Accepted Manuscript

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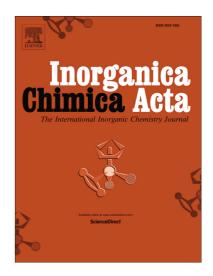
PII: S0020-1693(18)30126-9

DOI: https://doi.org/10.1016/j.ica.2018.05.018

Reference: ICA 18269

To appear in: Inorganica Chimica Acta

Received Date: 26 January 2018 Revised Date: 12 April 2018 Accepted Date: 14 May 2018



Please cite this article as: J-P. Li, Y-C. Shi, Synthesis, Characterization and Electrocatalytic H₂ Production of Base-containing Fe/S and Fe/Se Clusters, *Inorganica Chimica Acta* (2018), doi: https://doi.org/10.1016/j.ica.2018.05.018

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ACCEPTED MANUSCRIPT

Synthesis, Characterization and Electrocatalytic H₂

Production of Base-containing Fe/S and Fe/Se Clusters

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Abstract: Reaction of base-containing diphosphine ligand NP^{S_2} ($NP_2 = (Ph_2PCH_2)_2NCH_3$) with

 $[Fe_3(CO)_{12}]$ in THF at room temperature forms $[\{Fe_2(CO)_6(\mu-PPh_2)\}_2(\mu_4-S_2)]$ (1) and $[Fe_3(CO)_8(\mu_3-E_2)]$

S)₂{kP-PPh₂CH₂N(CH₃)CH₂P(=S)Ph₂}] (2) resulting from P=S bond cleavage and P-Fe bond

formation. Similarly, $NP_2^{Se}_2$ reacts with $Fe_3(CO)_{12}$ to provide $[\{Fe_2(CO)_6(\mu-PPh_2)\}_2(\mu_4-Se_2)]$ (3) and

 $[Fe_3(CO)_7(\mu_3-Se)_2\{k^2P,P-(PPh_2CH_2)_2NCH_3\}]$ (4). All the compounds have been completely

characterized by elemental analyses, IR, ¹H NMR, ¹³C NMR and ³¹P NMR spectroscopy and structurally

determined by X-ray crystallography. Electrochemical studies reveal that when using HOAc or TFA as a

proton source, 2 and 4 exhibit catalytic H₂ production.

Keywords: Fe/S Cluster; Fe/Se Cluster; Cyclic voltammetry; H₂ production

1. Introduction

The development of renewable energy is nowadays one of the most important challenges as global

energy consumption is rising significantly. Molecular hydrogen is a carbon-free fuel that could become

the energy carrier of the future, thus making the reversible interconversion of protons to molecular

hydrogen a key process for future energy schemes. Nature is able to catalyze this process by a class of

1

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