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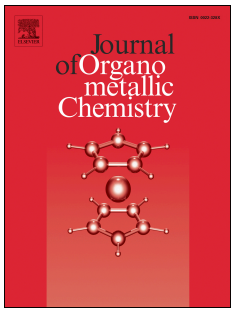

Nickel bis(dithiolene) complexes for electrocatalytic hydrogen evolution: A computational study

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Nickel Bis(dithiolene) Complexes for Electrocatalytic Hydrogen Evolution: A Computational Study

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Abstract

The substituent effect of dithiolene ligand on electrocatalytic activity of Ni bis(dithiolene) towards hydrogen evolution reaction (HER) are thoroughly explored with computational methods. A series of differently substituted dithiolene ligands are compared and evaluated. These computational results indicate that adding electron-withdrawing groups on the ligand or enlarging the conjugate π system of the ligand can greatly reduce the reduction potentials, while tuning the electronic structure of the non-innocent framework can optimize the geometry of dihydrogen species to achieve lower energy barrier for H_2 elimination. This study may benefit the deep understanding of the electrocatalyzed HER mechanism and give inspirations on the design of efficient nickel bis(dithiolene) electrocatalysts with low overpotential and improved efficiency.