Standard Operating Procedure

(SOP)

Measure H_2 Production and Gas (Mechanical) and Electrical Leakage

Test ID: HTE-P-27

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Date: 04/21/2020

Revision History

This page documents the revisions over time to the SOP. The most recent iteration should be listed in the row space, with consecutive versions following.

Date of Revision	Page(s)/Section(s) Revised	Revision Explanation
04/21/2020		First version completed

Outline/Table of Contents

1. Title Page
2. Table of Contents
3. Procedures
    a. Scope and Applicability
    b. Summary of Method
    c. Definitions
    d. Health & Safety Warning
    e. Cautions
    f. Interferences
    g. Personnel Qualifications / Responsibilities
    h. Equipment and Supplies
    i. Step by Step Procedure
        - Instrument or Method Calibration and Standardization
        - Sample Collection
        - Sample Handling and Preservation
        - Sample Preparation and Analysis
        - Troubleshooting
        - Data Acquisition, Calculations & Data Reduction Requirements
        - Computer Hardware & Software
    j. Data and Records Management
4. Quality Control and Quality Assurance Section
5. Reference Section

1. Title Page

This protocol presents the gas leakage and electrical leakage during efficiency measurement.

2. Table of Contents

n/a

3. Procedures

a. Scope and Applicability – measure gas leakage due to the existing imperfection in the cell and sealant.

b. Summary of Method – Determine the gas leakage by examining the change of the gas composition in the hydrogen electrode side.

c. Definitions –
OCV: open circuit voltage

d. Health & Safety Warning – as a combustion gas, hydrogen gas should be very carefully used with good ventilation and use steel pipe to ensure no leakage in all hydrogen stream. The leakage of hydrogen gas or the improper operation may cause combustion or other damages. Avoid contacting of high temperature surface on tube furnace.

e. Cautions – Hot sintering furnace and gas safety when using flammable gas.

f. Interferences – the leakage from the cell or sealant should be minimized to accurately measure the change of the gas composition.

g. Personnel Qualifications / Responsibilities – Basic skills on cell sealing, using electrochemical workstation, gas chromatography and flowmeter.

h. Equipment and Supplies – – flowmeter, cell testing fixture, tube furnace, Ag (or Au, Pt) wire, Ag (or Au, Pt) paste, electrochemical workstation, GC, stainless steel tube, hydrogen gas cylinder, oxygen gas cylinder, Ar gas cylinder

i. Step by Step Procedure <u>(LIST ALL PERTINENT STEPS, IN ORDER, AND THE MATERIALS NEEDED TO ACCOMPLISH)</u>

- Instrument or Method Calibration and Standardization – a standard complex gas (hydrogen included) cylinder is needed to calibrate the GC.

- Sample Collection – hydrogen electrode (cathode) support protonic ceramic electrolysis cell is fabricated by two steps. (1) cathode/electrolyte half cell is prepared by lamination of thick cathode and thin electrolyte green tape. The cells were cut out of the tapes and pre-sintered at 920 °C to remove the organic solvents. Finally, the half cells are sintered at high temperature to densify the electrolyte. (2) oxygen electrode is screen printed and sintered onto the electrolyte to obtain the final full electrolysis cells.

- Sample Handling and Preservation – (cell preparation) hydrogen electrode (cathode) support protonic ceramic electrolysis cell is fabricated by two steps. (1) cathode/electrolyte half cell is prepared by lamination of thick cathode and thin electrolyte green tape. The cells were cut out of the tapes and pre-sintered at 920 °C to remove the organic solvents. Finally, the half cells are sintered at high temperature to densify the electrolyte. (2) oxygen electrode is screen printed and sintered onto the electrolyte to obtain the final full electrolysis cells.

- Sample Preparation and Analysis – (sealing, configuration, before testing) (1) test fixture: it should contain the cell with capability of exposing electrodes to hydrogen and steam separately; (2) sample treatment:
    - The cell is sealed with glass powder followed by firing at the sealing temperature
    - The cell is reduced with hydrogen gradually
    - The gas leakage is examined by checking the gas flow rate and hydrogen concentration and then by GC to finalize the mechanical leakage

- Troubleshooting – A high open circuit voltage (>1.0 V) should be obtained when the cell is fully reduced with pure hydrogen. If the OCV cannot reach 1.0 V, severe leakage in the cell, sealing or the gas pipe may exist. A flow meter can be used to check the outlet gas in oxygen side and hydrogen side. If the flow rate is reduced, the gas leakage can be confirmed. If the voltage is abnormally high, e.g., larger than 1.2 V, it should be some problem with broken lead wires or peeled electrode.

- Data Acquisition, Calculations & Data Reduction Requirements –

- **- gas leakage:**
  The overall hydrogen gas leakage can be noted as *l* sccm. The tested baseline is always lower than settled hydrogen concentration (*x*>*y*). With an assumption that only hydrogen will be leaked due to the electrolyte membrane permeation, the Ar gas keeps a flow rate of $(1 - x) \times a$ while the hydrogen reduced to $x \times a - l$. As the Ar gas ratio

changed from $1-x$ to $1-y$,the overall gas flow is decreased to $\frac{(1-x)\times a}{1-y}$. A flowmeter is used to double-check the outlet gas is closed to calculated flow rate. The hydrogen leakage can be calculated as $l = a - \frac{(1-x)\times a}{1-y}$. The hydrogen leakage rate r is the drop ratio of hydrogen in the outlet, which can be calculated as $r = \frac{l}{a\times x}$.

- Computer Hardware & Software – relevant software for gas chromatography

j. Data and Records Management –Obtained GC information must be recorded clearly stating the conditions of the testing and the identification of the sample used.

4. Quality Control and Quality Assurance Section

The well-calibrated gas chromatography, fixed gas flow rate and concentration into reaction chamber, and good seal are the keys to obtain accurate measurement results. The leak examination is always required to check the accuracy before and after the measurements. The concentration of inert balance gas Ar in oxygen side is a key criterion to exam the sealing and evaluate the mechanical leakage. The Ar gas in oxygen side cannot permeate to the oxygen side, only mechanical leakage may cause the leakage, which result in sharp decrease in tested hydrogen concentration. [1] Using GC to measure the Ar in oxygen side, if the tested Ar gas concentration is δ with oxygen flow rate b, the Ar gas leakage is calculated to be $\delta \times b$ (sccm). The extent of the mechanical leakage can be characterized by $L = \frac{\delta\times b}{(1-x)\times a}$ with the assumption that the leaked gas has the same composition as the feed gas in hydrogen side[2]. When L is tested less than 5%, a good sealing can be assured.

5. Reference Section

R1. Choi, S.; Davenport, T. C.; Haile, S. M., Protonic ceramic electrochemical cells for hydrogen production and electricity generation: exceptional reversibility, stability, and demonstrated faradaic efficiency. *Energy & Environmental Science* 2019, 12 (1), 206-215.

R2. Ding, H. and Ding, D. et al., Self-sustainable protonic ceramic electrochemical cells using a triple conducting electrode for hydrogen and power production. *Nature Communications* 2020, 11, 1907.

R3. Chen, Chu-Sheng et al., Oxygen permeation through $La_{0.4}Sr_{0.6}Co_{0.2}Fe_{0.8}O_{3-\delta}$ membrane." *Chemistry of Materials* 2001, 13, 2797.

R4. Shao, Xin et al., Microstructure control of oxygen permeation membranes with templated microchannels." *Journal of Materials Chemistry A* 2014, 2, 410.