Development of measurement methodology at single cell and stack level for unstable hydrogen fuel impurity studies

HFC Nordic Conference
Sandviken, 26.10.2016
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Hydrogen Contaminant Risk Assessment

3-year EU project (FCH JU), 2014-17, coordinated by VTT
6 European partners, total budget of 3.907 M€.

The objectives of the project are

- to provide information to reduce cost of hydrogen fuel quality assurance (QA)
- to provide recommendations for revision of existing ISO 14687-2:2012 standard for hydrogen fuel in automotive applications

http://hycora.eu/deliverables.htm
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- HyCoRA strategy for cost reduction of H2 quality assurance (QA)
  - **Risk Assessment**, qualitative and quantitative, requires information from

  a) Real susceptibility for various poisonous species specifically for automotive applications

  b) Probabilities for QA failure in hydrogen production site and/or at HRS

  a) Concentration correlations between contaminant species in fuel
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The logical tree studying the contaminants that poison the catalyst

- Impurity
  - non-adsorbing
    - no effect
  - adsorbing
    - not reacting
      - Water soluble
        - Measure/estimate adsorption, wash-out, accumulation
    - reacting
      - Water insoluble
        - Measure/estimate adsorption, accumulation
      - New impurity and new analysis

SCONH₃, HCHO/HCOOH
CO NH₃, HCHO/HCOOH
NH₃, HCHO/HCOOH
HCHO/HCOOH permeating
Measurements with unstable H2 impurities

Unstable contaminants, such as HCHO/HCOOH, may not only to absorb to catalyst sites, but also

- **Accumulate** - into the H2 feed (recirculation system)
- **Decompose** - and form other, possibly harmful species
  - Contaminant concentration entering the cell should be monitored
- **Dissolve in water** - and exit the system
  - Collection and analyse of purge water needed
- **Permeate through Nafion membrane** - e.g. membrane gas dryer in gas analysis loop
  - Causing possible loss of contaminants before GC
  - A drying method 100% selective for water would be needed
  - Part may end up in the cathode side and oxidize there
Test systems

- **Single cell test station** (Green Light G60)
- Recirculation + GC Agilent 6890N

- **Stack test station** + recirculation + GC
- PowercellS2 10-cell stack (SN025)
- Low anode loading MEA: 0,05 mg$_{Pt}$/cm$^2$
Test system bench - stack

- Miniature automotive 1-2-kW system
- Three main subsystems: anode, cathode and coolant
- Additional instrumentation: contaminant injection line and gas sampling loop and N2 flushing system for corrosive impurities
- Anode is operated in dead end (constant fuel feed pressure) with purge
Impact of formaldehyde – stack measurement

- Two runs (4 and 3 hours) with ~1.6 ppm HCHO (and ~0.3 ppm CO) and 0.6 Acm⁻² using fuel utilisation of 99.5-99.6% (contaminant enrichment factor of 200-250)

- A very small (~ 10 mV) average voltage drop in 4 hours due to HCHO
  - In CO reference poisonings 1.86 ppm leads 50 mV average voltage drop in 67-71 min

- Current limit for HCHO (ISO 14687-2:2012) is 0.01 ppm

- A large CH4 increase (0 to 200 ppm) in anode recirculation loop → methanation of HCHO

- Some minor CO₂ increase/fluctuation in anode recirc loop. No changes in CO level.
Measurement conclusions with HCHO

- Measurement with HCHO are difficult as it decomposes quickly, is water soluble and permeable through membrane
  - Some question remains open regarding to the decomposition products (may decompose to CO)

- A major part (~50%) of HCHO seems to be methanated (still uncertain). Some part of HCHO (~10-15%) is dissolved in water and purged out with H2 purge.

- ~1.6 ppm (160 x limit in ISO 14687-2:2012) HCHO has probably a negligible effect
Acknowledgements
The research leading to these results has received funding from the European Union's Seventh Framework Programme (FP7/2007-2013) for the Fuel Cells and Hydrogen Joint Technology Initiative under grant agreement n° 621223.

Thank you

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