Ni-Based Molecular Electrocatalysts for Hydrogen Oxidation

Dan DuBois
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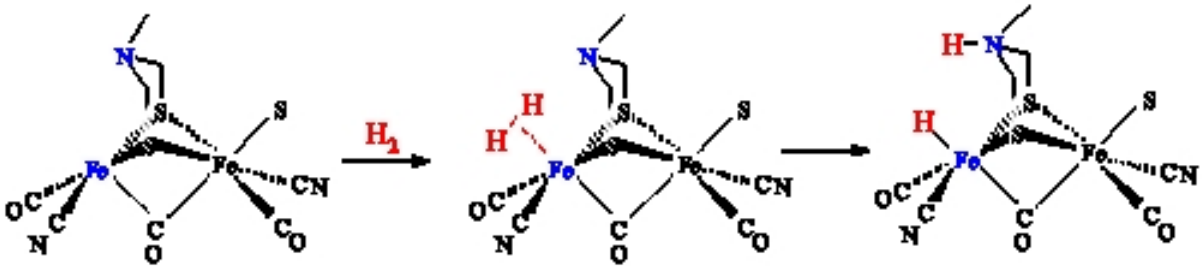
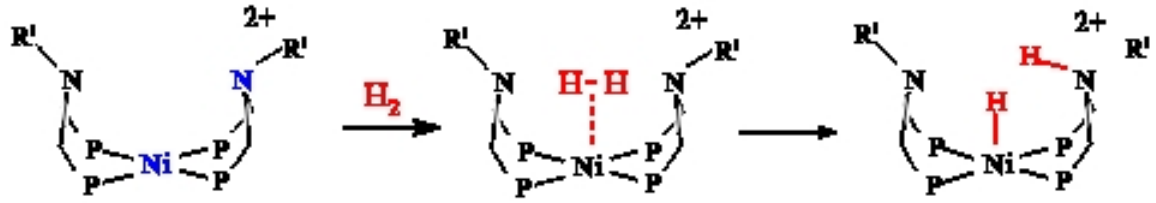
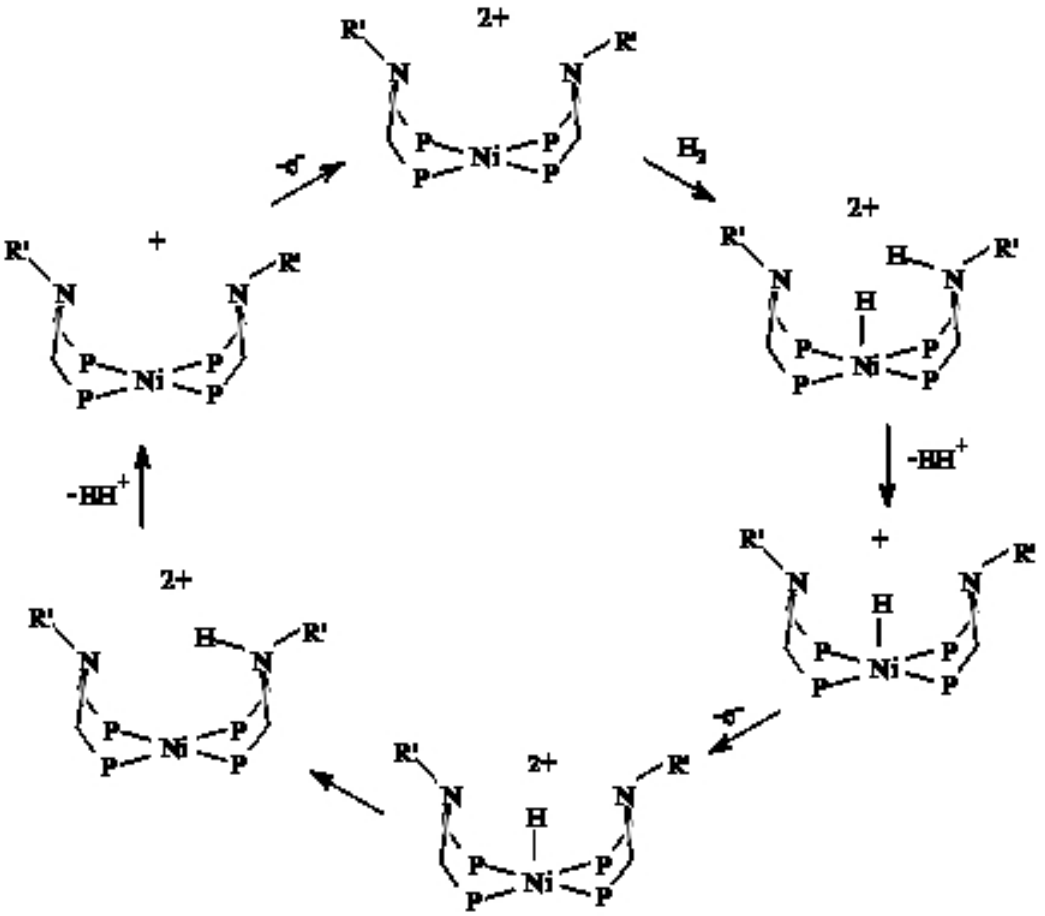


Nicolet, Y.; de Lacey, A. L.; Vernède, X.; Fernandez, V. M.; Hatchikian, E. C.; Fontecilla-Camps, J. C. *J. Am. Chem. Soc.* **2001,** *123*, 1596-1601.



Molecules Containing Both Hydride Acceptor and Base

Catalytic Cycle for H_2 Oxidation



Curtis, C. J.; Miedaner, A.; Ciancanelli, R. F.; Ellis, W. W.; Noll, B. C.; DuBois, M. R.; DuBois, D. L. *Inorg. Chem.* **2003,** *42*, 216-227.

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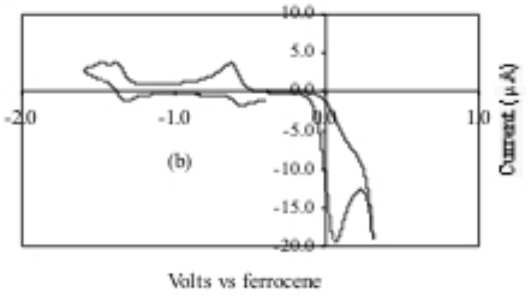
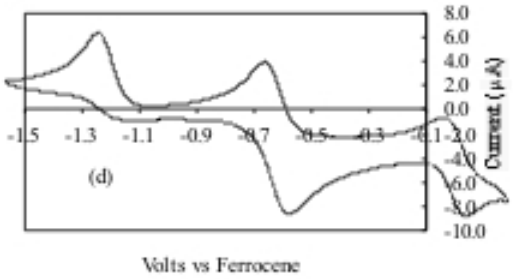
Electrochemical Results



Volts vs ferrocene



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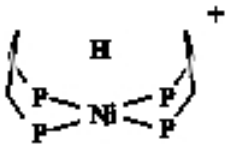
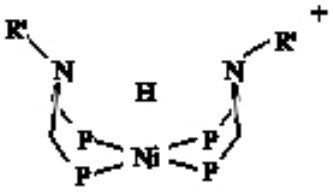
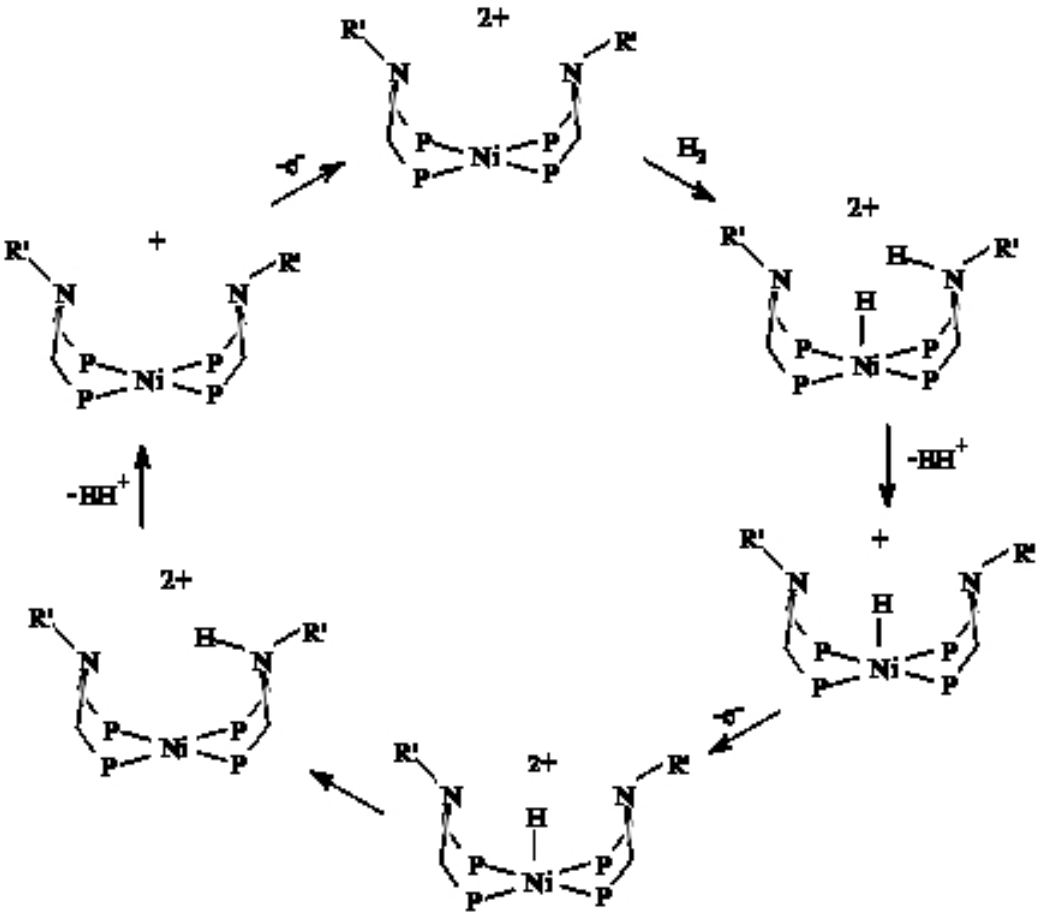


$[HNi(depp)_2]^+$



$[HNi(PNP)_2]^+$

Catalytic Cycle for H_2 Oxidation



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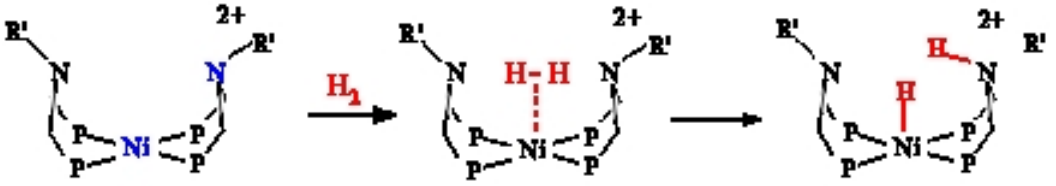
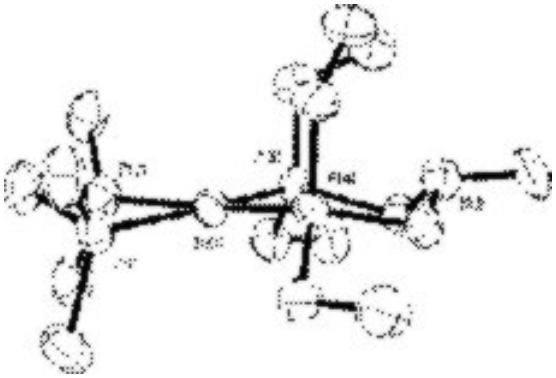
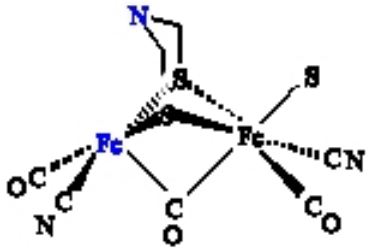
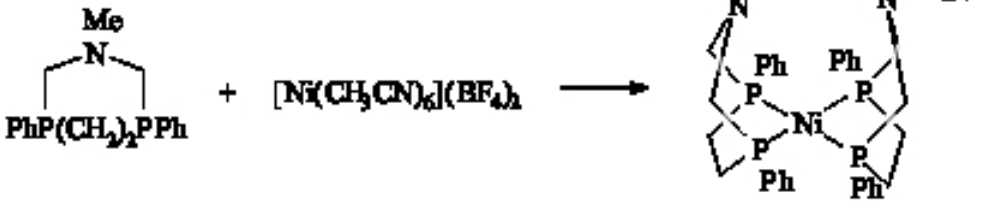
Design of Catalysts with Lower Barriers for H_2 Activation









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Summary

- Nickel-based molecular electrocatalysts for hydrogen oxidation have been developed
- They exhibit fast proton-coupled electron-transfer processes
- Designed catalysts need to be developed that will increase the rate of H_2 activation from 10 $M^{-1}s^{-1}$ to 10^3 - 10^4 $M^{-1}s^{-1}$

Acknowledgements

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