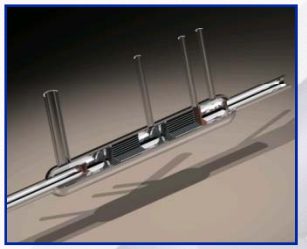

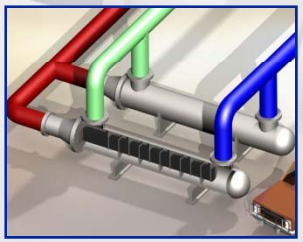

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Perovskite Adsorbents for Warm-Gas Arsenic and Phosphorus Removal

Erick J. Schutte, David A. Gribble, Jr., Sara L. Rolfe and Douglas S. Jack

11th Annual SECA Workshop

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Pittsburgh, PA.

Eltron Research & Development

- **Eltron Research & Development Inc.**
  - 60 patents (25 licensed)
  - Eltron Water Systems LLC
    - Commercialization of water purification technologies
  - Continental Technologies LLC
    - Design and fabrication of engineered systems and pilot plants
- **Eltron Areas of Expertise**
  - Energy: Fuels, Fuel Reforming, Membranes
  - Materials & Catalysts
  - Air and Water Purification
  - Electrochemistry

ELTRON WATER SYSTEMS

CONTINENTAL TECHNOLOGIES

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DOE SBIR Project Objectives

- **Formulate and synthesize perovskite-based adsorbents containing elements that very strongly bind arsenic and phosphorus, two elements known to irreversibly poison nickel catalysts in SOFCs.**
- **Demonstrate rapid uptake of arsenic and phosphorus by the adsorbents.**
- **Demonstrate retention of arsenic and phosphorus by the adsorbents under conditions expected upstream of commercial SOFCs.**

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Specific Project Goals

- Reduce arsenic and phosphorus in gas streams from ppmv to low ppbv quantities.
- Employ synthetic water-gas-shift mixtures during testing.
- Operate reactor at pressures up to five atmospheres.
- Operate reactor at temperatures between 250-450°C.

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Key Results*

***All data acquired during 3-hour test runs, with a 1-hour H_2 reduction phase, at ambient pressure and 300°C using an argon sweep stream containing ~550 ppm arsenic and ~140 ppm phosphorus flowing at 2,000 hr^{-1}.**

- **Preferred sorbent demonstrated As and P adsorption efficiency of at least 73%.**
- **Preferred sorbents demonstrated As and P adsorption capacity of at least 4.5%.**
- **Commercial Cu/ZnO sorbent yielded 47% A.E. and 1% A.C. respectively.**
- **Preferred sorbents contain no Noble metals and are projected to cost <$5/lb.**

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Contaminant Background

- **Affect ability of Ni in SOFCs to promote electrochemical reactions.**
  - Binding on Ni surface reduces active sites for H_2 and CO adsorption and inhibits dissociation of H_2.
- **Affect the electrical conductivity in SOFCs**
  - As and P form irreversible Ni-As and Ni-P solid phases which leads to a loss of electrical percolation in anode support.
- **DOE Polishing Filter Technical Targets**
  - Arsenic and Phosphorus - <20 ppbv
  - Sulfur - <60 ppbv (Previous Phase I and II at Eltron using similar adsorbents has proven successful for regenerable sulfur polishing)

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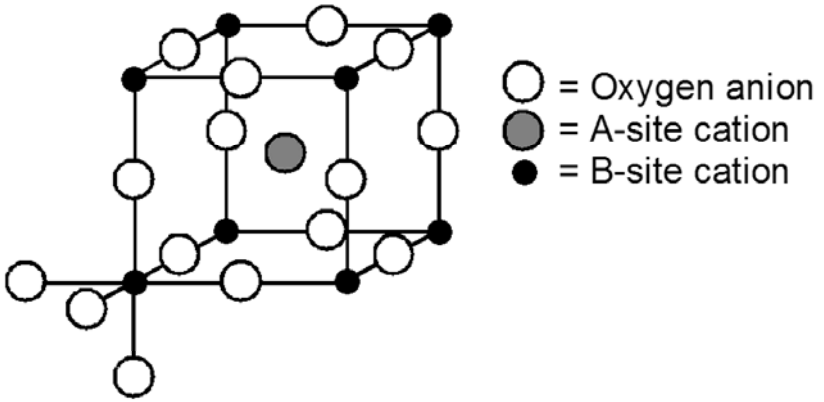
Design of Sorbents

- Focus on incorporating metals which form stable arsenides and phosphides into Perovskite (ABO_3) based materials.
- Different A-site (large metal cation) and/or B-site (small) atoms in same sorbent leading to disordered variants and mixed phase ceramics.



- High oxygen mobility, and thus the lower stability of oxides, within Perovskite structures increases driving force for formation of M-As and M-P solid phases on adsorbents.

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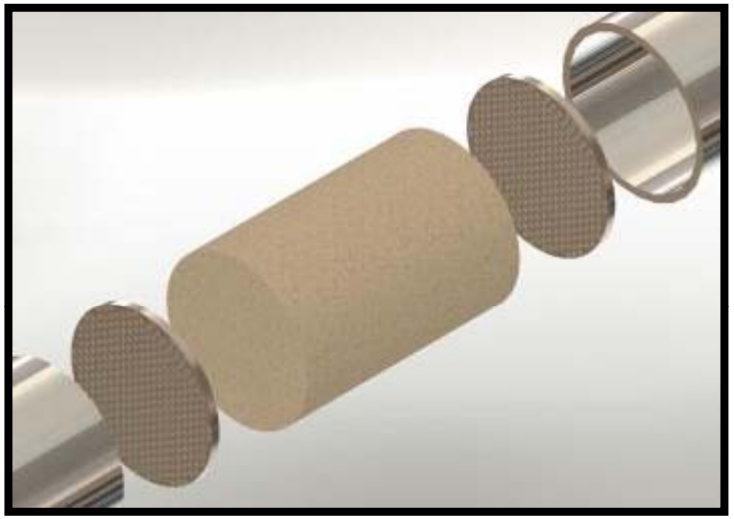
Initial Testing Reactor Designs

- **Breakthrough Reactor**
  - Employ Elemental As and P.
  - Vaporize Elements, Use Inert Sweep.
  - Ni-coated Coupons both Pre- and Post-Sorbent Bed.



- **High Pressure/WGS Reactor**
  - Same Testing Strategy as Breakthrough Reactor
  - Capable of Handling up to 5 Atm Pressure
  - Capable of Employing Simulated WGS Stream.

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Initial Testing Reactors

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Early Phase I Successes and Issues

- **Successes**
  - Synthesized and Characterized 16 Perovskite and Perovskite-like Adsorbents.
  - Preferred Sorbents Left No Trace of As or P on Post-Reactor Coupon.
  - Preferred Sorbents Out-Performed Commercial Cu/ZnO Sorbent.
- **Primary Issues Which Developed**
  - Contaminant Control – Difficulty in Generating Consistent Concentrations of As and P.
  - Quantifiable Data – Techniques for Quantifying Post-Sorbent Bed As and P Concentrations Needed Refining.

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Late Project Reactor Re-Design

- **Focus on Steady, Consistent Flow and Quantifiable Contaminant Concentration.**
  - **Replaced elemental As and P with arsine and phosphine gas (each 10ppm in hydrogen).**
  - **Equip tanks with mass flow control.**
  - **Acquired arsine sensor for post-reactor flow sampling.**
  - **Upgraded safety measures (hydrogen sensors, NaOH bubbler for scrubbing post-reactor gas).**

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State of Technology and Phase II Focus

- **Reactor and Instrumentation Upgraded**
  - More Precise Flow Control Using Arsine and Phosphine
  - Novel Detection System Capable of Measuring As and P Below 20ppbv
- **Phase II Focus**
  - Simulated WGS Stream Addition
  - Sulfur Addition (H_2S)
  - Independent Testing by Leading SOFC Company
  - Pilot Plant Sorbent Scale-Up and Evaluation by Commercial Sorbent Production Company

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Acknowledgments

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DE-FG02-07ER84666, Perovskite Adsorbents for Warm-Gas Removal of Sulfur

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