

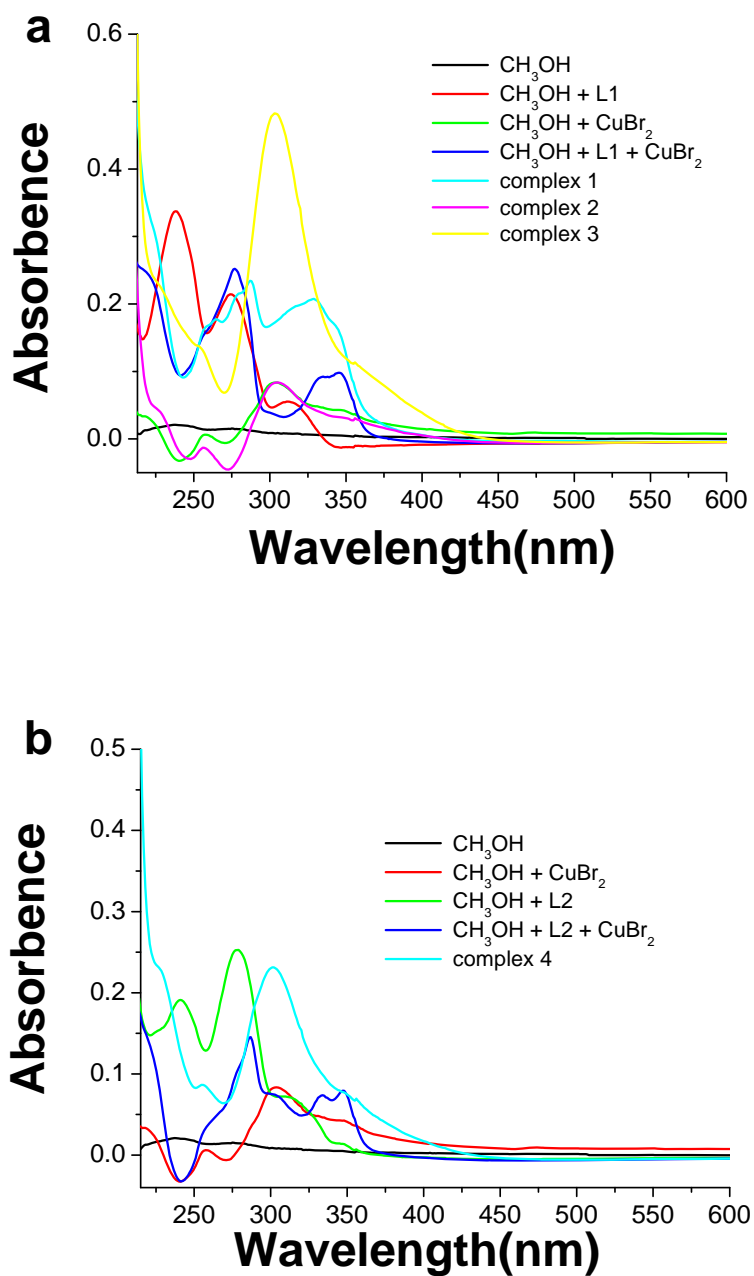
*Electronic Supplementary Information*

**In Situ Selective *N*-alkylation of Pendant Pyridyl Functionality in Mixed-Valence Copper Complexes with Methanol and Copper(II) Bromide**

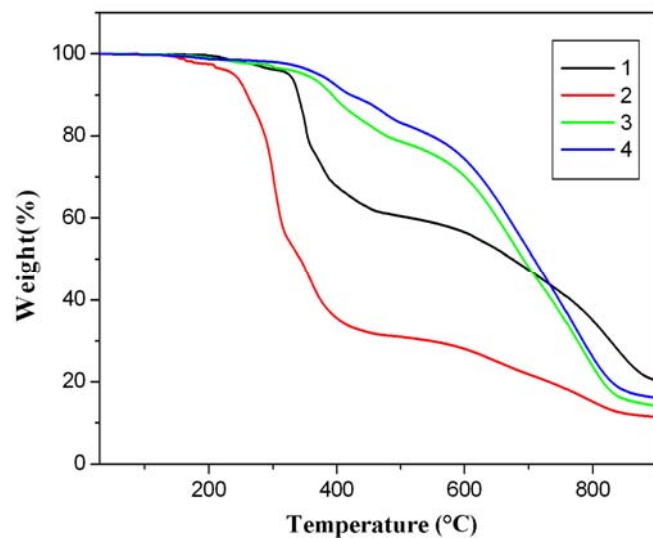
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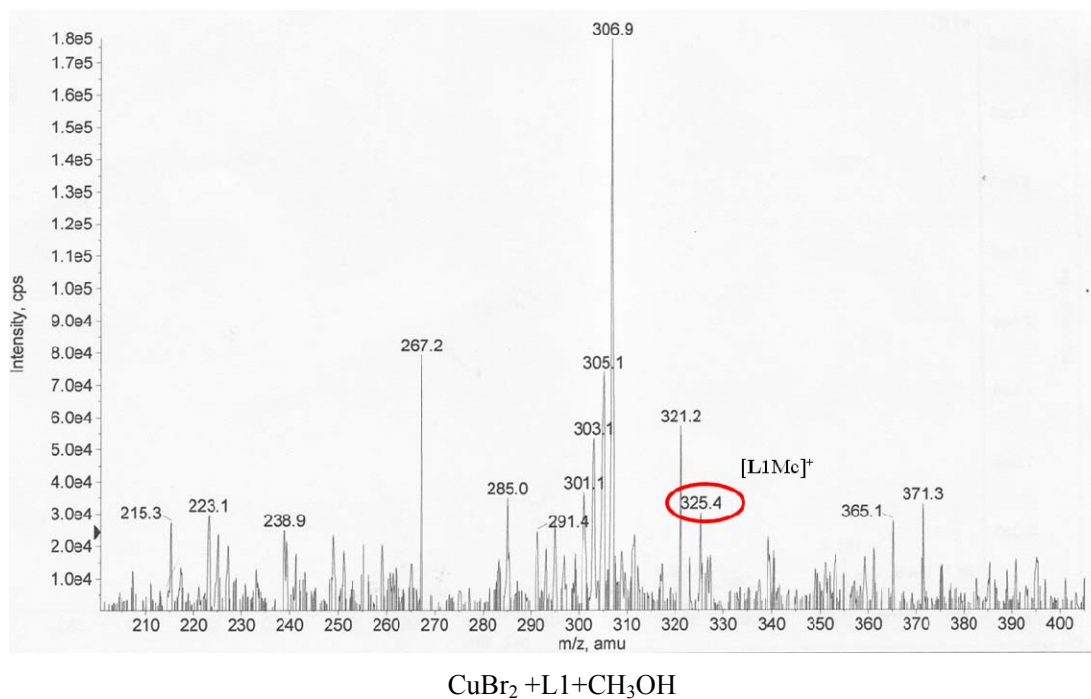
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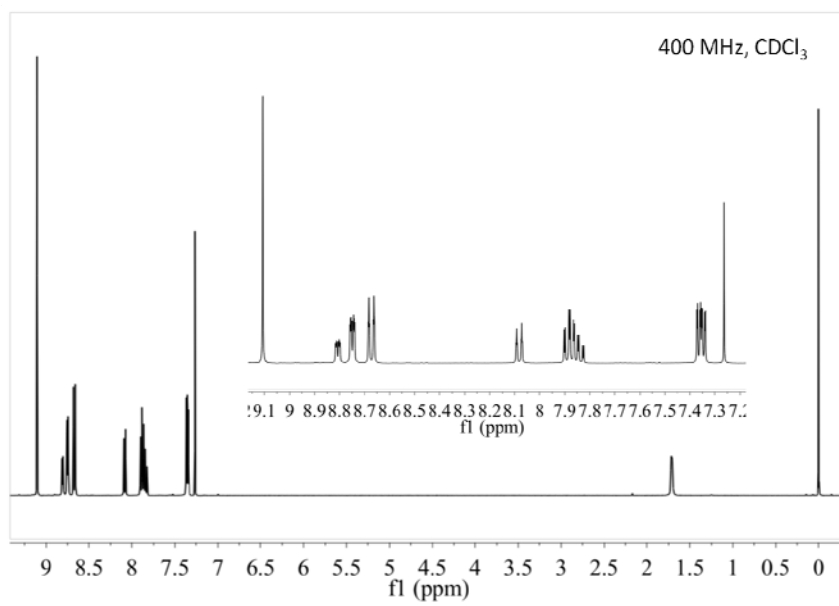
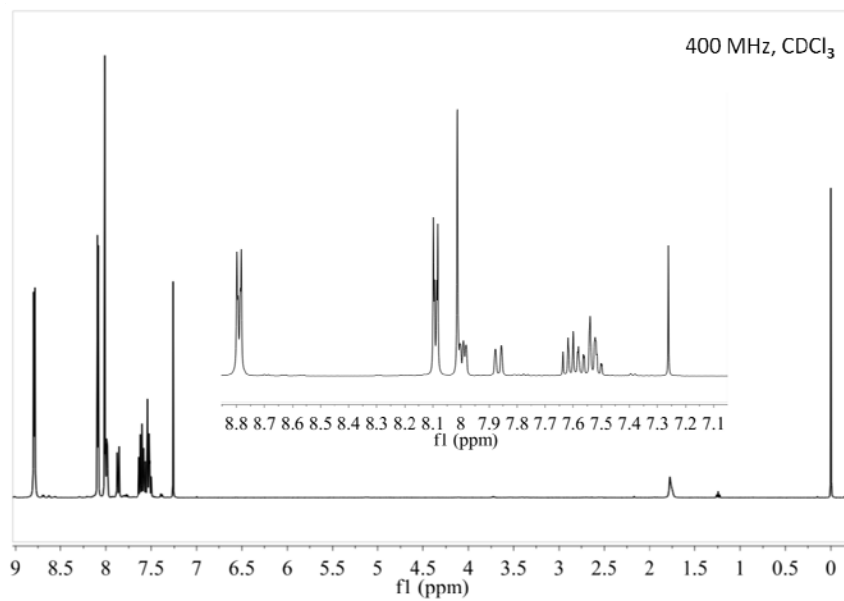
**Fig. S1.** The UV-VIS spectra of different reaction filtrates. (a) The ratio between Ligand L1 and CuBr<sub>2</sub> is 1:2 (complex 1), 1:4 (complex 2), and 1:8 (complex 3); (b) The ratio between Ligand L2 and CuBr<sub>2</sub> is 1:4 (complex 4).



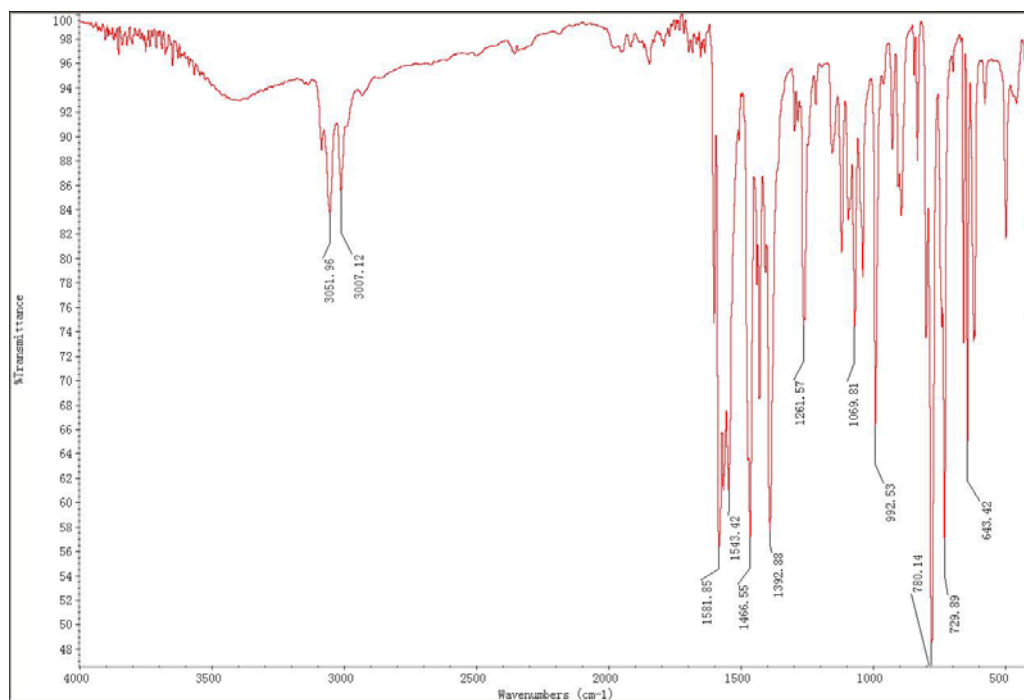
**Fig. S2.** TGA curves of complexes 1–4.



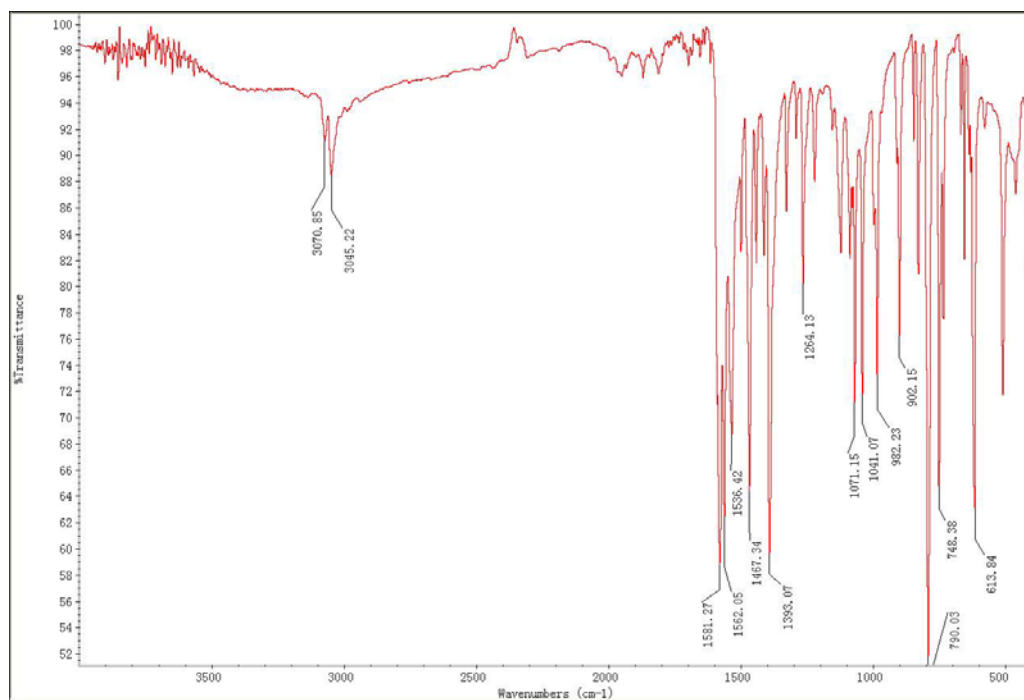
**Fig. S3.** ESI-Mass spectra of reaction filtrate of complex 1.



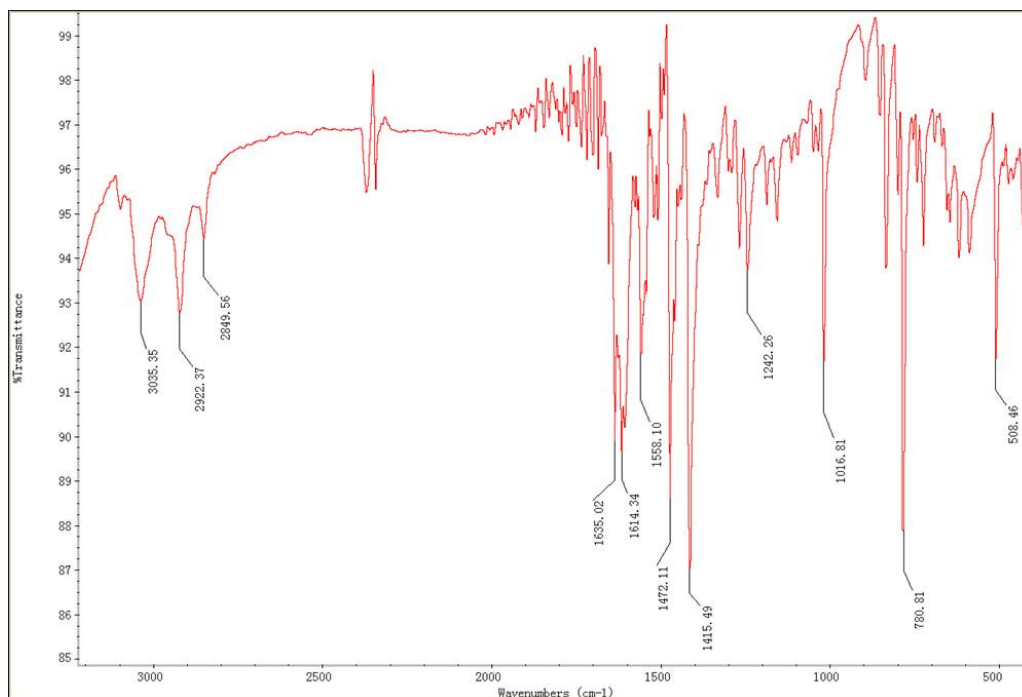
**Fig. S4.** <sup>1</sup>H NMR spectra of L1 (top) and L2 (bottom).



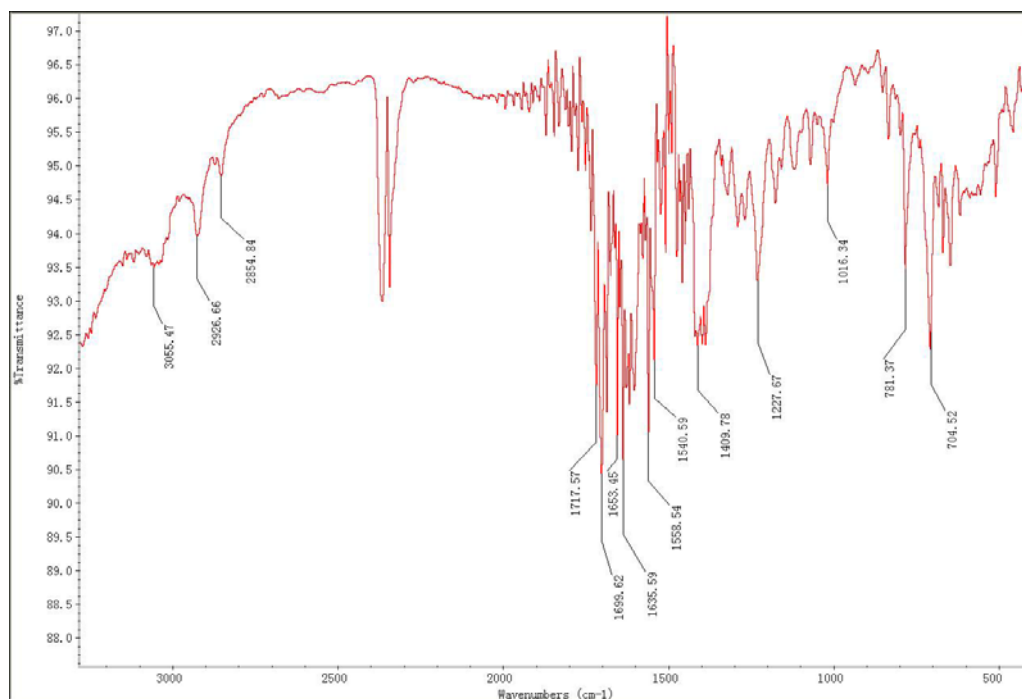
(a) L1



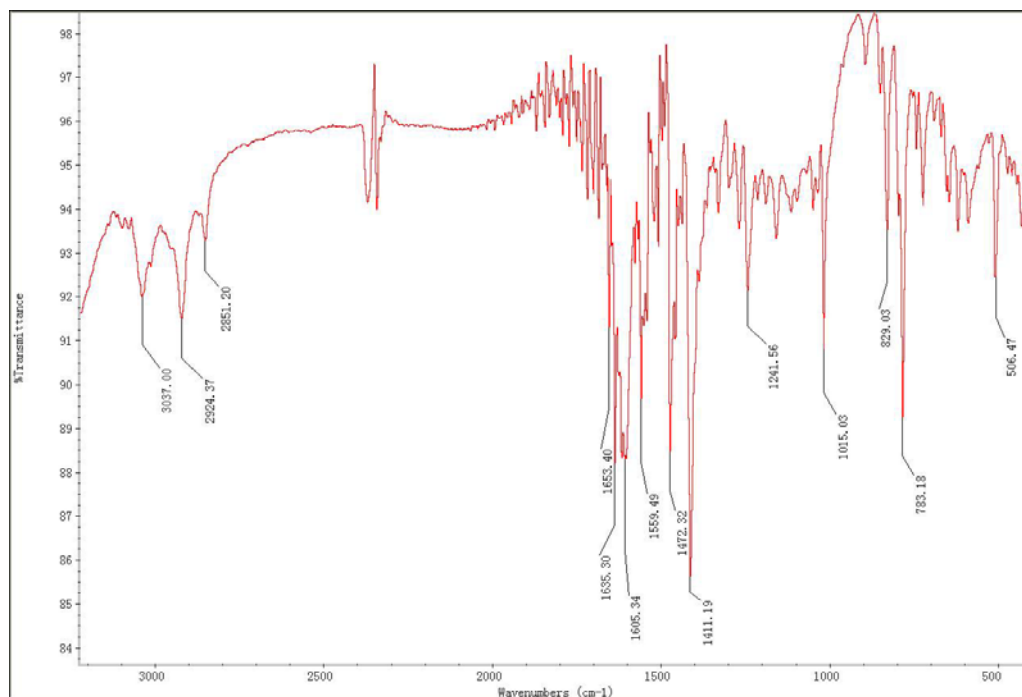
(b) L2



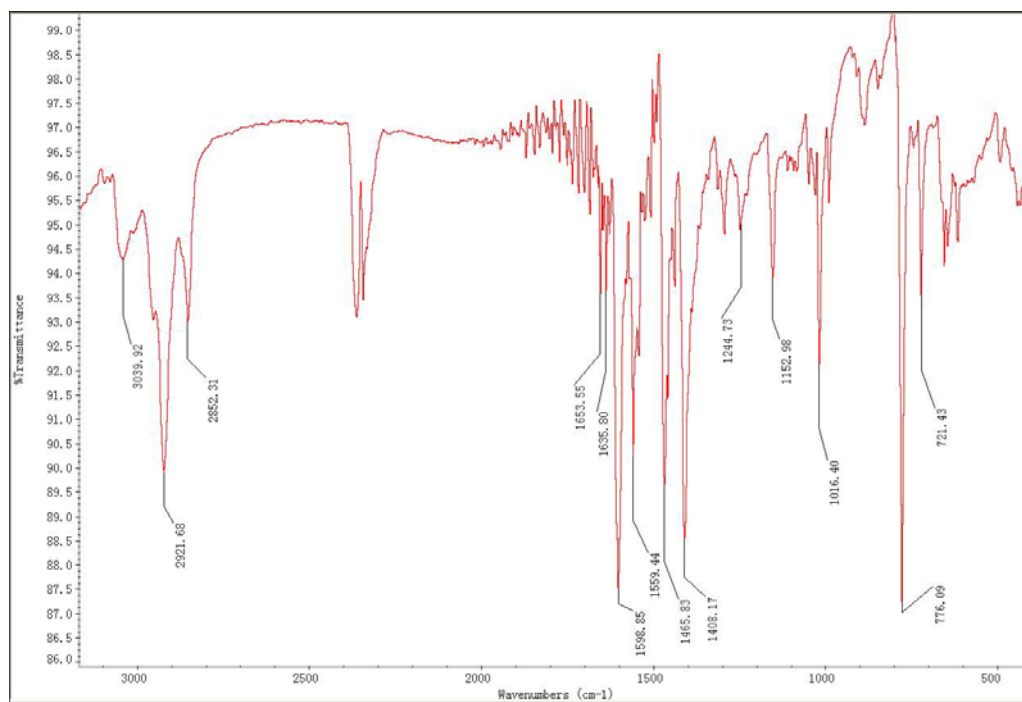
(c) complex 1



(d) complex 2



(e) complex 3



(f) complex 4

Fig. S5. IR spectra of ligands and complexes.