Undirected C-H Bond Activation in Aluminium   
Hydrido Enaminonates

Chijioke Kingsley Amadi,Ufuk Atamtürk, Andreas Lichtenberg, Aida Raauf and Sanjay Mathur \*

Institute of Inorganic Chemistry, Department of Chemistry, University of Cologne, Greinstr. 6, 50939 Cologne, Germany; camadi@smail.uni-koeln.de (C.K.A.); ufuk.atamtuerk01@gmail.com (U.A.); andreas.lichtenberg@uni-koeln.de (A.L.); ajamil@uni-koeln.de (A.R.).

**\*** Correspondence: sanjay.mathur@uni-koeln.de (S.M.)

Figure S1: 1H NMR Spectrum of **(ETFB)** in CDCl3 at RT with schematic and arbitrary numbering.....................................................................................................................................................................2

Figure S2: 19F NMR Spectrum of **(ETFB)** in CDCl3 at RT with schematic and arbitrary numbering………...3

Figure S3: 1H NMR Spectrum of (**H-TFB-TBA**) in CDCl3 at RT with schematic and arbitrary numbering…..................................................................................................................................................................4

Figure S4: 19F NMR Spectrum of **(H-TFB-TBA)** in CDCl3 at RT with schematic and arbitrary numbering......................................................................................................................................................................5

Figure S5:1H NMR spectra of complex **3** recorded in C6D6. Inset shows a magnified portion of the spectrum of (**3**) to illustrate the signal due to hydride ligands attached to aluminium…………………………………...6

Figure S6: 19F NMR spectra of complex **3** recorded in C6D6....................................................................................7

Figure S7**.**19F NMR spectra of **4b** and **4b´** recorded in C6D6 at room temperature…………………………….8

Figure S8**.**13C NMR spectra of **4b** and **4b´** recorded in C6D6 at room temperature……………………………..8

Figure S9**.** 1H−13C heteronuclear multiple bond correlation (HMBC) spectra of 4b and 4b´ at room temperature………………………………………………………………..…………………………………………..9

Figure S10**.** 1H−13C Heteronuclear multiple quantum coherence (HMQC) spectra of 4b and 4b´ at room temperature……………………………………………………………………….…………………………………...9

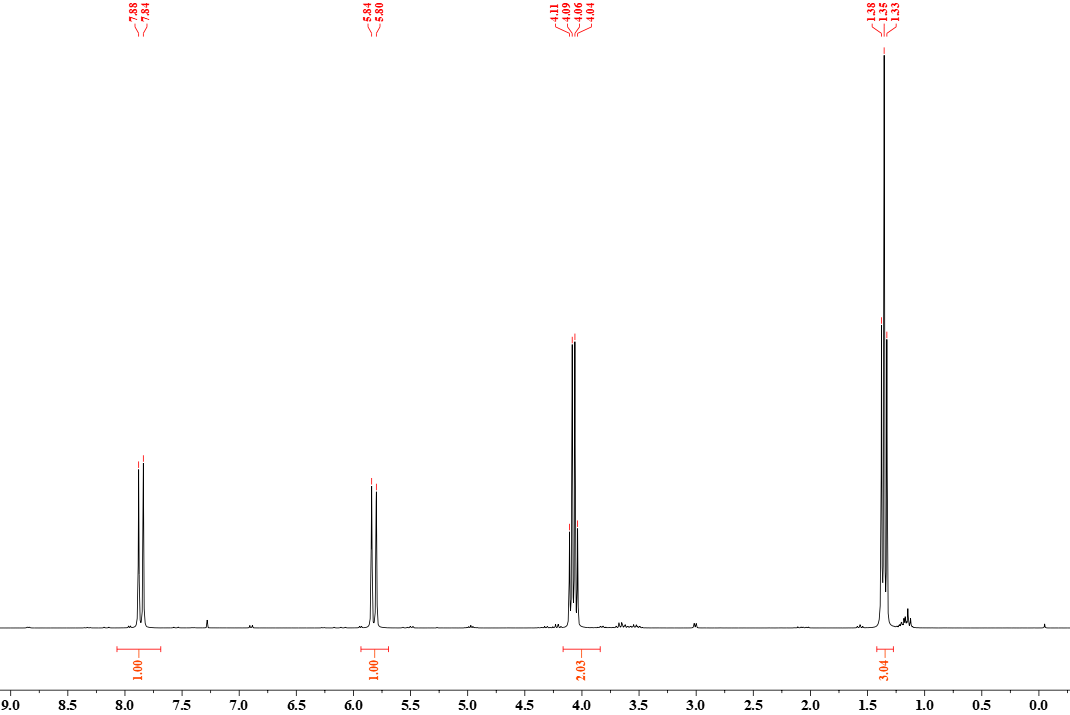
Figure S11:Solution 27Al NMR spectra of complex **3** recorded in C6D6 at room temperature………………...10

Figure S12:Solution 27Al NMR spectra of complexes **4b and 4b´** recorded in C6D6 at room temperature………………………………………………………………………………………….....…………….10

Figure S13. FT-IR spectra of the enaminone ligand (black), compound **3** (red) and compound (blue)…………………………………………………………………………………………………………………11

Figure S14:Obtained XRD pattern of the residue **3** and **4a** after TG…………………….……….……………11

Figure S15. Representative SEM images of post/annealed thin films deposited using **4** as precursor in a CVD process.…………………………………………………………………………………………………………...….12



**1**

**2**

**3**

**4**

**5**

**6**

**6-H**

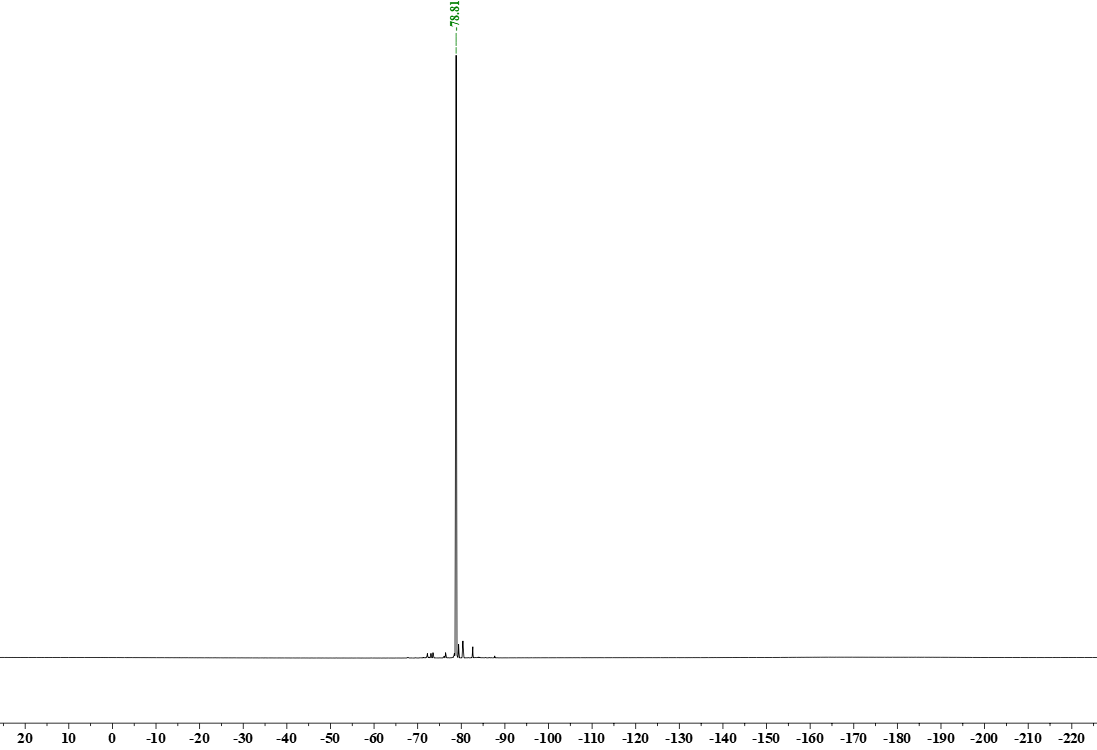
**5-H**

**4-H**

**3-H**

**ppm**

**Figure S1:** 1H NMR Spectrum of **ETFB** **1** in CDCl3 at RT with schematic and arbitrary numbering**.**



**1**

**2**

**3**

**4**

**5**

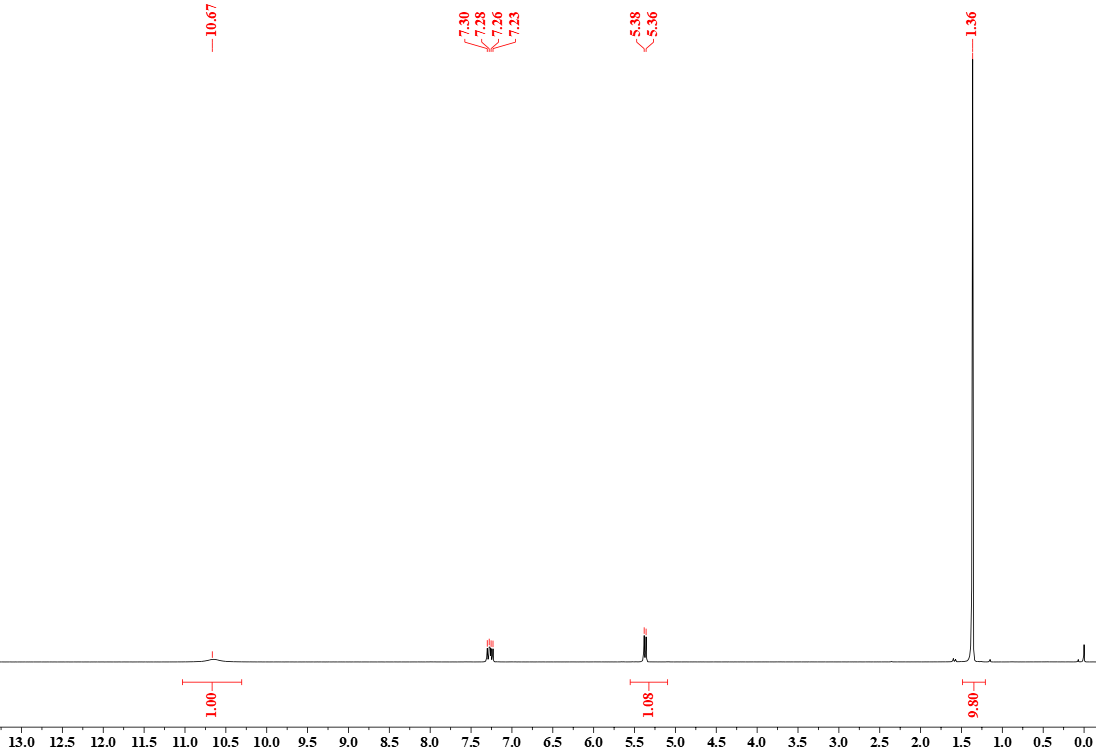
**6**

**ppm**

**1-F**

**1-F**

**Figure S2**: 19F NMR Spectrum of **ETFB** **1** in CDCl3 at RT with schematic and arbitrary numbering**.**



**ppm**



**1**

**2**

**3**

**4**

**5**

**NH**

**5-H**

**4-H**

**3-H**

**Figure S3**: 1H NMR Spectrum of **H-TFB-TBA** **2** in CDCl3 at RT with schematic and arbitrary numbering**.**



**ppm**



**1**

**2**

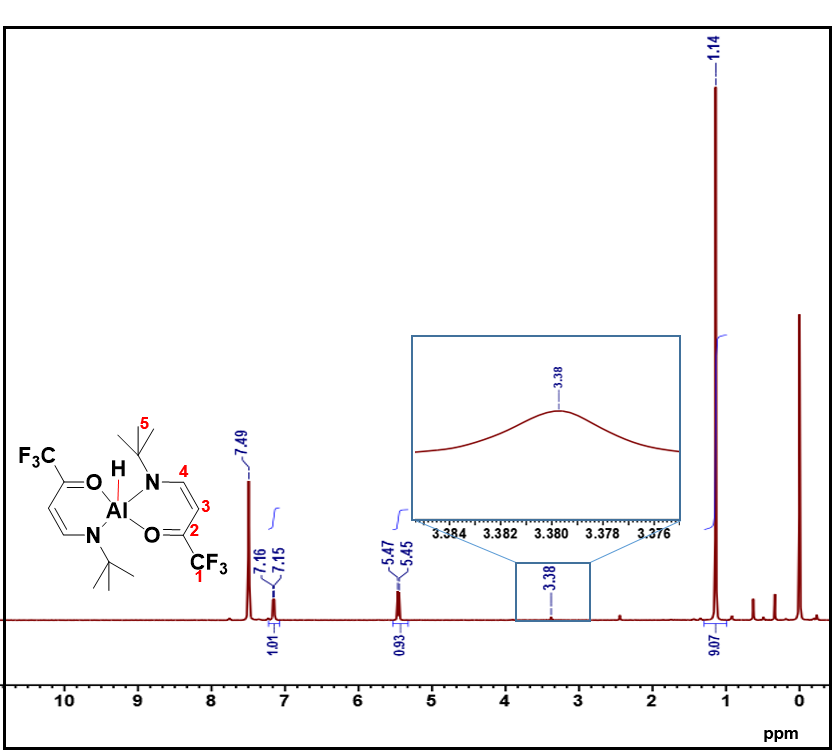
**3**

**4**

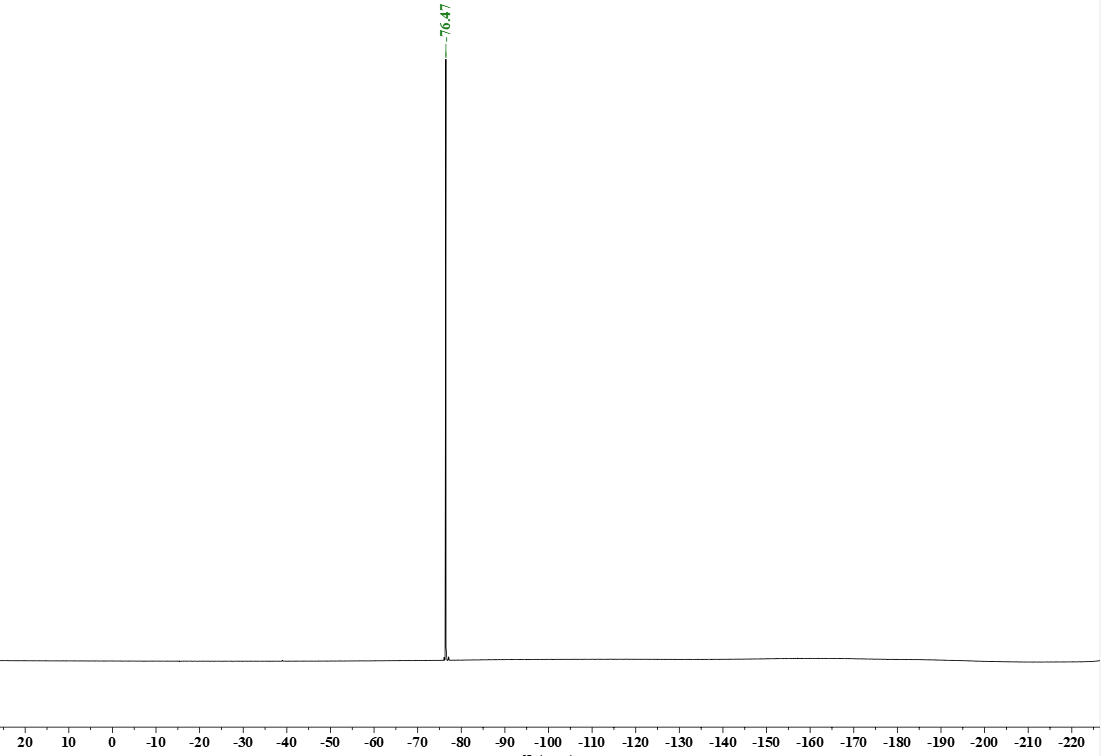
**5**

**1-F**

**Figure S4:** 19F NMR Spectrum of **H-TFB-TBA** **2** in CDCl3 at RT with schematic and arbitrary numbering**.**



**Figure S5.**1H NMR spectra of complex (**3**) recorded in C6D6. Inset shows a magnified portion of the spectrum of (**3**) to illustrate the signal due to hydride ligands attached to aluminium.



**ppm**



**3**

**5**

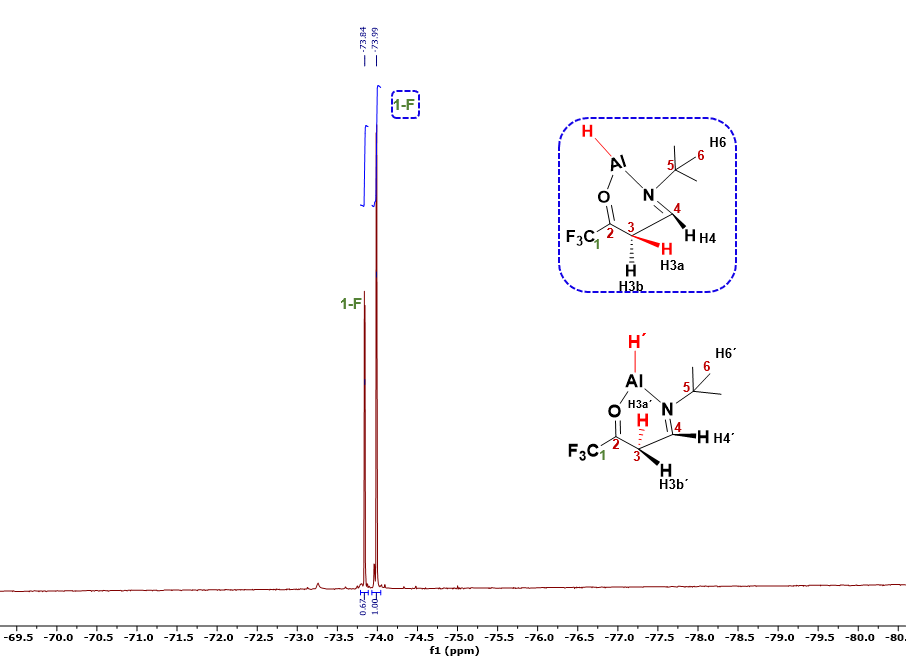
**4**

**1**

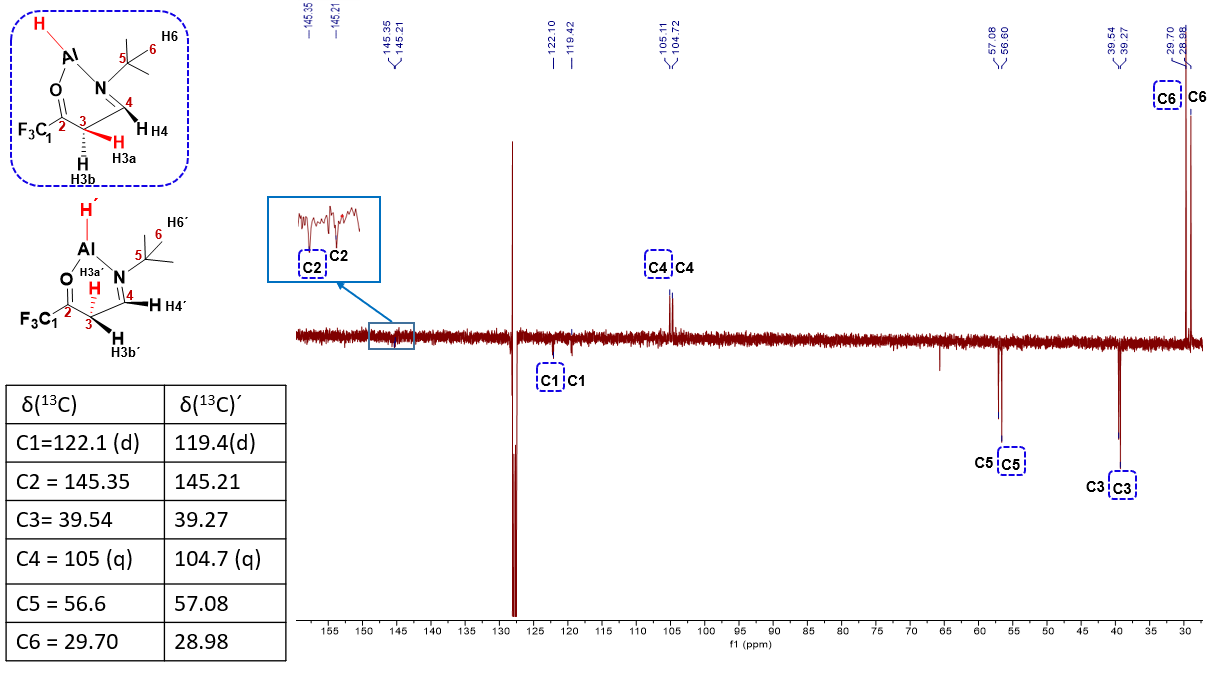
**1-F**

**2**

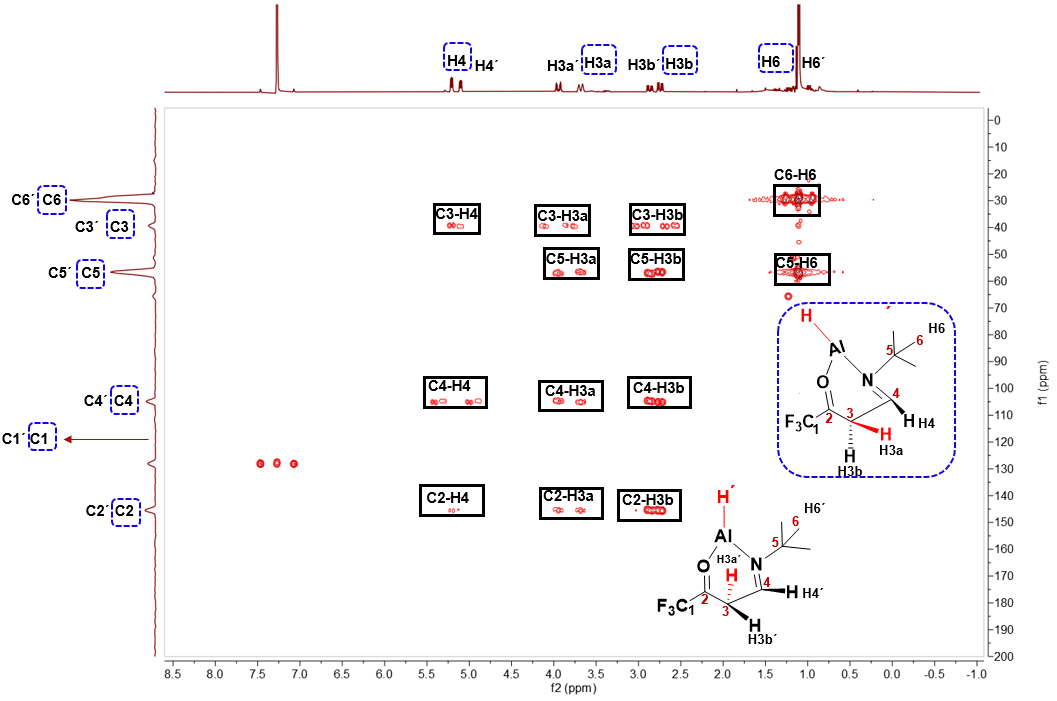
**Figure S6.**19F NMR spectra of complex (**3**) recorded in C6D6.

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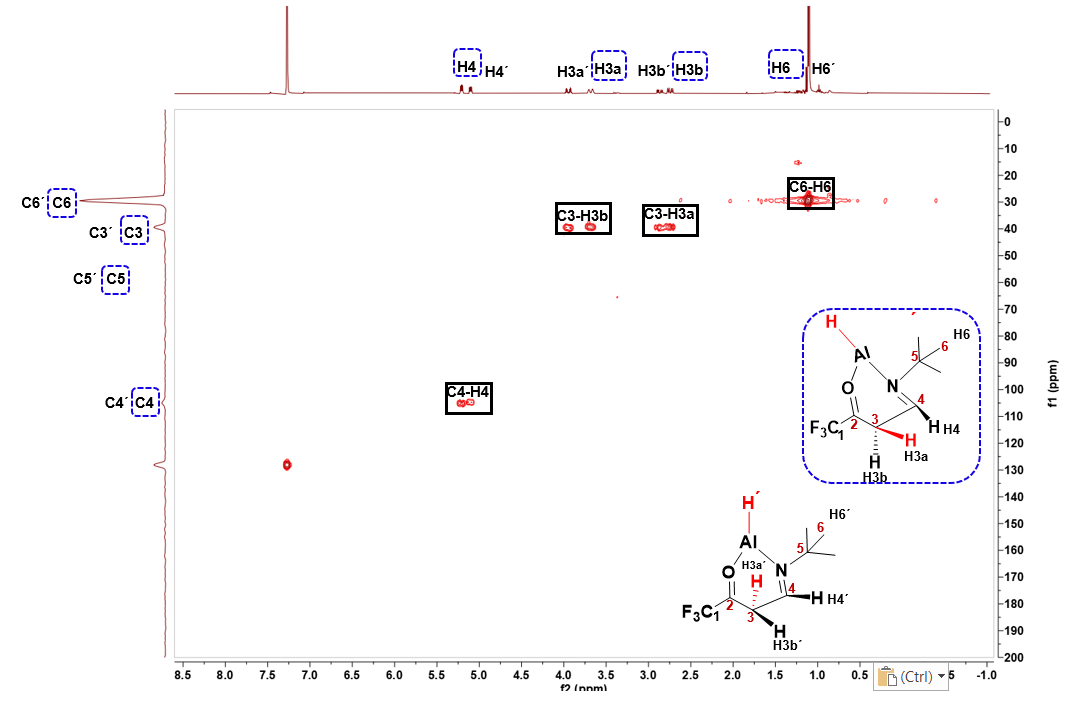
**Figure S7.**19F NMR spectra of **4b** and **4b´** recorded in C6D6 at room temperature.

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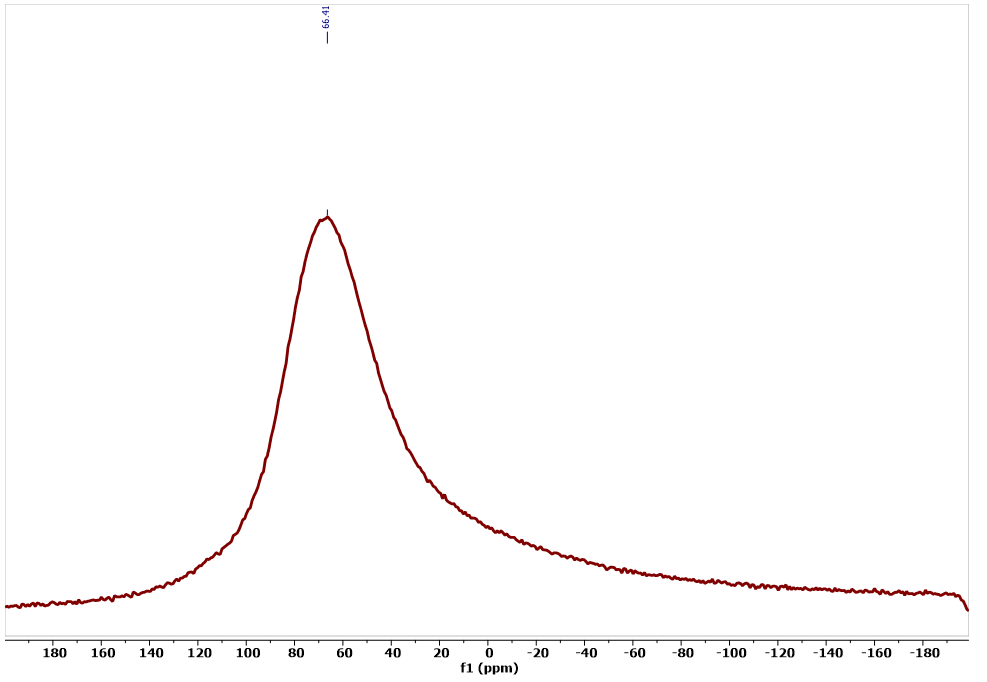
**Figure S8.**13C NMR spectra of **4b** and **4b´** recorded in C6D6 at room temperature.

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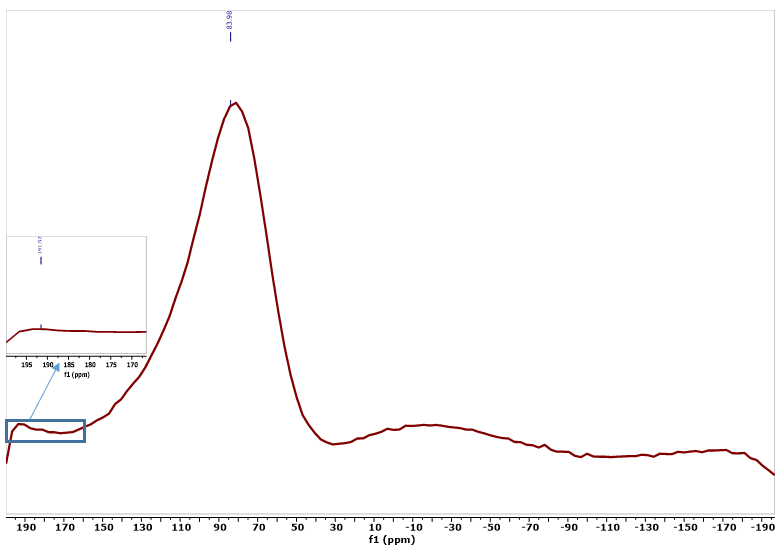
**Figure S9.** 1H−13C heteronuclear multiple bond correlation (HMBC) spectra of **4b** and **4b´** at room temperature.

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**Figure S10.** 1H−13C Heteronuclear multiple quantum coherence (HMQC) spectra of **4b** and **4b´** at room temperature.

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**Figure S11**:Solution 27Al NMR spectra of complex **3** recorded in C6D6 at room temperature

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**Figure S12**:Solution 27Al NMR spectra of complexes **4b** and **4b´** recorded in C6D6 at room temperature



**Al-H**

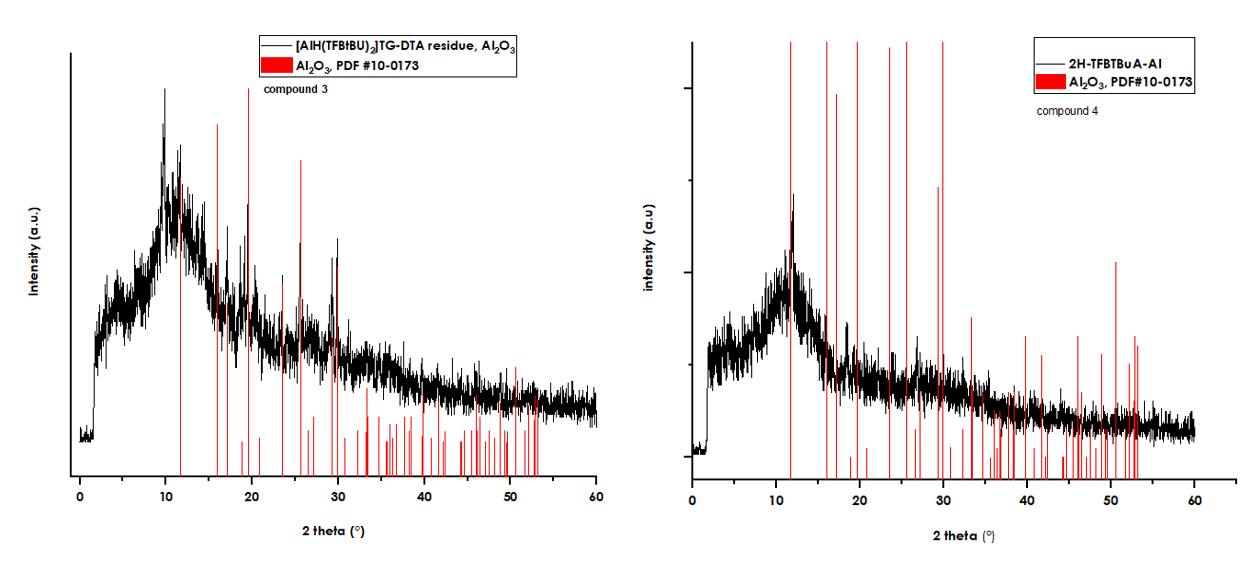
**4b 4b´**

**Ligand**

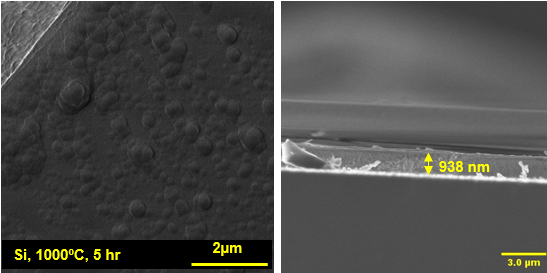
**3**

**Wavenumber (cm-1)**

**Figure S13**. FT−IR spectra of the enaminone ligand (black), compound **3** (red) and compound **4b and 4b´** (blue).



**Figure S14**:Obtained XRD pattern of the residue of **3** and **4a** after TG.



**Figure S15**. Representative SEM images of post-annealed thin films deposited using **4a** as precursor in a CVD process.