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Sulfuric Acid Decomposition Experiments for Thermochemical Production from Nuclear Power

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NNSA National Nuclear Security Administration

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Sandia Programmatic Goals

- Develop and construct a H_2SO_4 decomposition process with a production capacity of

  ~ 200 L-SO_2/hour (equivalent 200 L-H_2/hour).

- Integrate the Sandia H_2SO_4 decomposition process with the two other sections of the S-I cycle, being developed by General Atomics Corporation and the French Commissariat a l'Energy Atomique (CEA), into a demonstration scale process.

- The full S-I cycle is being assembled at the General Atomics Facility in San Diego, and will be operated by the end of FY07.

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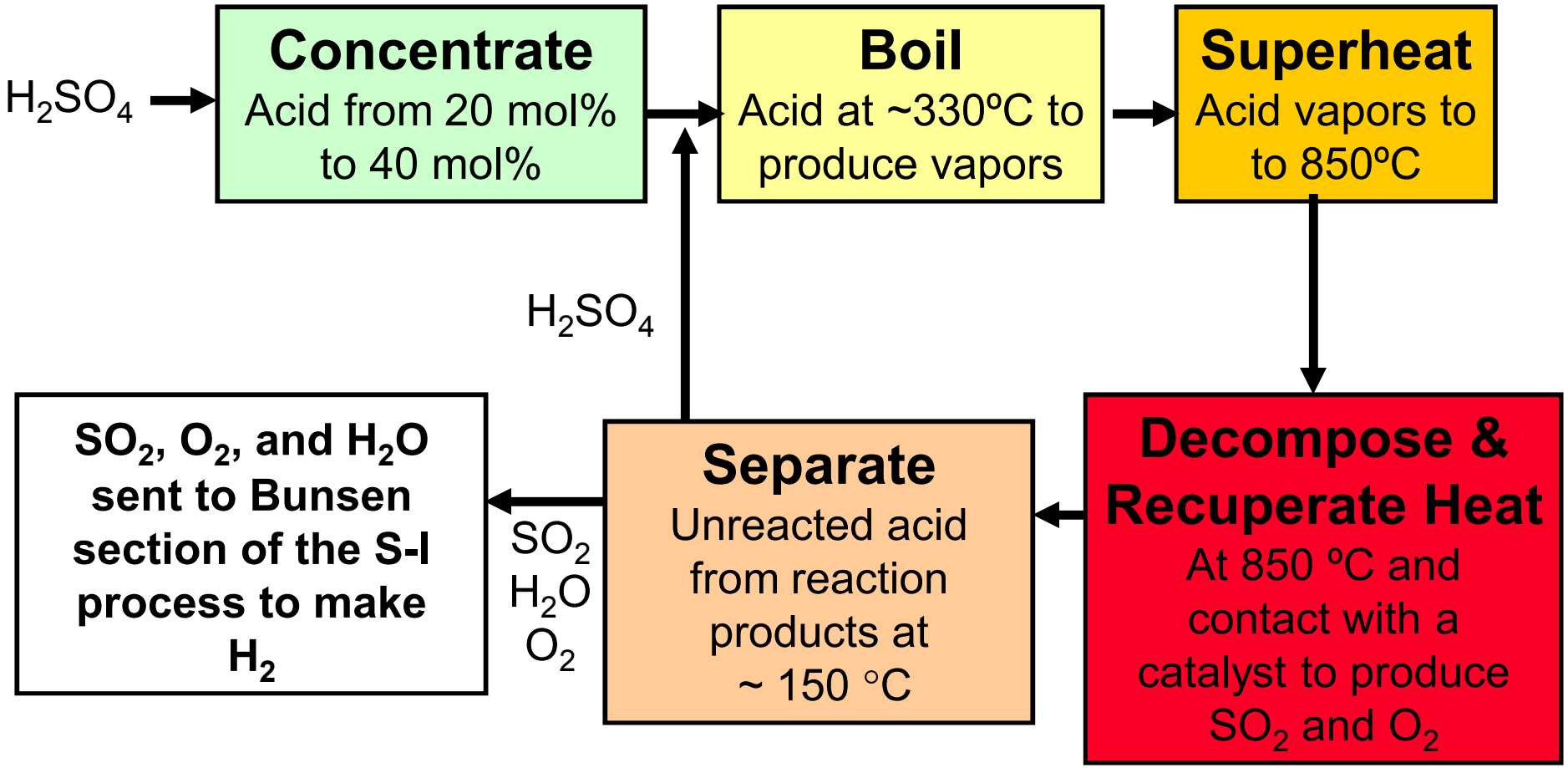
Sulfuric Acid Section
Five Step Process



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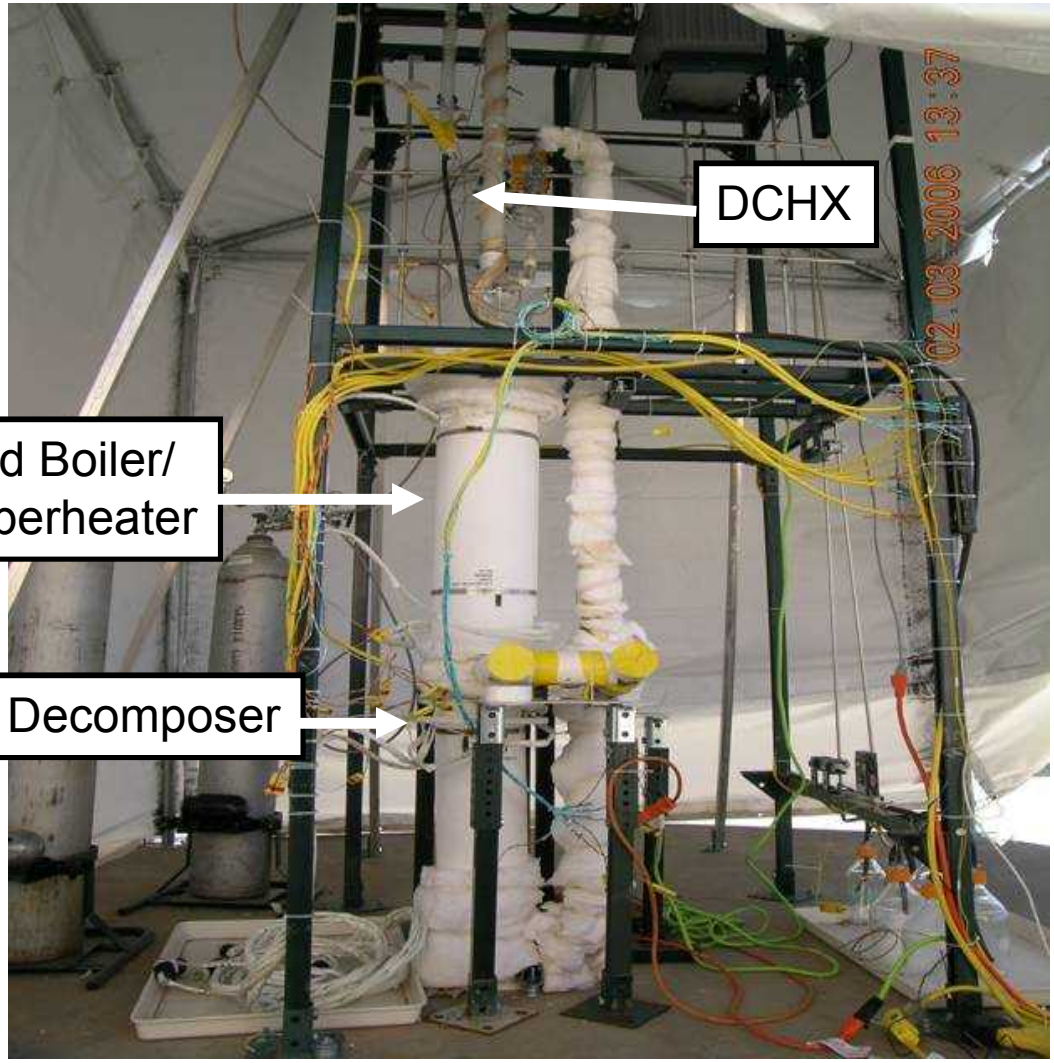
Past Experiments: Apparatus Constructed Mostly of Metals for Production of Sulfur Dioxide Using Direct Contact Heat Exchanger to Recycle Unreacted Acid and Recuperate Heat

- Three sections of the process were bolted or swag-lock fitted together for ~ 500 L-H_2/hr

- Constructed of Hastelloy, glass, and ceramics

- No observable corrosion, but delicate and difficult to operate and scale up.



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Sandia Silicon Carbide Integrated Acid Boiler/Superheater/Catalytic Decomposer

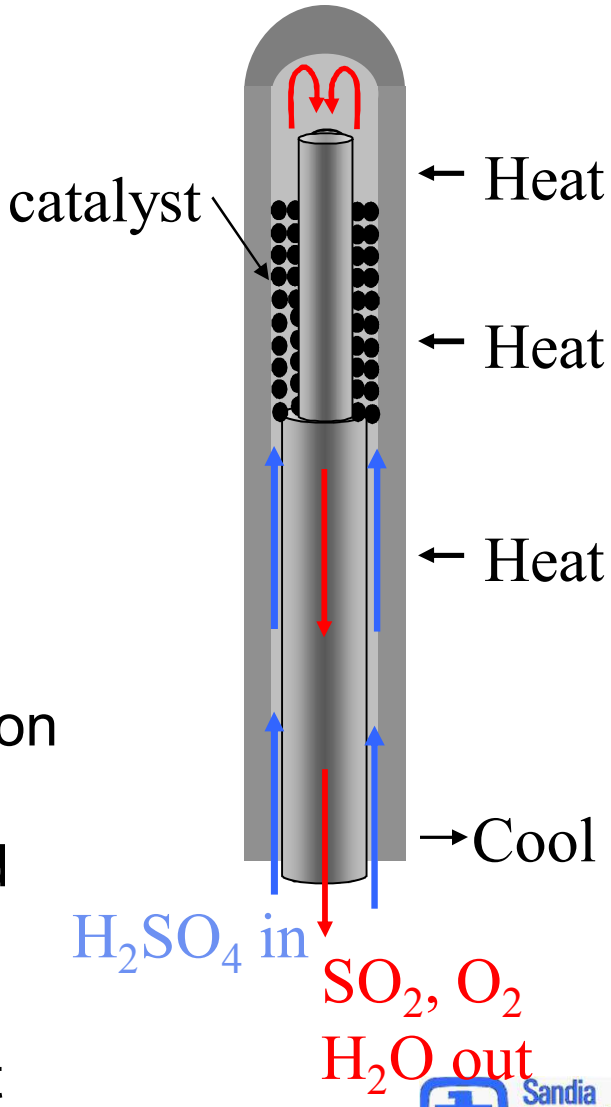
- Silicon carbide is extremely resistant to corrosion by acids

- Bayonet HX design. Heat from gaseous products traveling down the center tube is recuperated

- Bayonet design eliminates all high temperature connections.

- All other equipment in the acid decomposition process operates at low temperature. This allows use of commercial glass & teflon-lined components

- Solves corrosion problem but limits catalyst access for measurements and replacement



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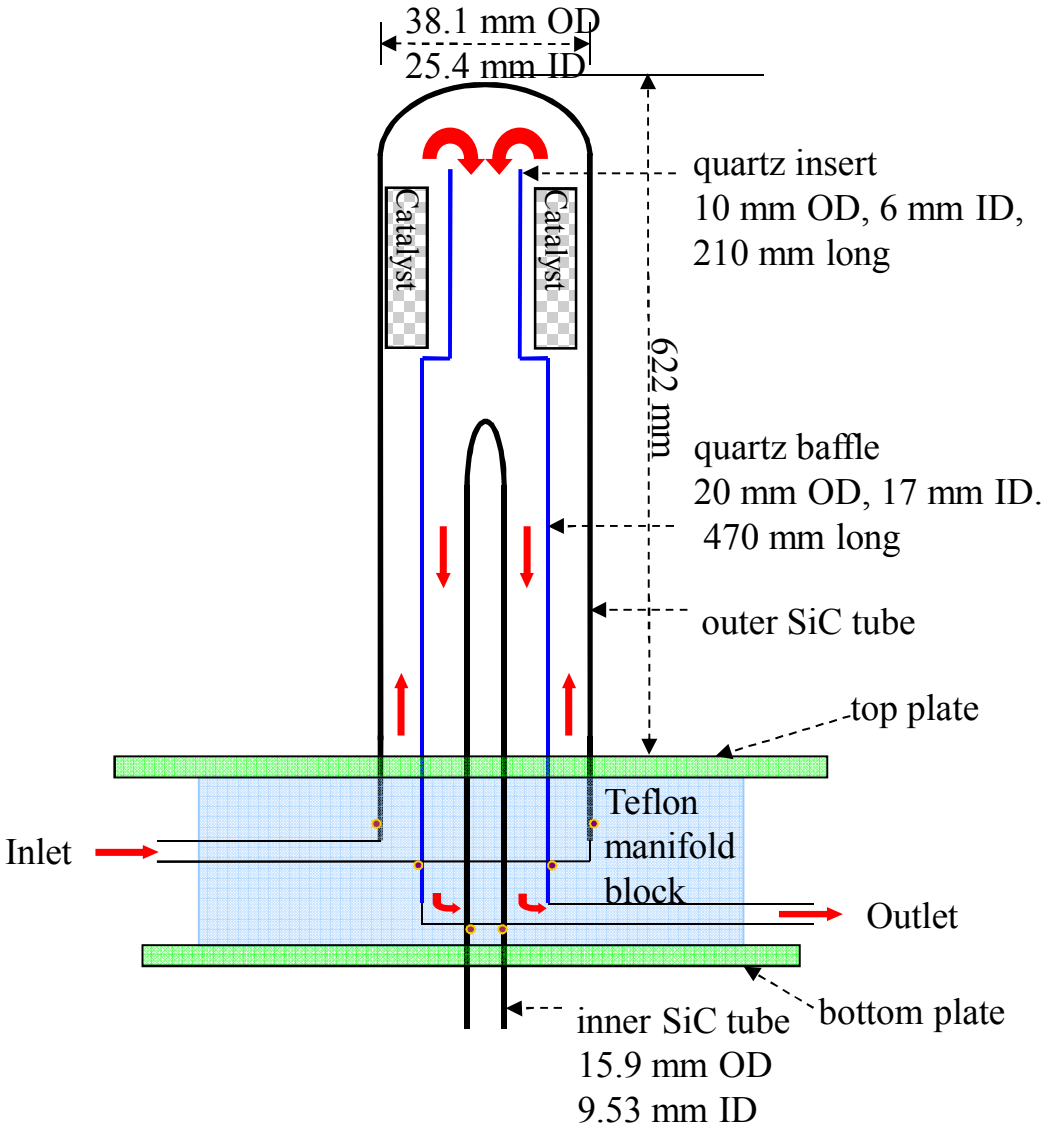
Bayonet & Teflon Manifold Schematic



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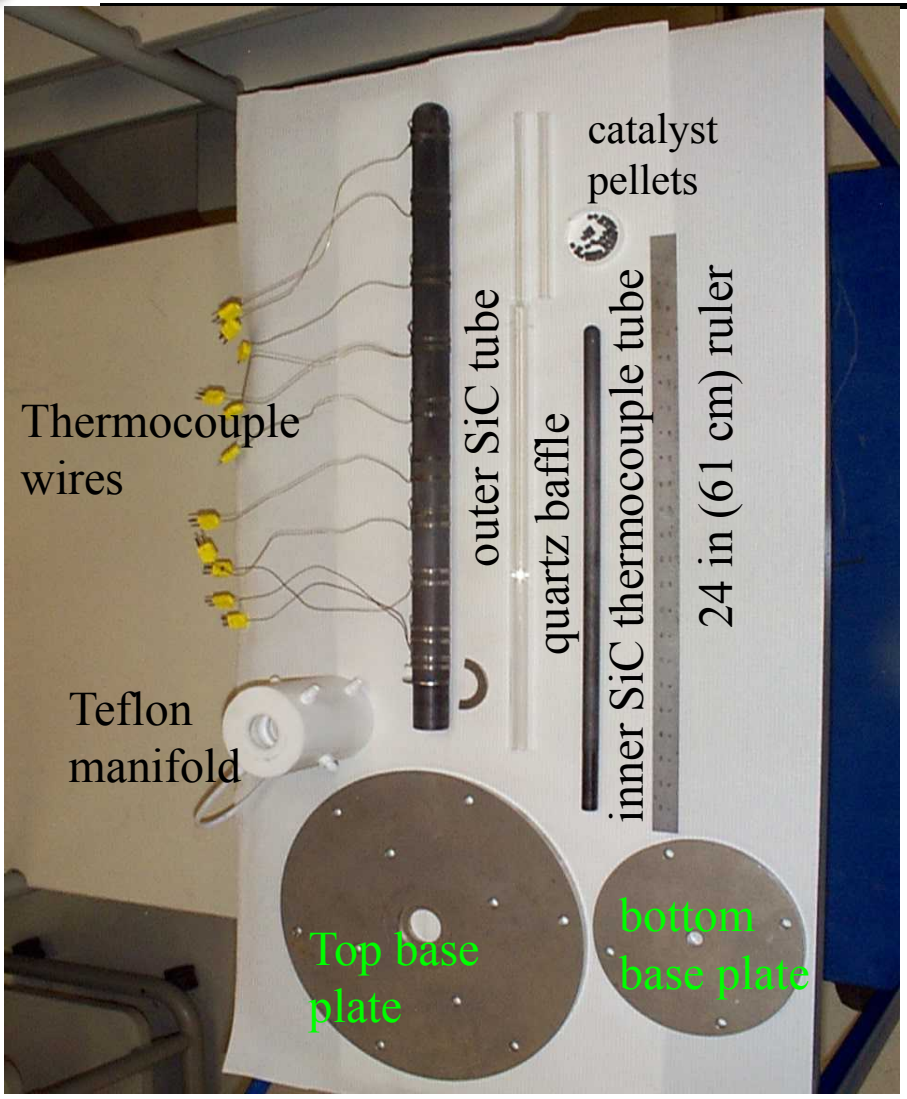
Major Components & Assembly (Half-Scale Unit)



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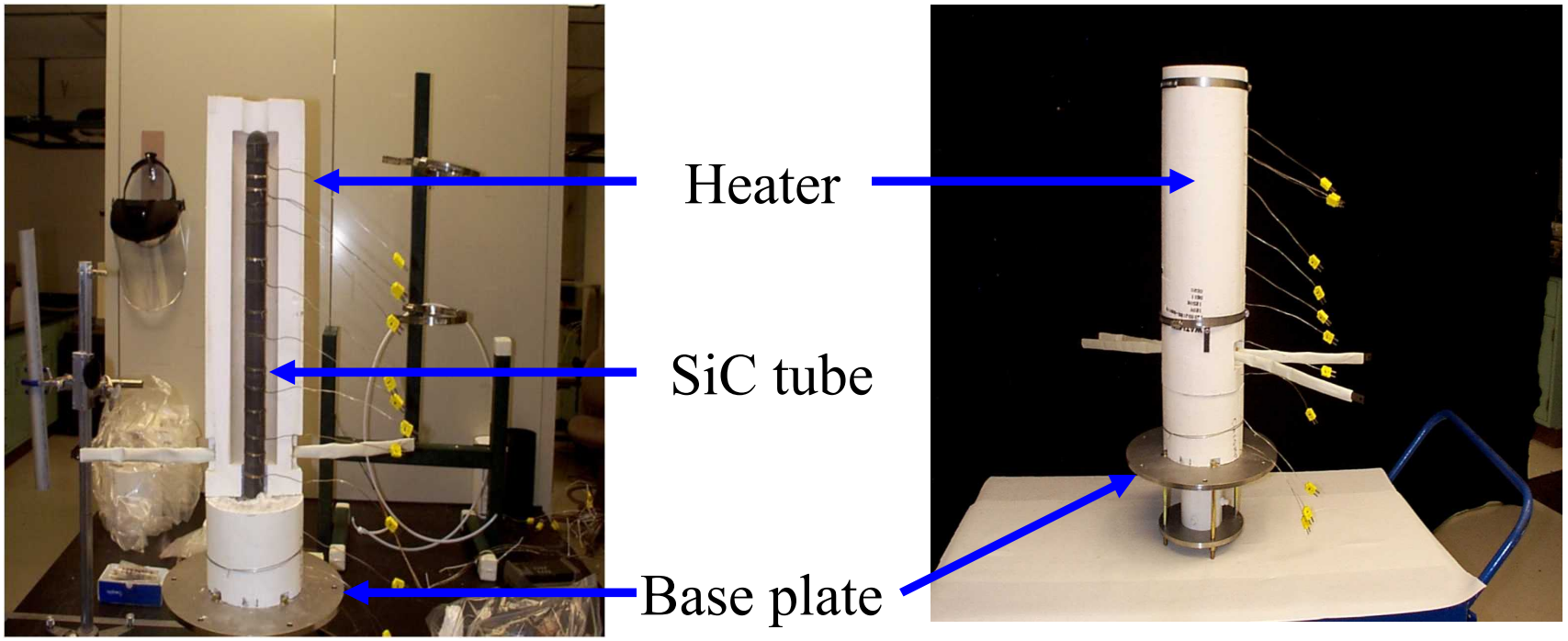
Sandia Silicon Carbide Integrated Acid Boiler/Superheater/Catalytic Decomposer



Initial Test Configuration

- 686 mm experimental apparatus (Half-scale)
- Heat provided by ceramic fiber heaters
- Tested at 850ºC, 4 bar with 40 mole percent sulfuric acid

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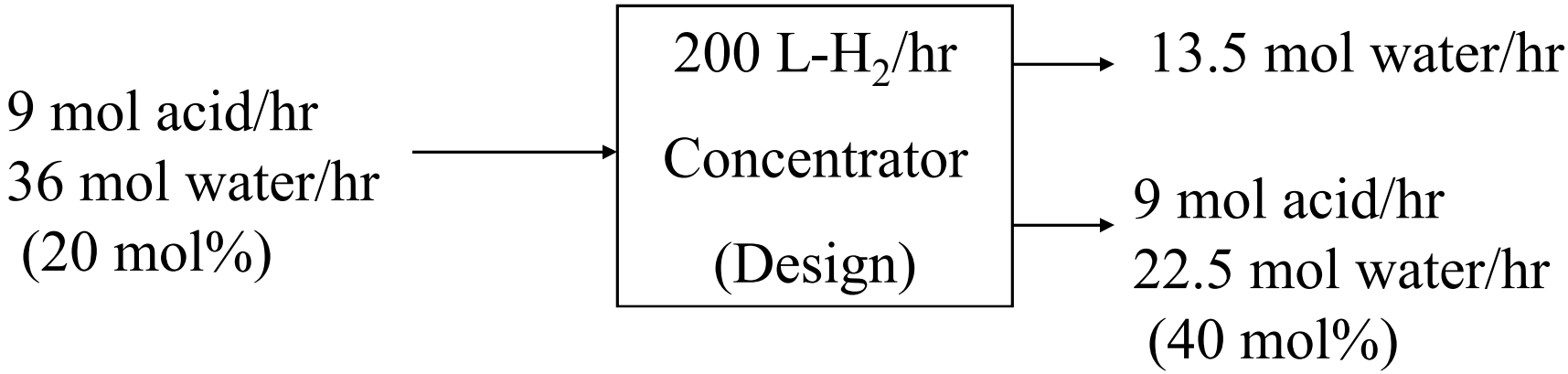
First Test of Coupled Vacuum-Concentrator and SID

- **Concentrator operated at 150 °C and 0.1 bar. Acid concentrated from 20 to 38 mol% at 14 mol/hr. This flow rate and concentration is adequate to process acid for 200 L-H2/hr.**



- **Operated at exterior temperature of 850 °C and 3 – 5 bar.**

- **Expected ~ 55% maximum conversion at 750 °C. Measured ~37% conversion which will require additional recycling.**

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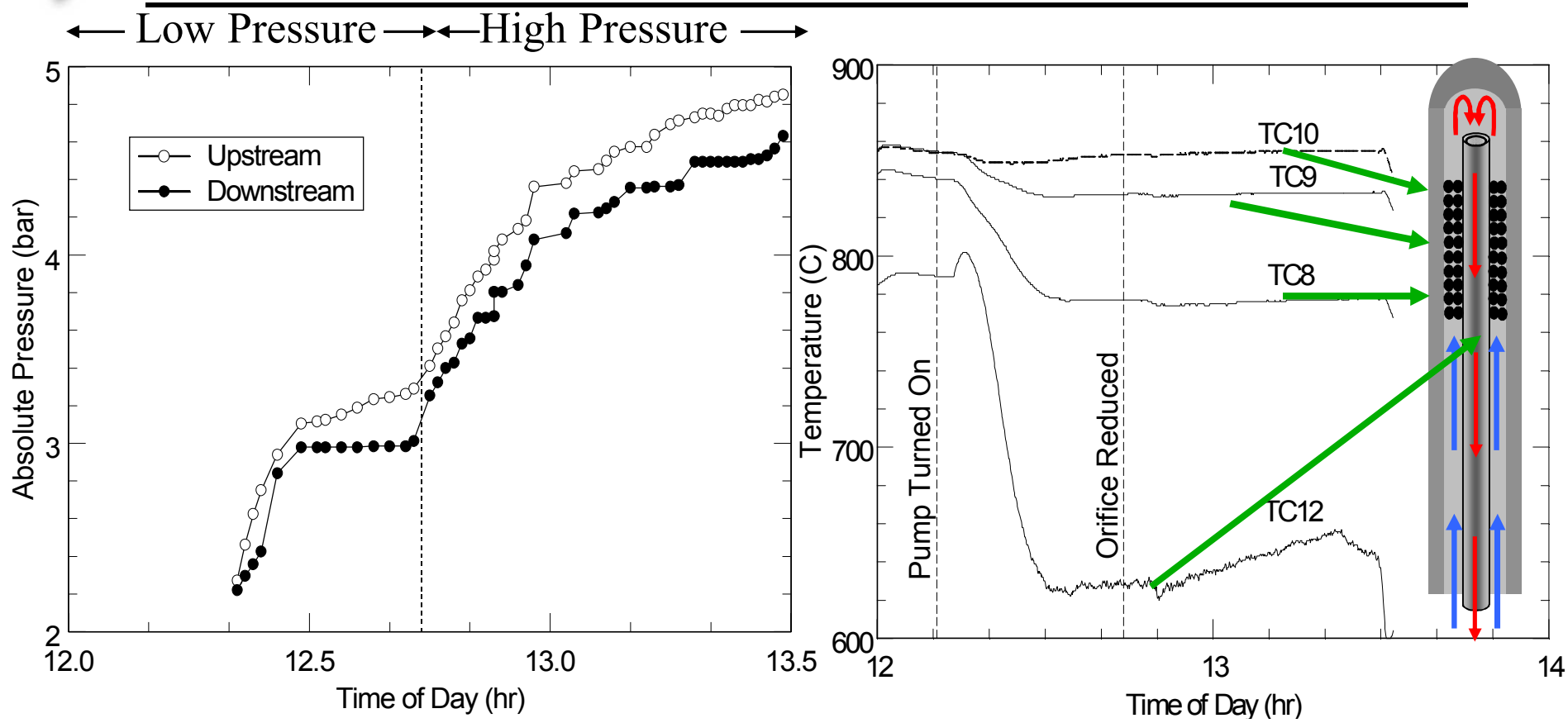
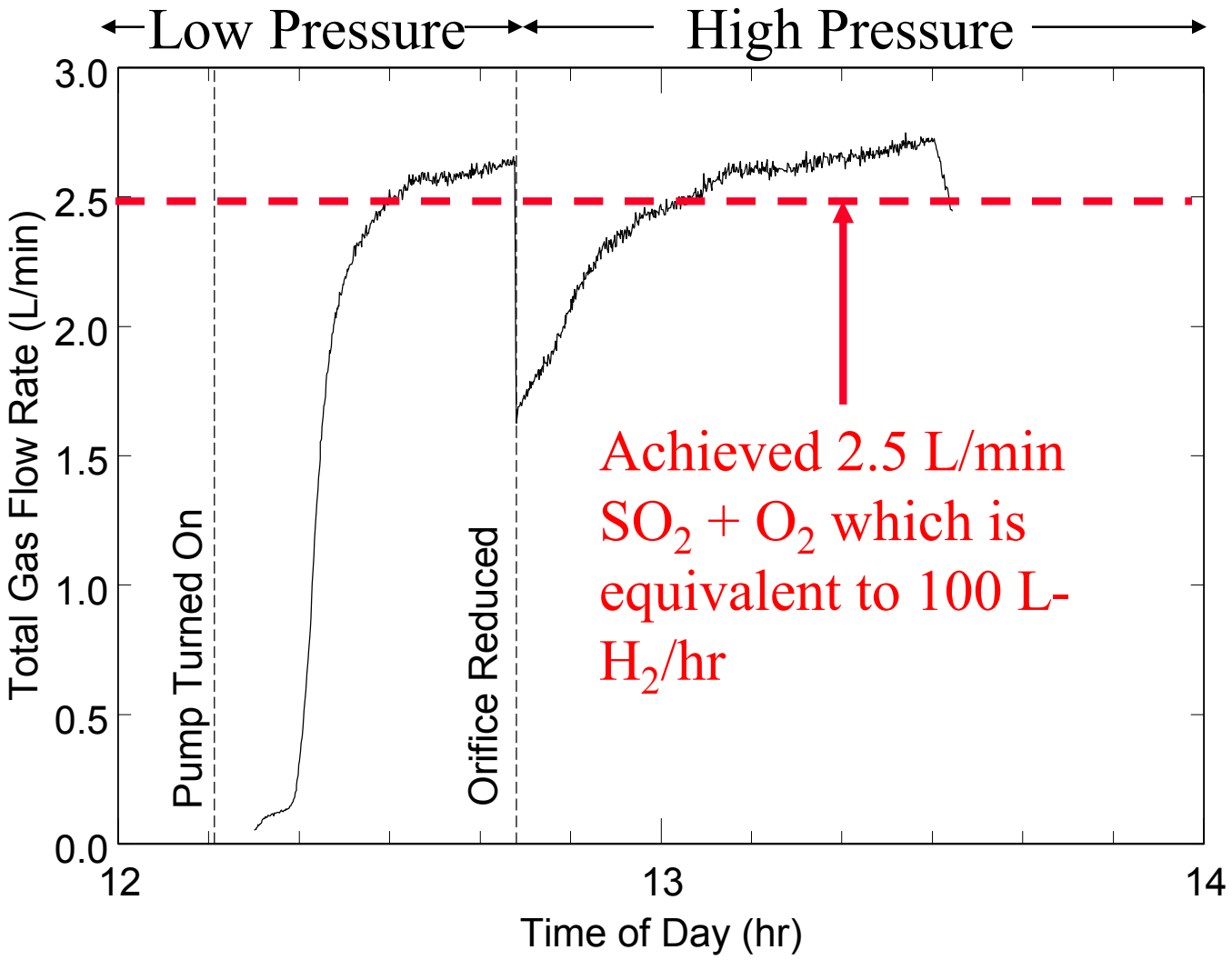
SID Pressure & Temperature



- Down stream pressure can be set, but upstream pressure building may be due to catalyst degradation. Catalyst and internal flow paths to be examined and reconfigured as needed.

- Exterior temperatures used to drive model. Internal temperature in down-flow agrees

Gas Flow Rate of $SO_2 + O_2$



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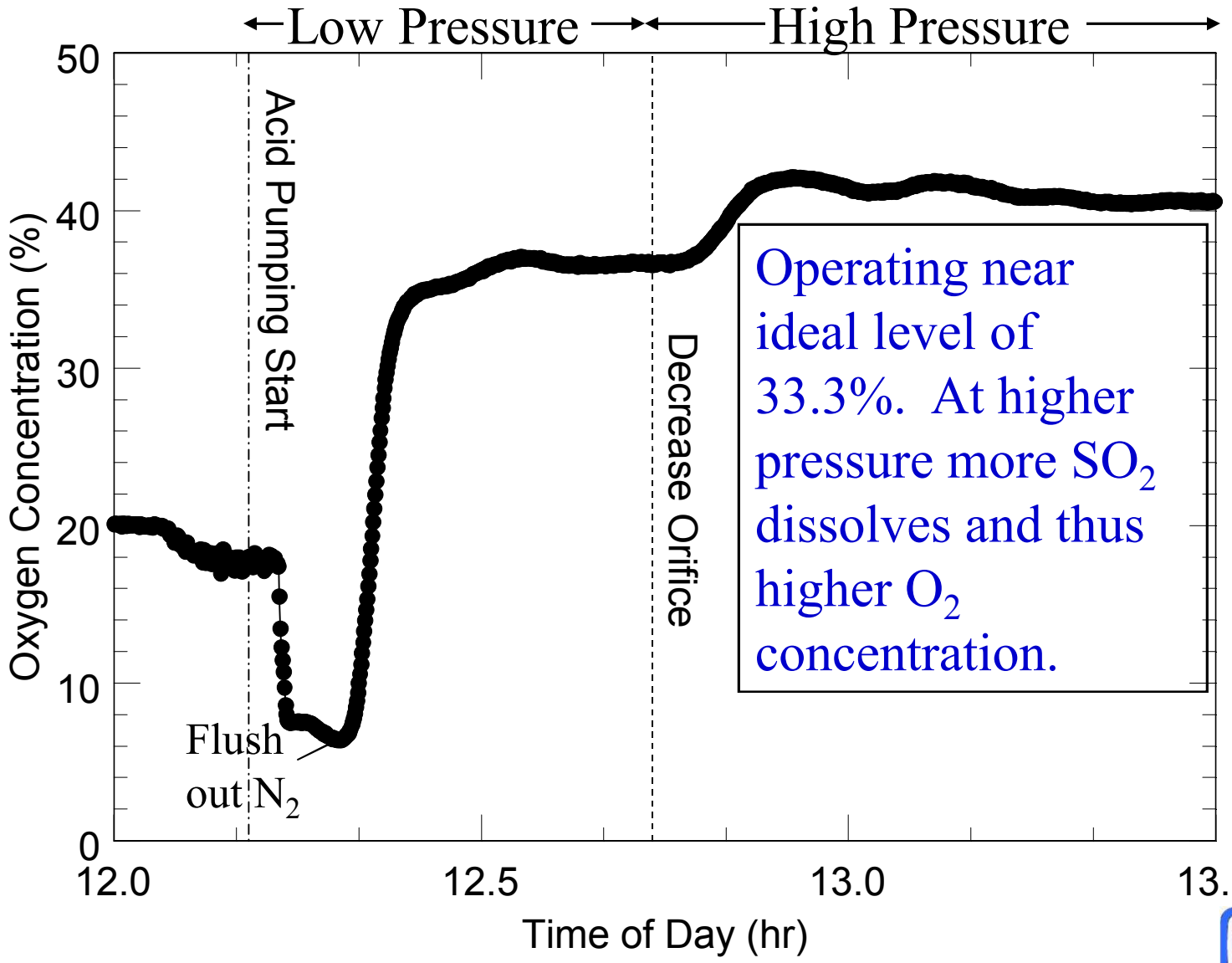
Effluent Oxygen Concentration



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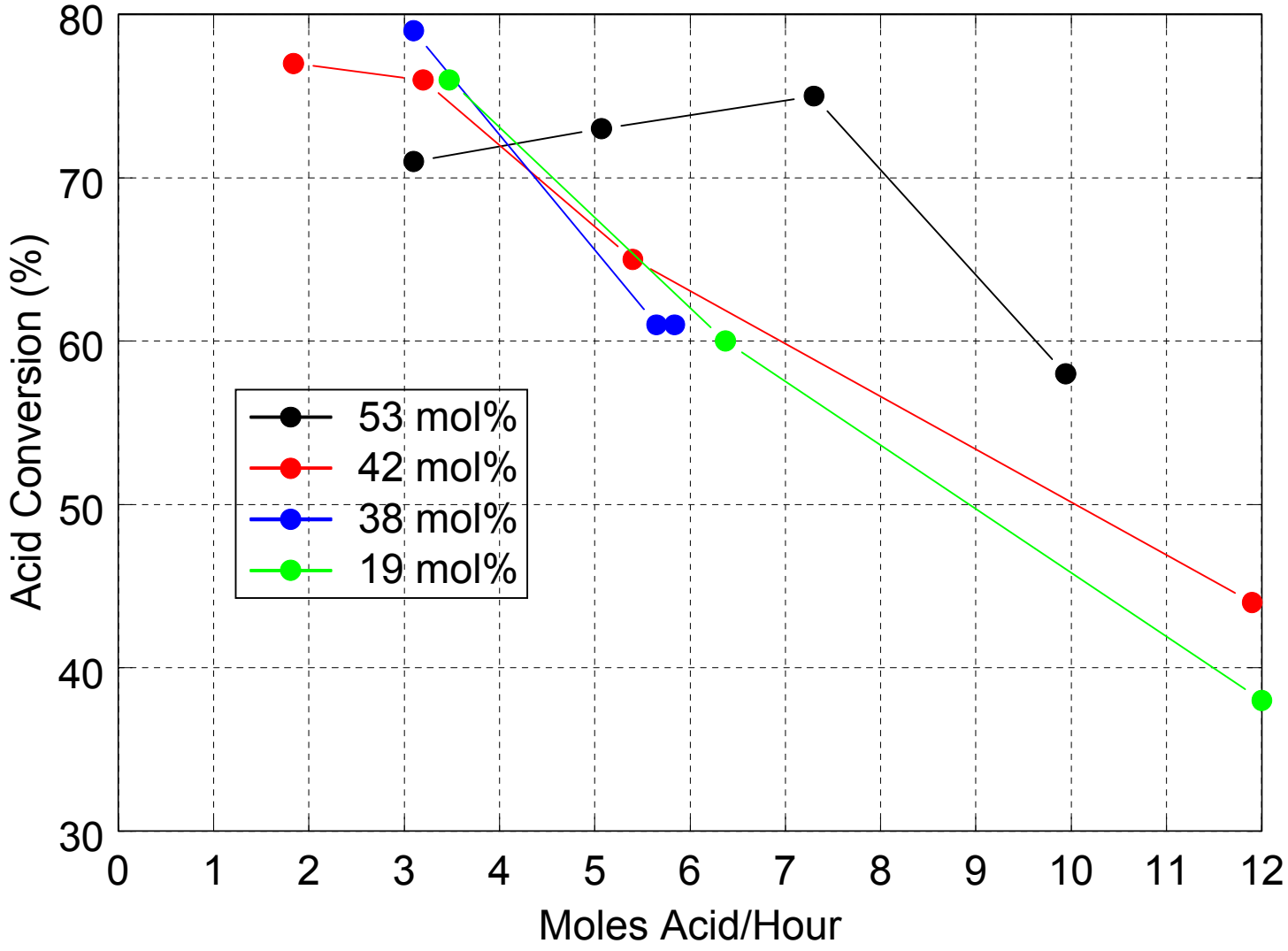
Performance Testing of Half-Scale SID

- Vary acid concentrations from 19 mol% to 53 mol%
  - ILS designed to receive from Bunsen reactor 20 mol% at 9 moles/hr for 200 L/hr of H2 production
  - Sulfuric acid section will concentrate acid to 40 mol% for bayonet decomposer.

- Operated at exterior temperature of 850 °C and atmospheric pressure.

- Maximum theoretical conversion ~ 86% at 850 °C. Measured ~78% conversion at slowest flow rate.

Acid Conversion
(Half-Scale, Ambient Pressure)



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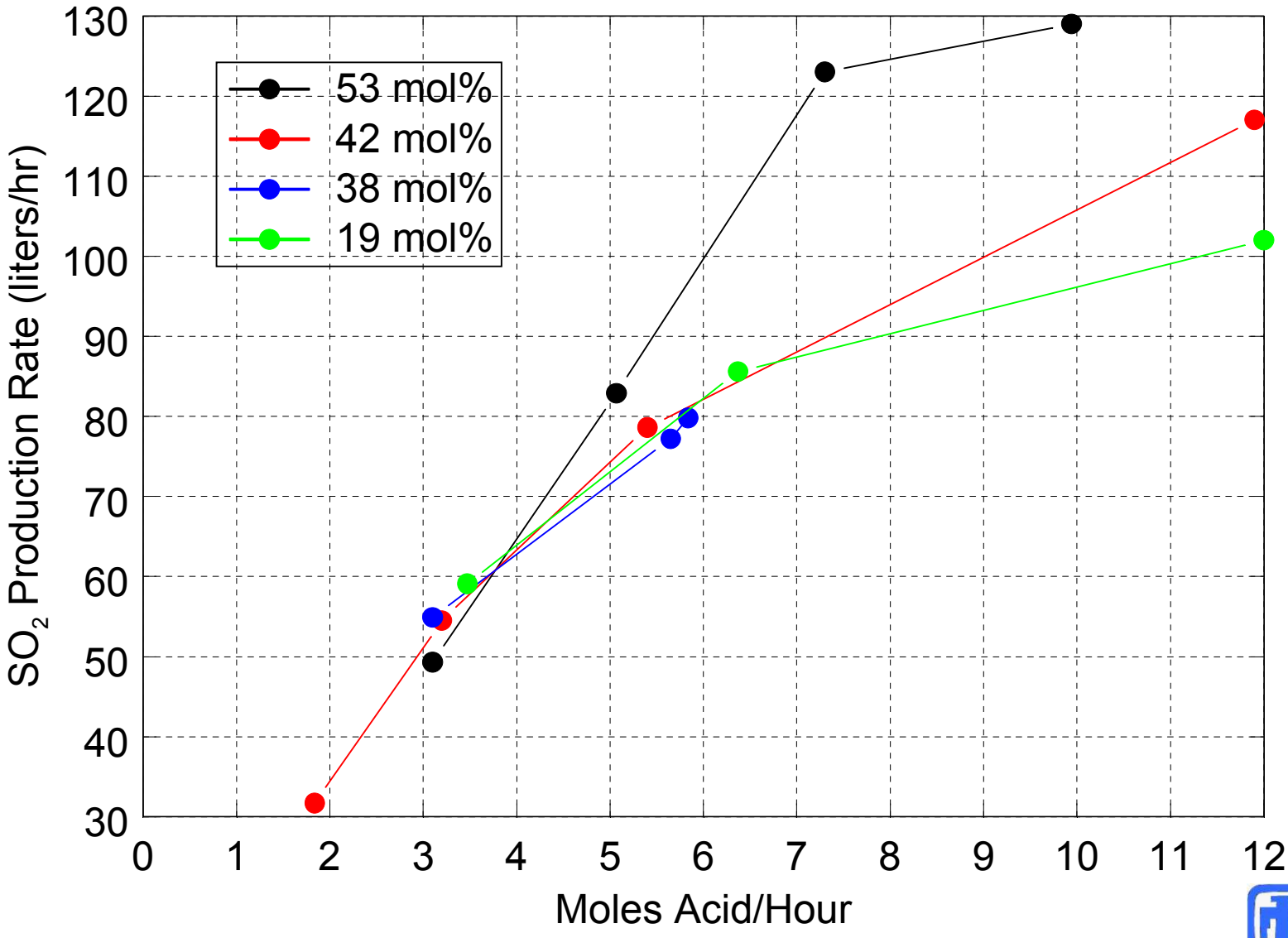
SO_2 Production Rate (Half-Scale, Ambient Pressure)



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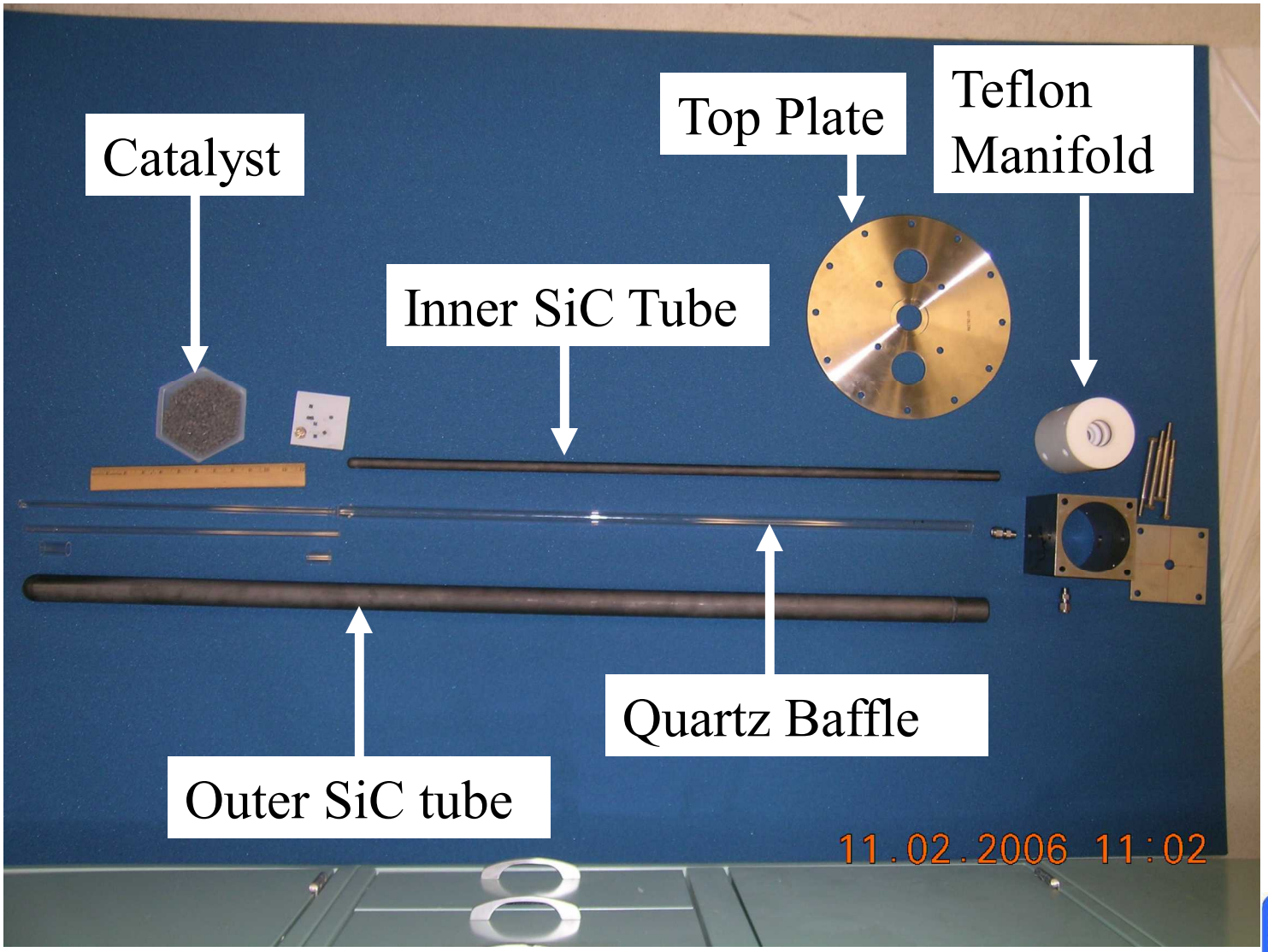
Disassembled 1372 mm Bayonet (Integrated Lab-Scale)



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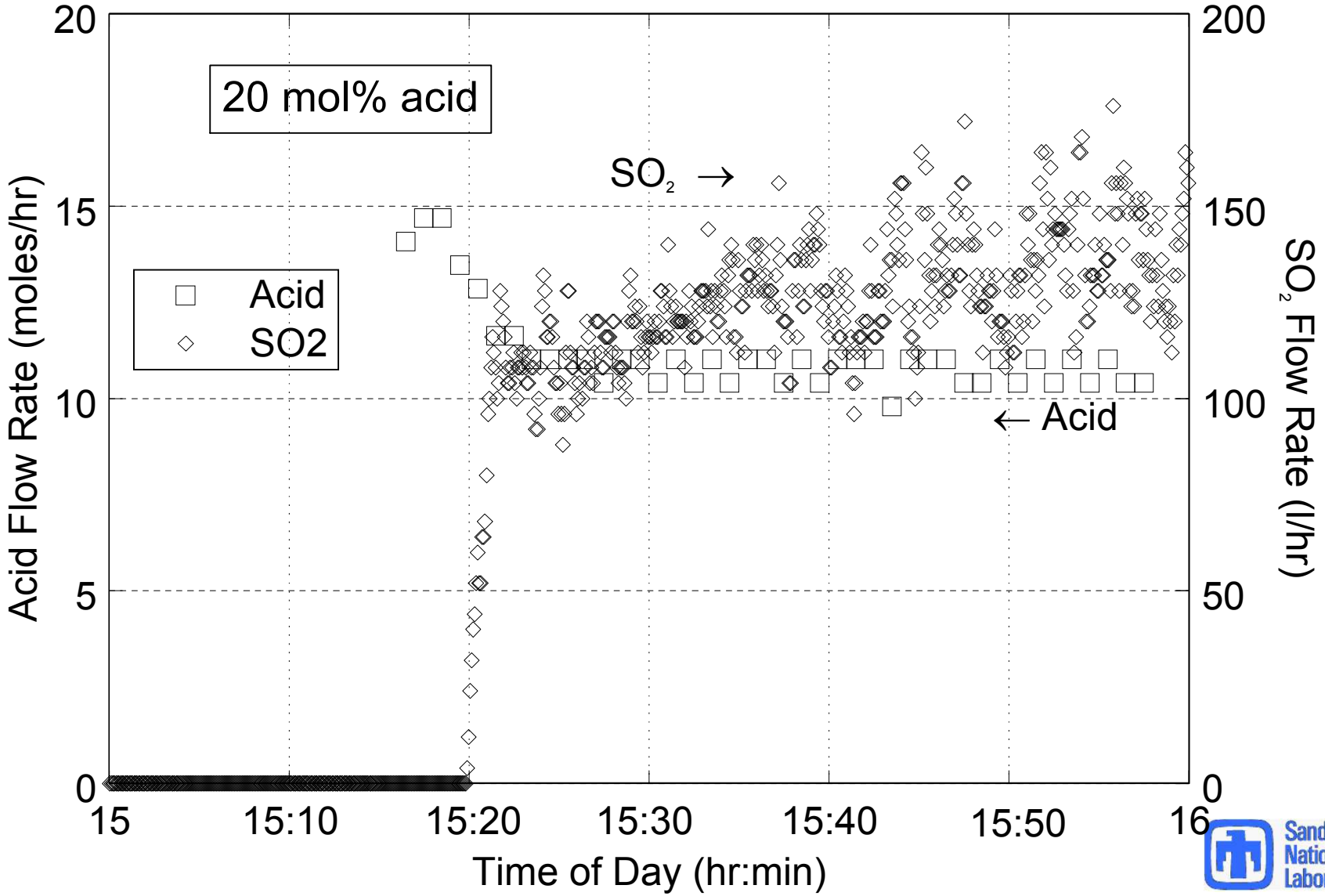
1372 mm Bayonet
20 mol % Acid Feed



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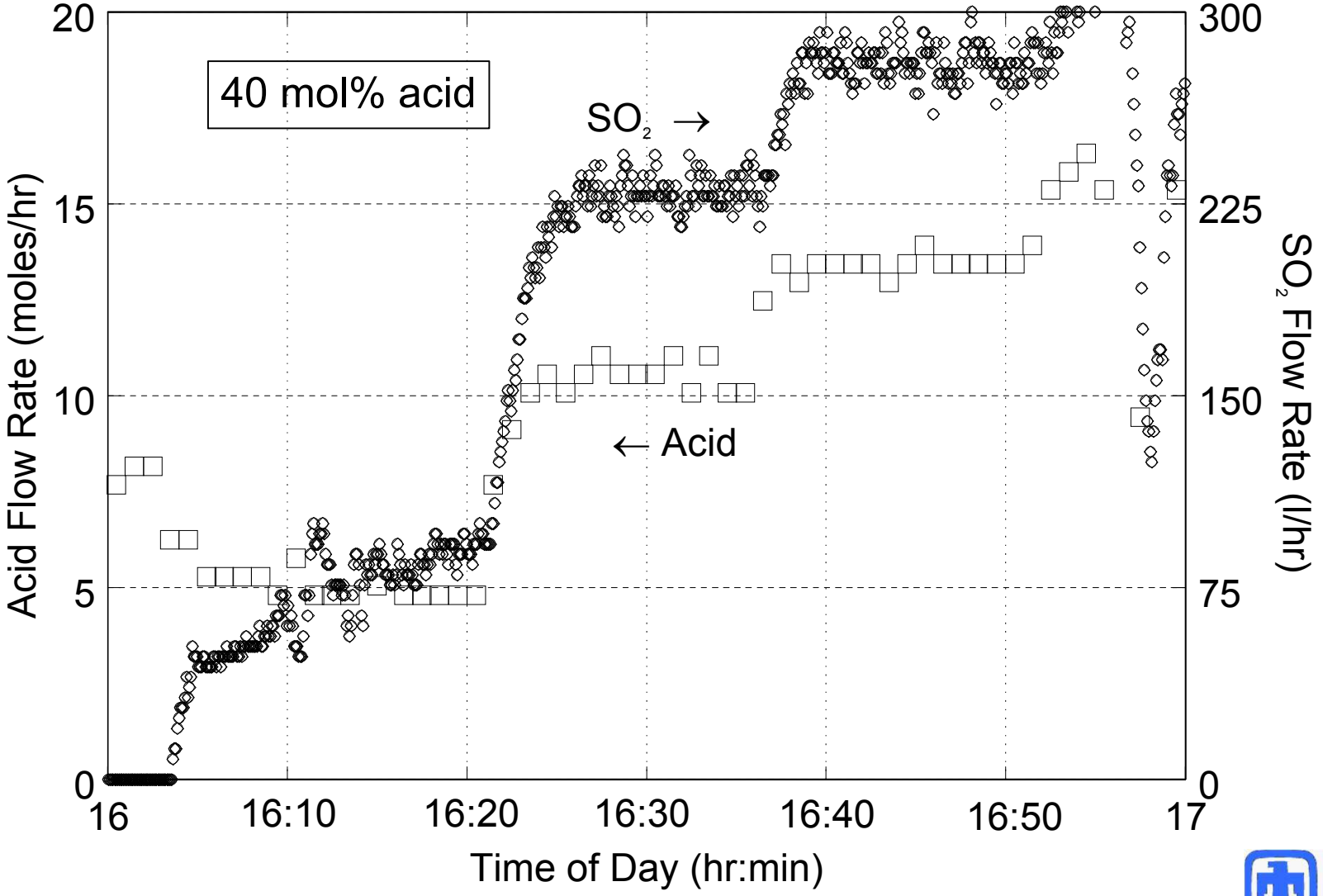
1372 mm Bayonet
40 mol % Acid Feed



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Technical Accomplishments/Progress
Summary (1 of 3)

- **Several half-scale (686 mm) SID acid decomposition experiments performed at 850 °C and pressures from ambient to 5 bar.**
  - No observable corrosion of SiC, glass-lined steel, Teflon, or Viton components.
  - Acid conversion 37% - 78%
    - Higher conversion ⇔ slower flow
    - Higher conversion ⇔ more concentrated acid feed
  - 686 mm half-scale SID capable of ~ 100 L-H_2/hr (limited by heat transfer), for concentrations of 19 mol% and higher.

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Technical Accomplishments/Progress
Summary (2 of 3)

- **Full-scale (1372 mm) SID acid decomposition experiments performed at 850 °C and ambient pressure.**
  - Experiments performed with 20 and 40 mol% acid feed.
  - Acid conversion 40% - 75%
    - Higher conversion ⇔ slower flow
    - Higher conversion ⇔ more concentrated acid feed
  - 1372 mm SID used for Integrated Lab-Scale capable of ~ 200 L-H_2/hr.

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Technical Accomplishments/Progress
Summary (3 of 3)

- **Integrated Lab-Scale Skid progress**
  - Lexan enclosure: 1.2 m x 1.2 m base, and 2.4 m tall.
    - Continuously vented through charcoal filtration system
    - Secondary lexan enclosure with charcoal filtration within primary enclosure to minimize SO_2 hazard
  - Skid construction and assembly completed
  - Completing wiring and hardware/software interface
  - Preparing for final safety reviews.

Next Steps

- June 2007- Assembly, wiring, and flow connections
- July 2007 - Shakedown tests
- August 2007 – Stand alone acid tests
- FY 2007 - Complete integration with GA-CEA
- FY 2008 - Integrated S-I cycle testing

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