Electrochemical Hydrogen Separation Via Solid Acid Membranes

Alexander B. Papandrew^a, David L. Wilson^a and Thomas A. Zawodzinski^{a,b}.

> ^aChemical and Biomolecular Engineering University of Tennessee Knoxville, TN 37996

^bPhysical Chemistry of Materials Group Oak Ridge National Laboratory Oak Ridge, TN 37831

In the absence of a widespread and economically viable water electrolysis strategy based on renewables, the vast majority of the hydrogen produced for industry remains derived from reformed hydrocarbons. The separation of hydrogen from the undesirable byproducts in the reformate stream is a crucial operation in determining the cost of hydrogen fuel.

Hydrogen separation technologies must be highly selective for hydrogen, tolerant to reformate components (CO, CH₄), and above all, inexpensive. A class of materials that may satisfy these requirements are the superprotonic solid acids, a class of materials distinguished by thermally-activated polymorphic phase transitions to high-symmetry crystal systems associated with large increases in proton conductivity¹. One such material, CsH₂PO₄ (CDP), has been successfully deployed in air-breathing cells operating at 240 °C on fuel streams containing as much as 20% CO².

We investigated the suitability of CDP-based systems for hydrogen separation by constructing electrochemical hydrogen pumps operating at temperatures from 230 °C to 260 °C. Initial experiments were performed with symmetrical Pt-based electrodes under pure H₂. The response of these electrodes was well-described by the Butler-Volmer equation, and the temperature dependence of the electrode response yielded an activation energy for the HOR/HER of 21 kJ/mol and a minimum exchange current density of 1.5 mA/cm²_{Pt}.

Using the performance of Pt electrodes in pure H_2 as a baseline, we probed the effects of hydrogen dilution and CO poisoning on cell performance by using simulated reformate streams containing varying amounts of CO and CH_4 .

Finally, the possibility of employing non-Pt catalysts was explored. Non-Pt metallic catalysts were identified for both the HOR and HER reactions. We evaluated these catalysts in both symmetrical and asymmetrical cells. In the asymmetrical case, Pt was used to establish a reversible hydrogen pseudo-reference electrode, allowing probing of the candidate non-Pt HOR/HER catalysts using linear sweep voltammetry and impedance spectroscopy.

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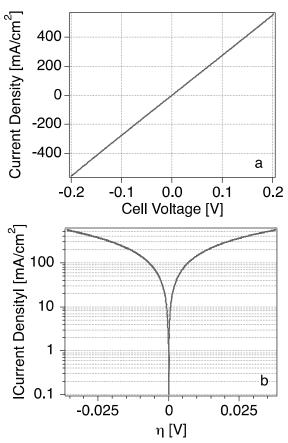


Figure 1. (a) Hydrogen evolution current from CDPbased MEA with symmetrical Pt-based electrodes operated at 250 °C in 30 sccm H_2/H_2 at atmospheric pressure and a dew point of 75 °C. (b) iR-free hydrogen evolution current from (a) in Tafel form.

References

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