

Supporting information for:

# A new approach to internal Lewis pairs featuring a phosphenium acid and a pyridine base

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**Figure S-1:** Stacked plot of the  $^{31}\text{P}\{^1\text{H}\}$  NMR spectra for **2**[I<sub>3</sub>], **2**Br, **2**[OTf], **2**•BH<sub>3</sub>[OTf] and **2**•DMAP[OTf]...S2

**Figure S-2:** Stacked plot of the variable temperature  $^{31}\text{P}\{^1\text{H}\}$  NMR spectra for **2**•DMAP[OTf]...S2

**Figure S-3:**  $^1\text{H}$  NMR spectrum of **2**[I<sub>3</sub>]...S3

**Figure S-4:**  $^1\text{H}$  NMR spectrum of **2**Br...S3

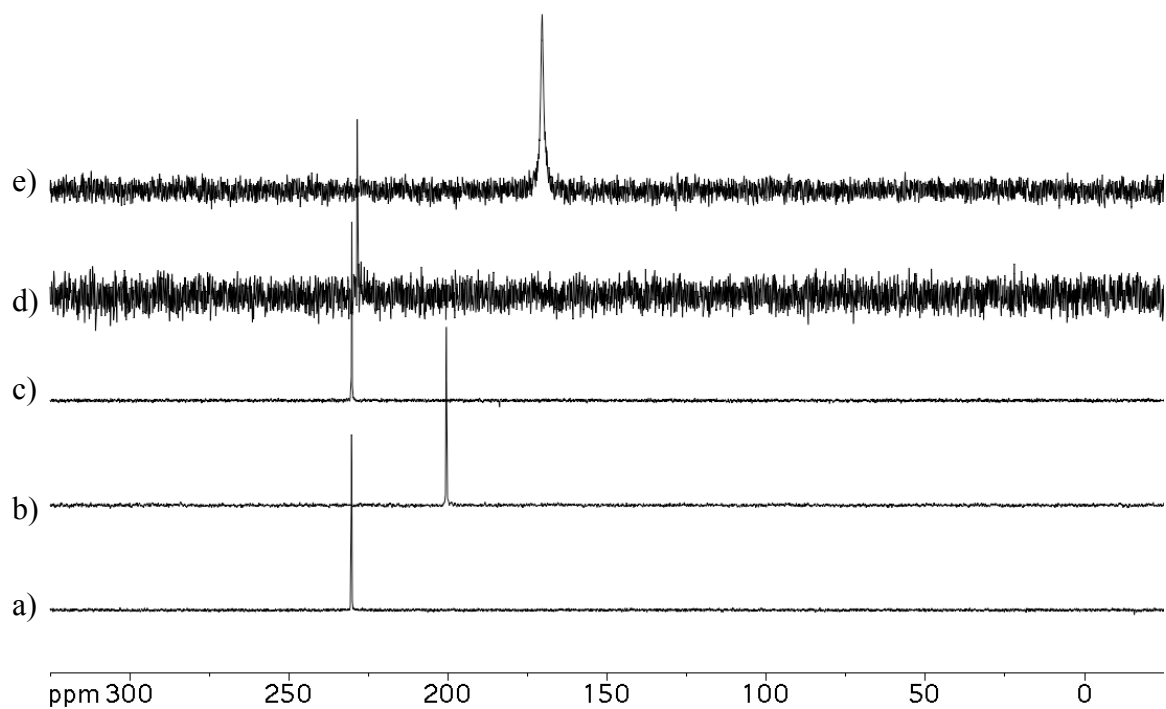
**Figure S-5:**  $^1\text{H}$  NMR spectrum of **2**[OTf]...S4

**Figure S-6:**  $^1\text{H}$  NMR spectrum of **2**•BH<sub>3</sub>[OTf]...S4

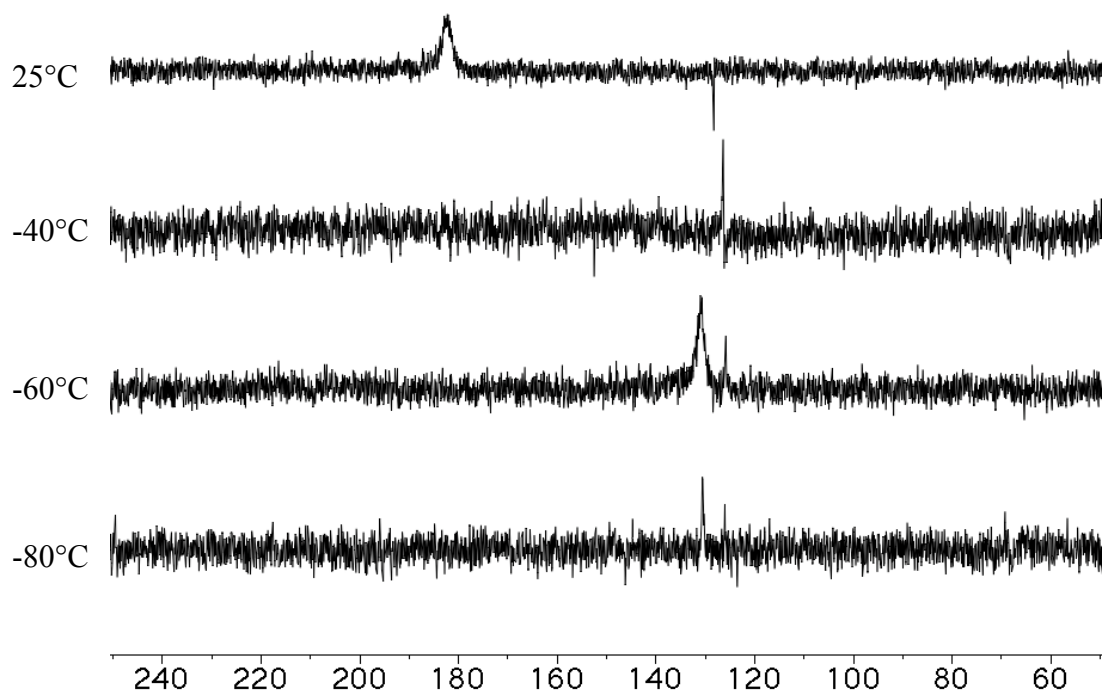
**Figure S-7:**  $^1\text{H}$  NMR spectrum of **2**•DMAP[OTf]...S5

**Figure S-8:** Stacked plot of the  $^1\text{H}$  NMR spectra of the alkyl region for **2**[OTf] and **2**•BH<sub>3</sub>[OTf]...S5

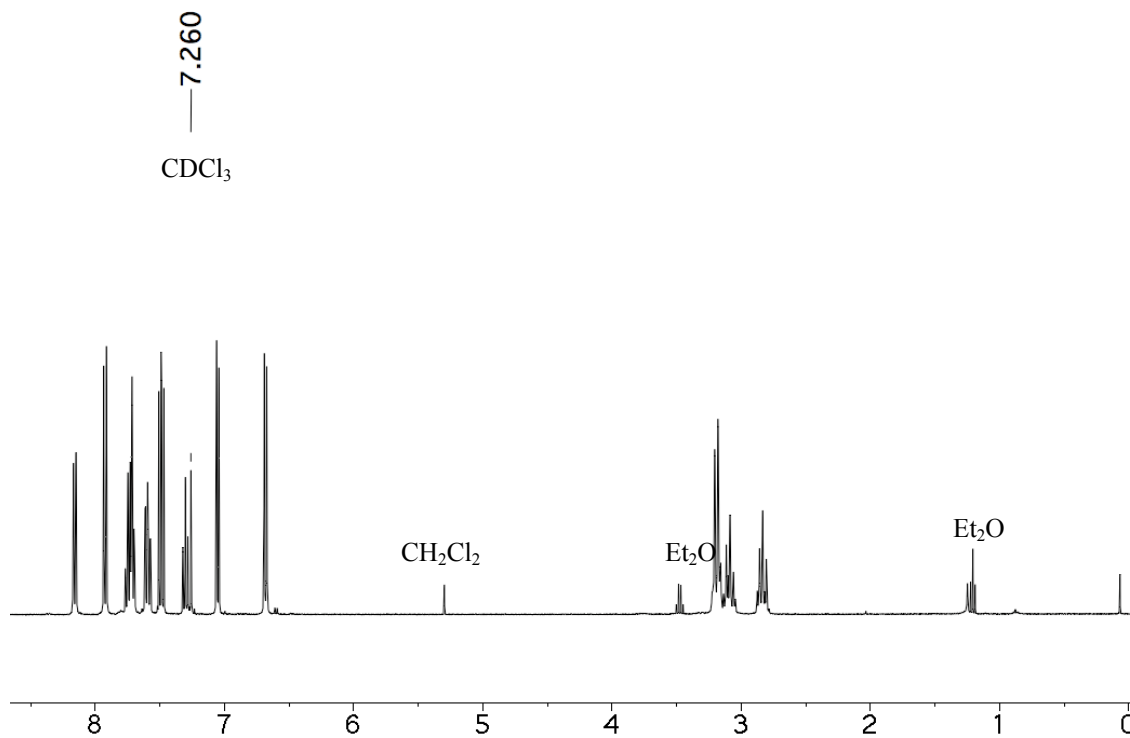
**Figure S-9:** Stacked plot of the FT-IR spectra of **2**[OTf] and **2**•BH<sub>3</sub>[OTf]...S6



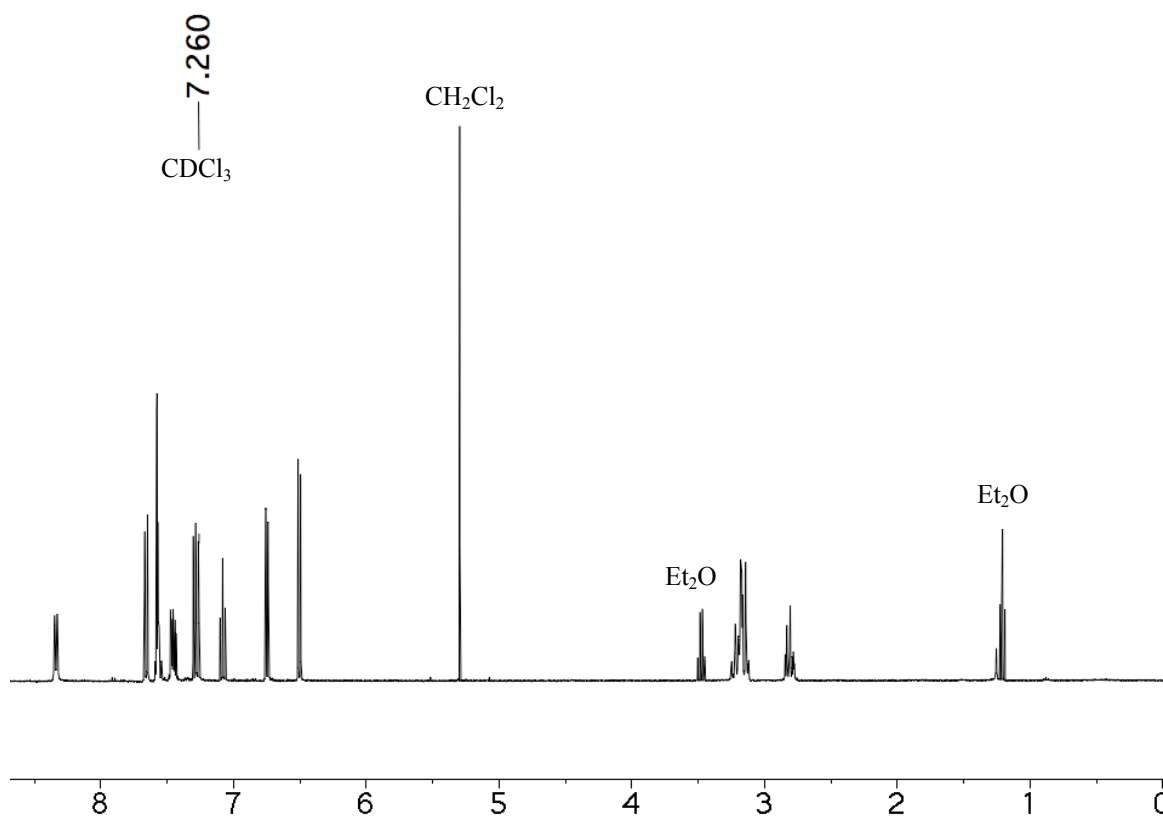
**Figure S-1:** Stacked plot of the 162 MHz  $^{31}\text{P}\{^1\text{H}\}$  NMR spectra for a)  $\mathbf{2}[\text{I}_3]$   $\delta_{\text{P}} = 230$ , b)  $\mathbf{2Br}$ :  $\delta_{\text{P}} = 200$ , c)  $\mathbf{2}[\text{OTf}]$ :  $\delta_{\text{P}} = 230$ , d)  $\mathbf{2}\bullet\text{BH}_3[\text{OTf}]$ :  $\delta_{\text{P}} = 228$  and e)  $\mathbf{2}\bullet\text{DMAP}[\text{OTf}]$ :  $\delta_{\text{P}} = 170$ .



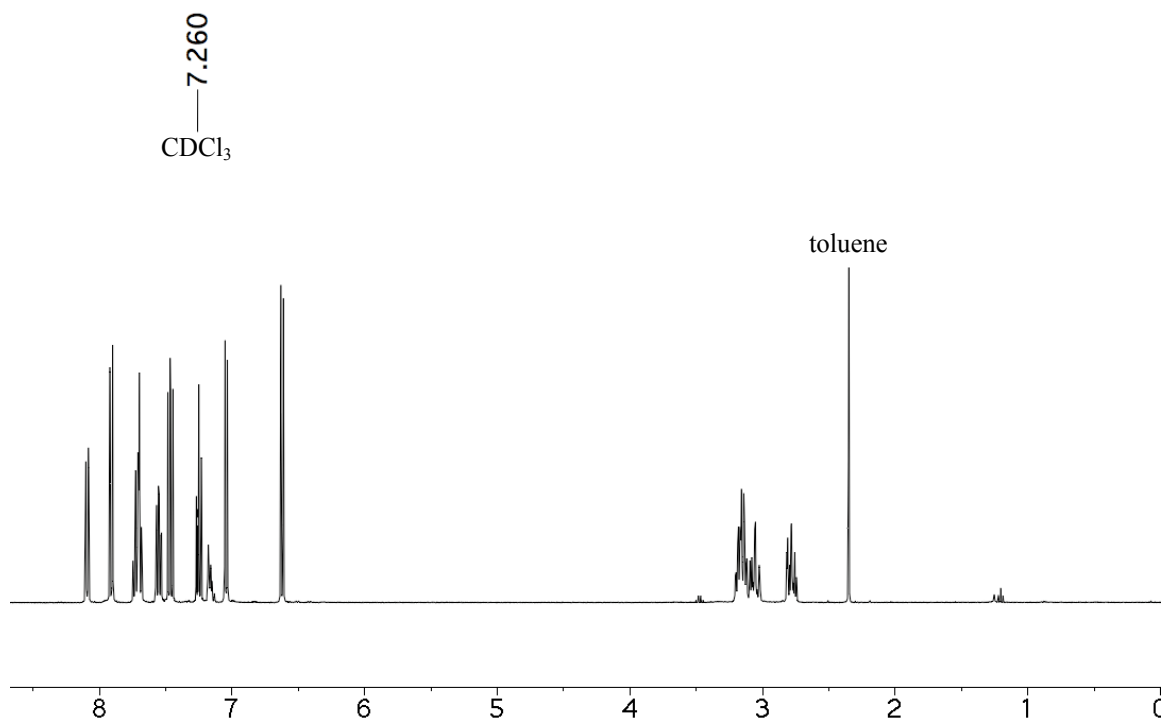
**Figure S-2:** Stacked plot of the variable temperature 162MHz  $^{31}\text{P}\{^1\text{H}\}$  NMR spectra for  $\mathbf{2}\bullet\text{DMAP}[\text{OTf}]$  in  $\text{CH}_2\text{Cl}_2$ .



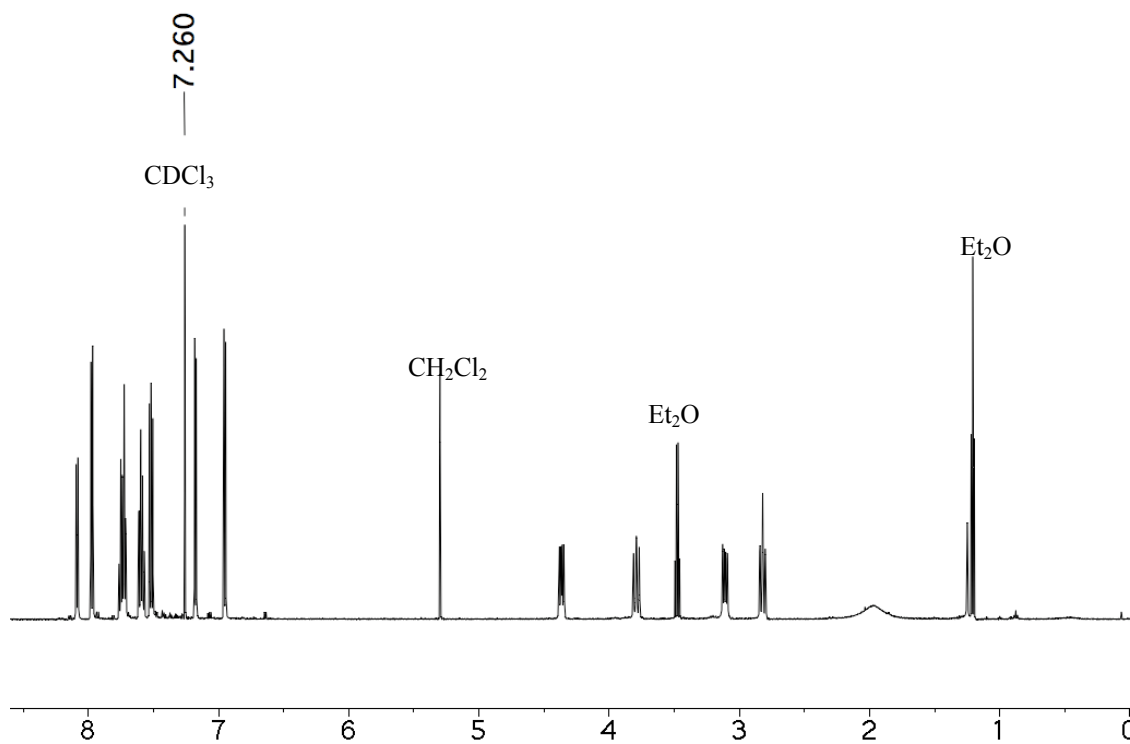
**Figure S-3:** 400 MHz <sup>1</sup>H NMR spectrum of 2[I<sub>3</sub>].



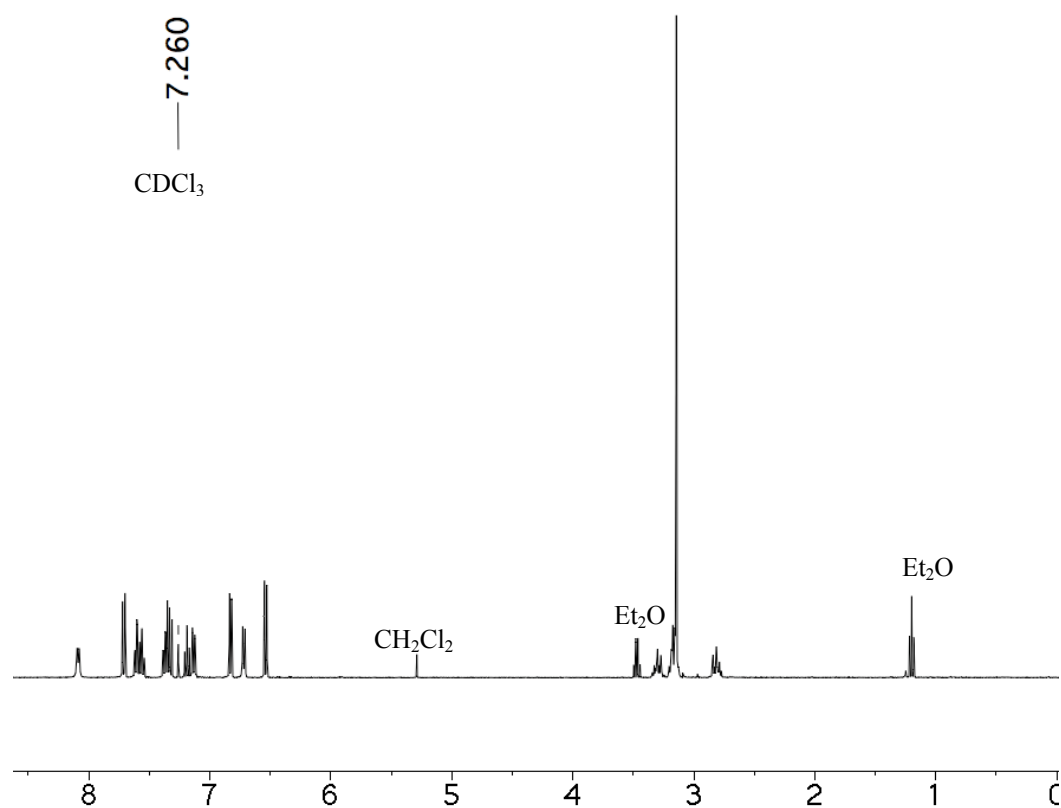
**Figure S-4:** 400 MHz <sup>1</sup>H NMR spectrum of 2Br.



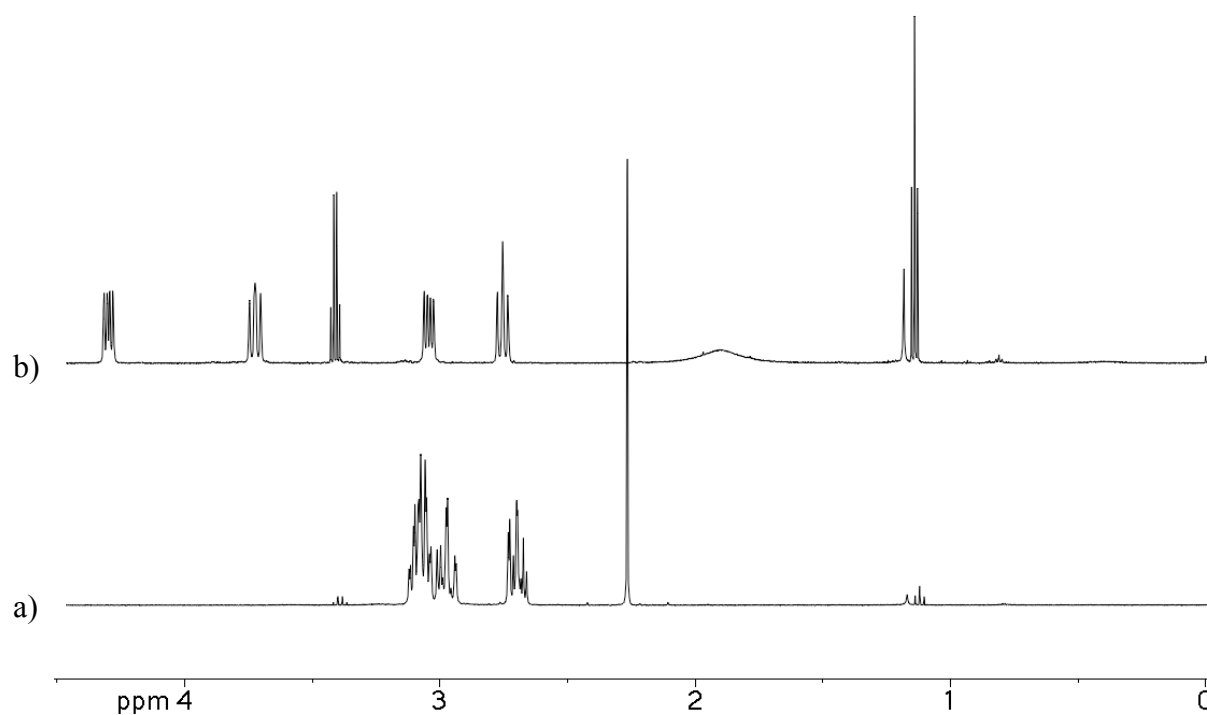
**Figure S-5:** 400 MHz <sup>1</sup>H NMR spectrum of **2**[OTf].



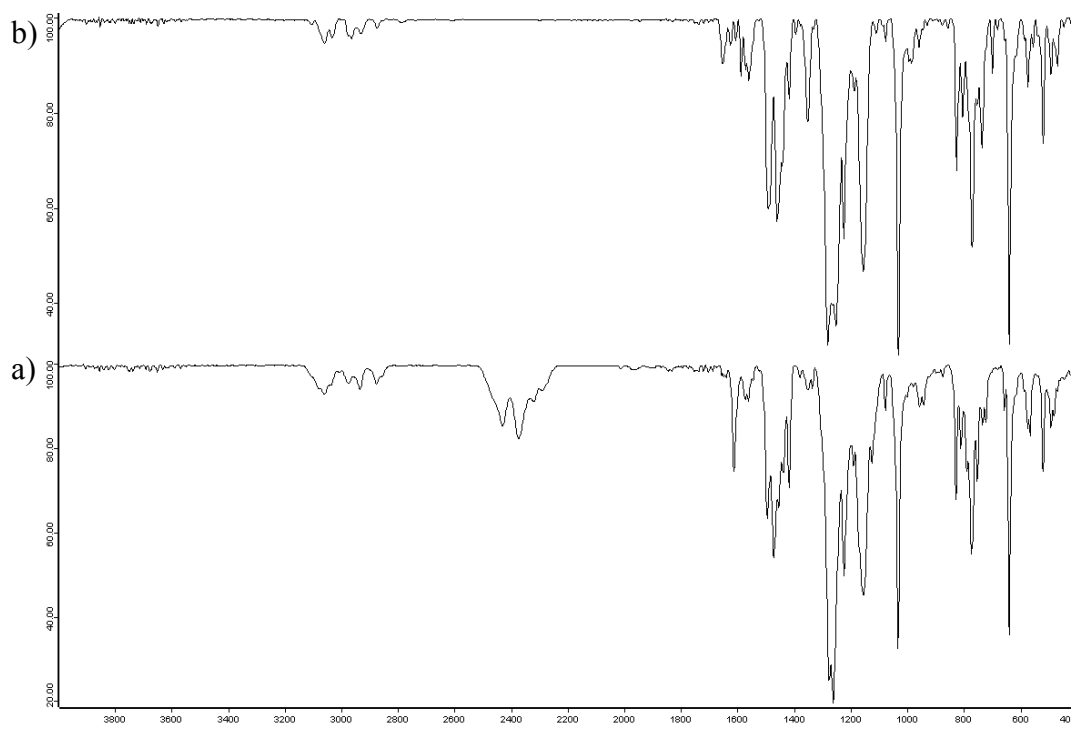
**Figure S-6:** 400 MHz <sup>1</sup>H NMR spectrum of **2**•BH<sub>3</sub>[OTf].



**Figure S-7:** 400 MHz <sup>1</sup>H NMR spectrum of **2**•DMAP[OTf].



**Figure S-8:** Stacked plot of <sup>1</sup>H NMR spectra of the alkyl region for a) **2**[OTf] and b) **2**•BH<sub>3</sub>[OTf].



**Figure S-9:** Stacked plot of the FT-IR spectra of a) **2**•BH<sub>3</sub>[OTf] and b) **2**[OTf].