Supporting Information

Solution-processable Homoleptic Aluminum(III) Catecholaldimine Complex as an Active Material for RRAM Switching Device

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Contents

Table S1. Crystal data and structure refinement for Complex_1.

Identification code	Complex_**1**
Empirical formula	$C_{40}H_{49}AlN_4O_8 \cdot CH_3OH \cdot 1.5\ H_2O$
Formula weight	799.38
Temperature/K	296.15
Crystal system	triclinic
Space group	P-1
a/Å	9.8115(7)
b/Å	11.8924(8)
c/Å	18.6058(13)
α/°	79.636(2)
β/°	84.703(2)
γ/°	85.766(2)
Volume/Å^3	2122.8(3)
Z	2
ρ_{calc}g/cm^3	1.159
μ/mm^{-1}	0.100
F(000)	788.0
Crystal size/mm^3	0.2 × 0.2 × 0.2
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	3.8 to 56.818
Index ranges	$-13 \leq h \leq 13$, $-15 \leq k \leq 15$, $-24 \leq l \leq 24$
Reflections collected	58132
Independent reflections	10582 [R_{int} = 0.0667, R_{sigma} = 0.0466]
Data/restraints/parameters	10582/0/492
Goodness-of-fit on F^2	1.101
Final R indexes [I>=2σ (I)]	R_1 = 0.0662, wR_2 = 0.1474
Final R indexes [all data]	R_1 = 0.0793, wR_2 = 0.1539
Largest diff. peak/hole / e Å^{-3}	0.73/-0.41

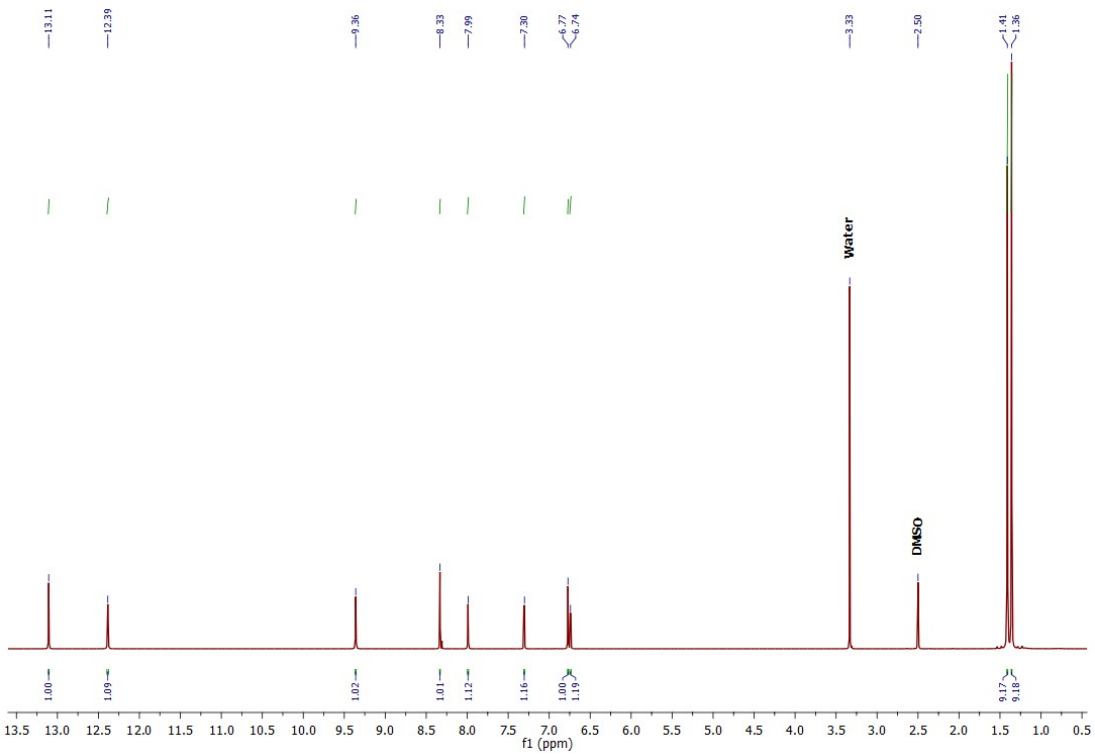


Figure S1. ^{1}H NMR spectrum of Ligand LH_3 in DMSO-d_6.

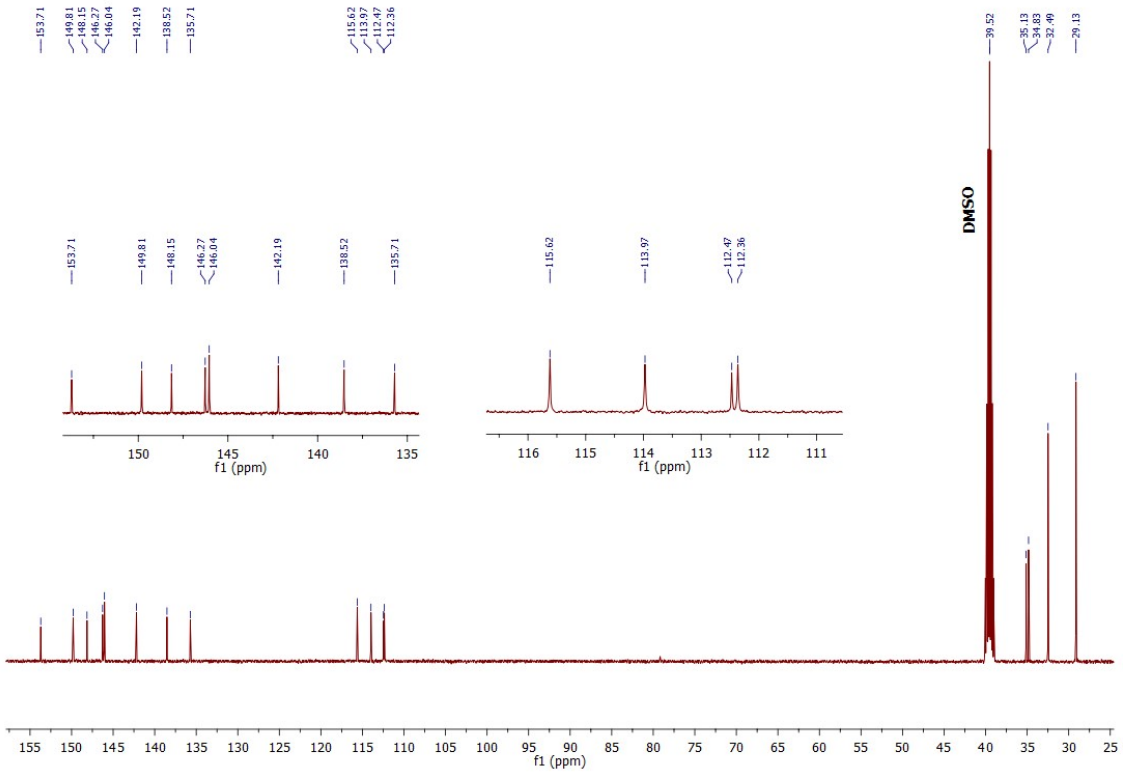


Figure S2. $^{13}C\{^{1}H\}$NMR spectrum of Ligand LH_3 in DMSO-d_6.

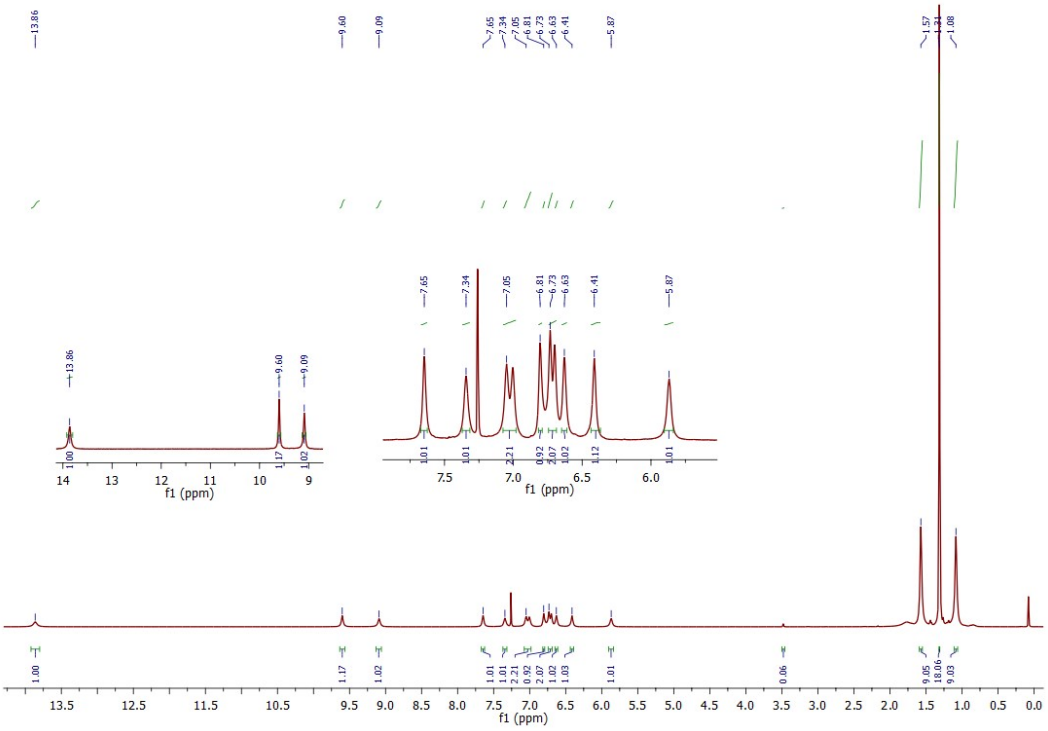


Figure S3. ^{1}H NMR spectrum of Complex **1** in $CDCl_3$**.**

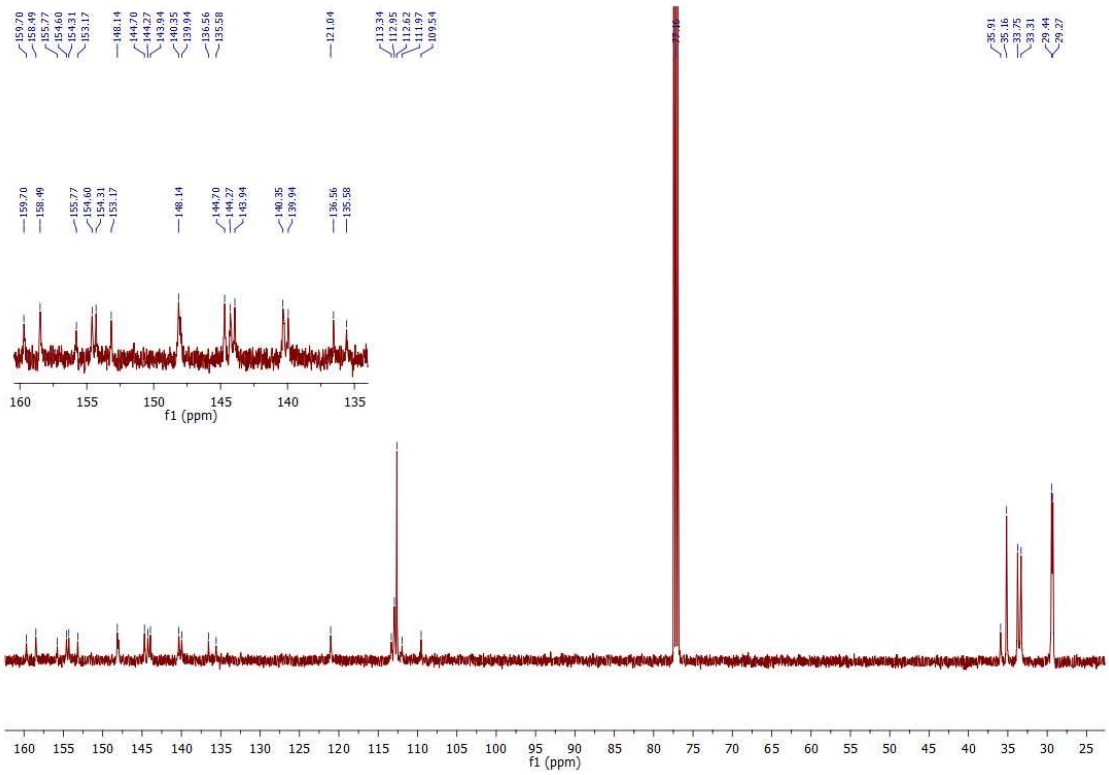


Figure S4. $^{13}C\{^{1}H\}$NMR spectrum of Complex **1** in $CDCl_3$.

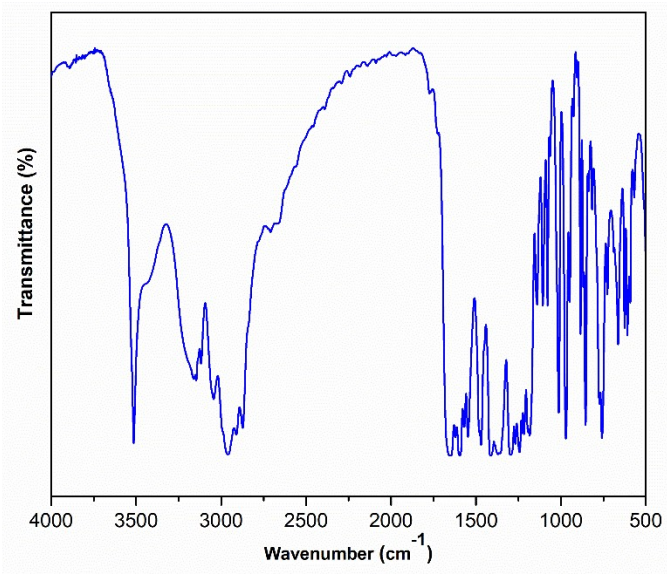


Figure S5. FT-IR Spectrum of Ligand LH_3.

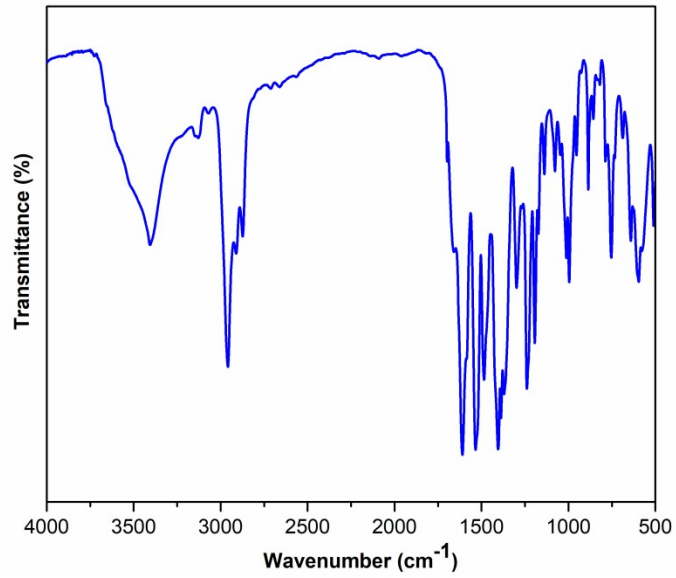


Figure S6. FT-IR Spectrum of Complex **1**.

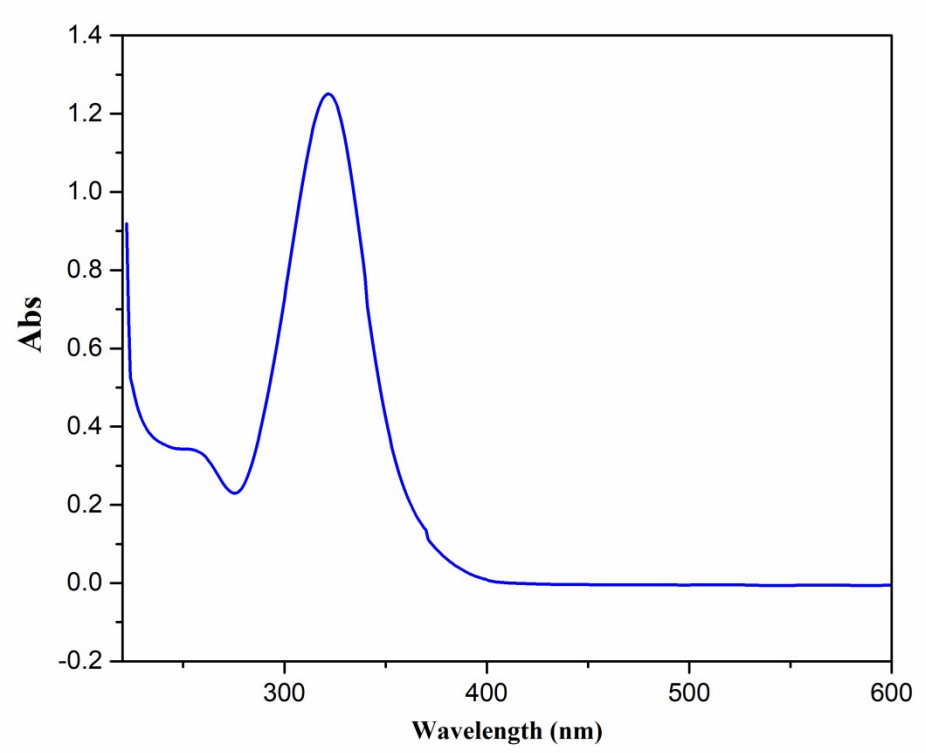


Figure S7. UV-Vis Absorption Spectrum of Ligand LH_3 in CH_2Cl_2 (DCM) Solution.

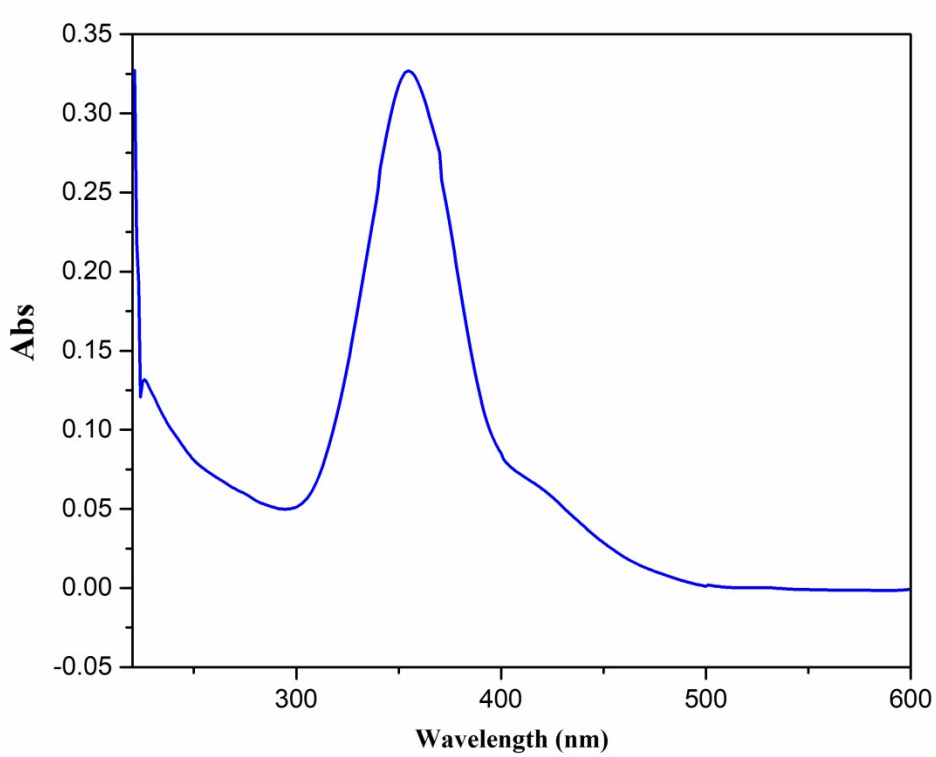


Figure S8. UV-Vis Absorption Spectrum of Complex **1** in CH_2Cl_2 (DCM) Solution.

Calculation of Optical band gap E_{opt} using UV-Vis spectrum of complex 1:

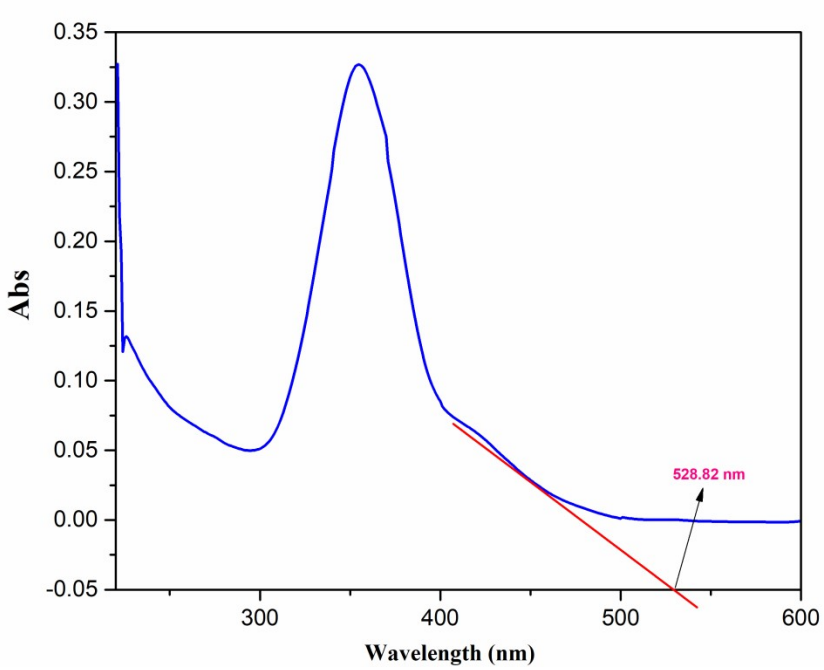


Figure S9. UV-Vis absorption spectrum for the Complex **1** in CH_2Cl_2 (DCM) Solution exhibiting onset optical absorbance.

$$E_{opt} = hc/\lambda_{onset}$$

Where, E_{opt} is the optical band gap of the complex **1**, h is the Planck constant (6.63×10^{-34} m^2kg/s), c is the speed of light (3×10^8 m/s), λ_{onset} is the onset optical absorbance wavelength.

$$E_{opt}\ (eV) = 1240/\lambda_{onset}\ (nm) = 1240/528.82 = 2.34\ eV$$

Calculation of molecular orbital energy levels using electrochemical studies and E_{opt}, optical band gap of complex 1:

$$E_{HOMO} = -[E_{ox,onset} - E_{ox,\,Fc}] - 4.8\ eV$$

$$E_{LUMO} = E_{HOMO} + E_{opt}$$

Where E_{opt} is the optical band gap of the complex **1**, E_{HOMO} and E_{LUMO} are the HOMO and LUMO energy levels, $E_{ox,onset}$ is the onset oxidation potential for the complex **1**, 4.8 eV is the ferrocene reference energy level and $E_{ox,Fc}$ is the potential when ferrocene starts to oxidize (0.40 eV vs. Ag/AgCl).

$$E_{HOMO} = -[0.52 - 0.40] - 4.8$$

$$= -4.92 \text{ eV}$$

$$E_{LUMO} = -4.92 + 2.34 = -2.58 \text{ eV}$$

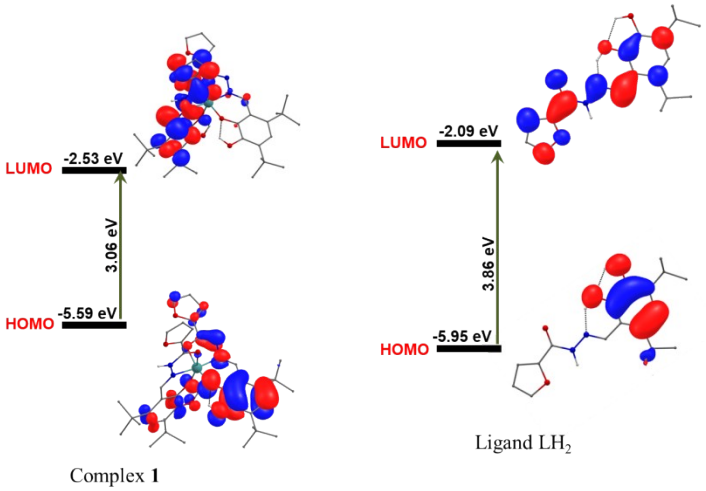


Figure S10. HOMO-LUMO energy gap of the complex **1** and uncoordinated hydrazine based catecholaldimine ligand (LH_3) calculated by DFT.

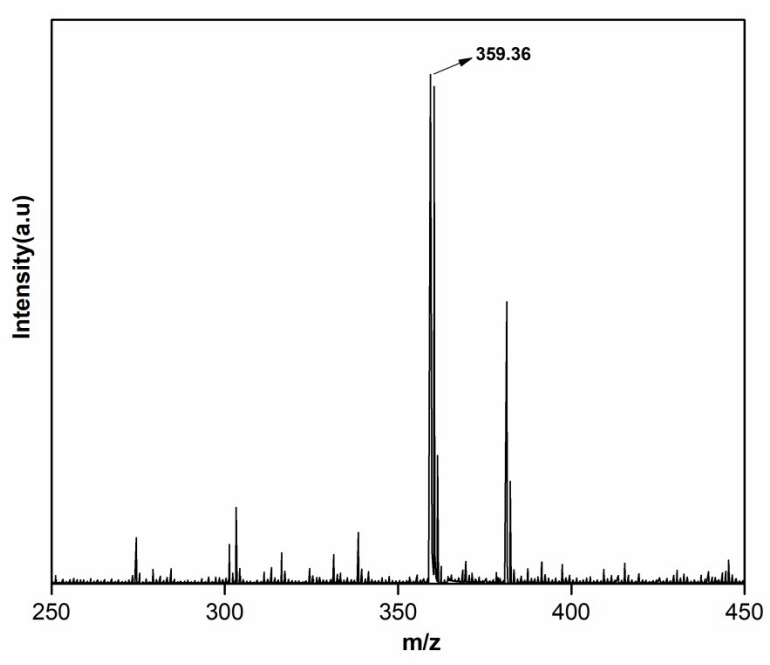


Figure S11. ESI-MS spectrum for LH_3.

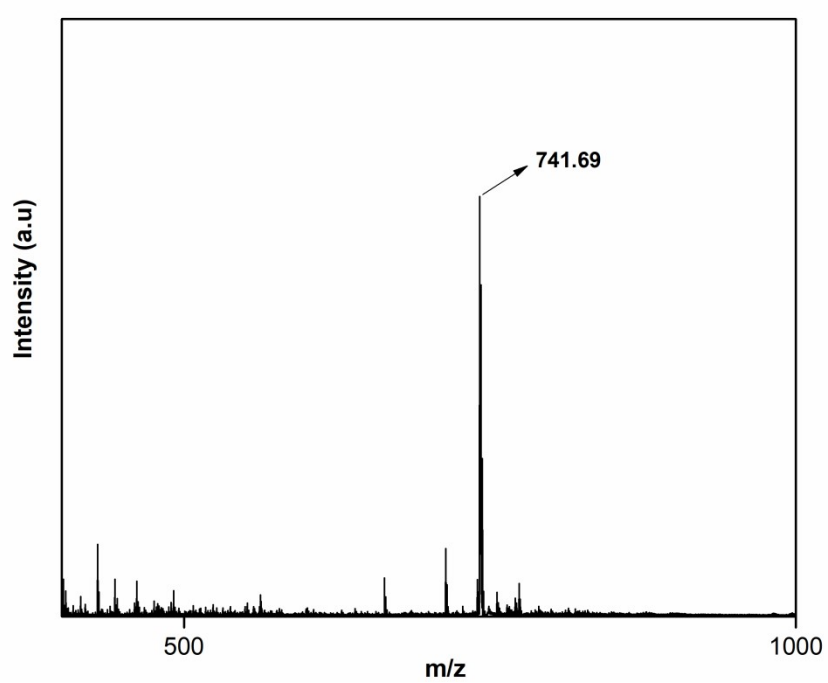


Figure S12. ESI-MS spectrum of Complex **1.**

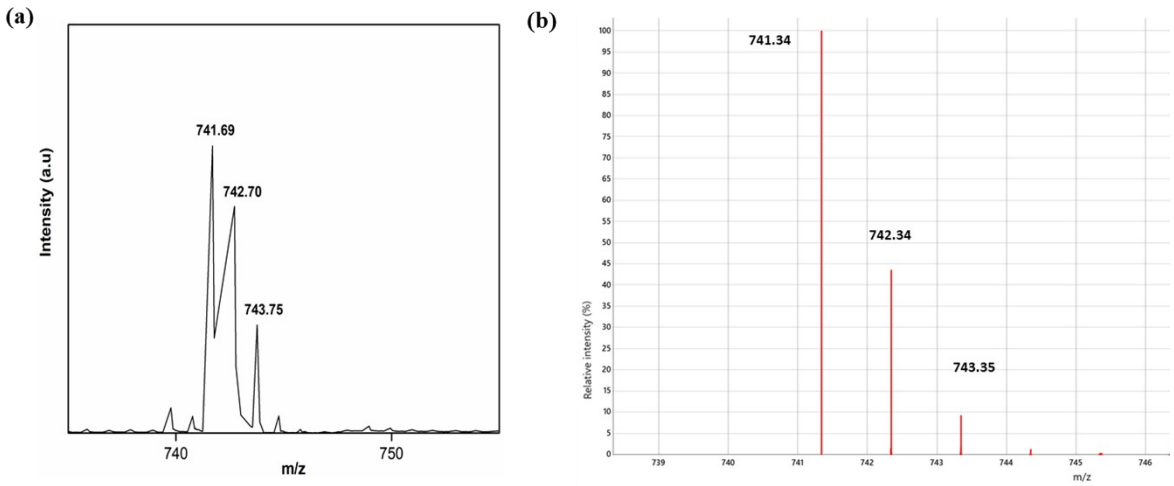
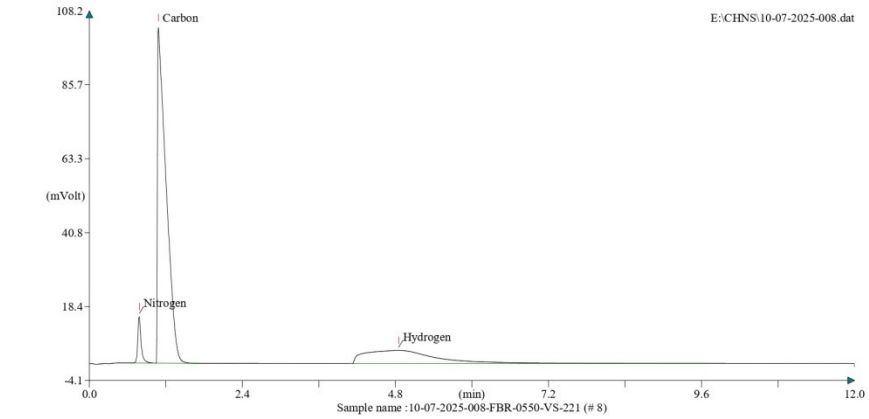
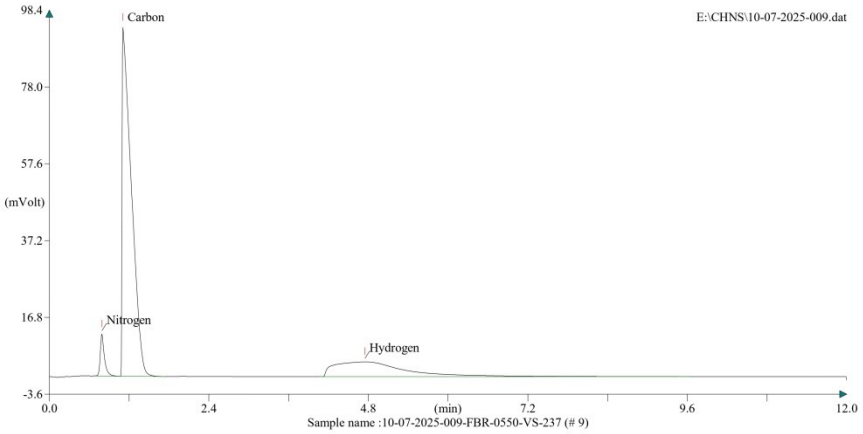


Figure S13. a) Experimental and b) Simulated isotopic distribution for Complex **1**.



Peak Number (#)	Retention Time (min)	Area (.1*uV*sec)	Element %	Component Name
1	0.783	510273	7.877	Nitrogen
2	1.083	9577476	67.295	Carbon
3	4.850	3198239	7.773	Hydrogen
		13285990	82.945	

Figure S14: Elemental CHN Analysis of Ligand LH_3**.**



Peak Number (#)	Retention Time (min)	Area (.1*uV*sec)	Element %	Component Name
1	0.792	478995	7.338	Nitrogen
2	1.108	8946320	62.828	Carbon
3	4.750	2961915	7.196	Hydrogen
		12387230	77.362	

Figure S15: CHN Analysis of Complex **1.**

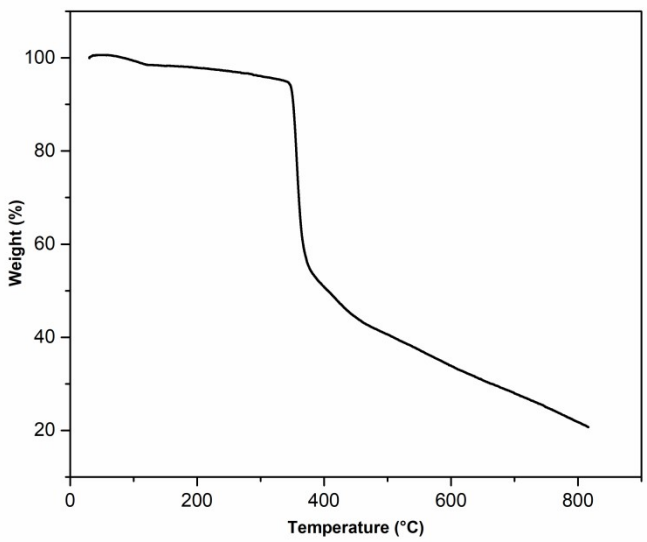


Figure S16: TGA Analysis of Complex **1.** Heating rate = 5^{o}C min−1 under a nitrogen atmosphere.

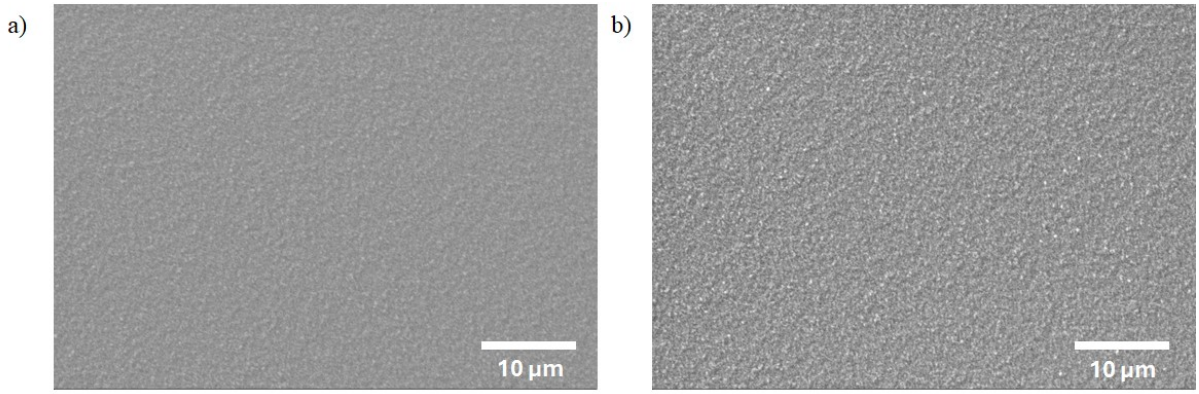


Figure S17: a) SEM image of device A2, FTO/HfO_2/Al complex **1**/Ag b) Device A1, FTO/Al Complex **1**/Ag on a scale bar of 10 µm.