



Evidence supporting the proposed mechanism

- A** The introduction of radical scavengers did not inhibit the reaction, suggesting that the free radicals are not major intermediates
- B** Cu(II) complex was characterized by EPR and UV-Vis spectra when *NH*-sulfilimine was subjected to the CuBr solution, supporting that *NH*-sulfilimine could facilitate the oxidation of Cu(I) to Cu(II)
- C** The H₂ gas was detected by headspace gas chromatography (GC) analysis and the hydrogen atom origin of H₂ was confirmed to be from alcoholic solvent by mass spectroscopy while CD₃OD was employed as the solvent
- D** The copper hydride pathway was plausible based on a density functional theory (DFT) computational study

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