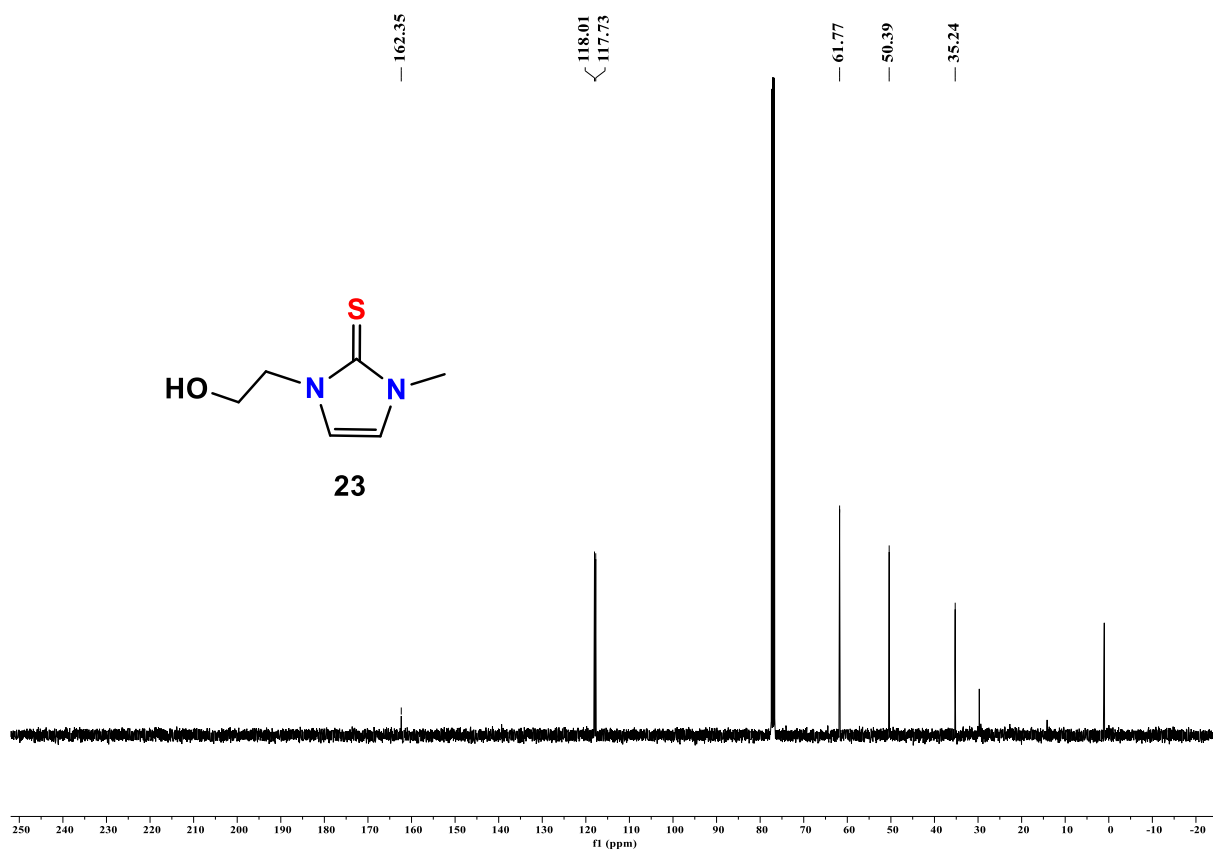


Fig. S66 ^{13}C NMR spectrum of **23**.

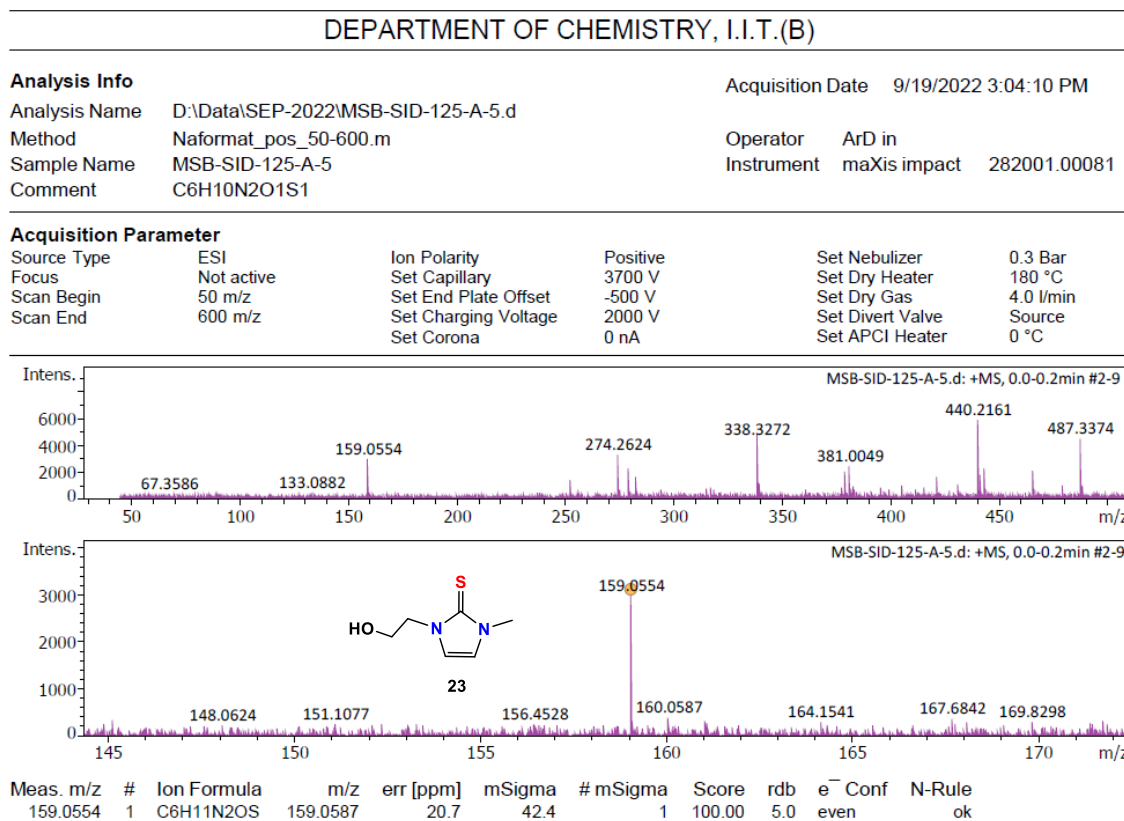


Fig. S67 HRMS spectrum of **23**.

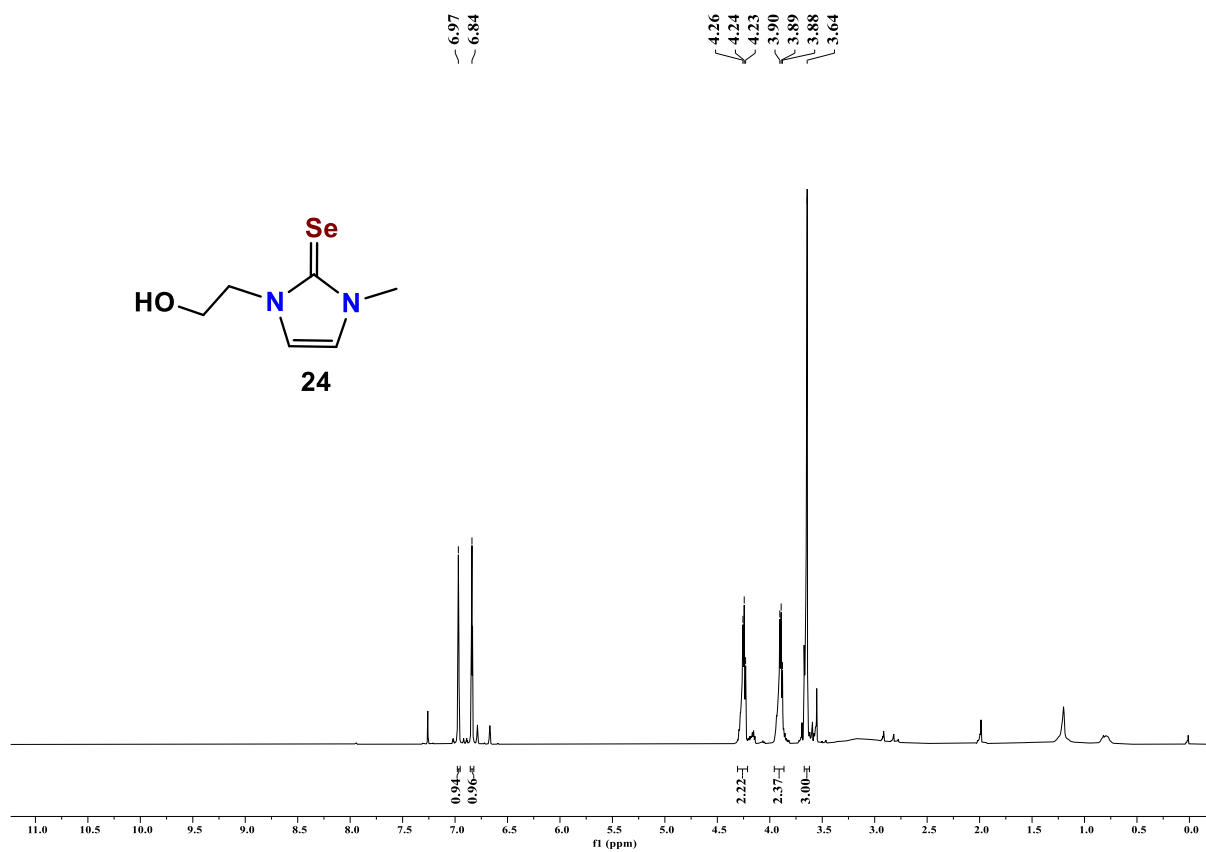


Fig. S68 ^1H NMR spectrum of **24**.

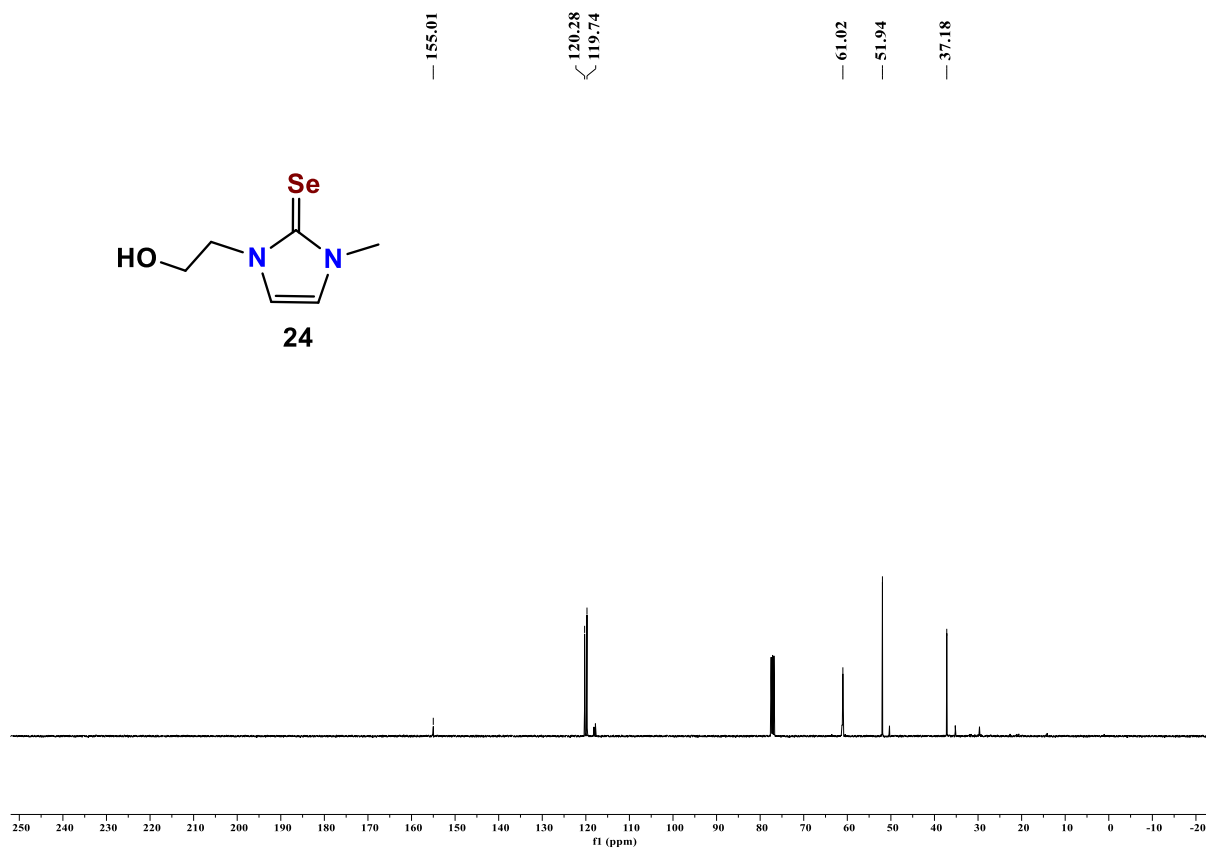


Fig. S69 ^{13}C NMR spectrum of **24**.

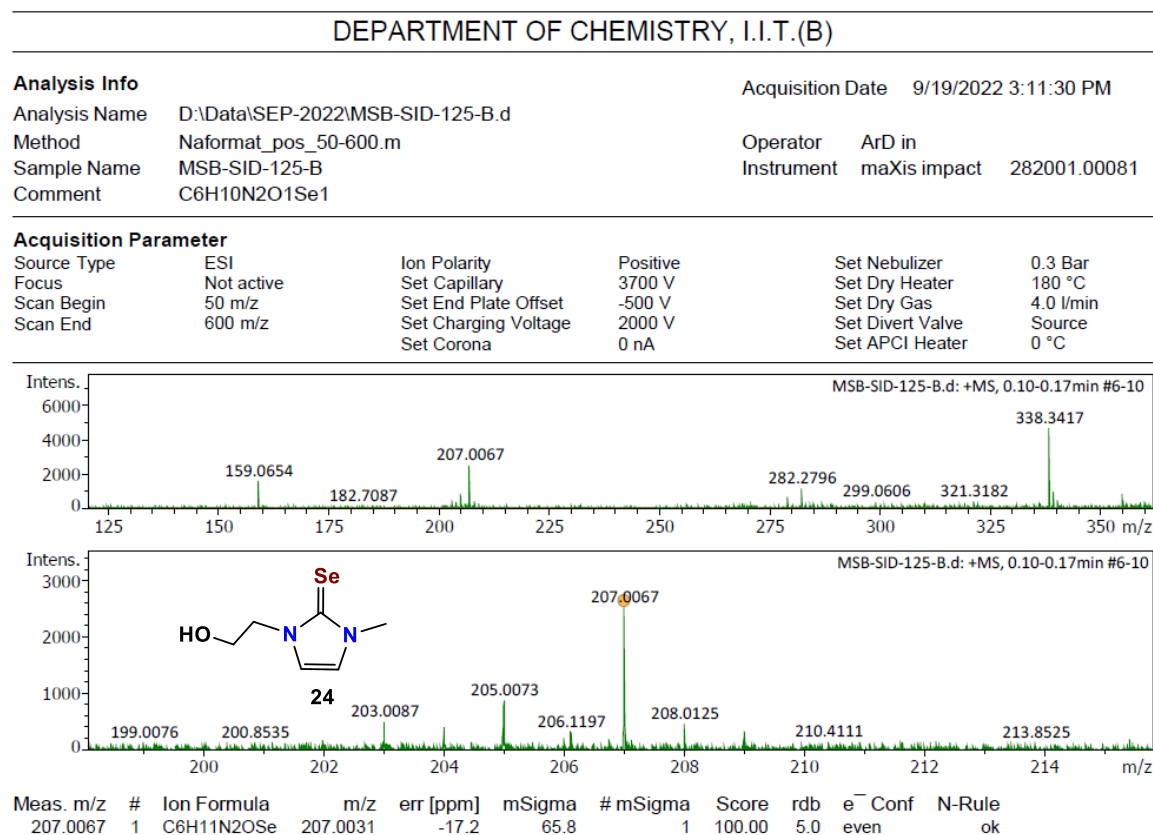


Fig. S70 HRMS spectrum of **24**.

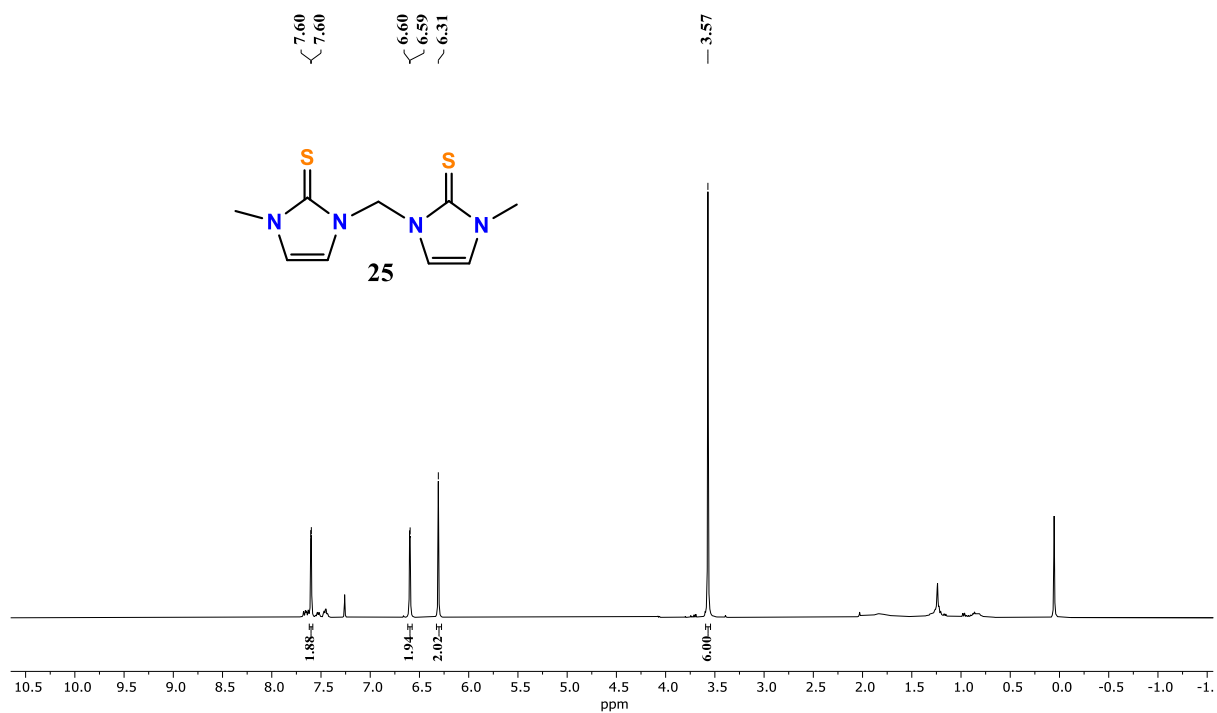


Fig. S71 ¹H NMR spectrum of **25**.

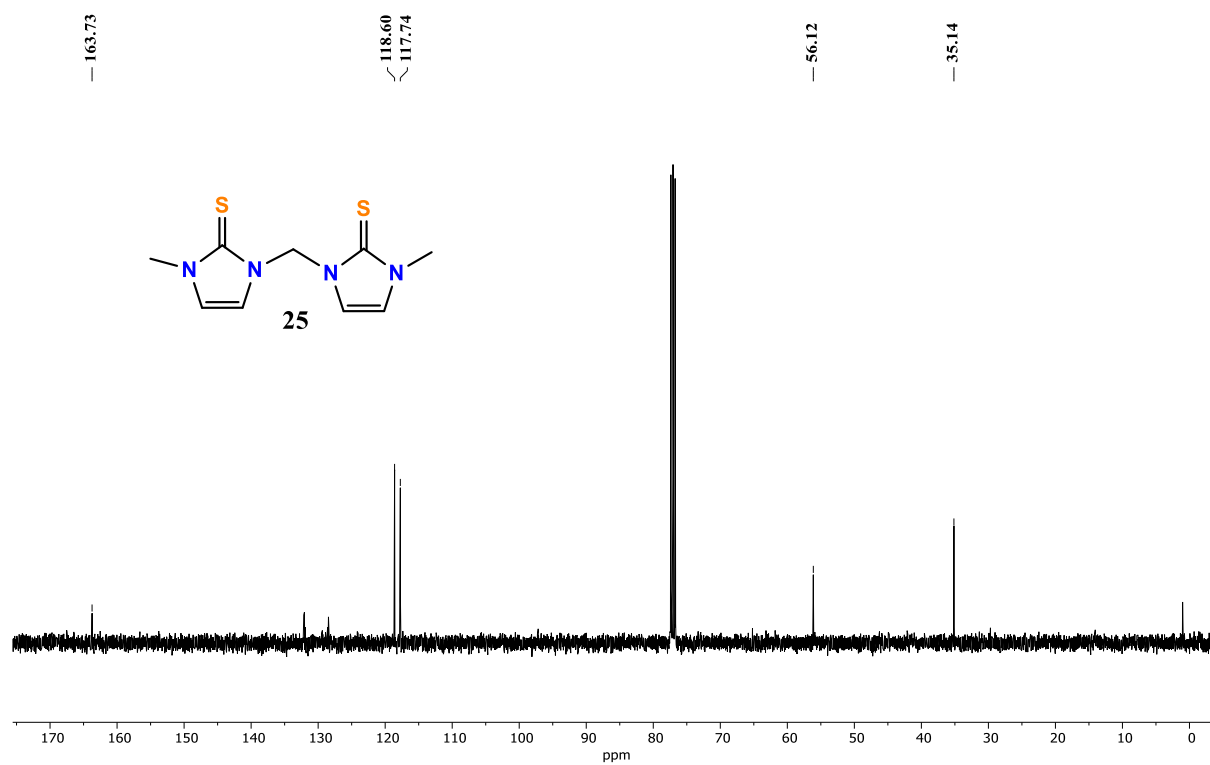


Fig. S72 ^{13}C NMR spectrum of **25**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

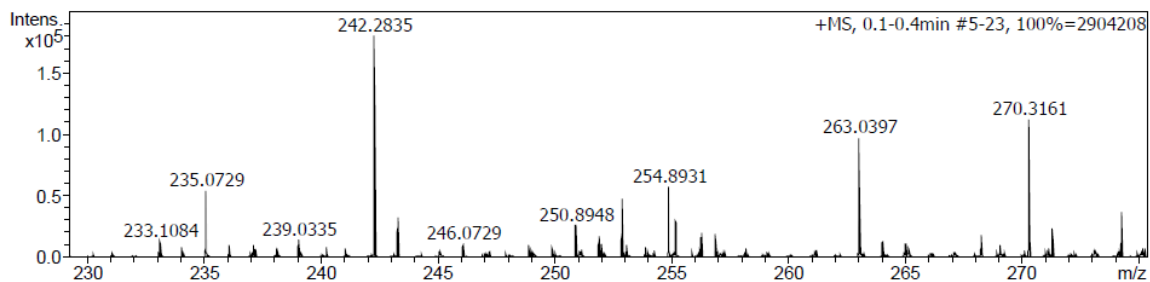
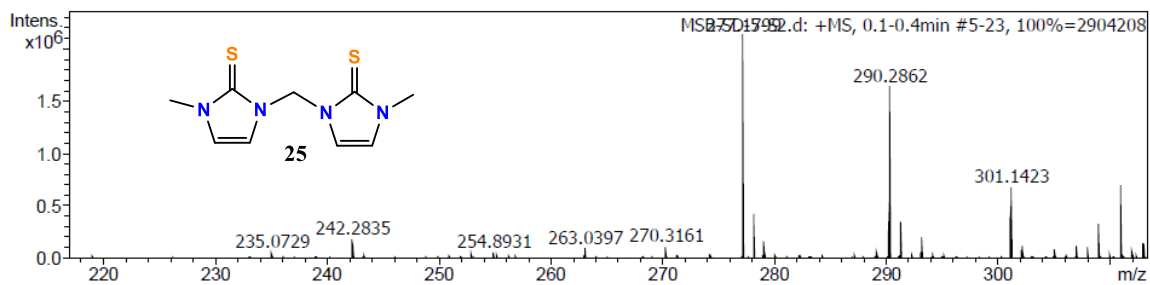
Analysis Name D:\Data\DEC-2019\MSB-SD-5-S2.d
 Method Tune_pos_500_NAF.m
 Sample Name MSB-SD-5-S2
 Comment C9H12N4S1S2

Acquisition Date 12/6/2019 8:59:35 PM

Operator CMRV-OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|---------|-----------------------|-----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 500 m/z | Set Collision Cell RF | 800.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 263.0397 | 1 | C9H12N4NaS2 | 263.0396 | 0.6 | 11.2 | 1 | 100.00 | 5.5 | even | ok |

Fig. S73 HRMS spectrum of **25**.

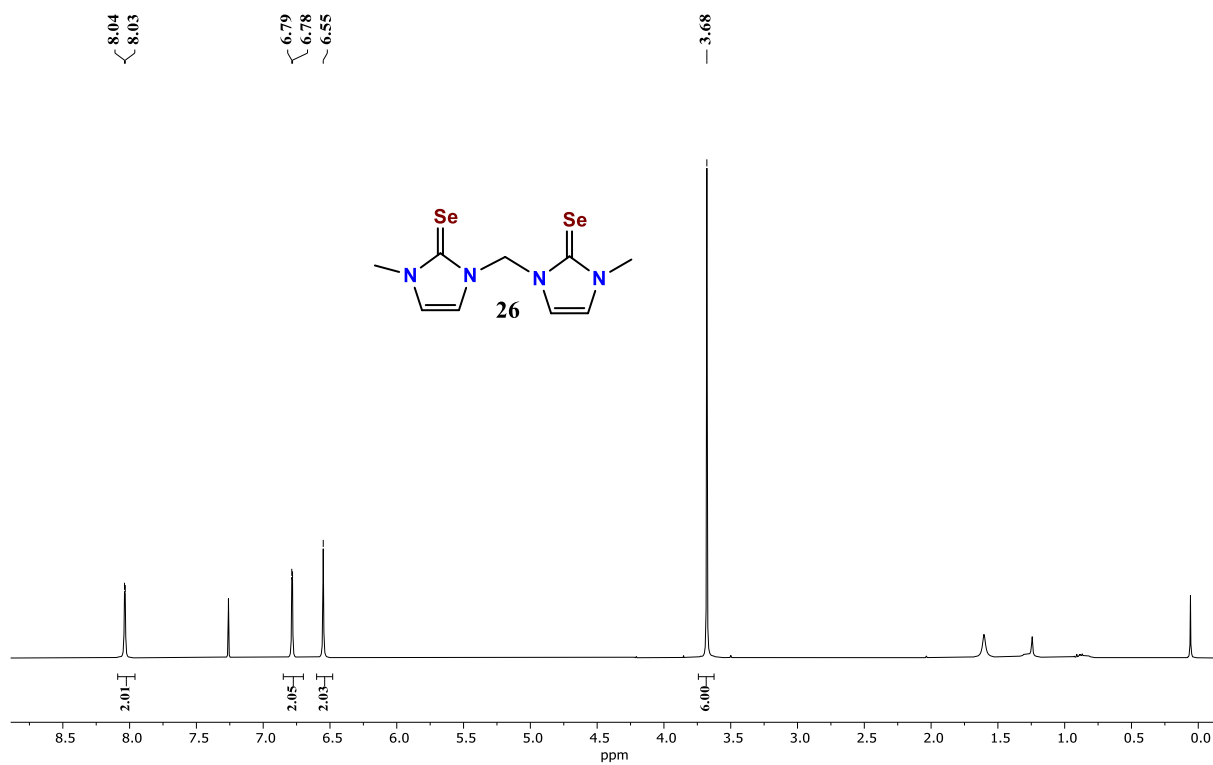


Fig. S74 ^1H NMR spectrum of **26**.

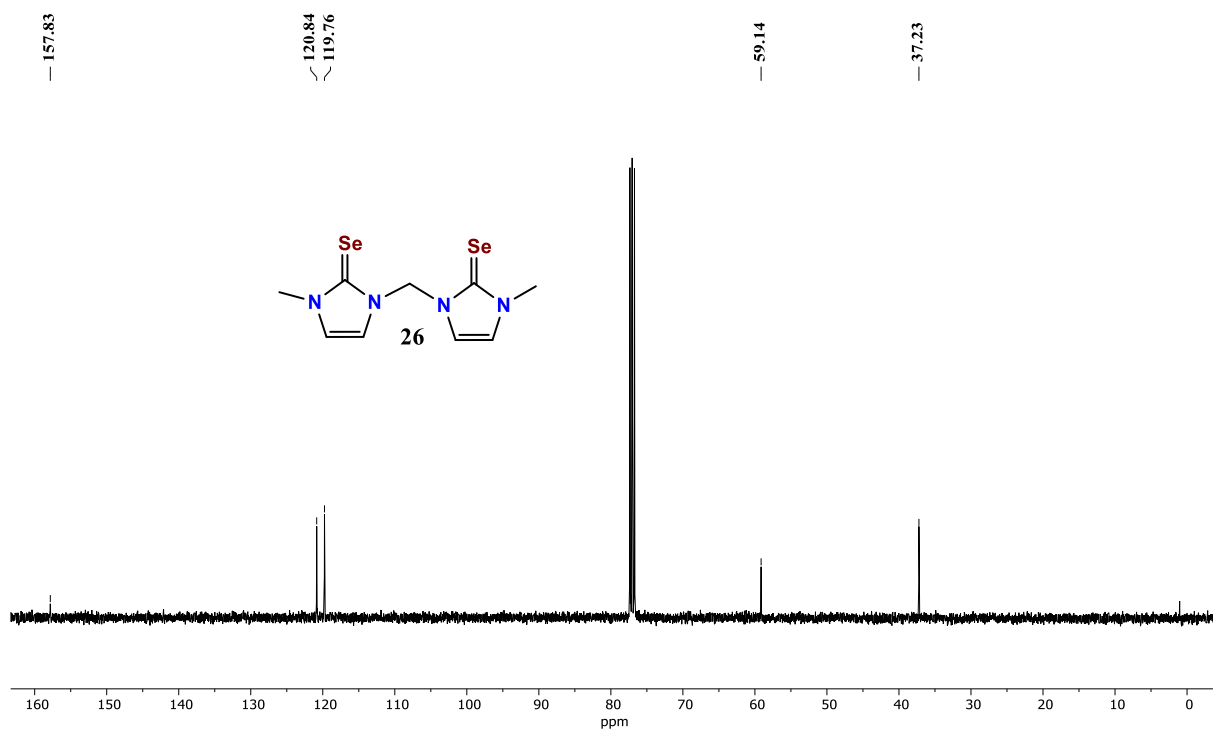


Fig. S75 ^{13}C NMR spectrum of **26**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

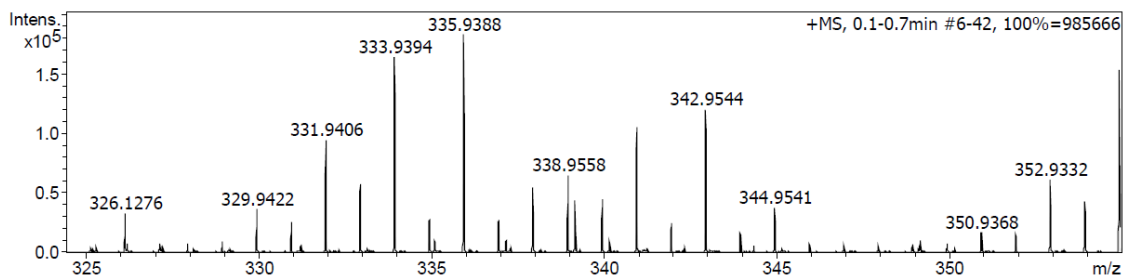
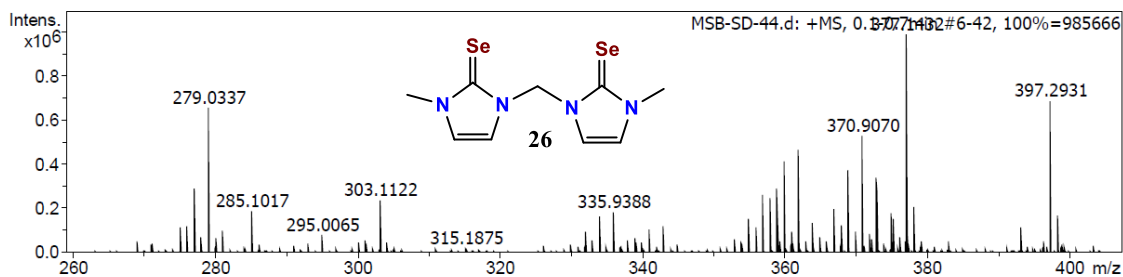
Analysis Info

Analysis Name D:\Data\NOV-2019\MSB-SD-44.d
 Method Tune_pos_NAICSI-1000.m
 Sample Name MSB-SD-44
 Comment C9H12N4Se2

Acquisition Date 11/26/2019 11:57:22 PM
 Operator INN-IN
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|----------|-----------------------|------------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Collision Cell RF | 1200.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 335.9388 | 1 | C9H12N4Se2 | 335.9389 | 0.3 | 14.3 | 1 | 100.00 | 6.0 | odd | ok |

Fig. S76 HRMS spectrum of **26**.

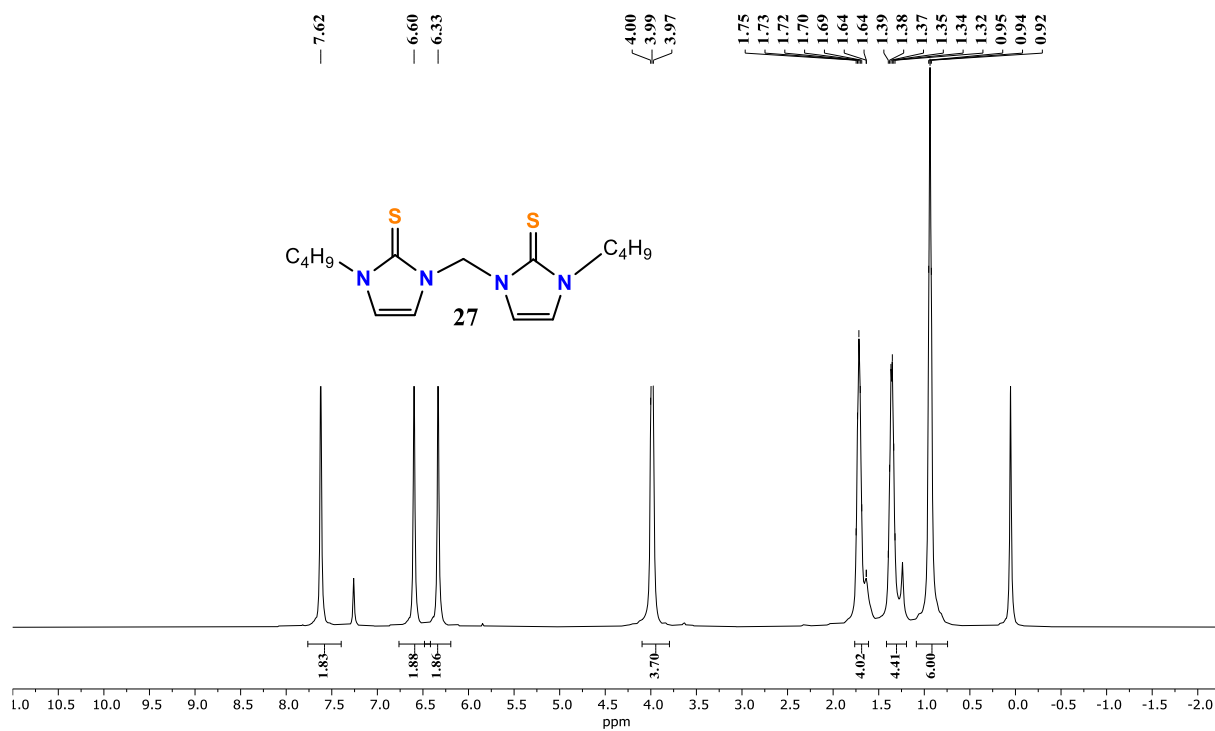


Fig. S77 ¹H NMR spectrum of **27**.

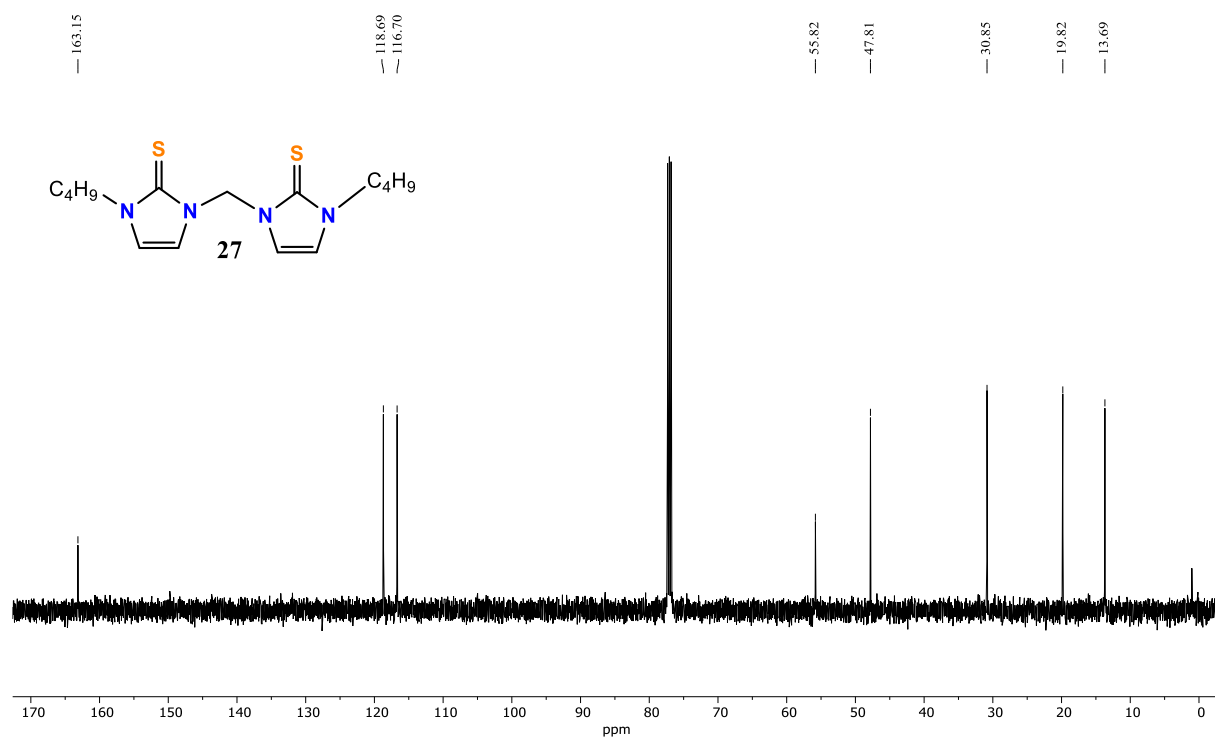


Fig. S78 ¹³C NMR spectrum of **27**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

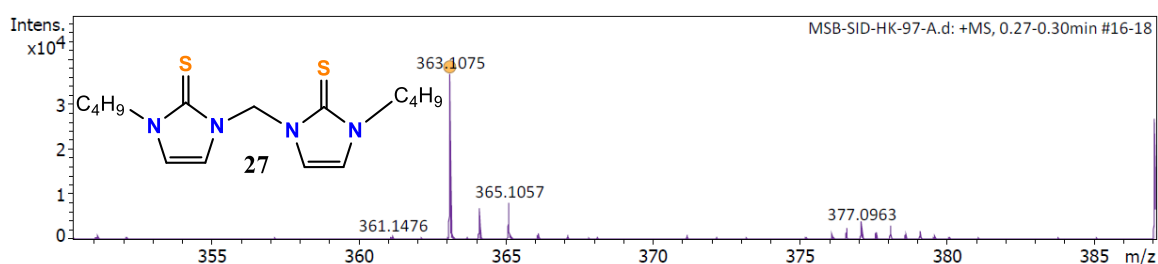
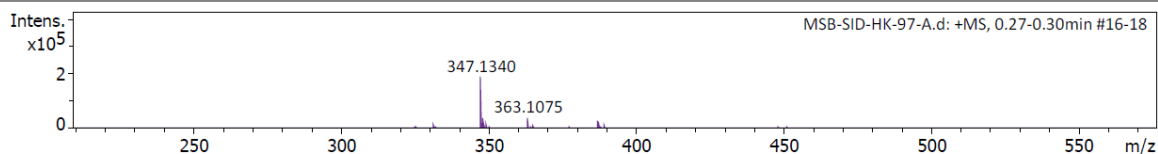
Analysis Name D:\Data\FEB-2022\MSB-SID-HK-97-A.d
 Method Naformat_pos_1000.m
 Sample Name MSB-SID-HK-97-A
 Comment C15H24N4S2

Acquisition Date 2/4/2022 8:12:50 PM

Operator SJGOUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1500 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ | Conf | N-Rule |
|-----------|---|-------------|----------|-----------|--------|----------|--------|------|----------------|------|--------|
| 363.1075 | 1 | C15H24KN4S2 | 363.1074 | -0.3 | 19.0 | 1 | 100.00 | 10.0 | even | | ok |

Fig. S79 HRMS spectrum of 27.

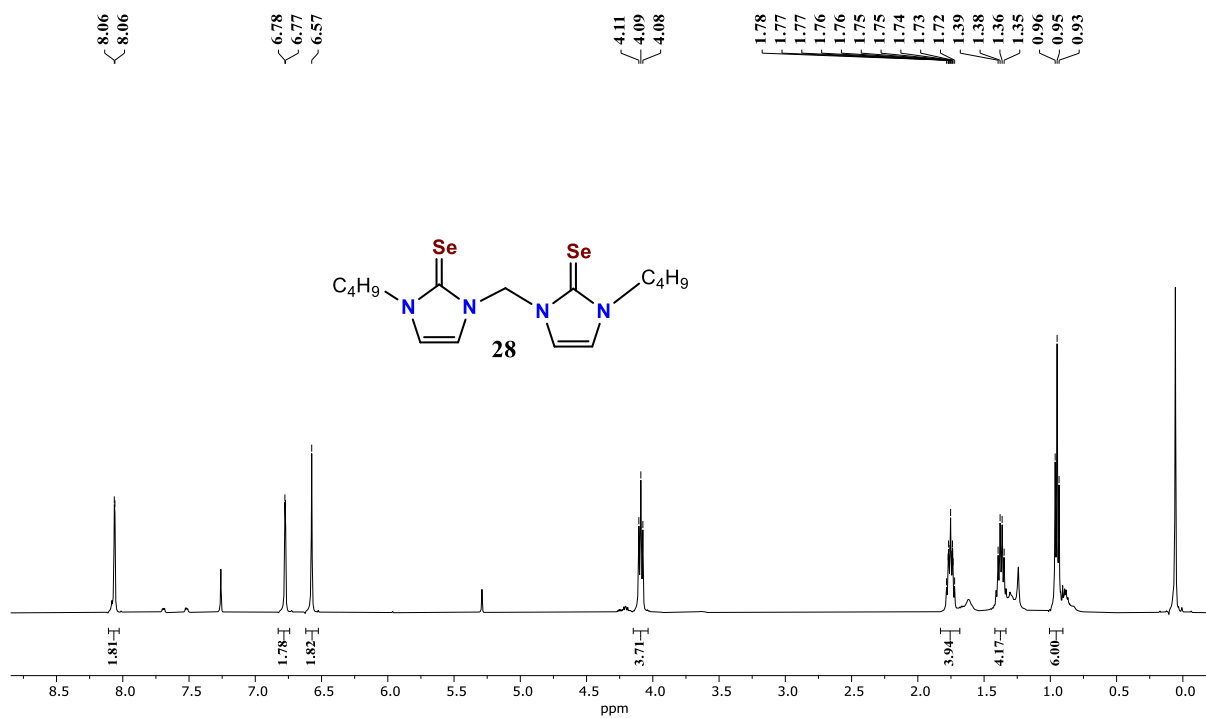


Fig. S80 ¹H NMR spectrum of **28**.

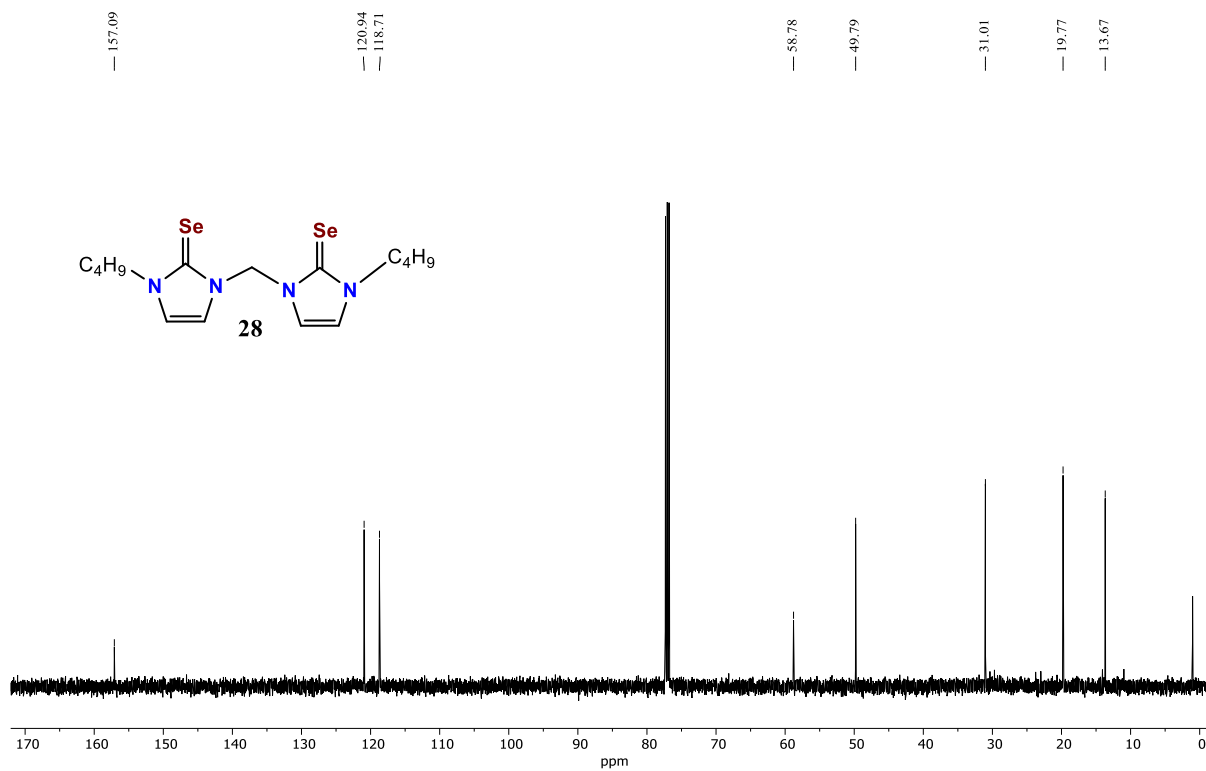


Fig. S81 ¹³C NMR spectrum of **28**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

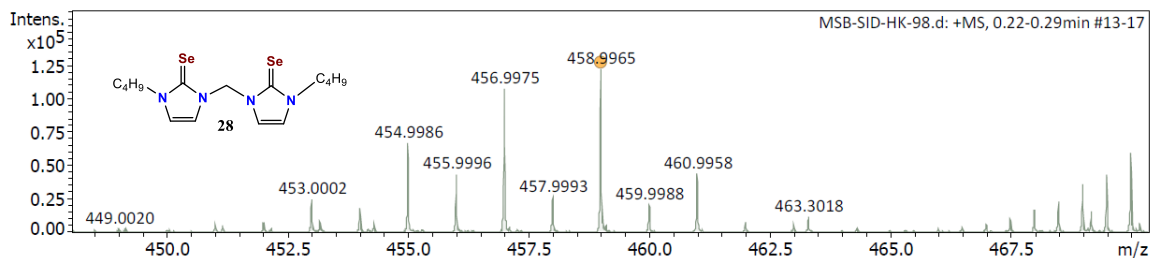
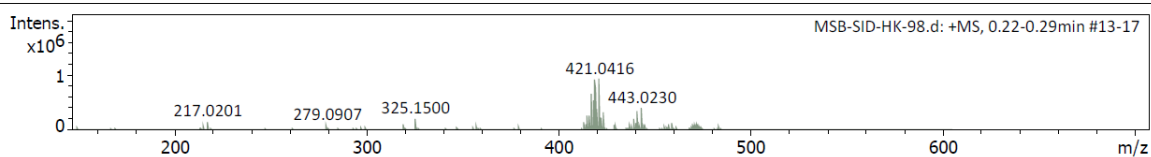
Analysis Info

Analysis Name D:\Data\FEB-2022\MSB-SID-HK-98.d
 Method Low_mass-NAF.m
 Sample Name MSB-SID-HK-98
 Comment C15H24N4Se2

Acquisition Date 2/4/2022 8:24:46 PM
 Operator SJGOUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 600 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|--------------|----------|-----------|--------|----------|--------|------|---------------------|--------|
| 458.9965 | 1 | C15H24KN4Se2 | 458.9963 | 0.1 | 9.6 | 1 | 100.00 | 10.0 | even | ok |

Fig. S82 HRMS spectrum of **28**.

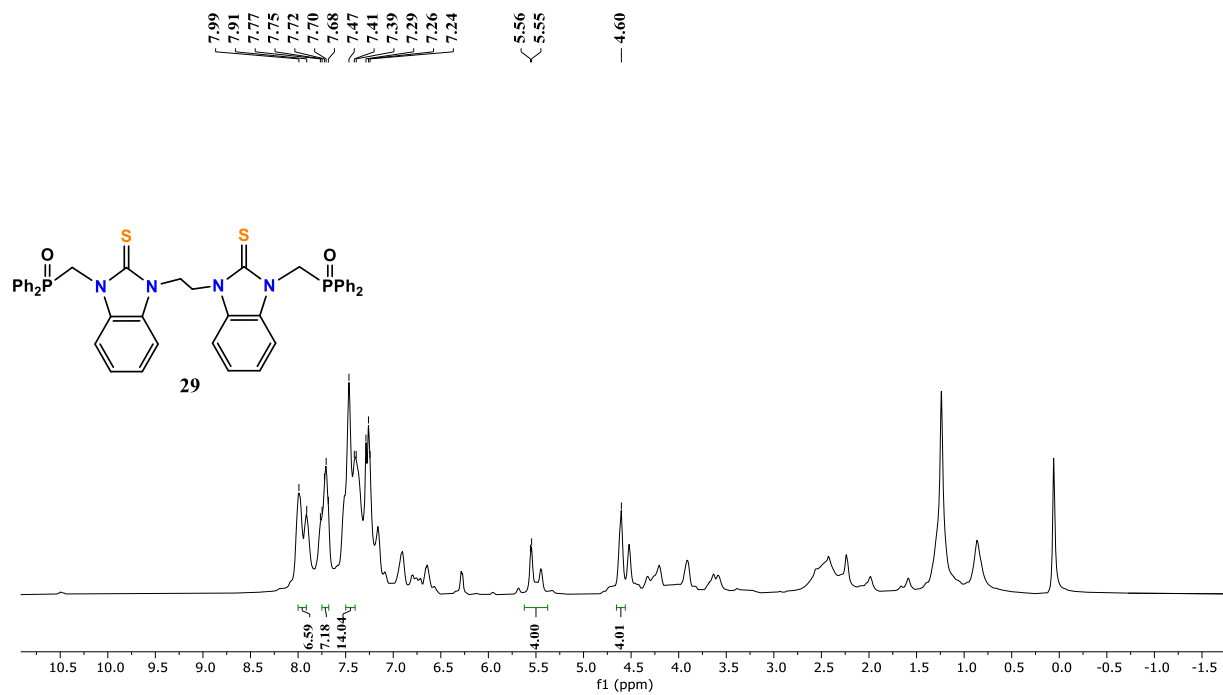


Fig. S83 ^1H NMR spectrum of **29**.

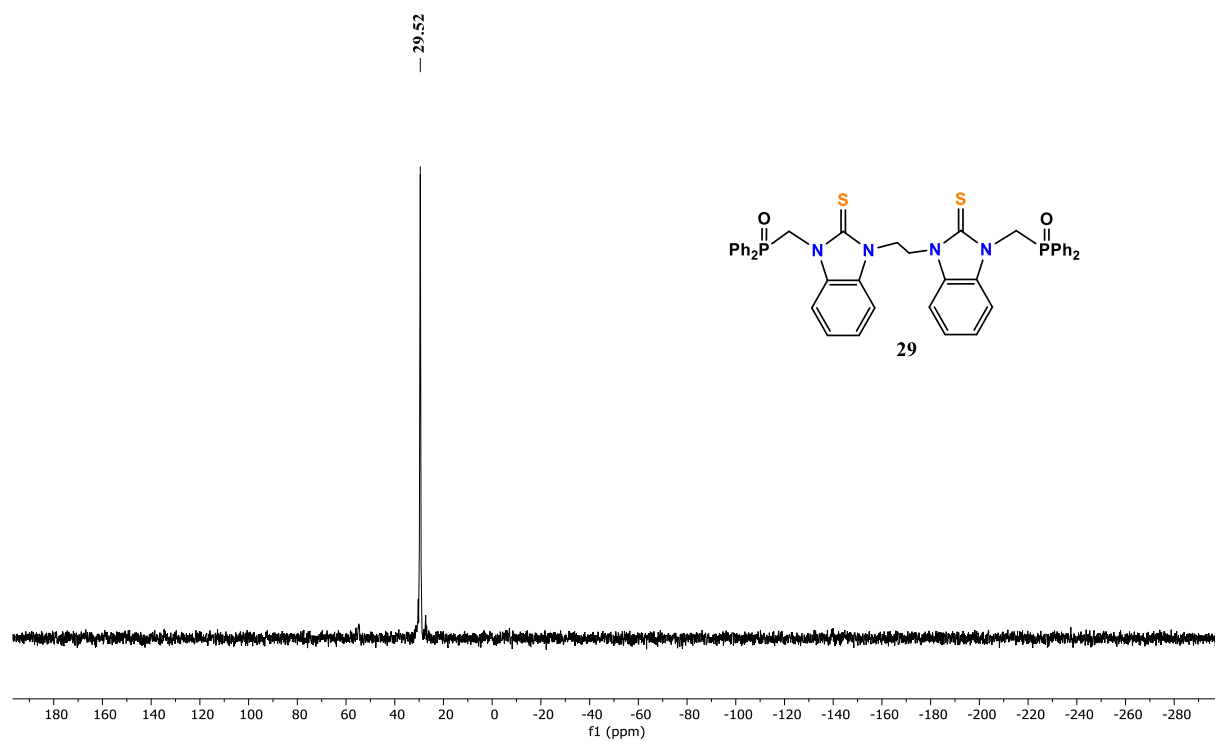


Fig. S84 $^{31}\text{P}\{^1\text{H}\}$ NMR Spectrum of **29**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

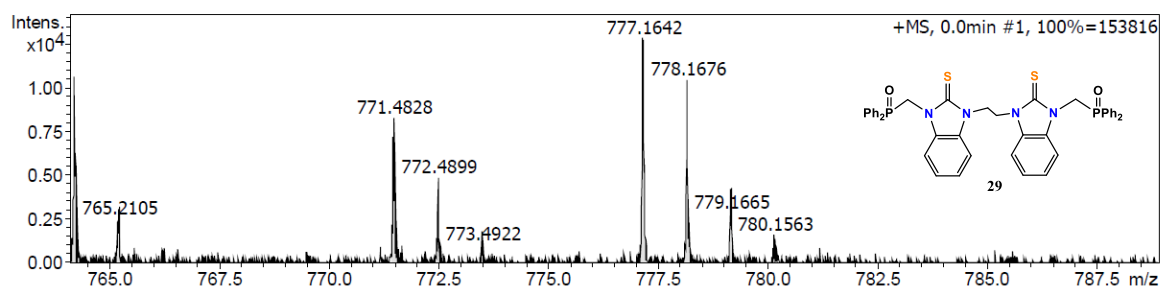
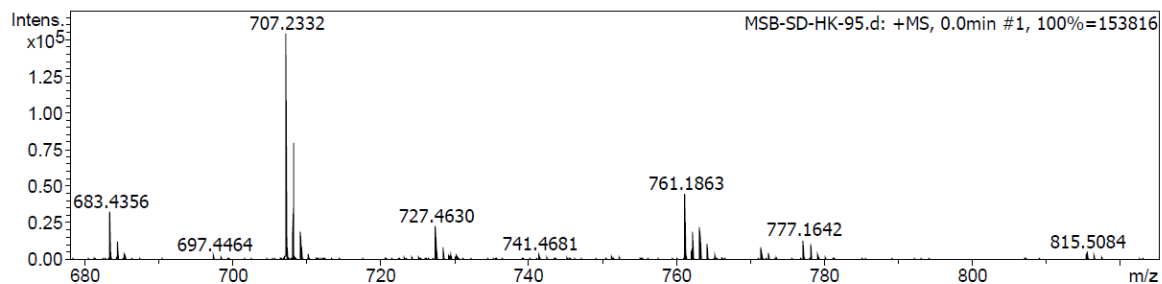
Analysis Info

Analysis Name D:\Data\MAR-2020\MSB-SD-HK-95.d
 Method Tune_pos_NAICSI-1500.m
 Sample Name MSB-SD-HK-95
 Comment C42H36N4O2P2S2

Acquisition Date 3/12/2020 11:10:35 AM
 Operator SJG-out
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|----------|-----------------------|------------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1500 m/z | Set Collision Cell RF | 1500.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|------------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 777.1642 | 1 | C42H36N4NaO2P2S2 | 777.1647 | 0.6 | 148.4 | 1 | 100.00 | 27.5 | even | ok |

Fig. S85 HRMS spectrum of **29**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

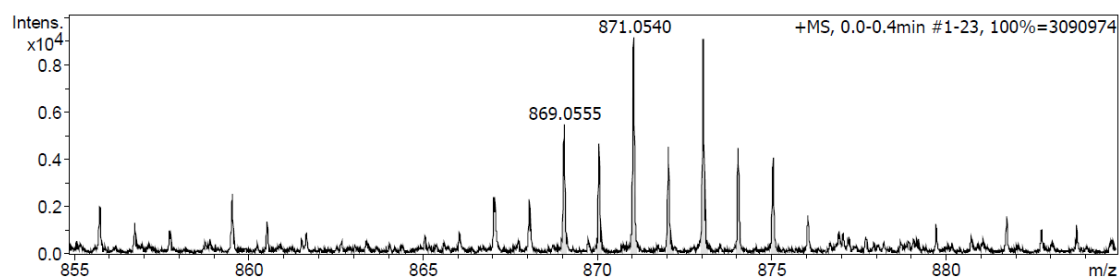
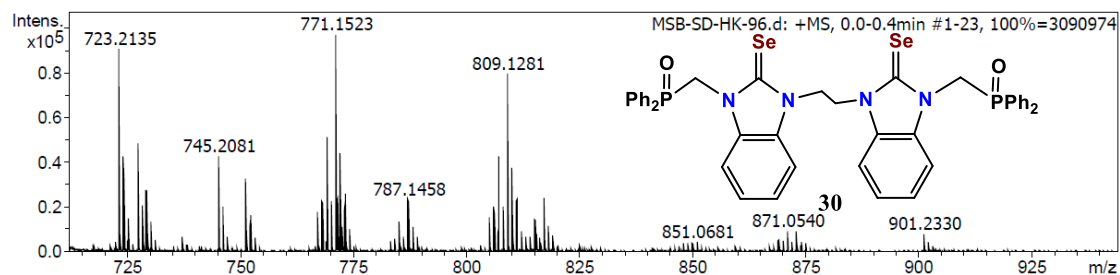
Analysis Name D:\Data\MAR-2020\MSB-SD-HK-96.d
 Method Tune_pos_NAICSI-1500.m
 Sample Name MSB-SD-HK-96
 Comment C42H36N4O2P2Se2

Acquisition Date 3/12/2020 10:41:16 AM

Operator SJG-out
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|----------|-----------------------|------------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1500 m/z | Set Collision Cell RF | 1500.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------------|----------|-----------|--------|---------|--------|------|---------------------|--------|
| 850.0648 | 1 | C42H36N4O2P2Se2 | 850.0648 | 0.0 | 285.2 | 1 | 100.00 | 28.0 | odd | ok |
| | 1 | C42H36N4O2P2Se2 | 850.0648 | 0.0 | 285.2 | 1 | 100.00 | 28.0 | odd | ok |
| | 1 | C42H36N4O2P2Se2 | 850.0648 | 0.0 | 285.2 | 1 | 100.00 | 28.0 | odd | ok |
| | 1 | C42H36N4O2P2Se2 | 850.0648 | 0.0 | 285.2 | 1 | 100.00 | 28.0 | odd | ok |
| 873.0520 | 1 | C42H36N4NaO2P2Se2 | 873.0546 | -3.0 | 91.5 | 1 | 100.00 | 27.5 | even | ok |

Fig. S88 HRMS spectrum of **30**.

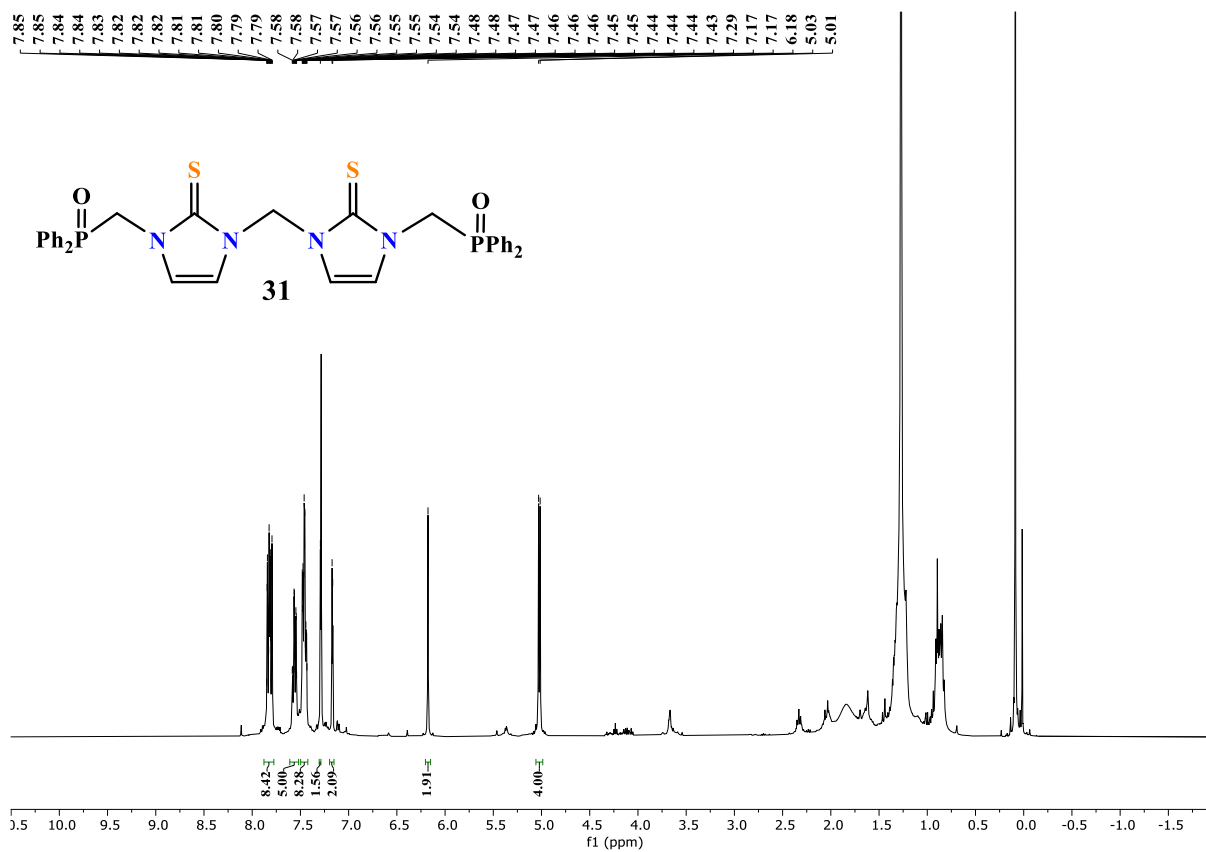


Fig. S89 ¹H NMR spectrum of **31**.

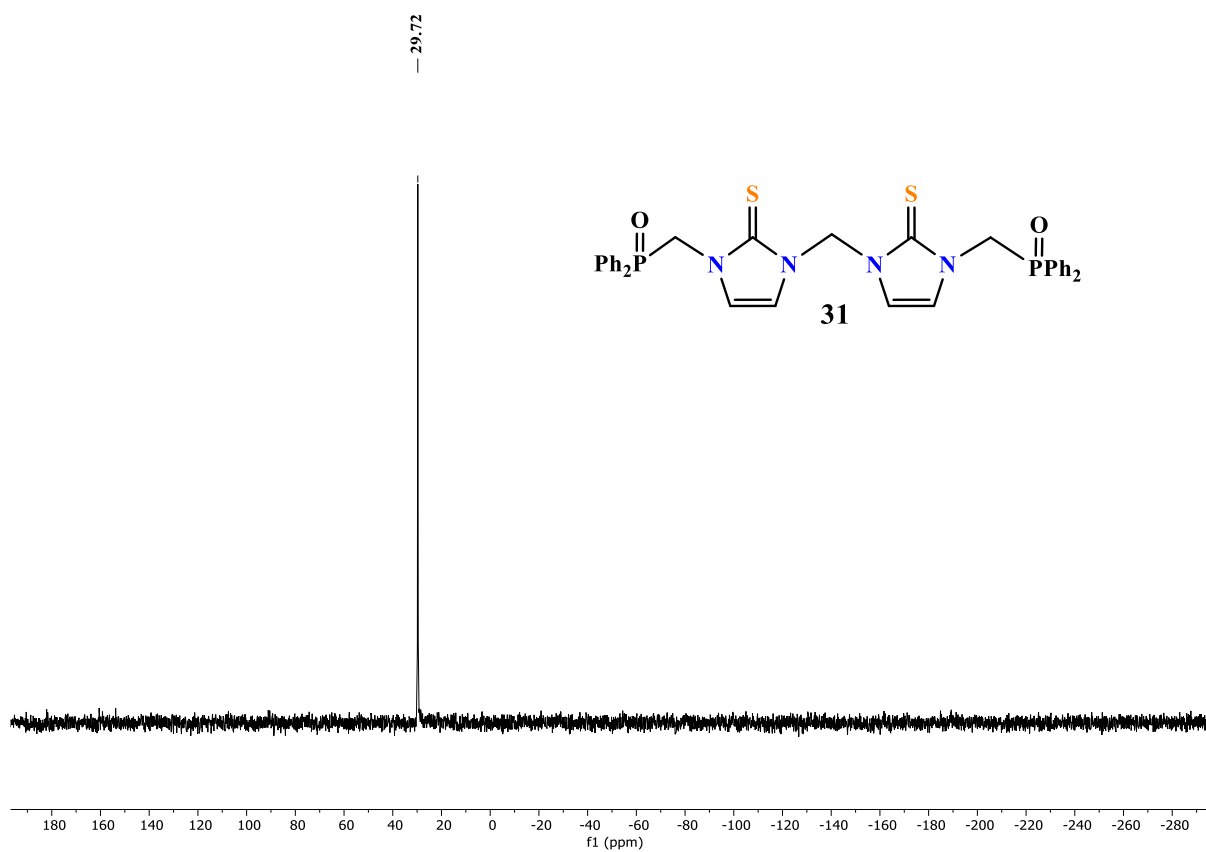


Fig. S90 ³¹P{¹H} NMR spectrum of **31**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

Analysis Name D:\Data\JAN-2022\MSB-SID-HK-104.d
 Method NaICSI_pos_1000-a.m
 Sample Name MSB-SID-HK-104
 Comment C33H30O2P2S2N4

Acquisition Date 1/21/2022 8:34:54 PM
 Operator SJG-IN
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |

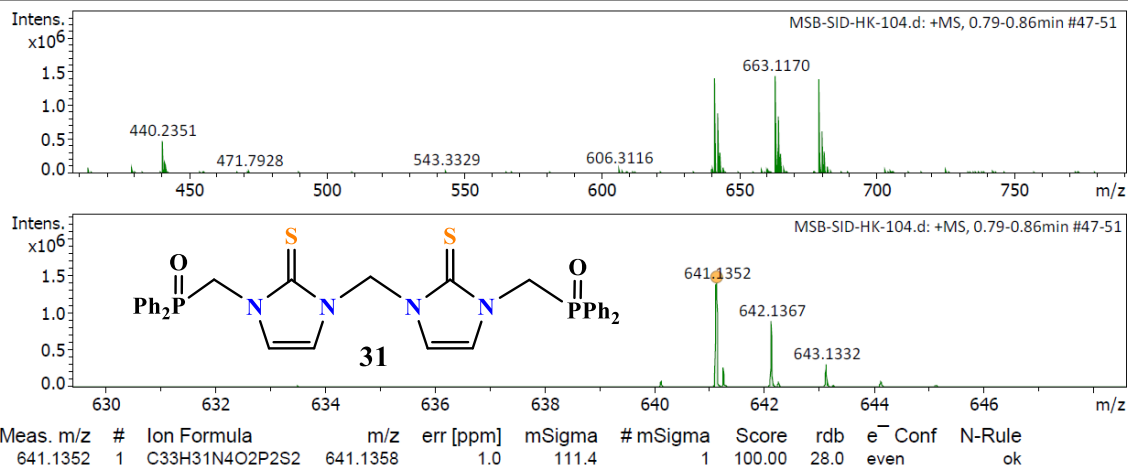


Fig. S92 HRMS spectrum of **31**.

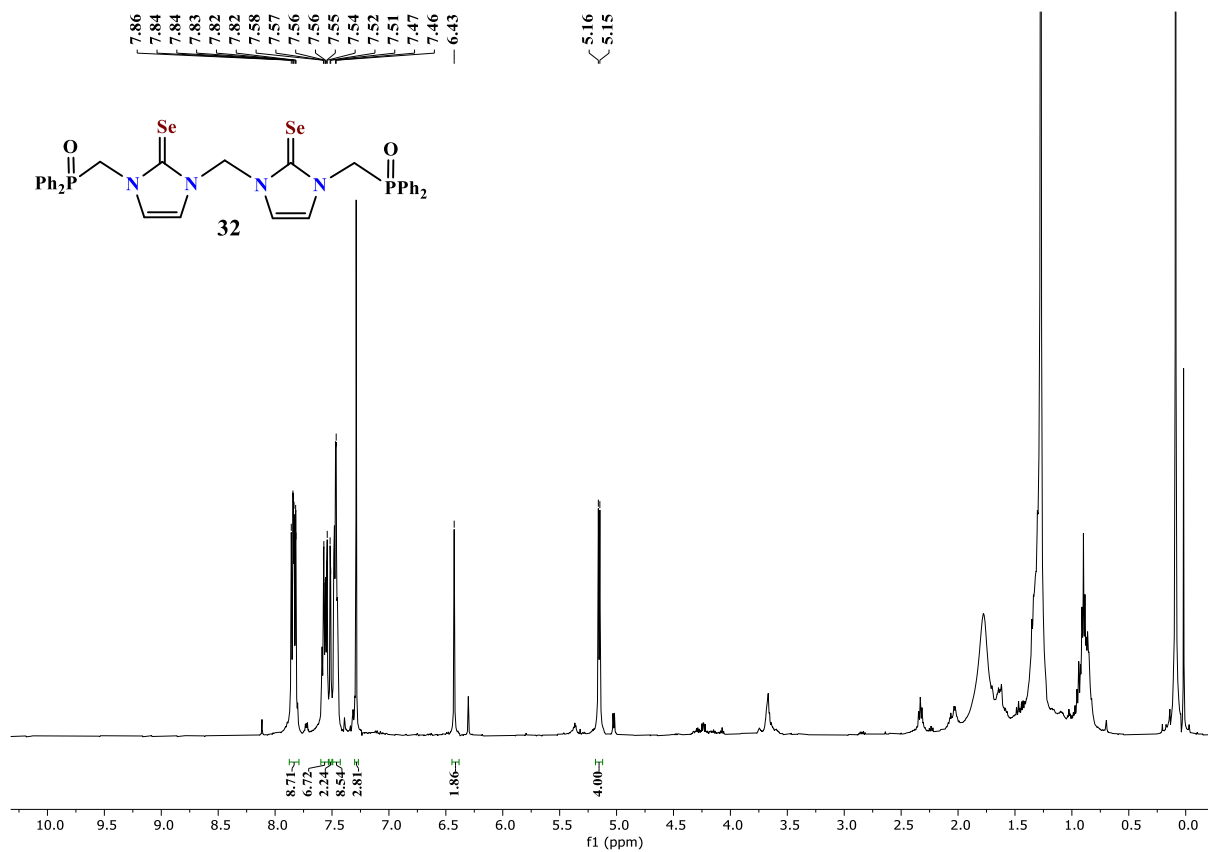


Fig. S93 ^1H NMR spectrum of **32**.

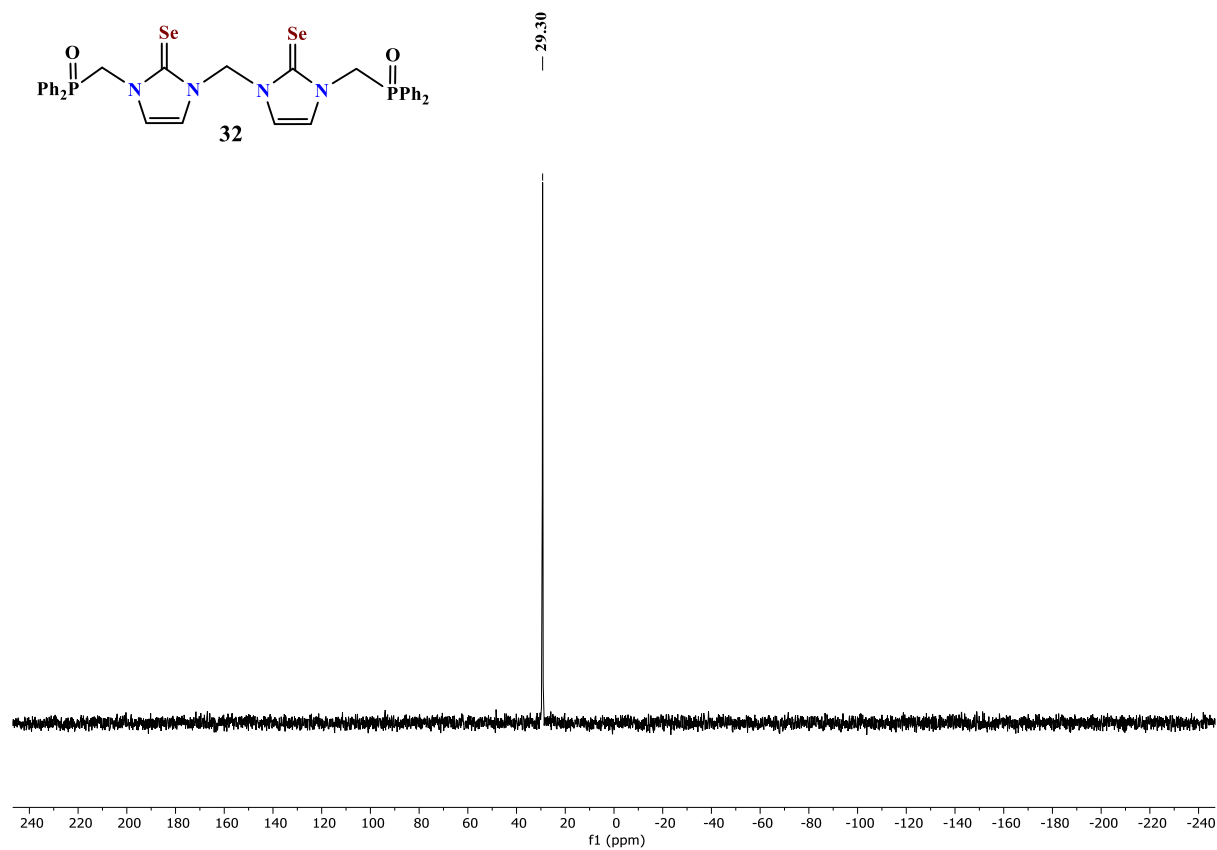


Fig. S94 $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of **32**.

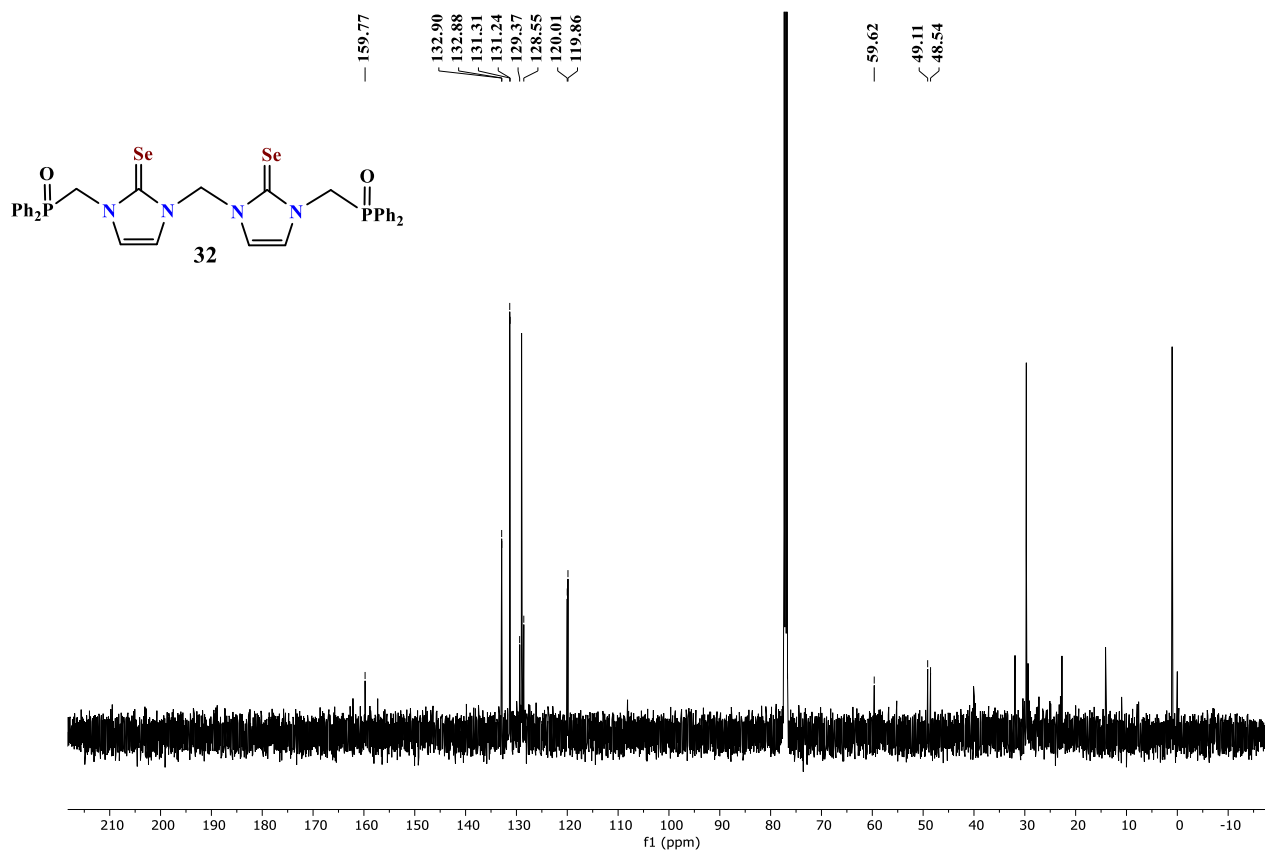
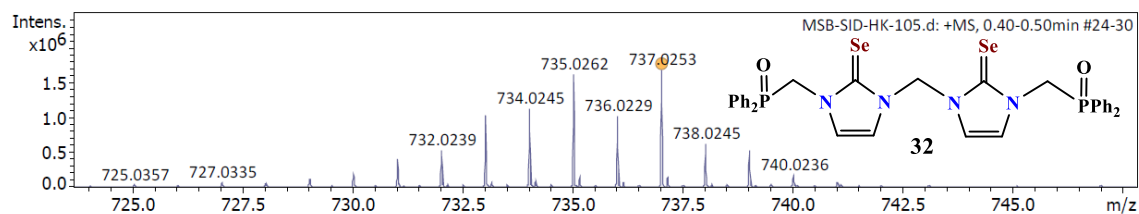
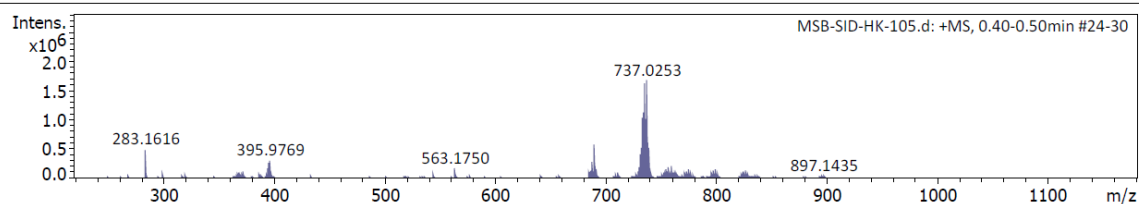


Fig. S95 ^{13}C NMR spectrum of **32**.

| DEPARTMENT OF CHEMISTRY, I.I.T.(B) | | | | | |
|------------------------------------|-----------------------------------|----------------------|---------------------------------------|---------------------------|-----------|
| Analysis Info | | | Acquisition Date 1/21/2022 8:27:45 PM | | |
| Analysis Name | D:\Data\JAN-2022\MSB-SID-HK-105.d | | Operator | SJG-IN | |
| Method | NaICsI_pos_1000-a.m | | Instrument | maXis impact 282001.00081 | |
| Sample Name | MSB-SID-HK-105 | | | | |
| Comment | C33H30O2P2Se2N4 | | | | |
| Acquisition Parameter | | | | | |
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-----------------|----------|-----------|--------|----------|--------|------|---------------------|--------|
| 737.0253 | 1 | C33H31N4O2P2Se2 | 737.0247 | 0.1 | 99.5 | 1 | 100.00 | 28.0 | even | ok |

Fig. S96 HRMS spectrum of **32**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

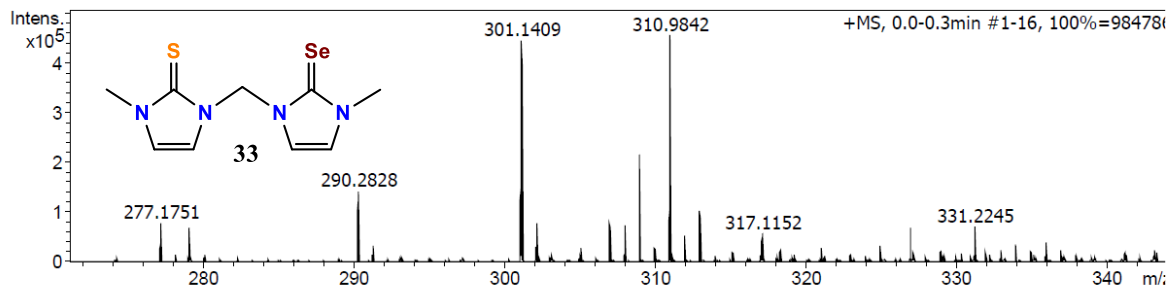
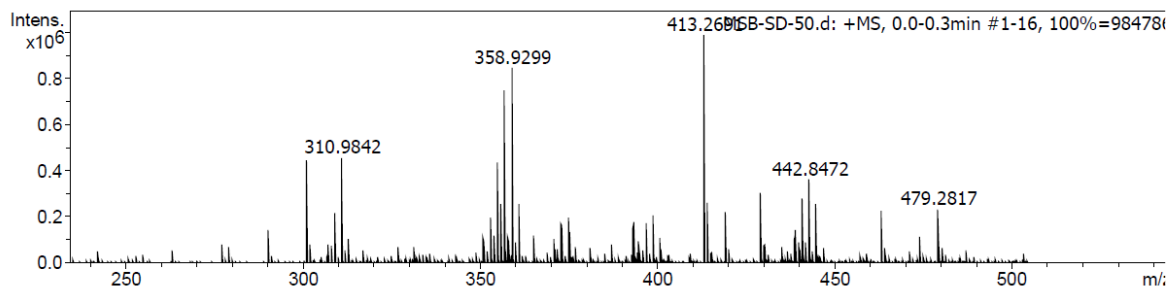
Analysis Info

Analysis Name D:\Data\DEC-2019\MSB-SD-50.d
 Method Tune_pos_500_NAF.m
 Sample Name MSB-SD-50
 Comment C9H12N4S1Se1

Acquisition Date 12/6/2019 8:46:27 PM
 Operator CMRV-OUT
 Instrument maXis impact 282001.0008

Acquisition Parameter

| | | | | | |
|-------------|---------|-----------------------|-----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Active | Set Capillary | 4500 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 500 m/z | Set Collision Cell RF | 800.0 Vpp | Set Divert Valve | Source |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # Sigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|--------------|----------|-----------|--------|---------|--------|-----|---------------------|--------|
| 310.9842 | 1 | C9H12N4NaSSe | 310.9840 | -0.9 | 8.5 | 1 | 100.00 | 5.5 | even | ok |

Fig. S97 HRMS spectrum of **33**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

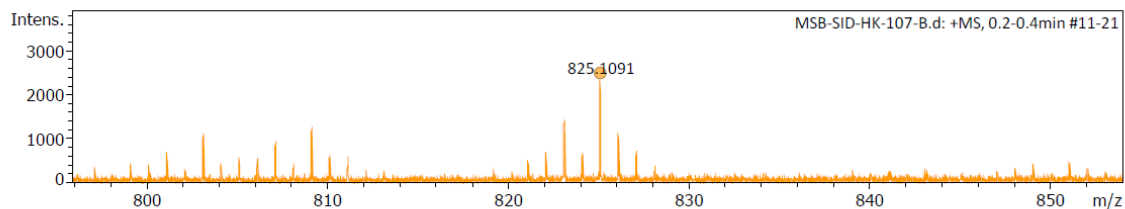
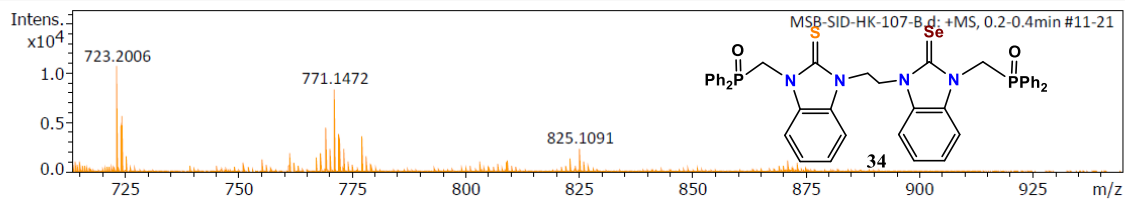
Analysis Name D:\Data\APR-2022\MSB-SID-HK-107-B.d
 Method NaICsl_pos_1500hplc.m
 Sample Name MSB-SID-HK-107-B
 Comment C42H36N4O2P2S1Se1

Acquisition Date 4/5/2022 6:40:22 PM

Operator SJG OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.4 Bar |
| Focus | Not active | Set Capillary | 3700 V | Set Dry Heater | 200 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.5 l/min |
| Scan End | 1500 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------------|----------|-----------|--------|----------|--------|------|---------------------|--------|
| 825.1091 | 1 | C42H36N4NaO2P2SSe | 825.1092 | 0.5 | n.a. | 1 | 100.00 | 34.0 | even | ok |

Fig. S98 HRMS spectrum of **34**.

DEPARTMENT OF CHEMISTRY, I.I.T.(B)

Analysis Info

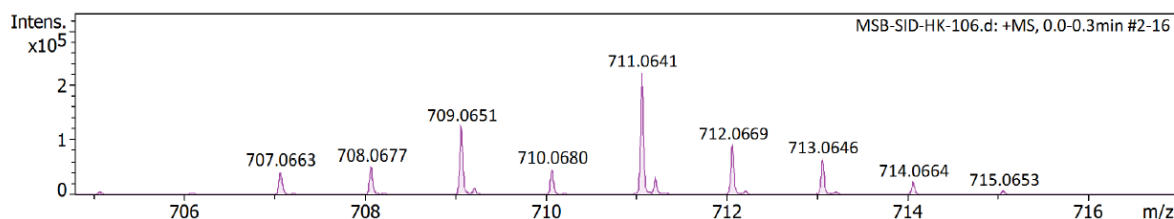
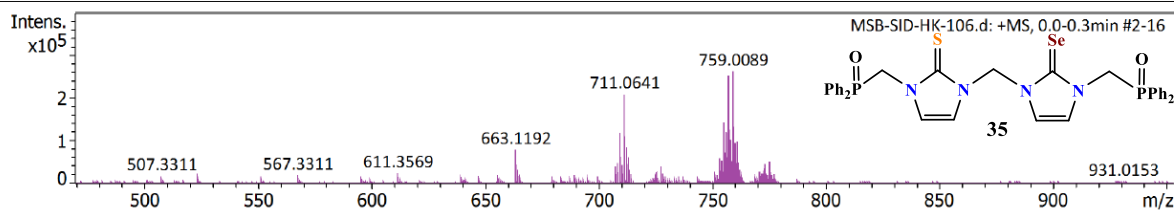
Analysis Name D:\Data\AUG 2021\MSB-SID-HK-106.d
 Method Naformat_pos_1000a.m
 Sample Name MSB-SID-HK-106
 Comment C33H30O2P2SSeN4

Acquisition Date 8/12/2021 3:15:03 AM

Operator MSB-OUT
 Instrument maXis impact 282001.00081

Acquisition Parameter

| | | | | | |
|-------------|------------|----------------------|----------|------------------|-----------|
| Source Type | ESI | Ion Polarity | Positive | Set Nebulizer | 0.3 Bar |
| Focus | Not active | Set Capillary | 4000 V | Set Dry Heater | 180 °C |
| Scan Begin | 50 m/z | Set End Plate Offset | -500 V | Set Dry Gas | 4.0 l/min |
| Scan End | 1000 m/z | Set Charging Voltage | 2000 V | Set Divert Valve | Source |
| | | Set Corona | 0 nA | Set APCI Heater | 0 °C |



| Meas. m/z | # | Ion Formula | m/z | err [ppm] | mSigma | # mSigma | Score | rdb | e ⁻ Conf | N-Rule |
|-----------|---|-------------------|----------|-----------|--------|----------|--------|------|---------------------|--------|
| 709.0651 | 1 | C33H30N4NaO2P2SSe | 711.0622 | -1.9 | 13.8 | 1 | 100.00 | 28.0 | even | ok |

Fig. S99 HRMS spectrum of **35**.

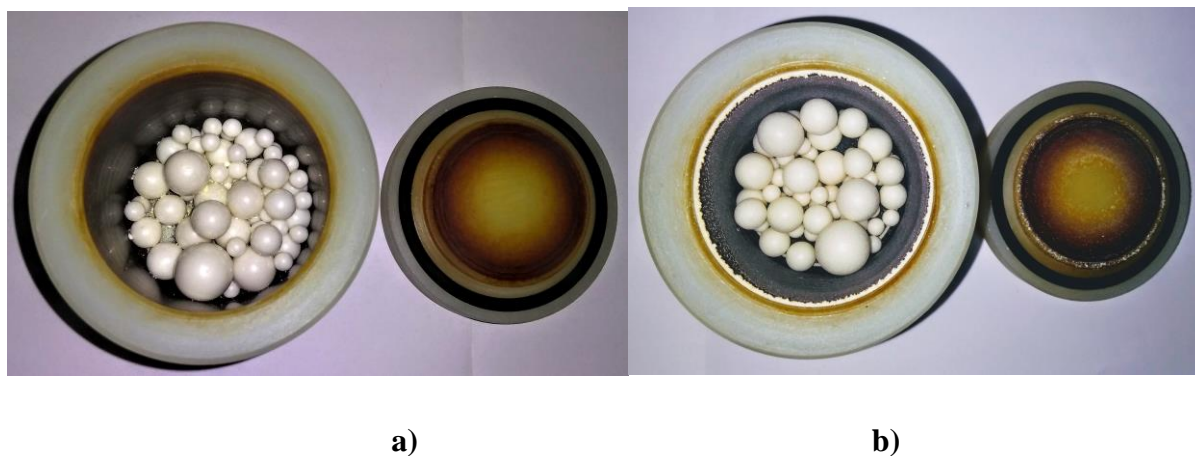


Fig. S100 Synthesis of thiones a) before the synthesis b) after the synthesis



a)

b)

Fig. S101 Synthesis of selones a) before the synthesis b) after the synthesis

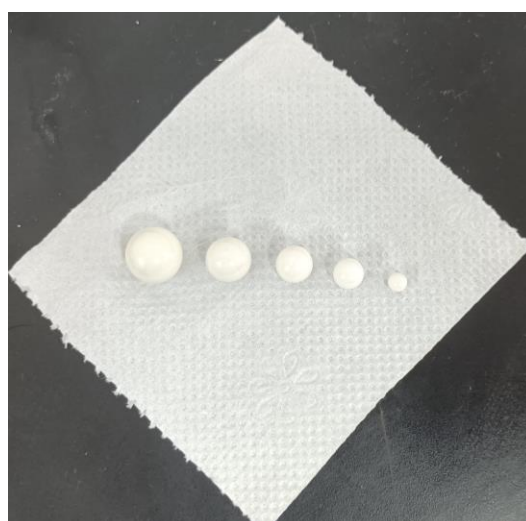


Fig. S102 Different sized balls used in the current work for efficient grinding.



Fig. S103 Ball-milling machine used in the current study.

References:

- 1 O. V. Dolomanov, L. J. Bourhis, R. J. Gildea, J. A. K. Howard and H. Puschmann, *J. Appl. Cryst.*, 2009, **42**, 339-341.
- 2 G. M. Sheldrick, *Acta Crystallogr., Sect. A: Found. Crystallogr.*, 2015, **71**, 3-8.
- 3 G. Sheldrick, *Acta Crystallogr., Sect. C: Struct. Chem.*, 2015, **71**, 3-8.