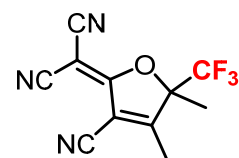


(Contents)

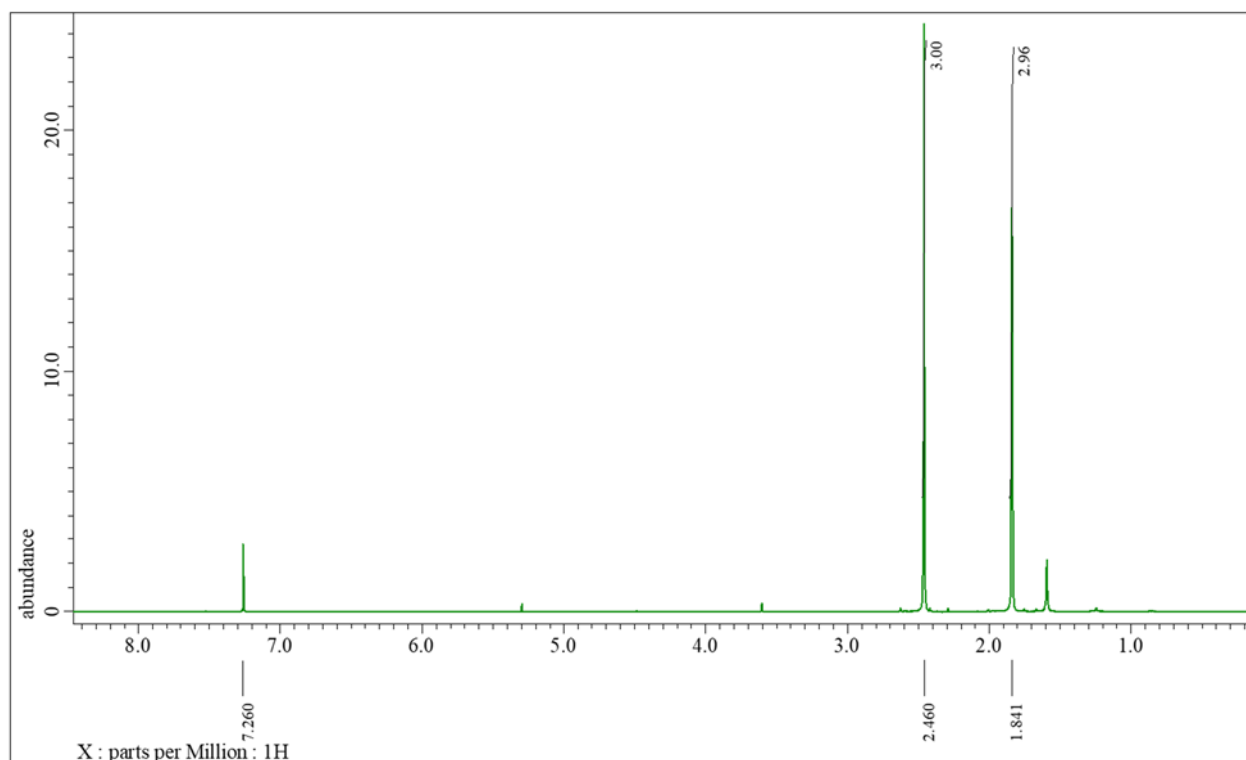
^1H , ^{13}C , ^{19}F NMR, IR and HRMS for 2a	S2–S4
^1H , ^{13}C NMR, IR and HRMS for 3 -.....	S5, S6
^1H , ^{13}C , ^{19}F NMR, IR and HRMS for 5a	S7–S9
^1H , ^{13}C NMR, IR and HRMS for 6	S10, S11
^1H , ^{13}C NMR, IR and HRMS for 7	S12, S13
^1H , ^{13}C , ^{19}F NMR, IR and HRMS for 8a	S14–S16
Result of CV measurement for 5a	S17
Results of TG–DTA experiments for 5a , 7a	S18

2-(2-Cyano-3,4-dimethyl-4-(trifluoromethyl)cyclopent-2-en-1-ylidene)malononitrile (2a) [1]

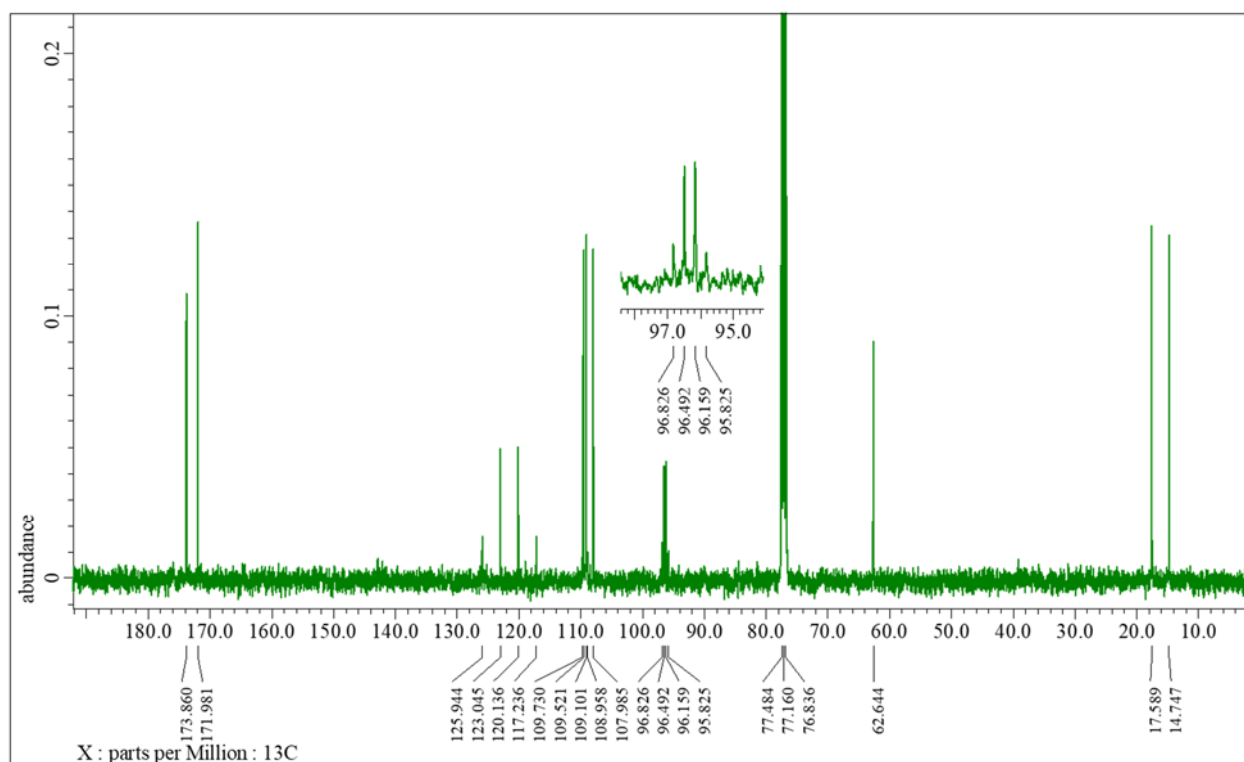


2a

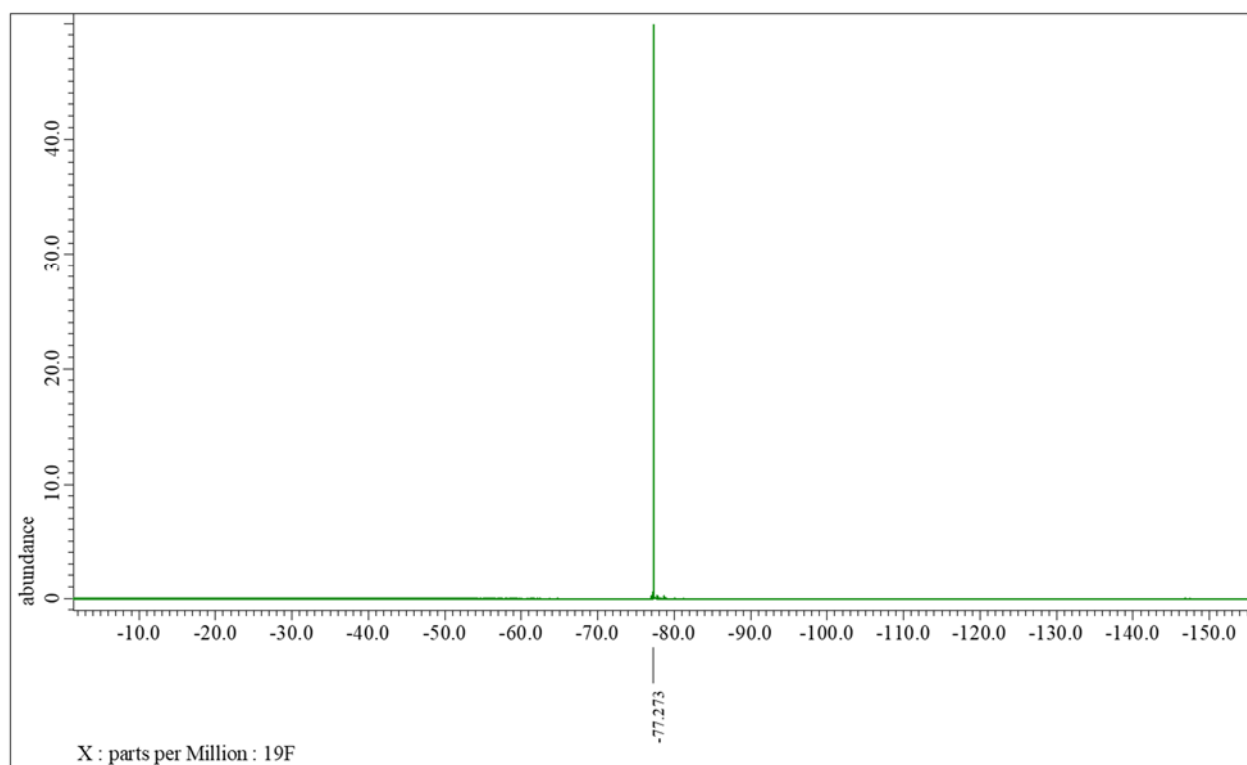
^1H NMR



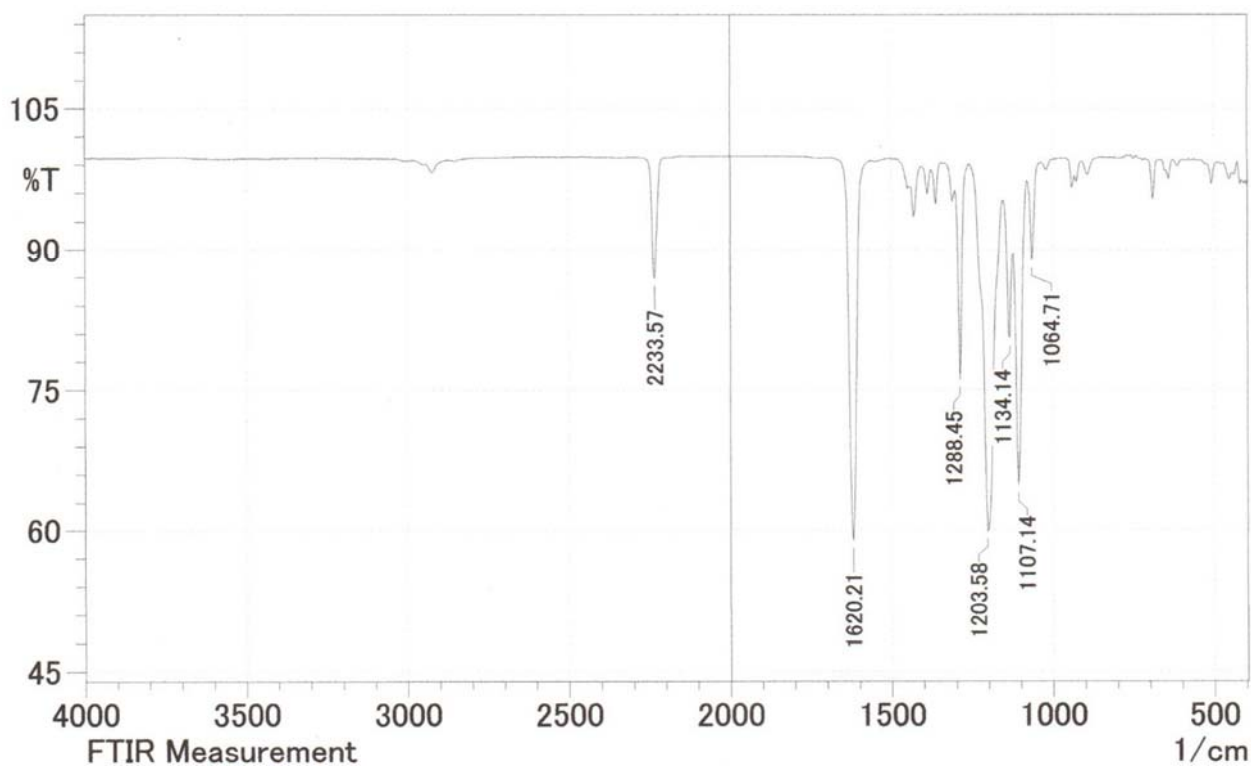
^{13}C NMR



^{19}F NMR



IR



Elemental Composition Report

Page 1

Single Mass Analysis

Tolerance = 50.0 mDa / DBE: min = -1.5, max = 100.0

Element prediction: Off

Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions

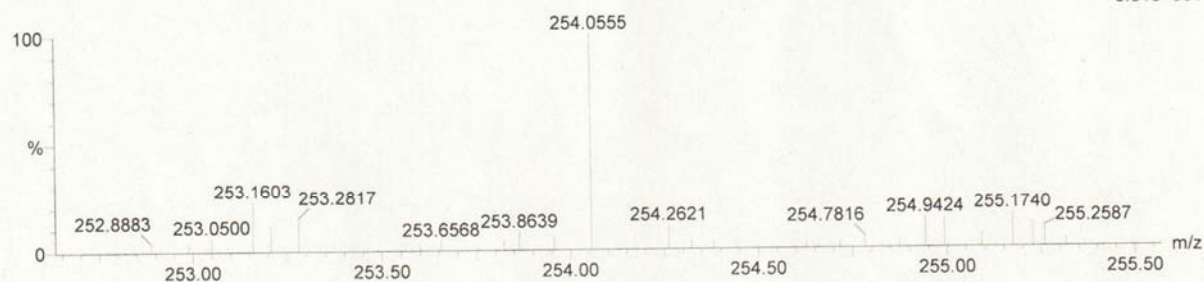
4 formula(e) evaluated with 1 results within limits (up to 50 best isotopic matches for each mass)

Elements Used:

C: 11-11 H: 6-7 N: 3-3 O: 1-1 F: 3-3 Na: 0-1 Cl: 0-1

20220422_2 15 (0.524)

1: TOF MS ES+

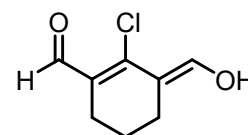


Minimum:

Maximum: 50.0 80.0 -1.5 100.0

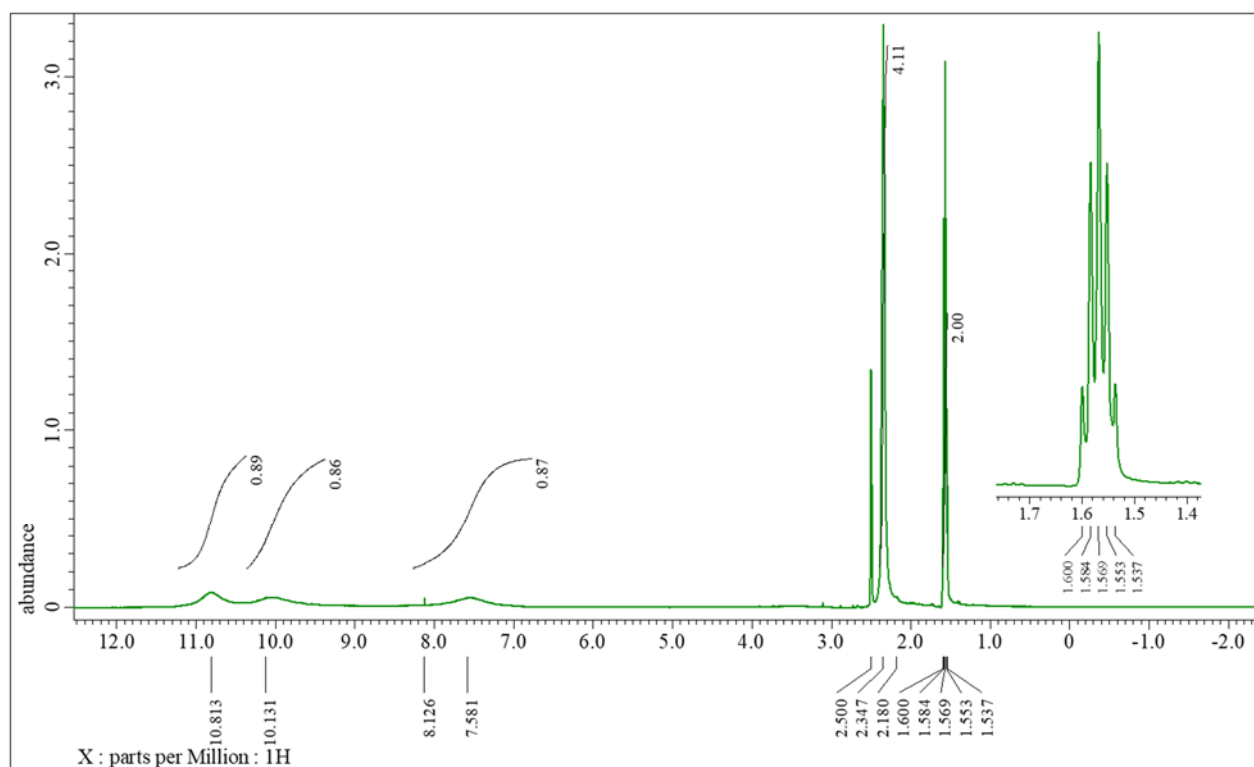
Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
254.0555	254.0541	1.4	5.5	8.5	67.1	0.0	C11 H7 N3 O F3

(E)-2-Chloro-3-(hydroxymethylene)cyclohex-1-ene-1-carbaldehyde (3) [2]

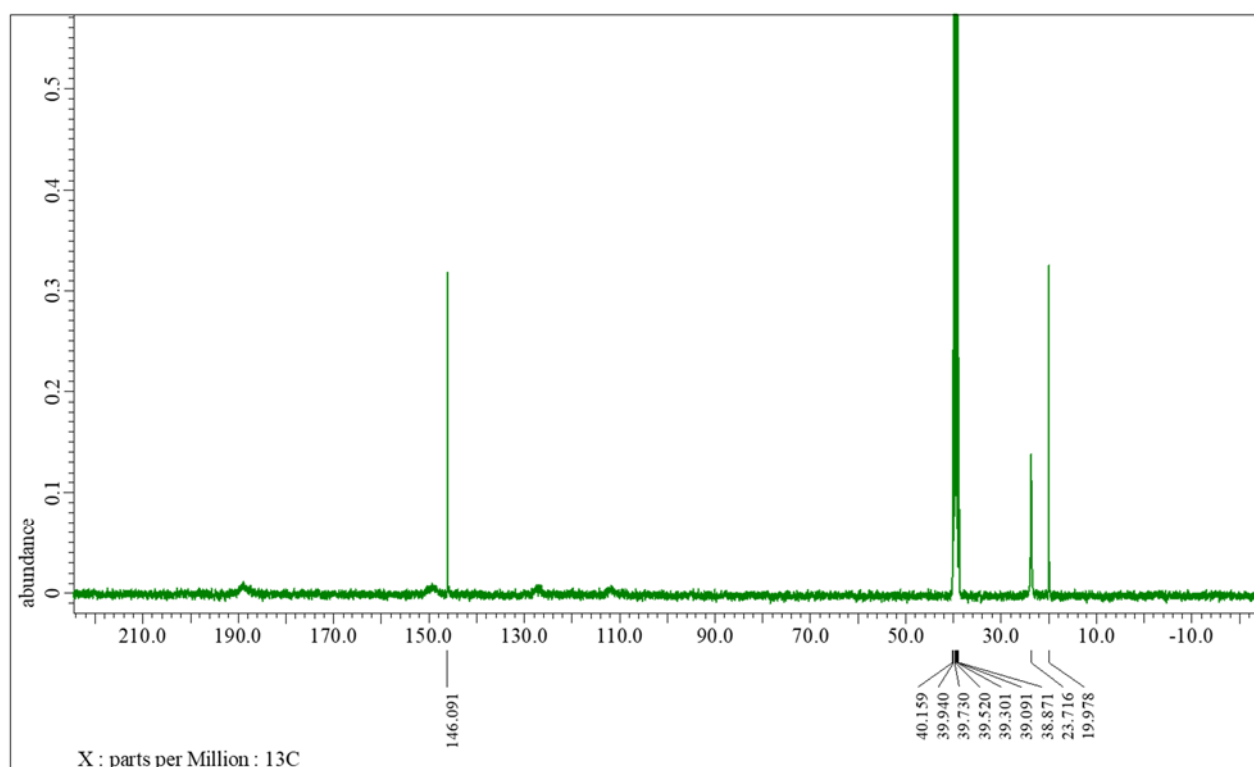


3

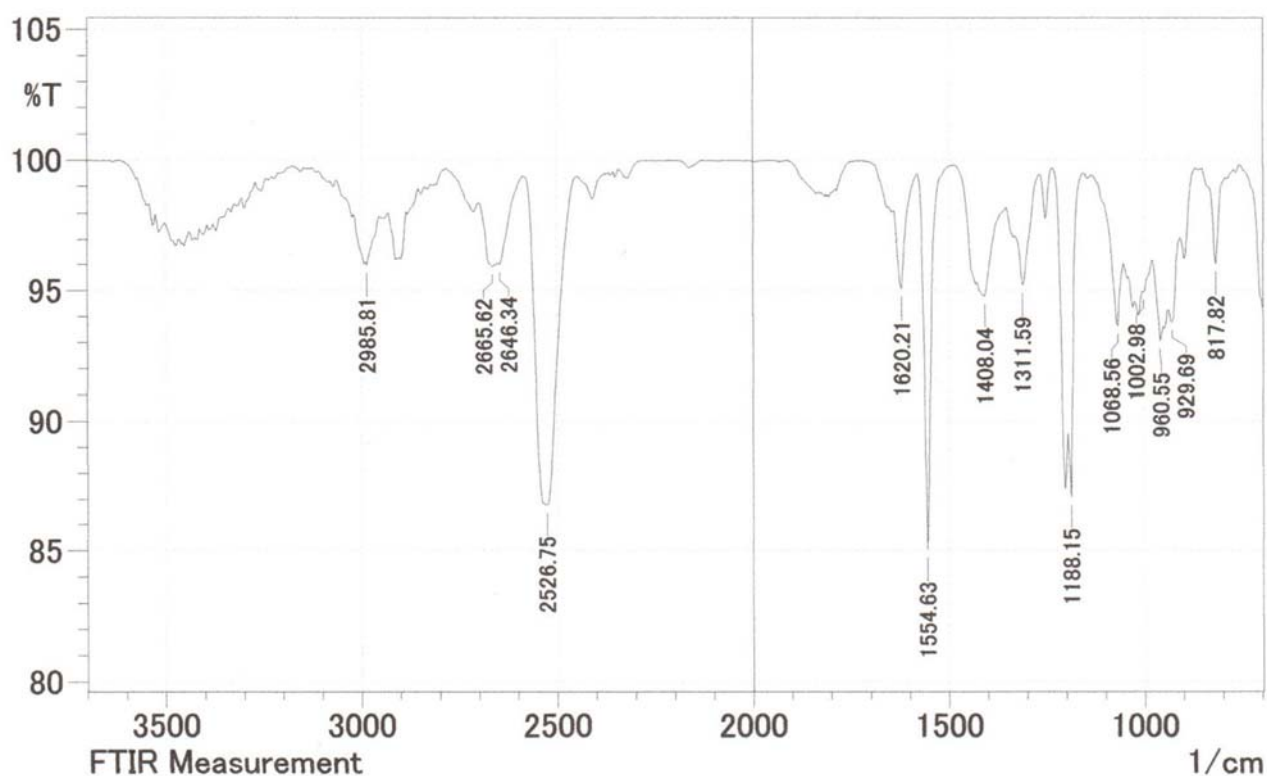
¹H NMR



¹³C NMR



IR



HRMS

Elemental Composition Report

Page 1

Single Mass Analysis

Tolerance = 50.0 mDa / DBE: min = -1.5, max = 100.0

Element prediction: Off

Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions

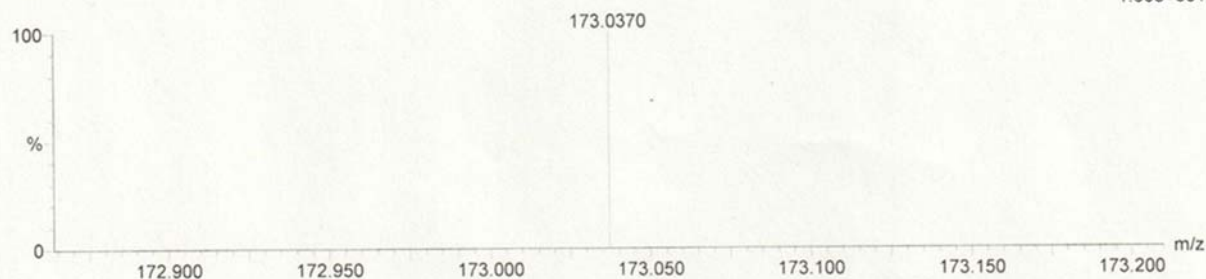
2 formula(e) evaluated with 1 results within limits (up to 50 best isotopic matches for each mass)

Elements Used:

C: 8-8 H: 9-10 O: 2-2 Na: 0-1 Cl: 1-1

20230422_8 8 (0.284)

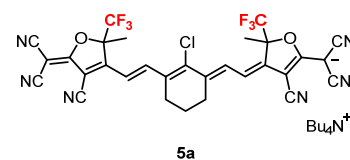
1: TOF MS ES+



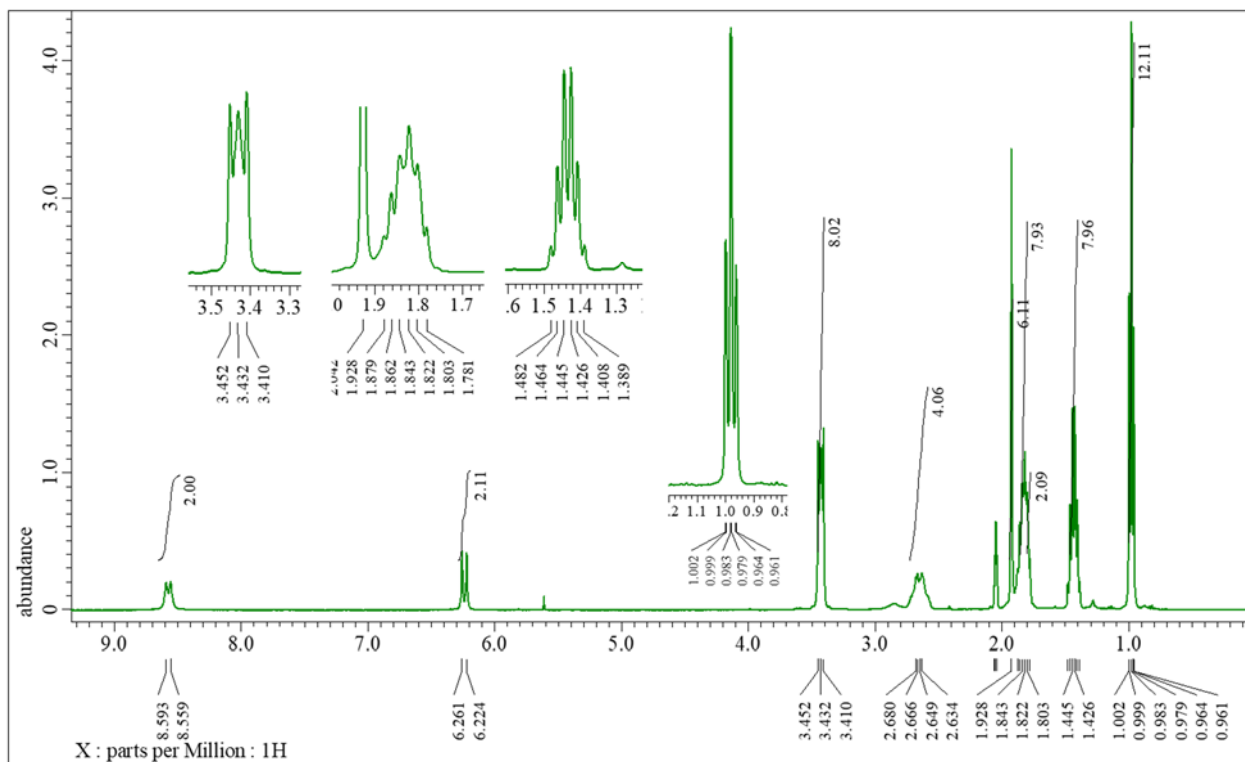
Minimum: -1.5
Maximum: 50.0 80.0 100.0

Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
173.0370	173.0369	0.1	0.6	3.5	11.6	0.0	C8 H10 O2 Cl

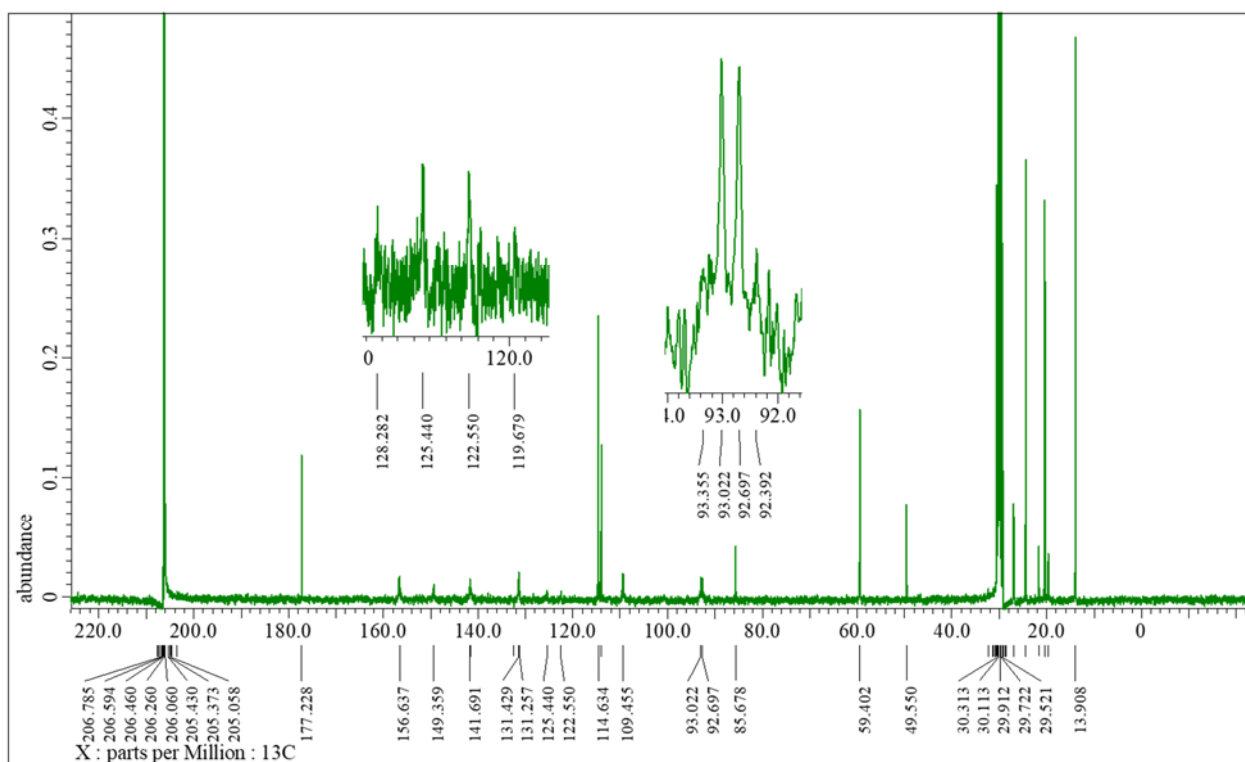
Tetrabutylammonium ((Z)-4-((E)-2-(2-chloro-3-((E)-2-(4-cyano-5-(dicyanomethylene)-2-methyl-2-(trifluoromethyl)-2,5-dihydrofuran-3-yl)vinyl)cyclohex-2-en-1-ylidene)ethylidene)-3-cyano-5-methyl-5-(trifluoromethyl)-4,5-dihydrofuran-2-yl)dicyanomethanide (5a)



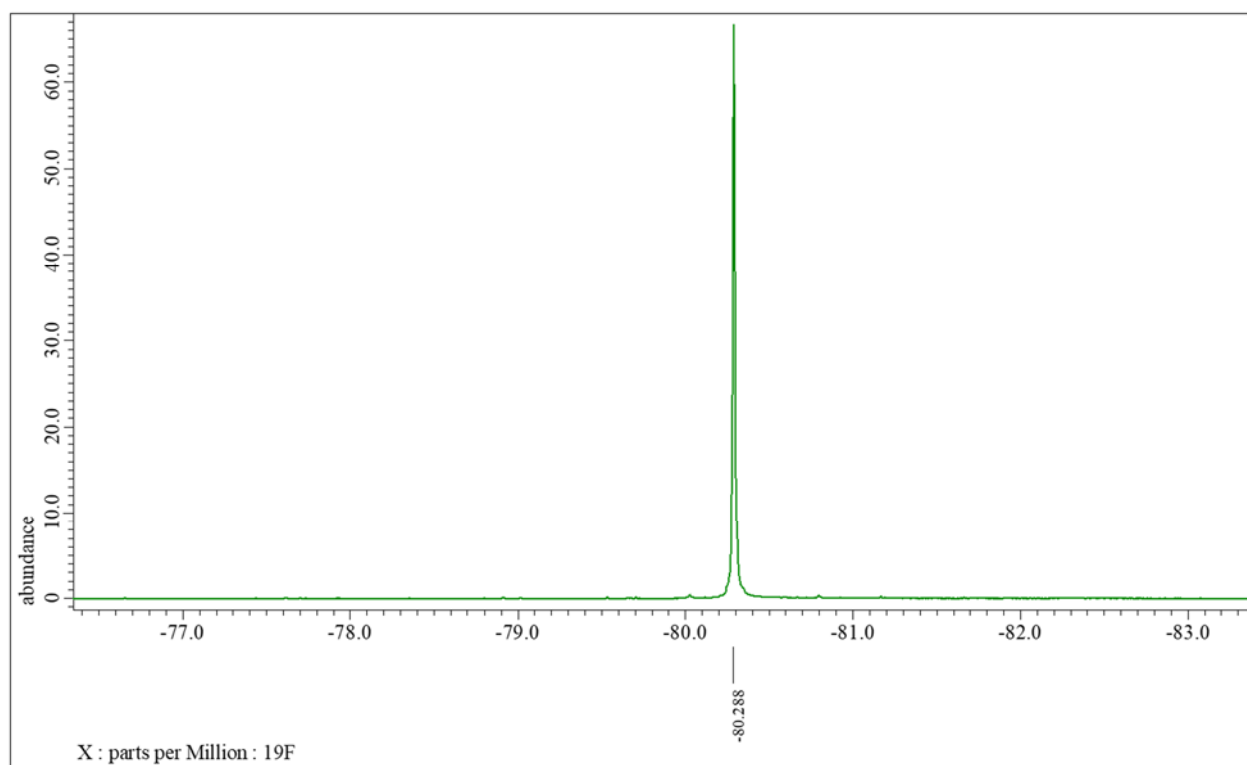
^1H NMR



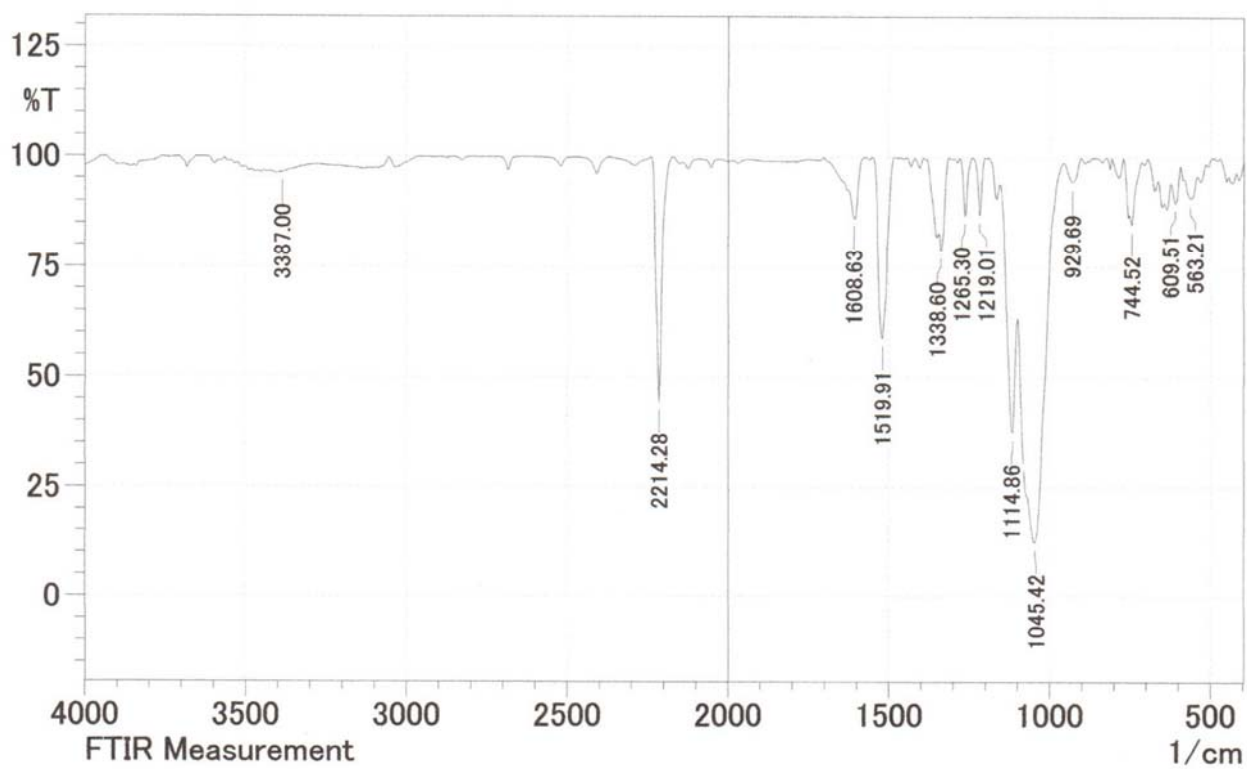
^{13}C NMR



^{19}F NMR



IR



Elemental Composition Report

Page 1

Single Mass Analysis

Tolerance = 50.0 mDa / DBE: min = -1.5, max = 100.0

Element prediction: Off

Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions

2 formula(e) evaluated with 1 results within limits (up to 50 best isotopic matches for each mass)

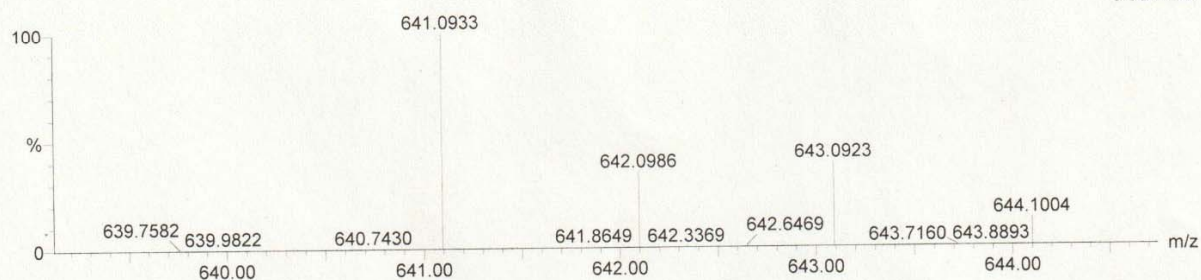
Elements Used:

C: 30-30 H: 16-16 N: 6-6 O: 2-2 F: 6-6 Na: 0-1 Cl: 1-1

20230422_4-1 13 (0.455)

1: TOF MS ES-

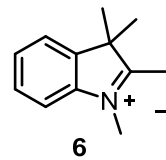
3.93e+003



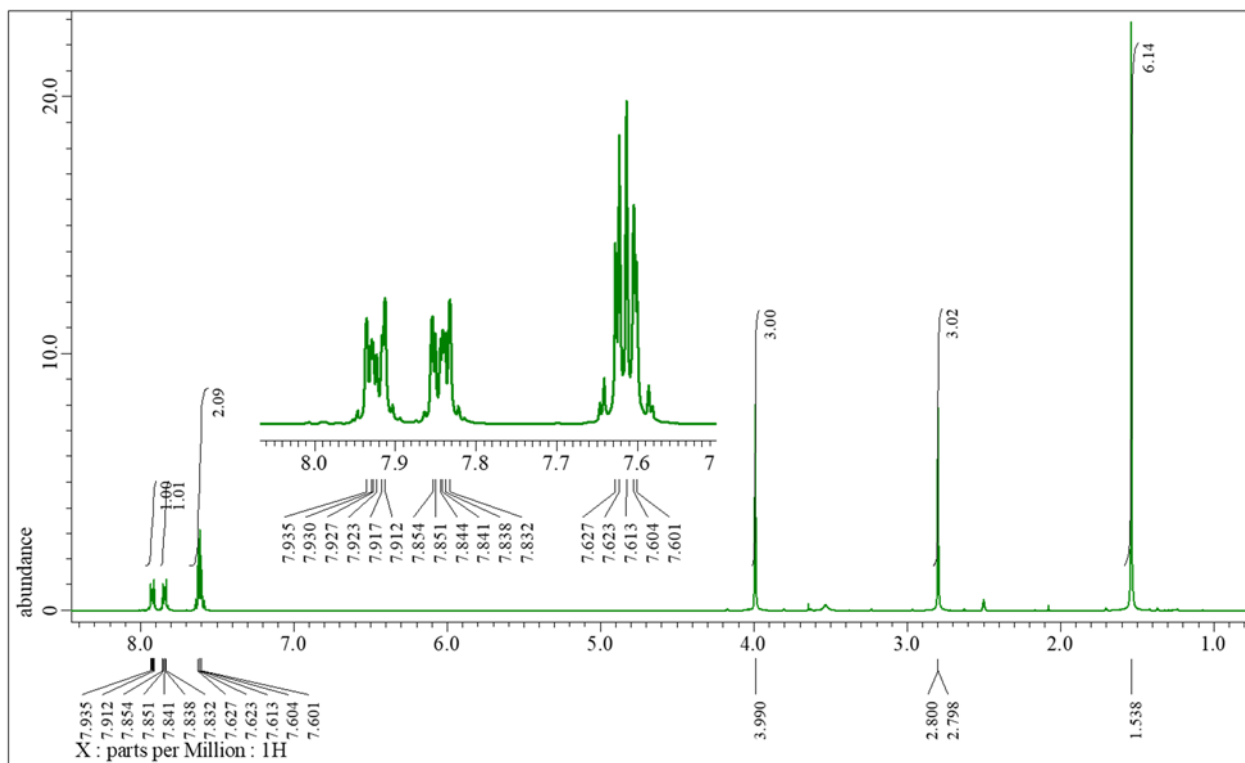
Minimum: -1.5
Maximum: 50.0 80.0 100.0

Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
641.0933	641.0927	0.6	0.9	22.5	32.8	0.0	C30 H16 N6 O2 F6 Cl

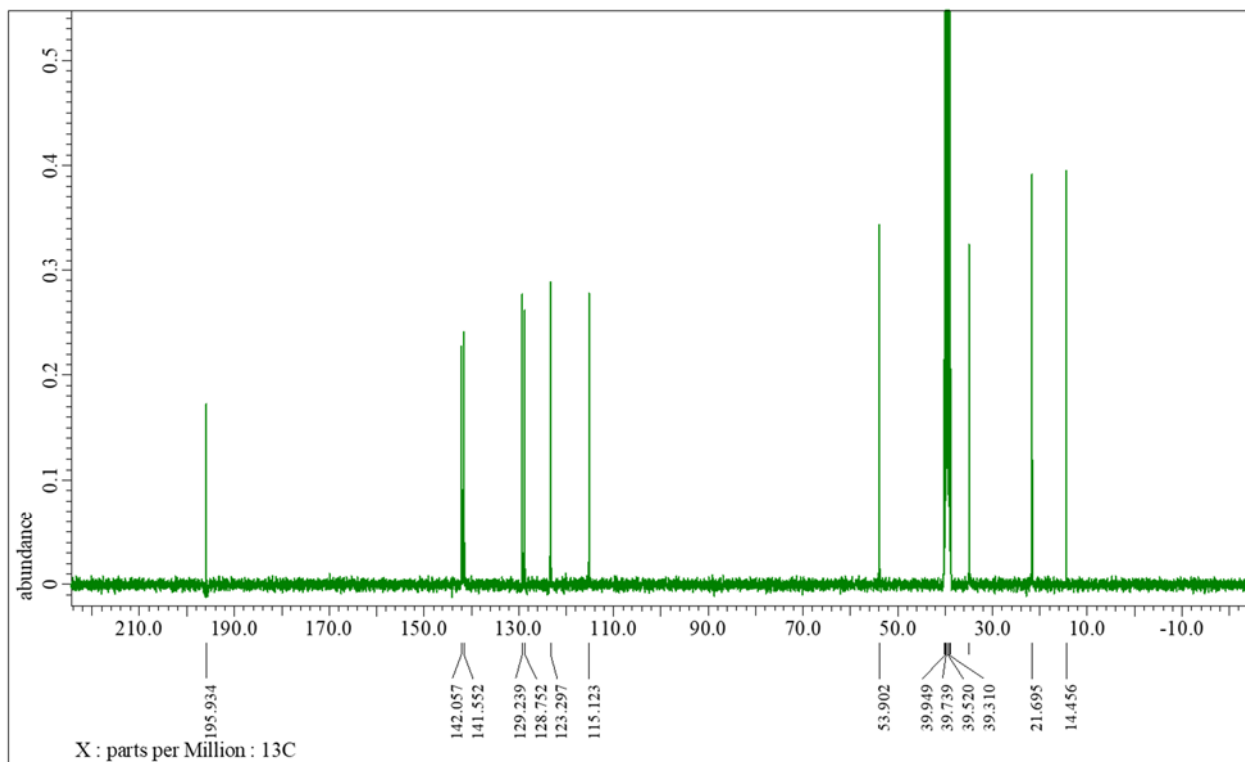
1, 2, 3, 3-tetramethyl-3*H*-indol-1-ium iodide (6) [3]



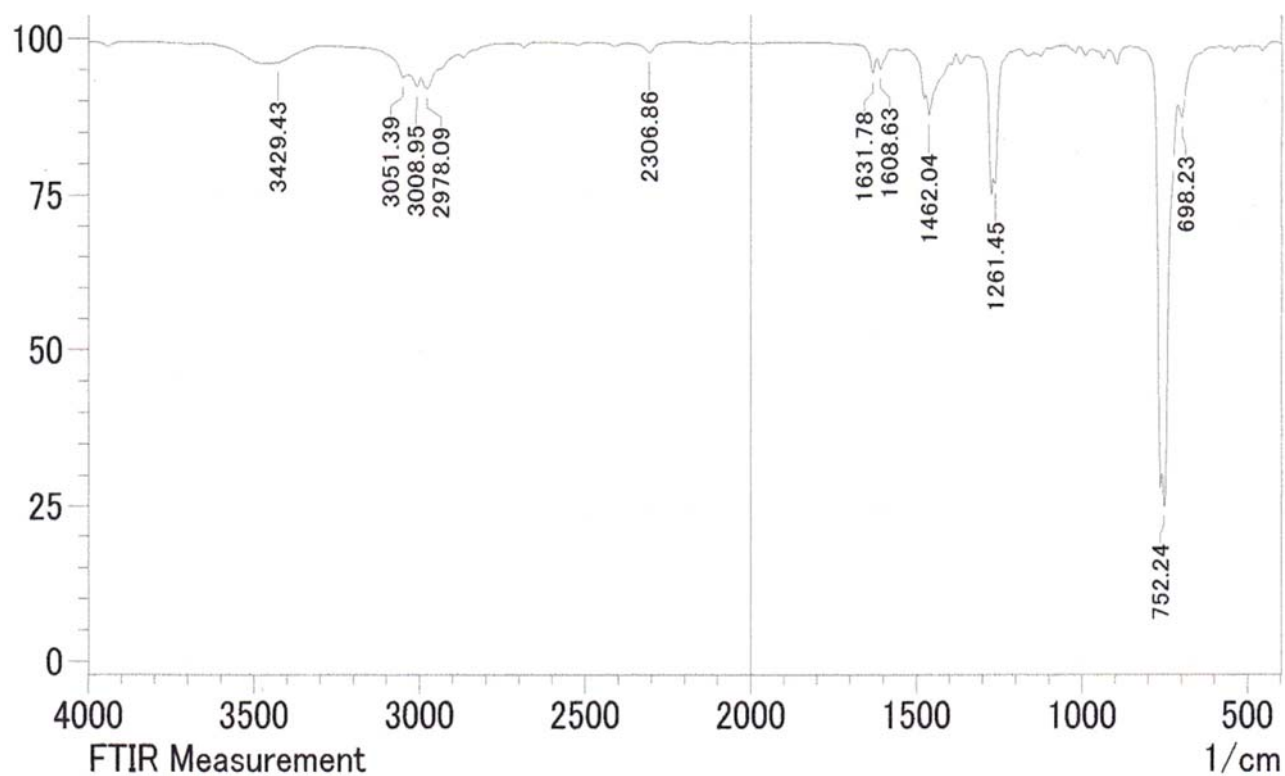
^1H NMR



^{13}C NMR



IR



HRMS

Monoisotopic Mass, Even Electron Ions

5 formula(e) evaluated with 1 results within limits (up to 50 closest results for each mass)

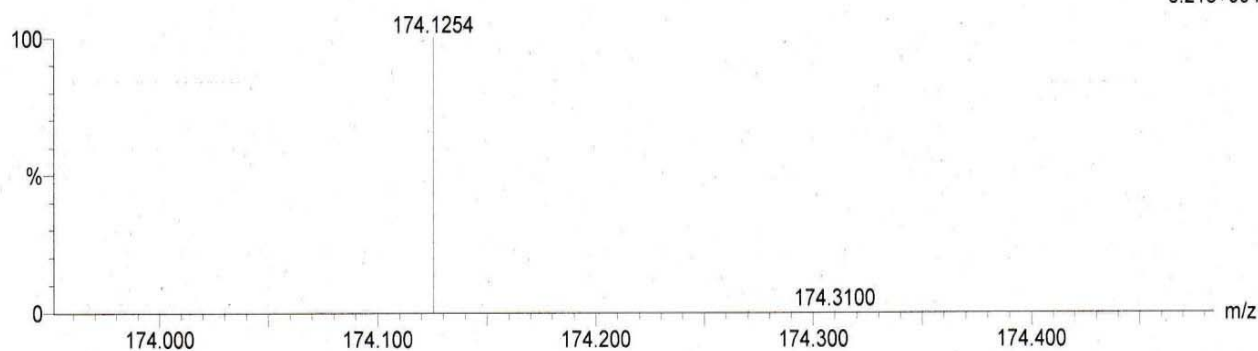
Elements Used:

C: 12-12 H: 16-16 N: 1-1 Na: 0-1 Se: 0-2

20211220_7 7 (0.250)

1: TOF MS ES+

6.21e+004



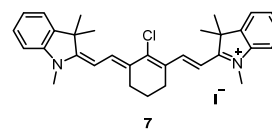
Minimum: -1.5
Maximum: 1000.0 10.0 50.0

Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
174.1254	174.1283	-2.9	-16.7	5.5	42.7	0.0	C12 H16 N

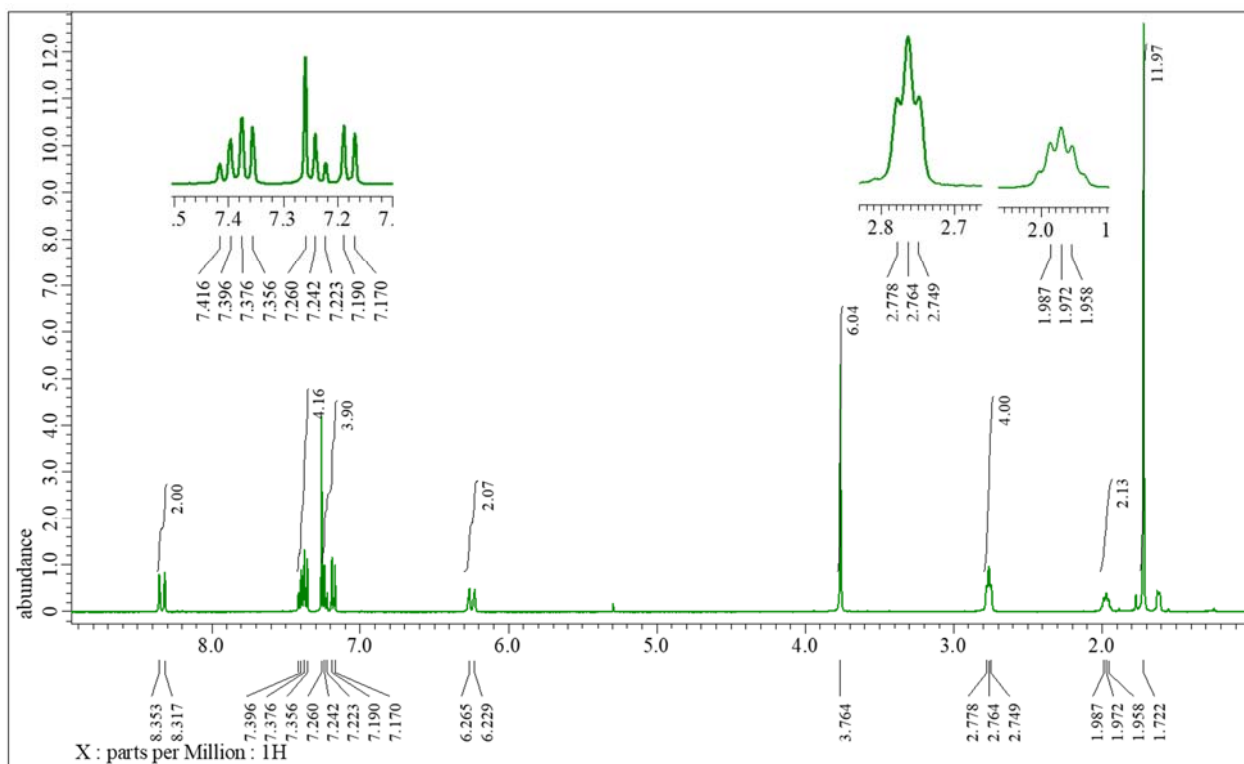
2-((*E*)-2-((*E*)-2-chloro-3-(2-((*E*)-1,3,3-trimethylindolin-2-

ylidene)ethylidene)cyclohex-1-en-1-yl)vinyl)-1,3,3-trimethyl-3*H*-indol-1-

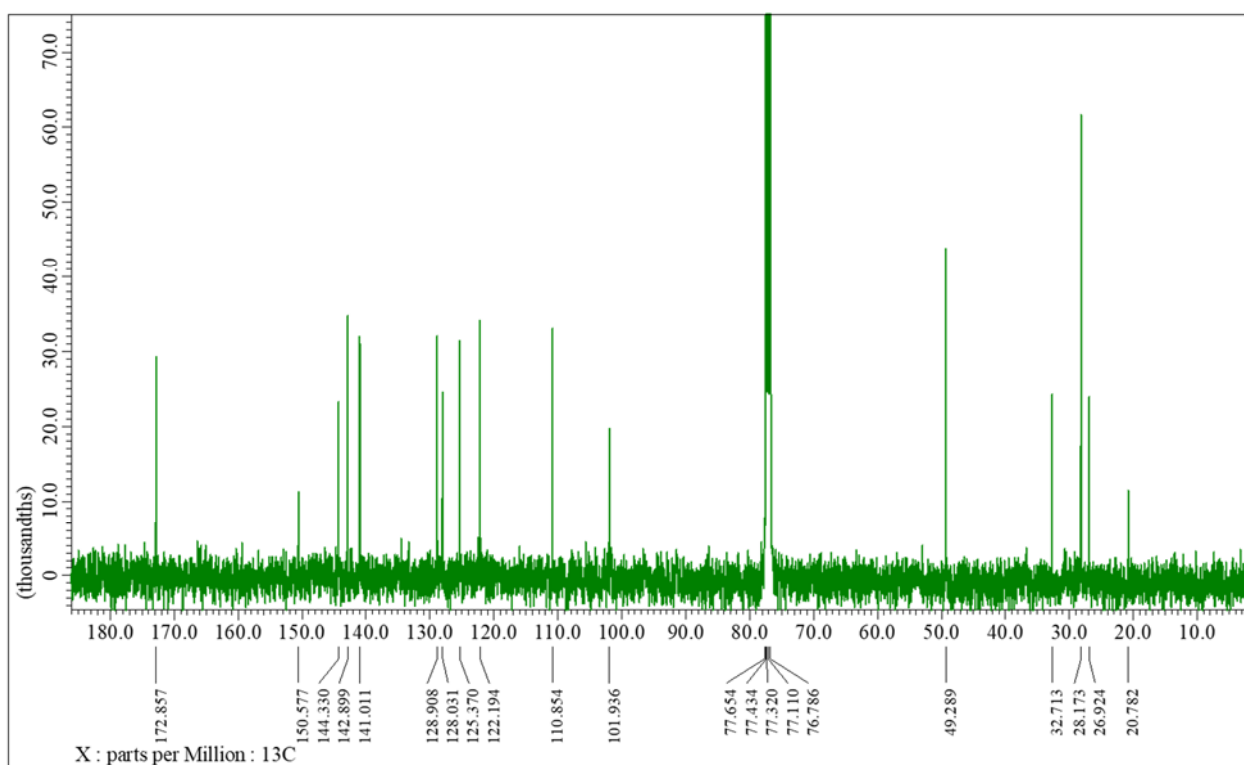
ium iodide (7). [4]



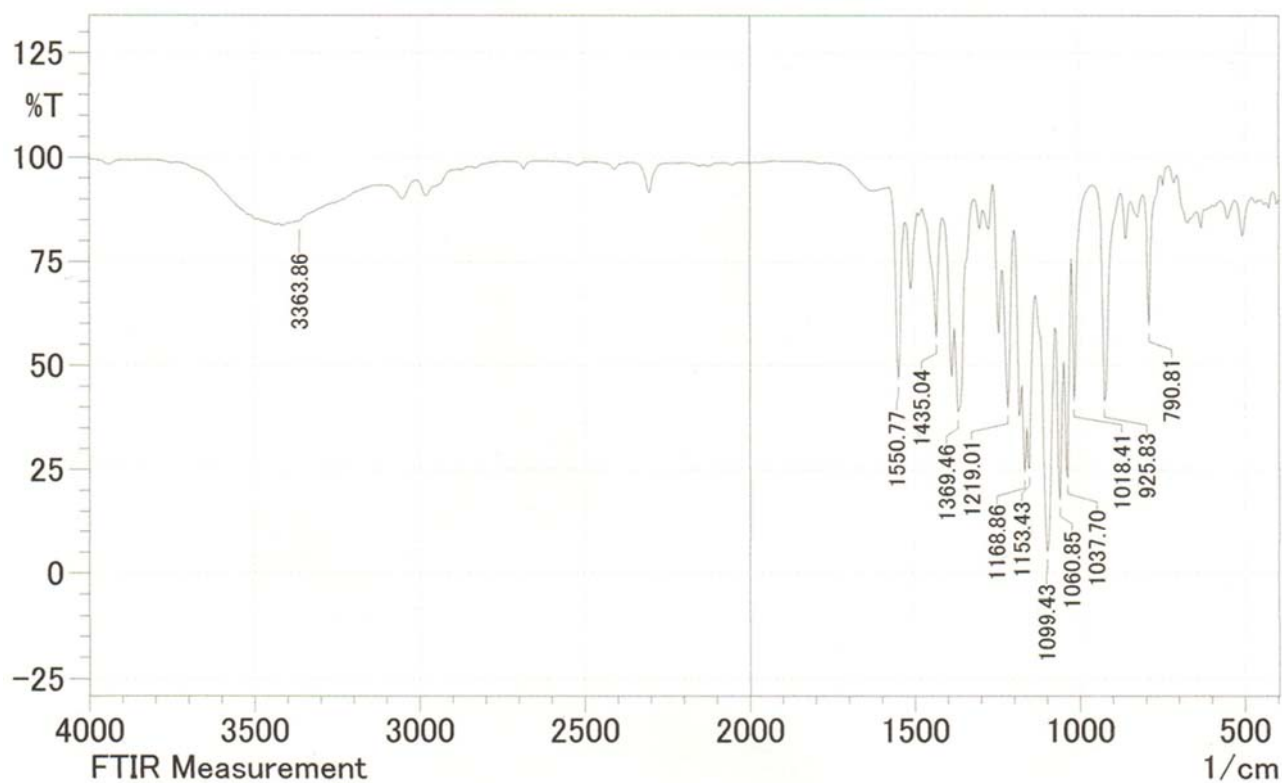
¹H NMR



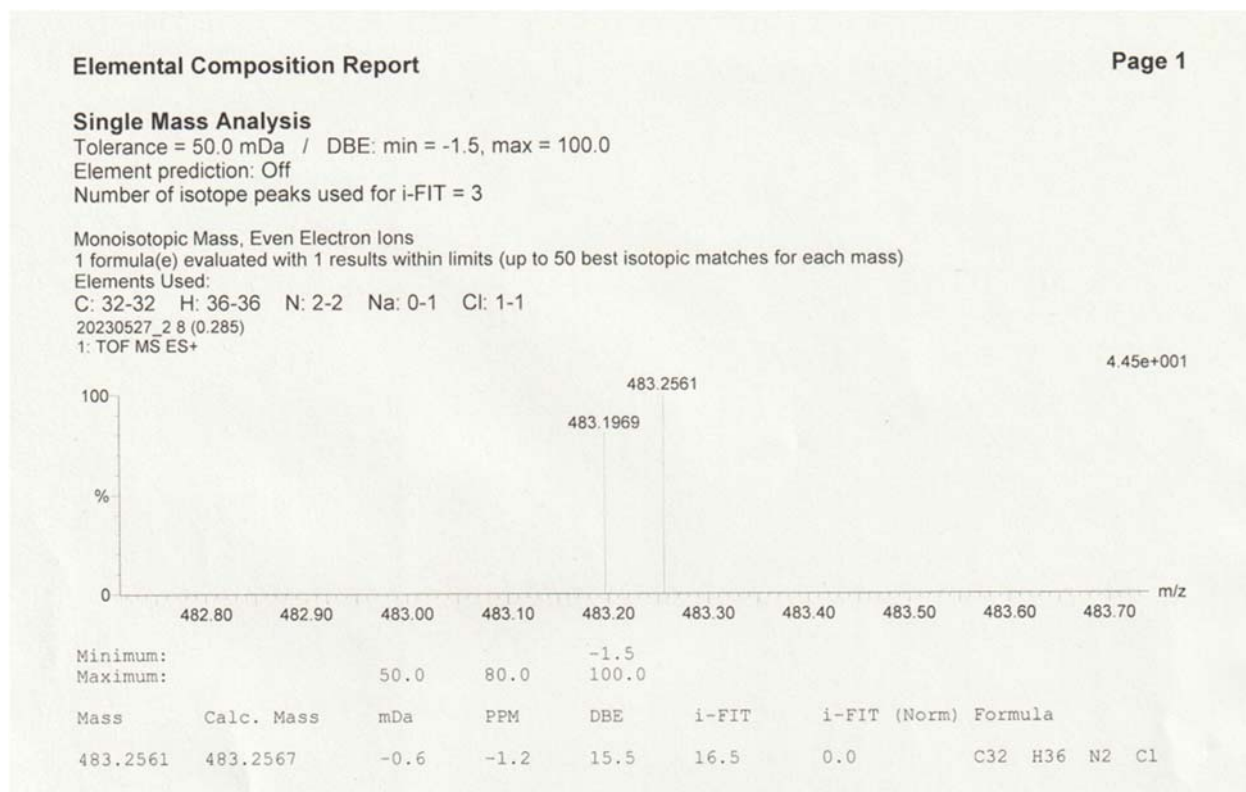
¹³C NMR



IR



HRMS



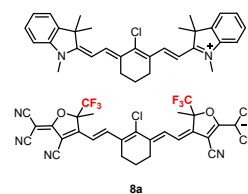
2-((*E*)-2-((*E*)-2-Chloro-3-(2-((*E*)-1,3,3-trimethylindolin-2-

ylidene)ethylidene)cyclohex-1-en-1-yl)vinyl)-1,3,3-trimethyl-3H-indol-1-

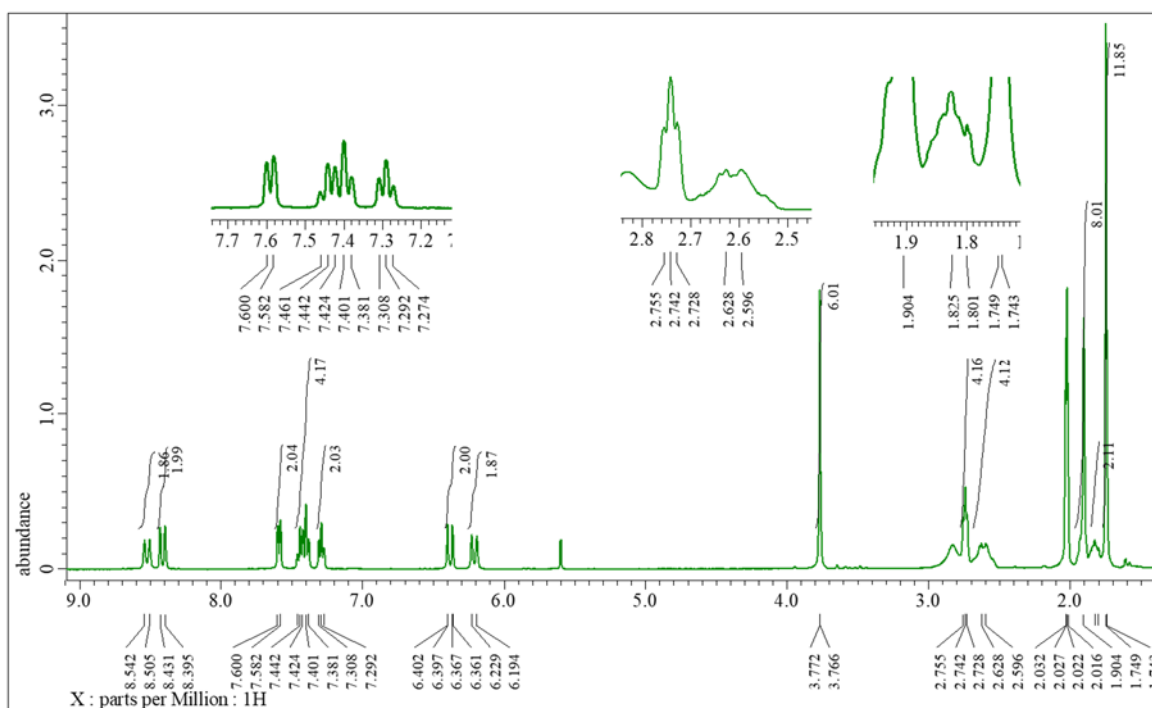
ium ((*Z*)-4-((*E*)-2-(2-chloro-3-((*E*)-2-(4-cyano-5-(dicyanomethylene)-2-

methyl-2-(trifluoromethyl)-2,5-dihydrofuran-3-yl)vinyl)cyclohex-2-en-1-ylidene)ethylidene)-3-

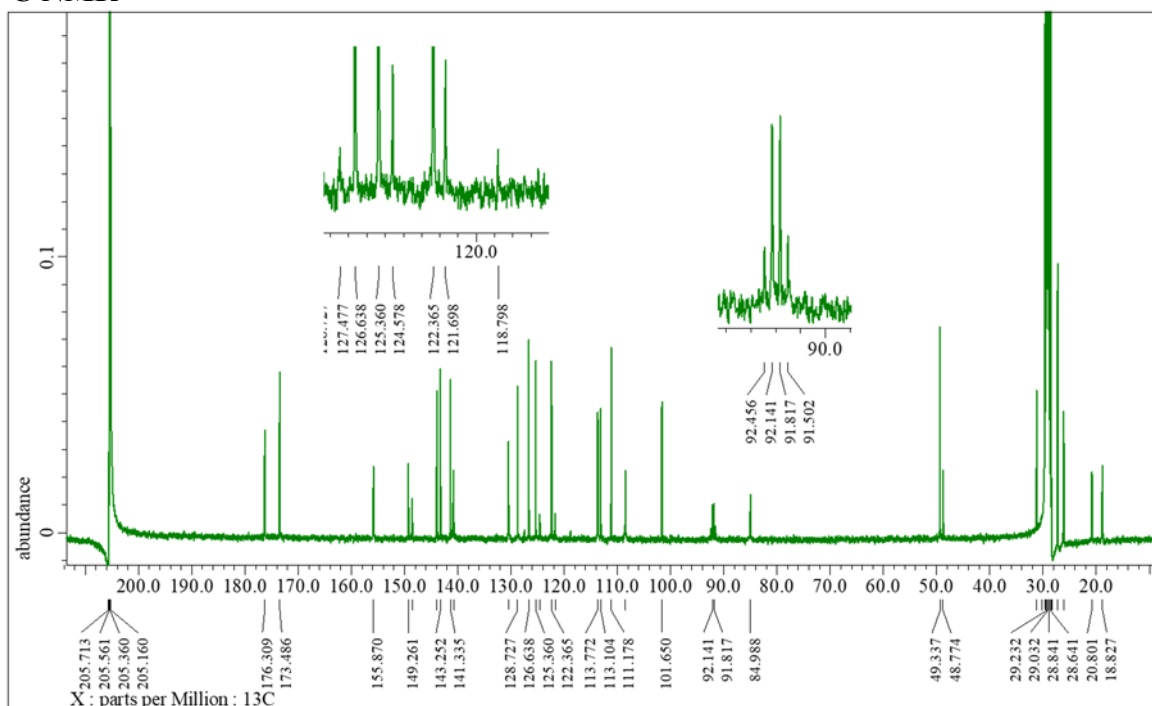
cyano-5-methyl-5-(trifluoromethyl)-4,5-dihydrofuran-2-yl)dicyanomethanide (8a)



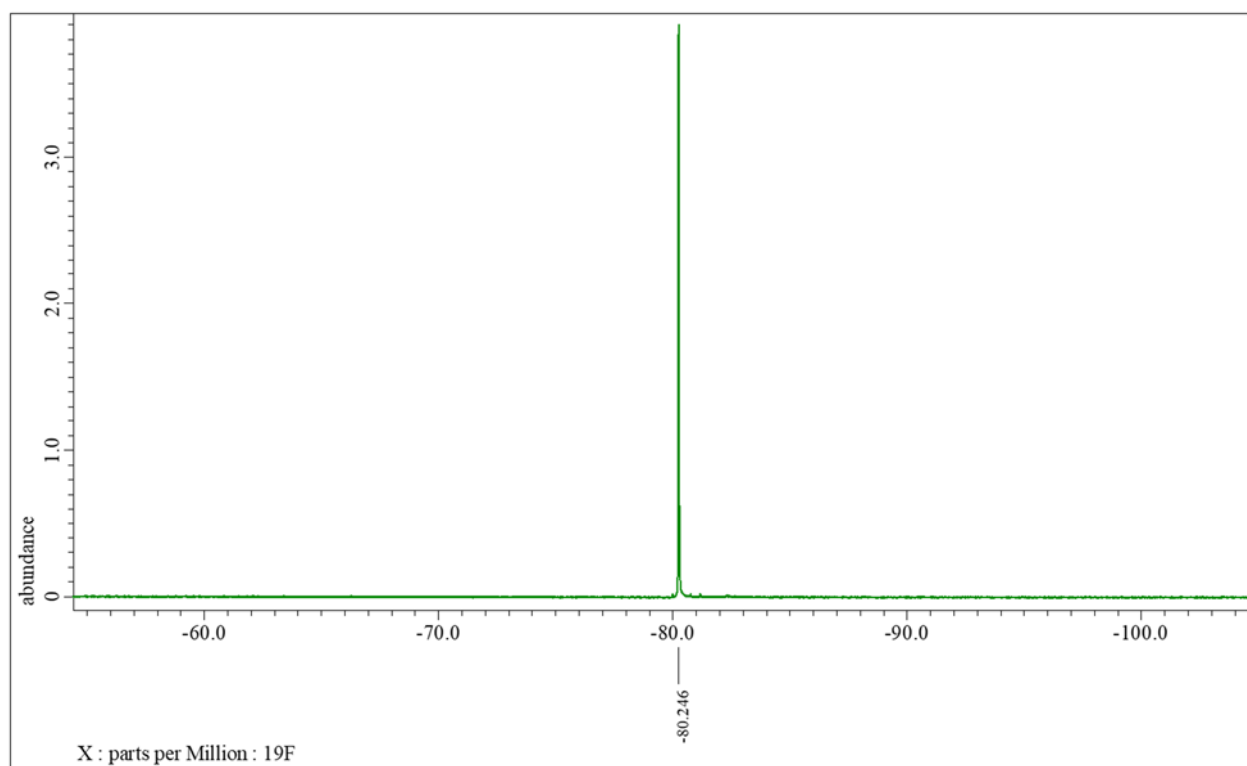
¹H NMR



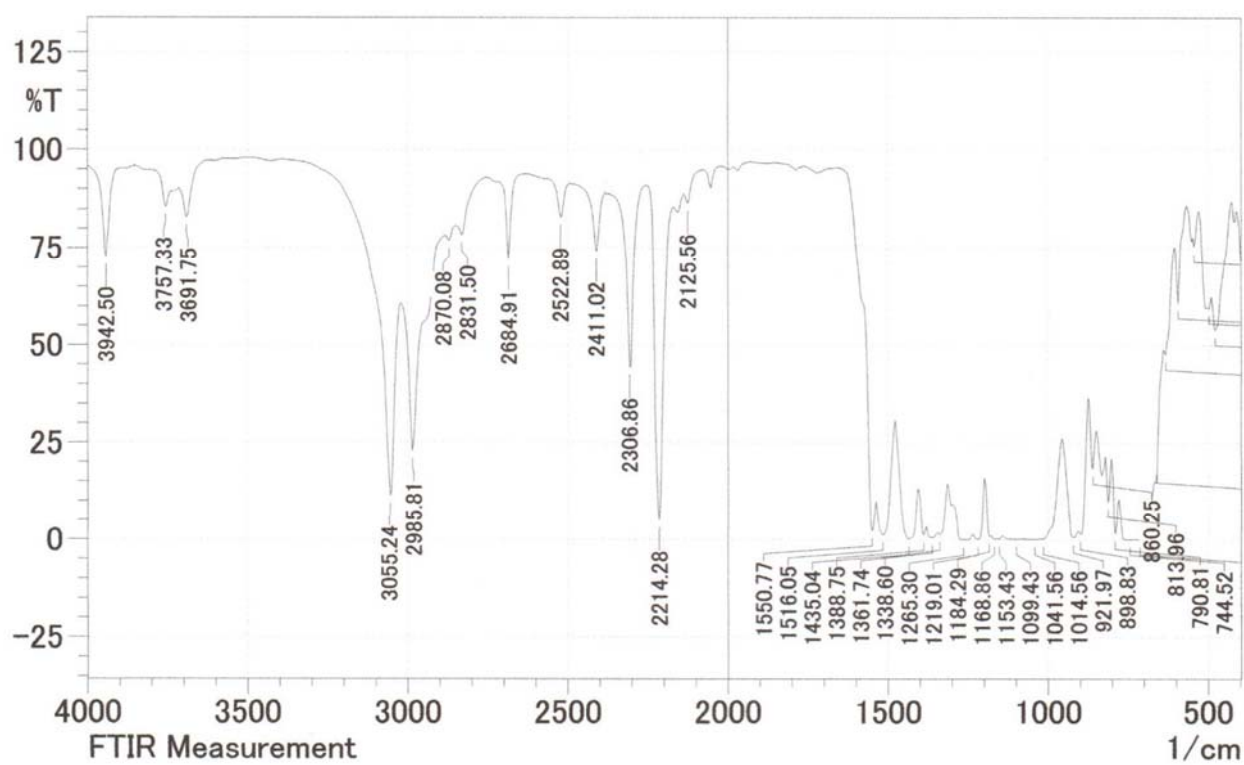
¹³C NMR



^{19}F NMR



IR



Elemental Composition Report

Page 1

Single Mass Analysis

Tolerance = 30.0 mDa / DBE: min = -1.5, max = 50.0

Element prediction: Off

Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions

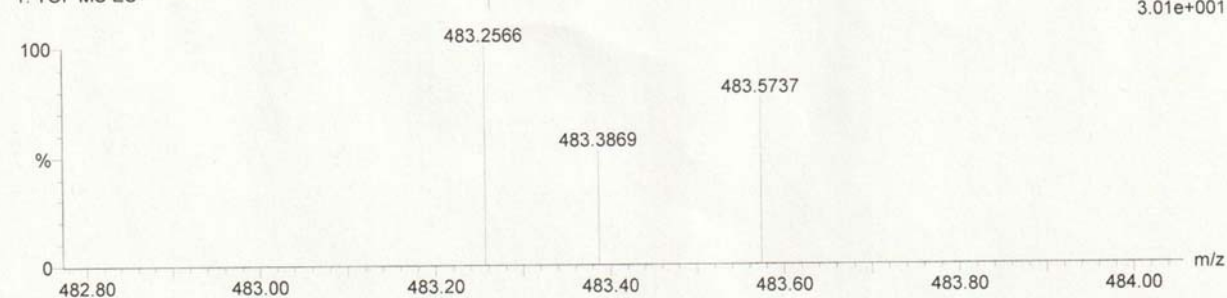
1 formula(e) evaluated with 1 results within limits (up to 50 best isotopic matches for each mass)

Elements Used:

C: 32-32 H: 36-36 N: 2-2 Na: 0-1 Cl: 1-1

20230422_6 23 (0.808)

1: TOF MS ES+



Minimum:				-1.5			
Maximum:	30.0	80.0	50.0				
Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
483.2566	483.2567	-0.1	-0.2	15.5	27.4	0.0	C32 H36 N2 Cl

Elemental Composition Report

Page 1

Single Mass Analysis

Tolerance = 50.0 mDa / DBE: min = -1.5, max = 100.0

Element prediction: Off

Number of isotope peaks used for i-FIT = 3

Monoisotopic Mass, Even Electron Ions

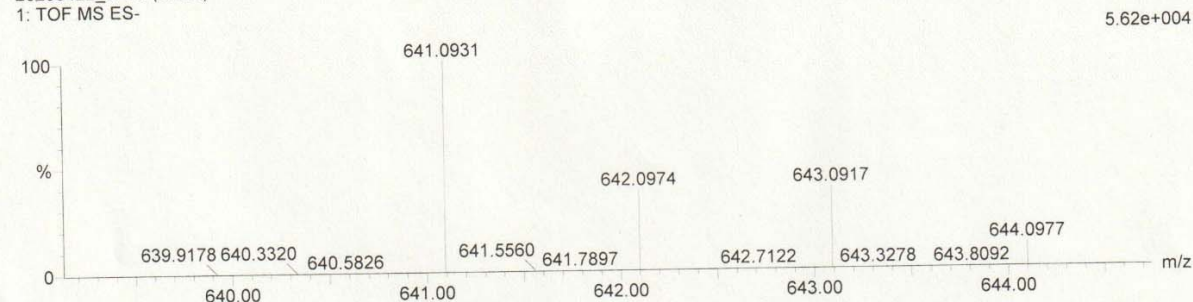
2 formula(e) evaluated with 1 results within limits (up to 50 best isotopic matches for each mass)

Elements Used:

C: 30-30 H: 16-16 N: 6-6 O: 2-2 F: 6-6 Na: 0-1 Cl: 1-1

20230422_5-1 8 (0.284)

1: TOF MS ES-



Minimum:				-1.5			
Maximum:	50.0	80.0	100.0				
Mass	Calc. Mass	mDa	PPM	DBE	i-FIT	i-FIT (Norm)	Formula
641.0931	641.0927	0.4	0.6	22.5	73.6	0.0	C30 H16 N6 O2 F6 Cl

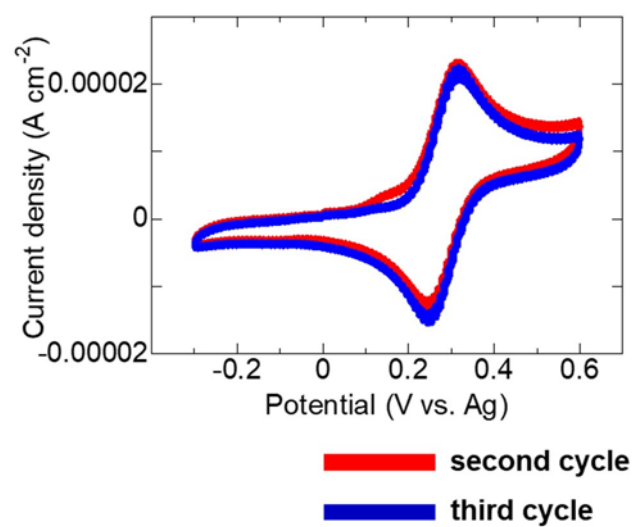


Figure S1. Cyclic voltammograms of the prepared anionic HMC dyes **5a** (1.0×10^{-3} M) in MeCN containing Bu₄NClO₄ (0.1 M) as the supporting electrolyte; scan rate of 200 mV s⁻¹

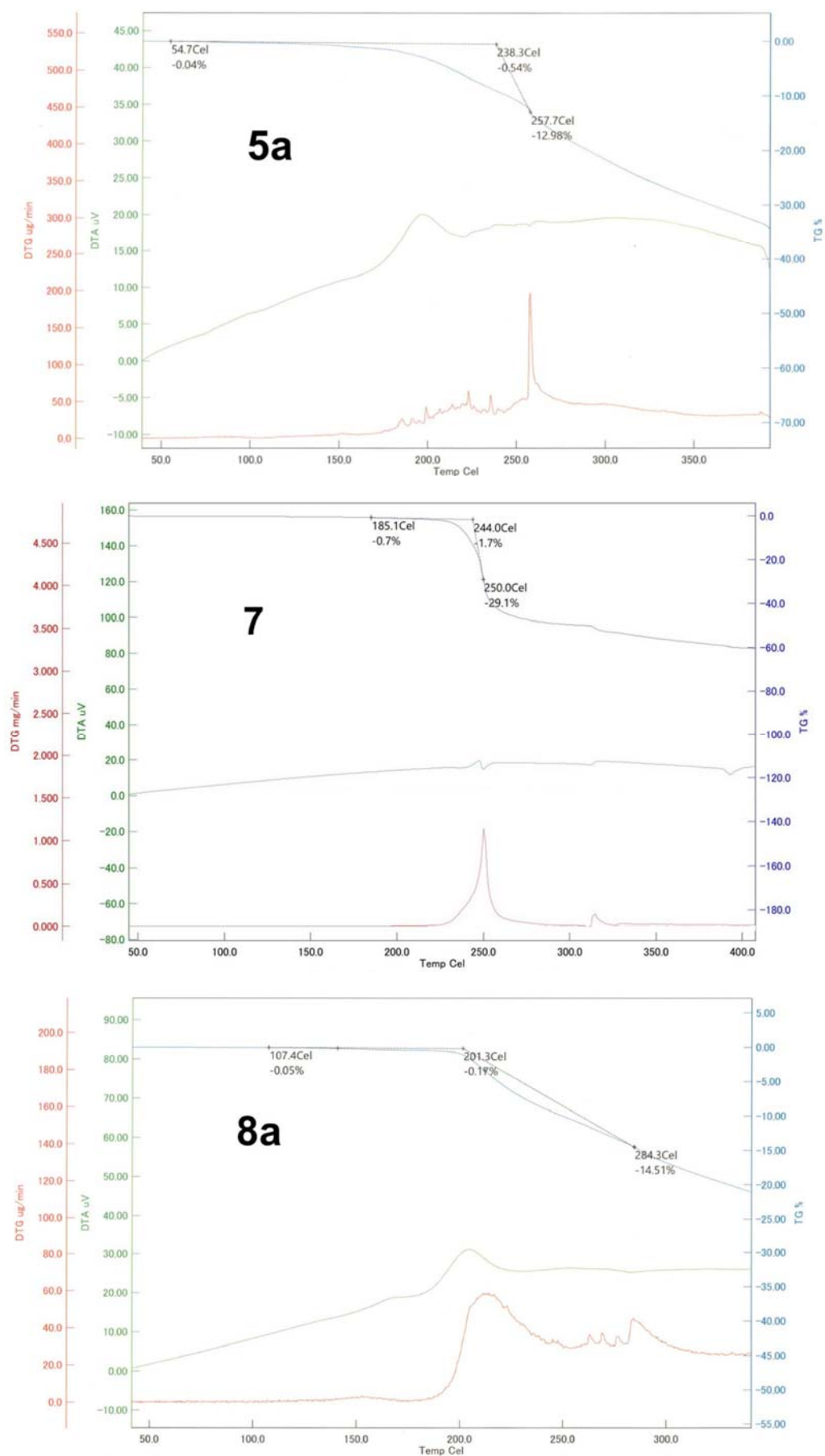


Figure S2. Results of TG-DTA measurements for **5a** and **7a**

References

1. Liu, S.; Haller, M.A.; Ma, H.; Dalton, L.R.; Jang, S.-H.; Jen, A. K.-Y. Focused Microwave-Assisted Synthesis of 2,5-Dihydrofuran Derivatives as Electron Acceptors for Highly Efficient Nonlinear Optical Chromophores. *Adv. Mater.*, **2003**, *15*, 603–607.
2. Mukherjee, A.; Saha, P. C.; Das, R. S.; Bera, T.; Guha, S. Acidic pH-Activatable Visible to Near-Infrared Switchable Ratiometric Fluorescent Probe for Live-Cell Lysosome Targeted Imaging. *ACS Sens.*, **2021**, *6*, 2141–2146.
3. Burdette, M. K.; Jenkins R.; Bandera, Y. P.; Jones, H.; Foulger, I. K.; Dickey, A.; Nieminen, A-L.; Foulger, S. H. Click-Engineered, Bioresponsive, and Versatile Particle–Protein–Dye System. *ACS Appl. Bio Mater.*, **2019**, *2*, 8, 3183– 3193.
4. Shibayama, M.; Uehashi, Y.; Ajioka, S.; Kubota, Y.; Inuzuka, T.; Funabiki, K. Vapochromism of indolenine-based heptamethine cyanine dye adsorbed on silica gel. *New J. Chem.*, **2023**, *47*, 5262- 5269.