SAND2006-3469C

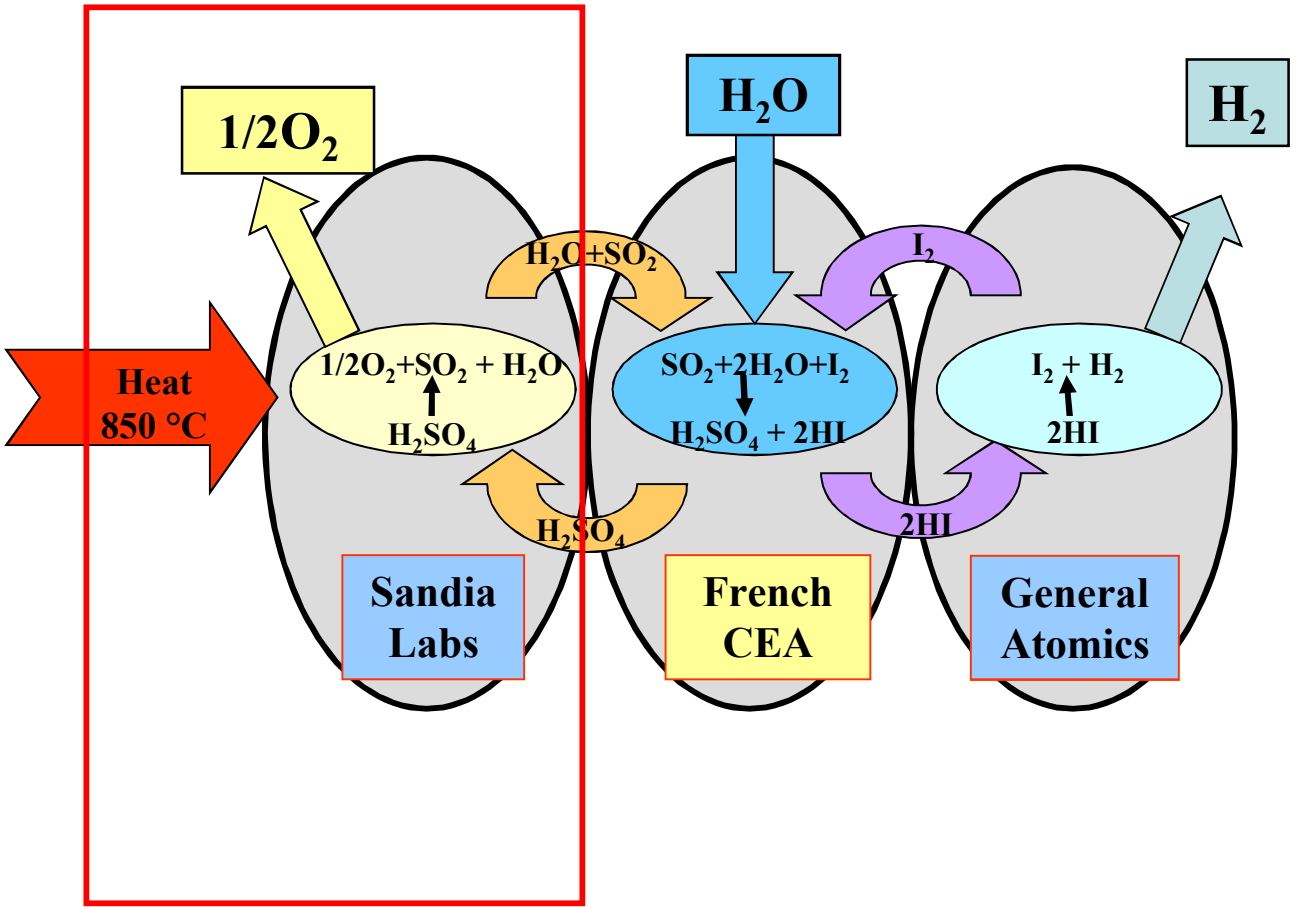
Pressurized Sulfuric Acid Decomposition Experiments for the Sulfur-Iodine Thermochemical Cycle

Fred Gelbard, Robert C. Moore, Milton E. Vernon, Edward J. Parma, Dion A. Rivera, James C. Andazola, Gerald E. Naranjo, Carlos E. Velasquez, and Andrew R. Reay

Sandia National Laboratories
Albuquerque, NM

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Sulfur-Iodine Thermochemical Cycle



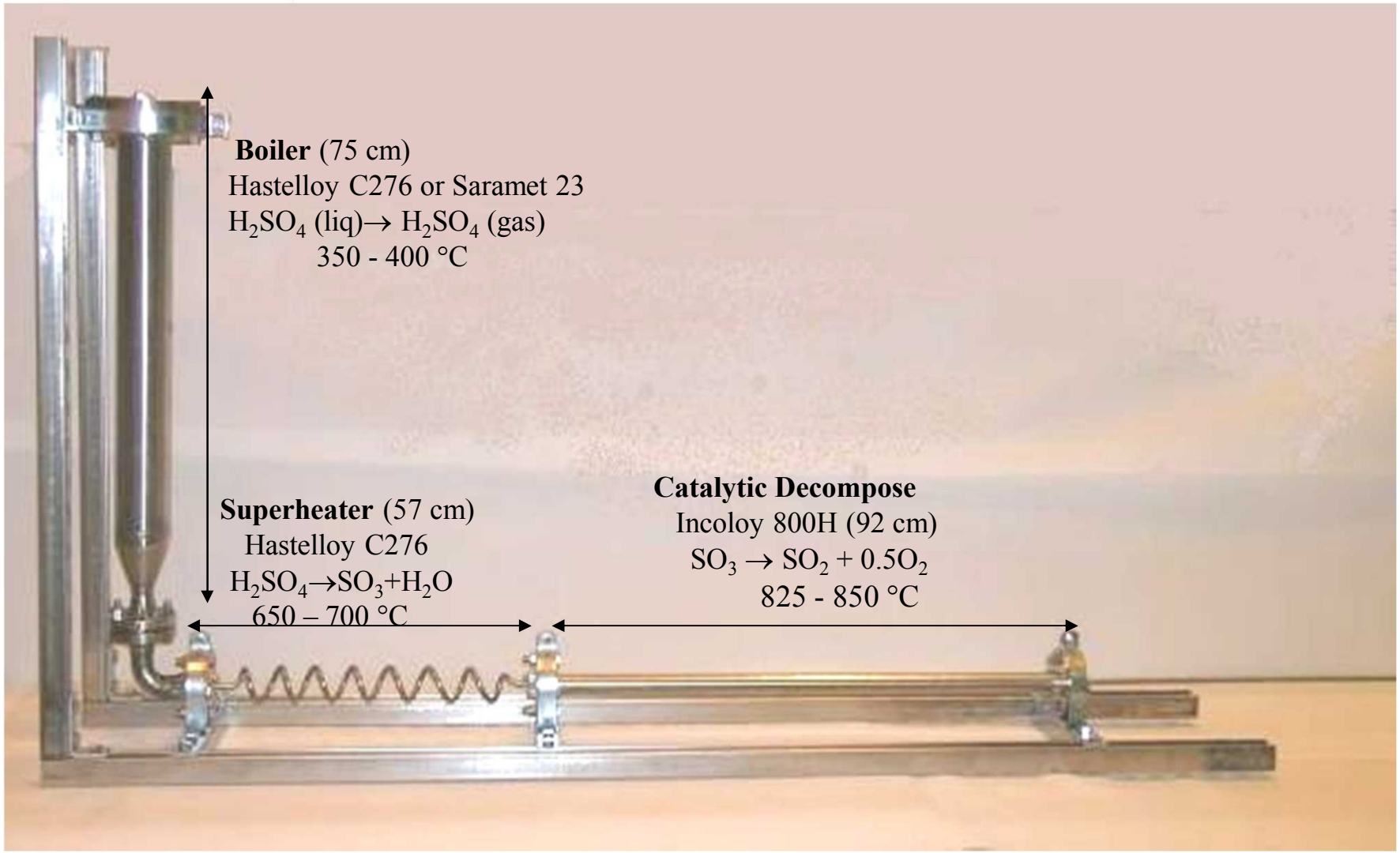
Sandia National Laboratories

Objectives

- **Demonstrate laboratory-scale sulfuric acid decomposition process with engineering materials**
- **Assess process performance and materials**
  - Variations in acid concentration, temperature, and pressure
  - Catalyst: conversion, poisoning, acid concentration, temperature, pressure
  - Engineering materials (starting with Hastelloy, Incoloy, ceramics, and glass)
- **Integrate into a closed-loop SI cycle: Integrated Lab-Scale (ILS) Experiments**
  - Real-time instrumentation for process control

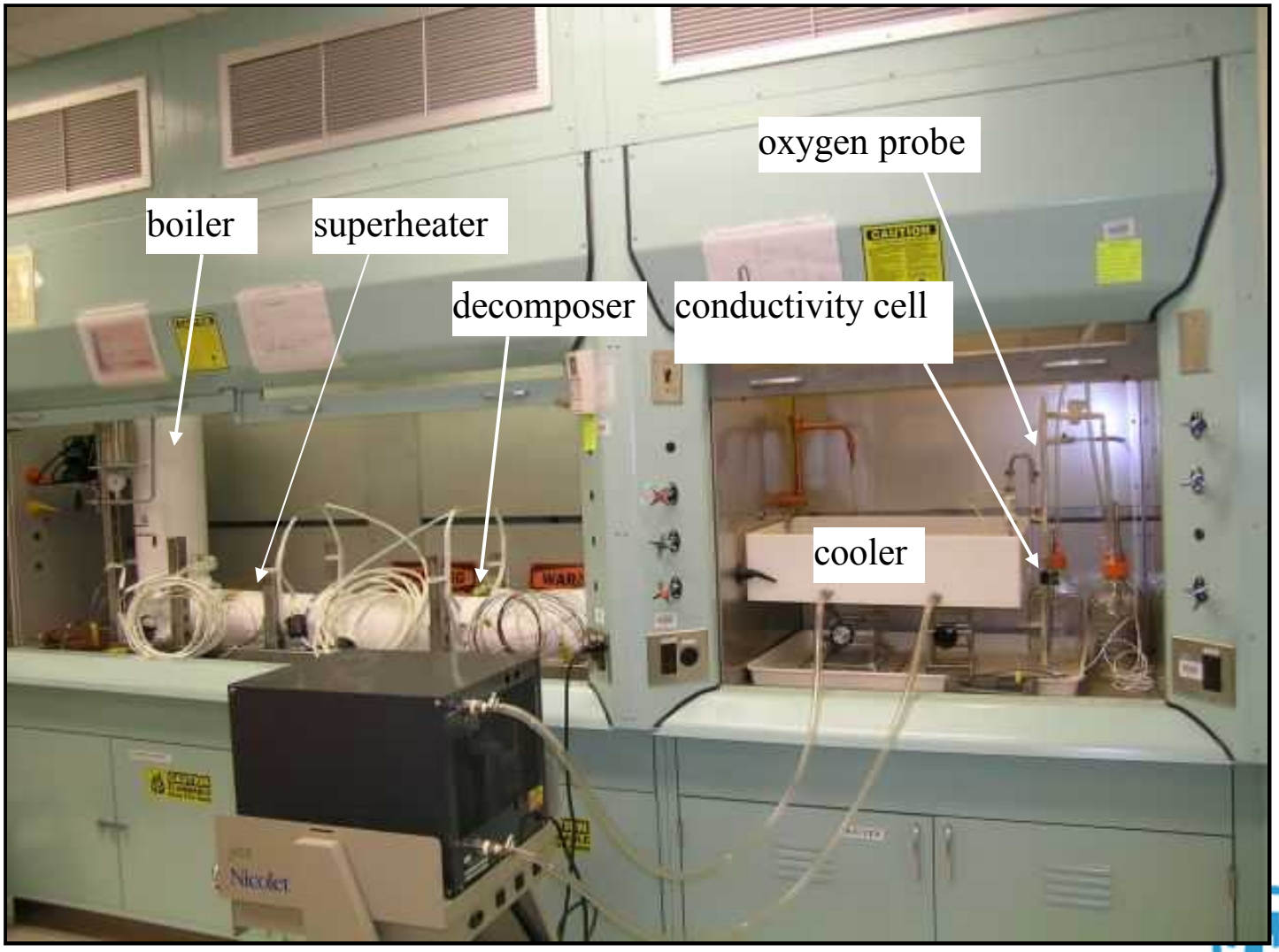
Sandia National Laboratories

Basic Lab-Scale Units



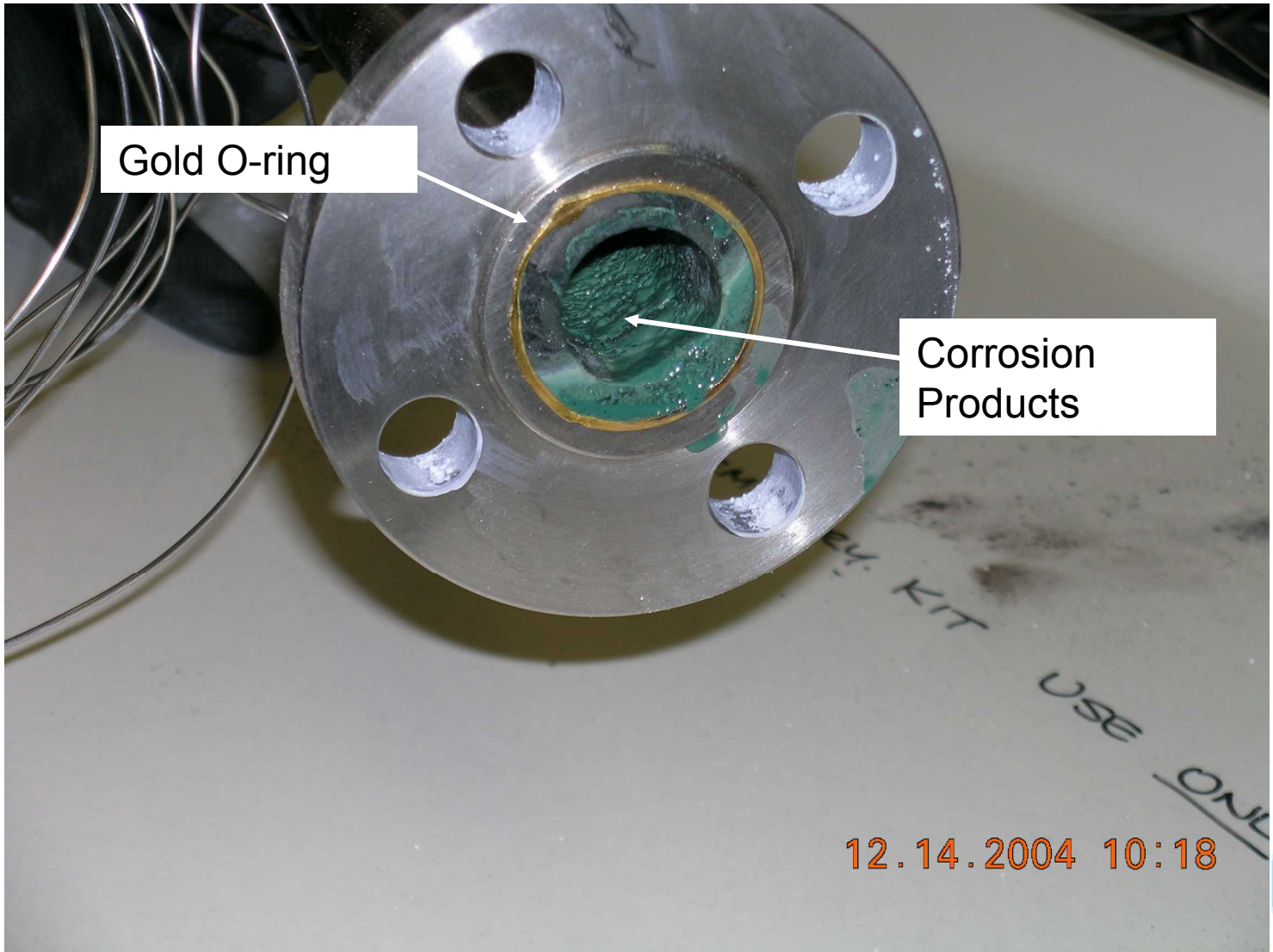
Sandia National Laboratories

Laboratory-Scale Acid Decomposer



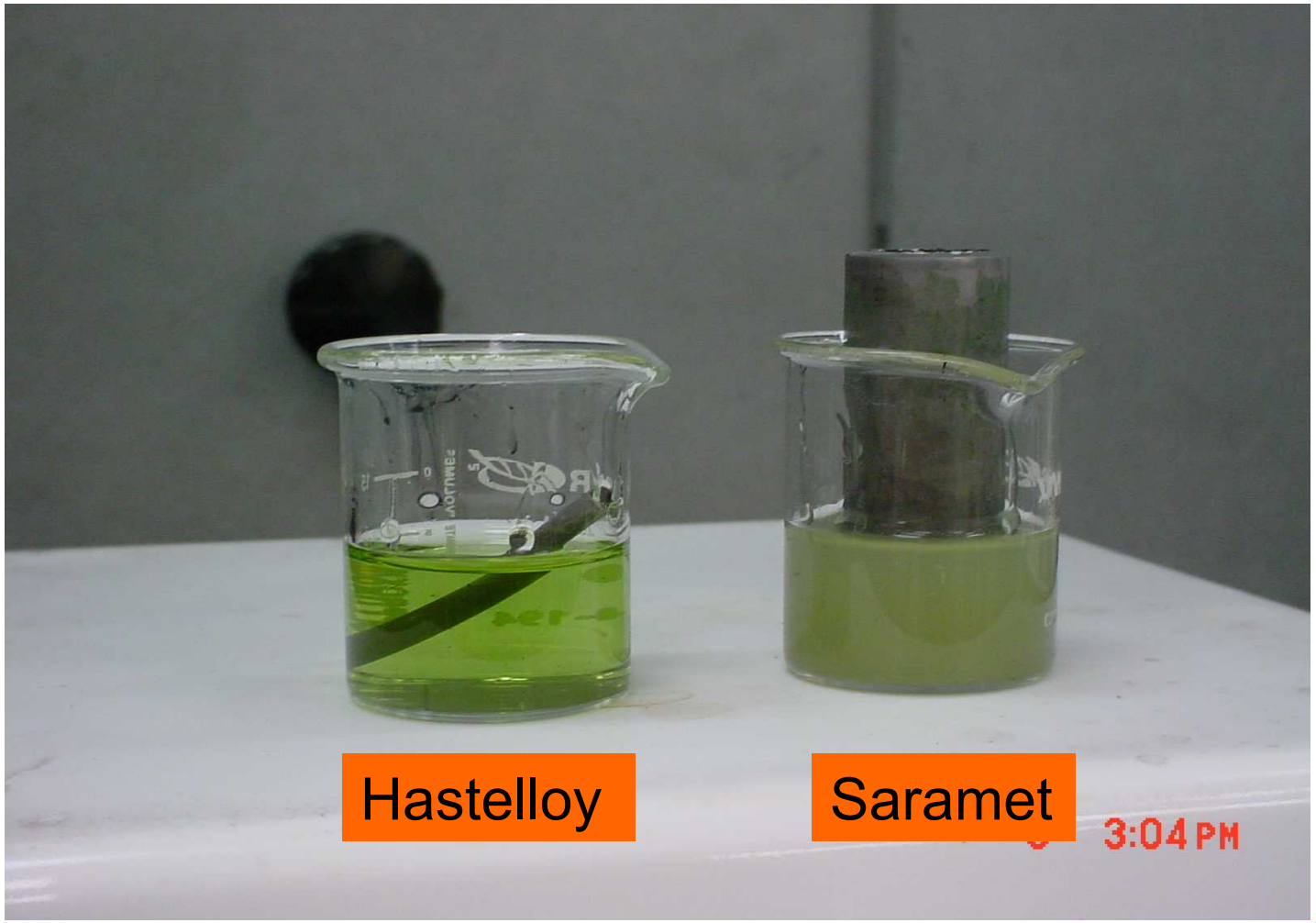
Sandia National Laboratories

Post Decomposer



Sandia National Laboratories

Simple Corrosion Test
40 mol% acid after 20 minutes

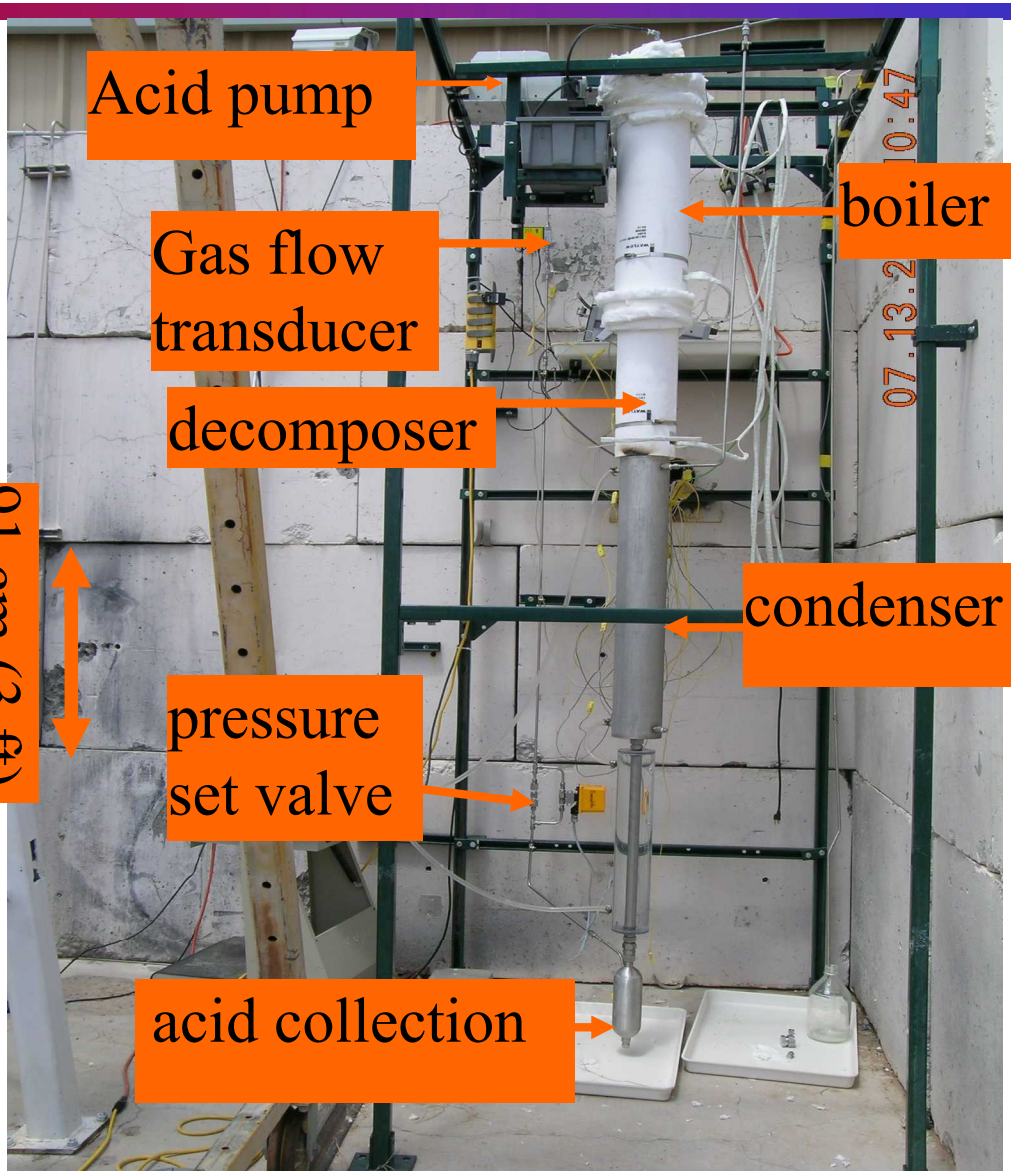


Sandia National Laboratories

Results of First Acid Test

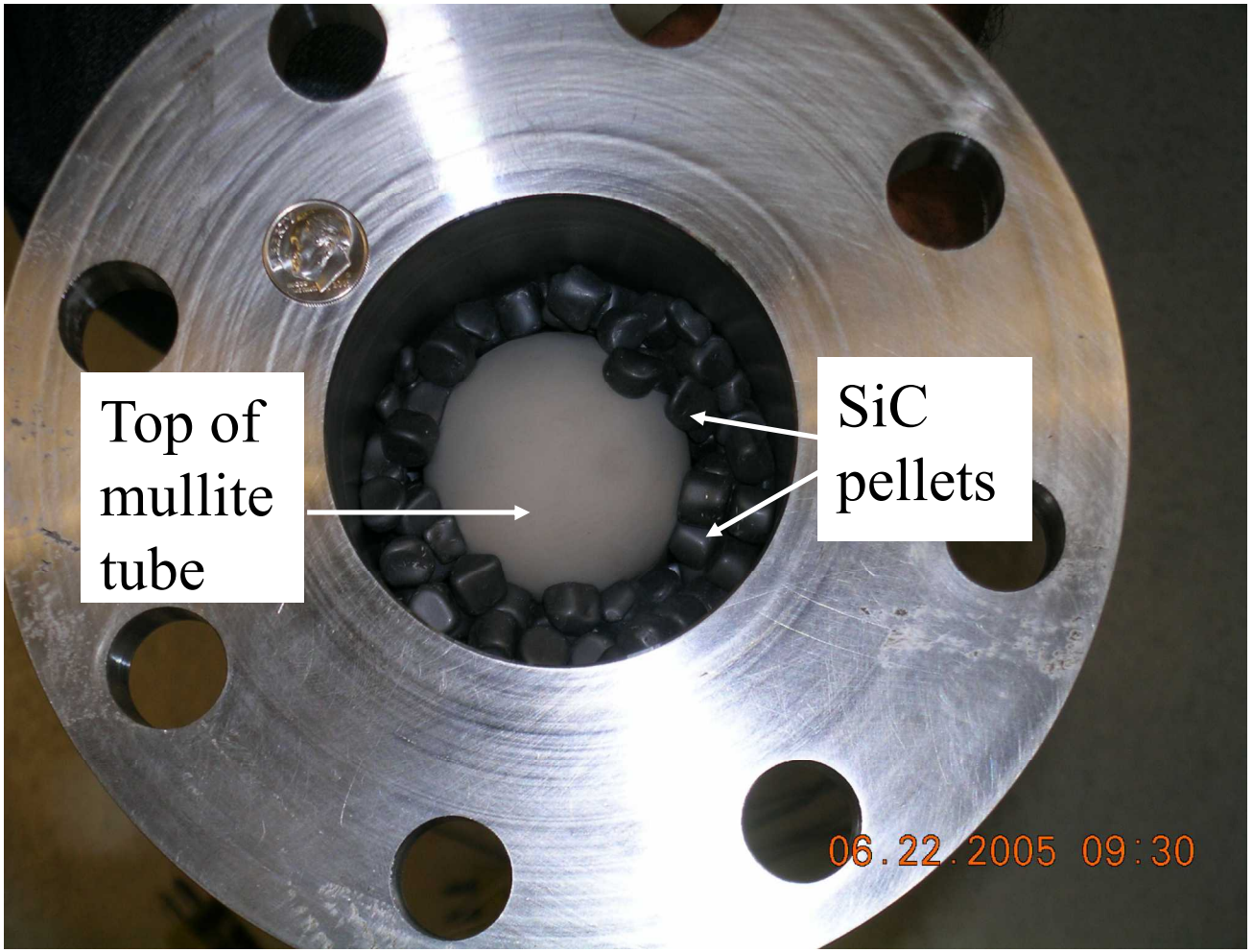
- **Corrosion products clogged apparatus in minutes**
  - Green deposits primarily in upper boiler and after decomposer. (Incoloy 800H flange severely corroded, ~1/2 mm).
  - Boiler thermocouple tube (Hastelloy) corroded slightly. Hastelloy flange blackened and bonded with gold O-ring.
  - Corrosion rates much higher than estimated from data with sulfuric acid coupon tests
    - Static versus flow experiments
    - SO_2 and O_2 may contribute to enhanced corrosion
    - Volume expansion: $FeSO_4 \cdot 7H_2O/Fe = 21$
- **New Strategy Based on Test Results**
  - Keep metallic walls either very hot (>600 C) or cold (<20 C)
  - Ceramics/glass needed for hot liquid acid vaporizing or condensing acid vapors

Single-Pass Sulfuric Acid Decomposition Apparatus



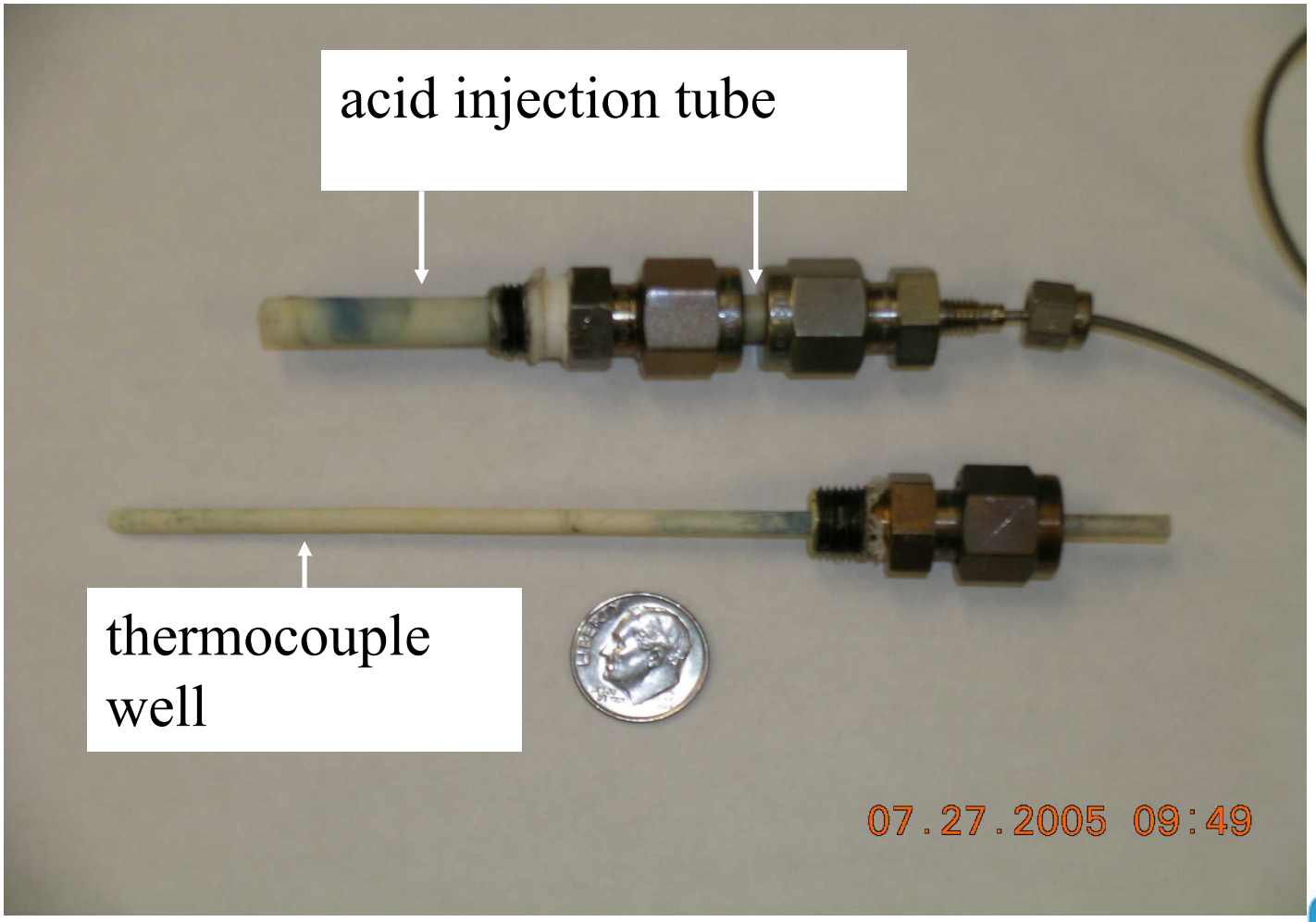
Sandia National Laboratories

Top of Boiler



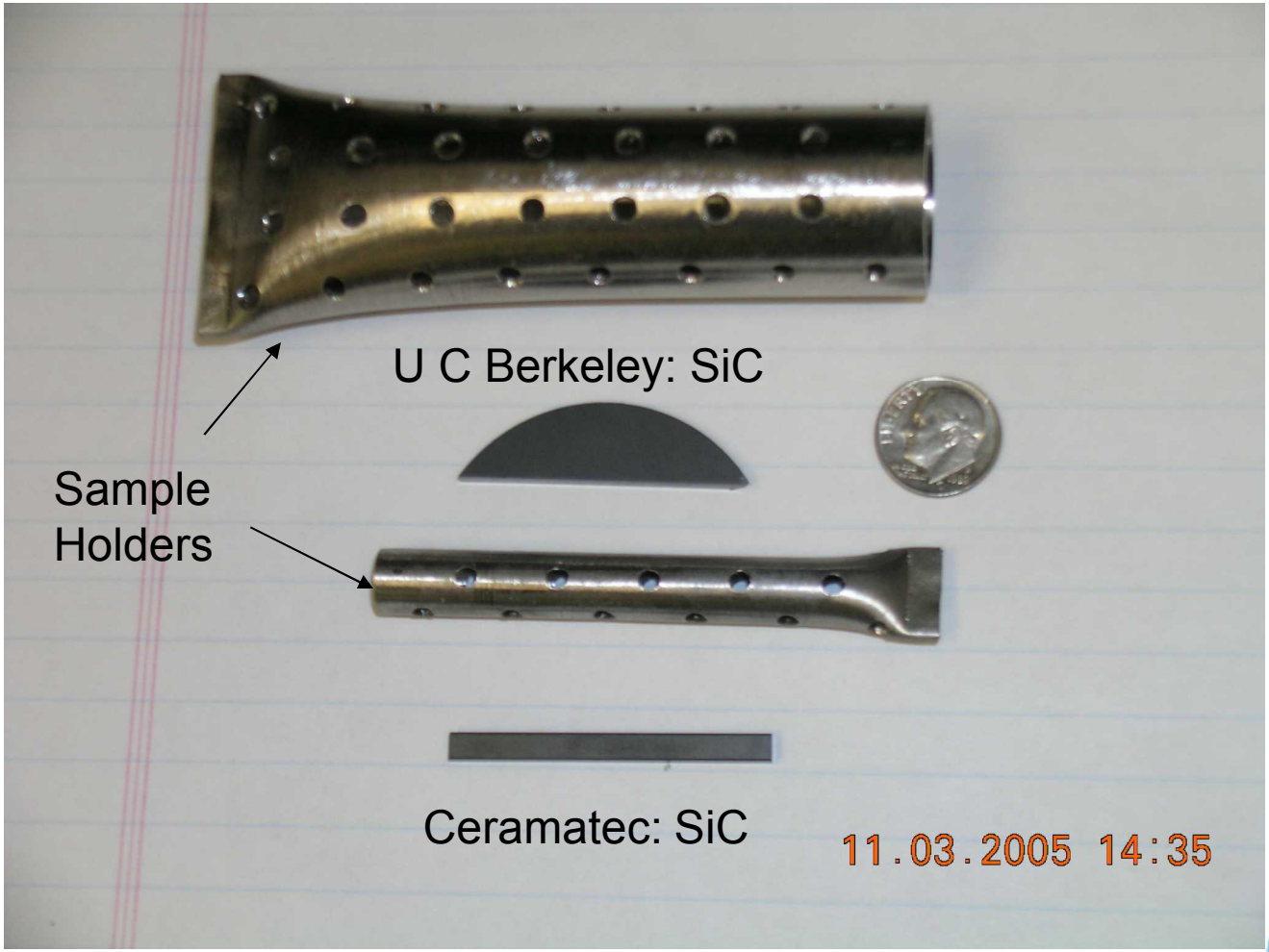
Sandia National Laboratories

Ceramic Injector and Thermocouple Well



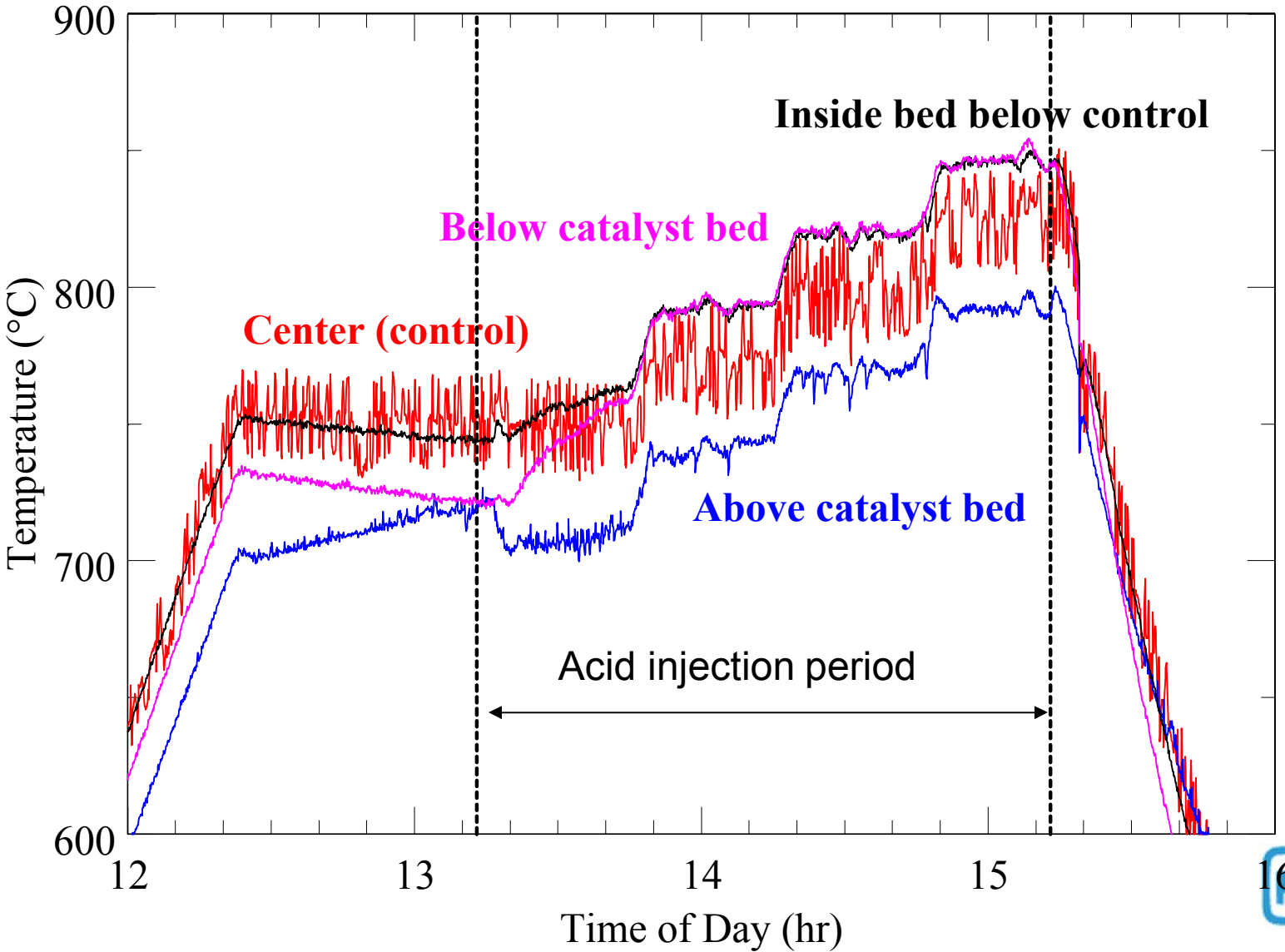
Sandia National Laboratories

Corrosion Coupon Testing



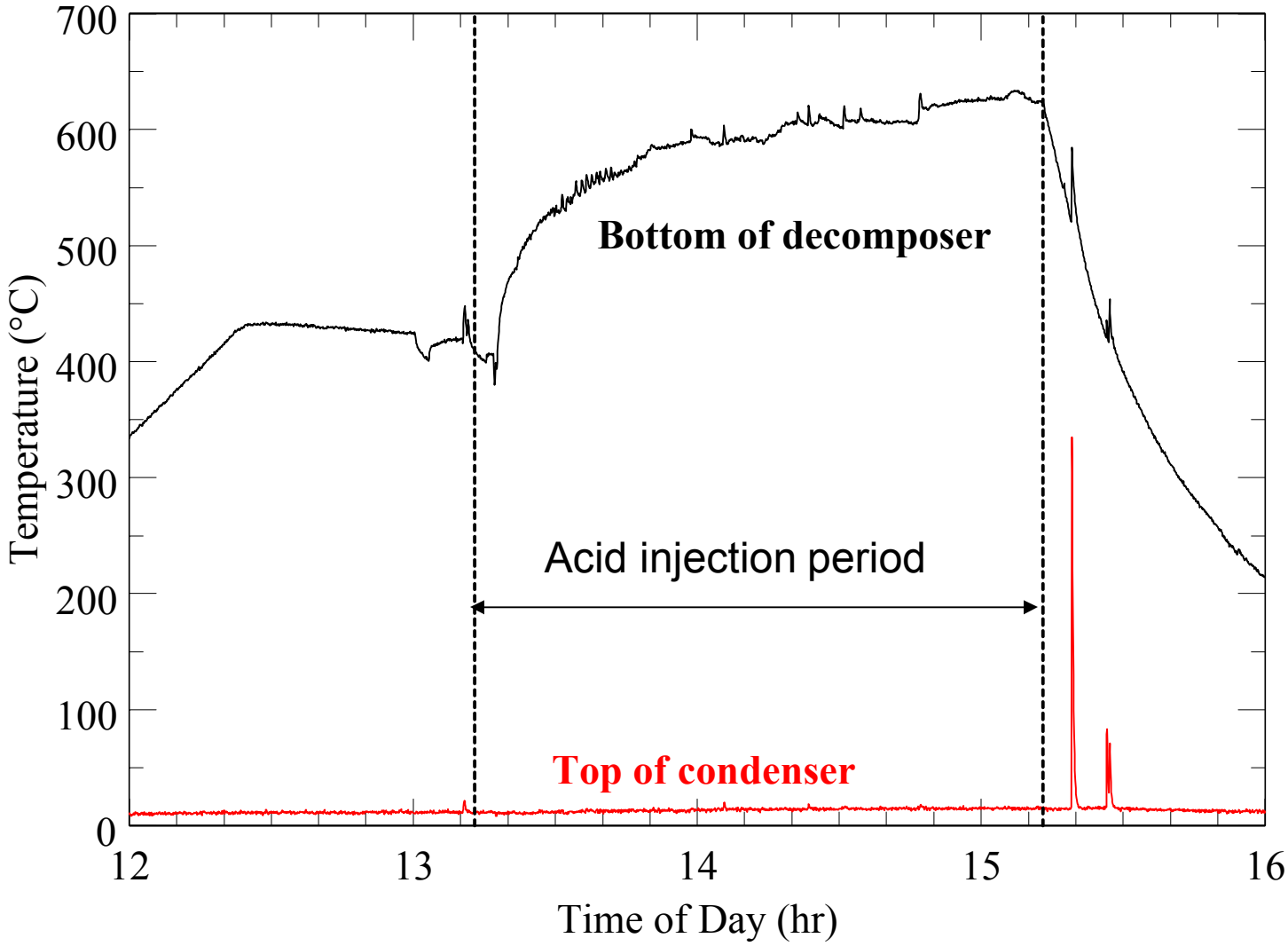
Sandia National Laboratories

Decomposer Temperature Profile



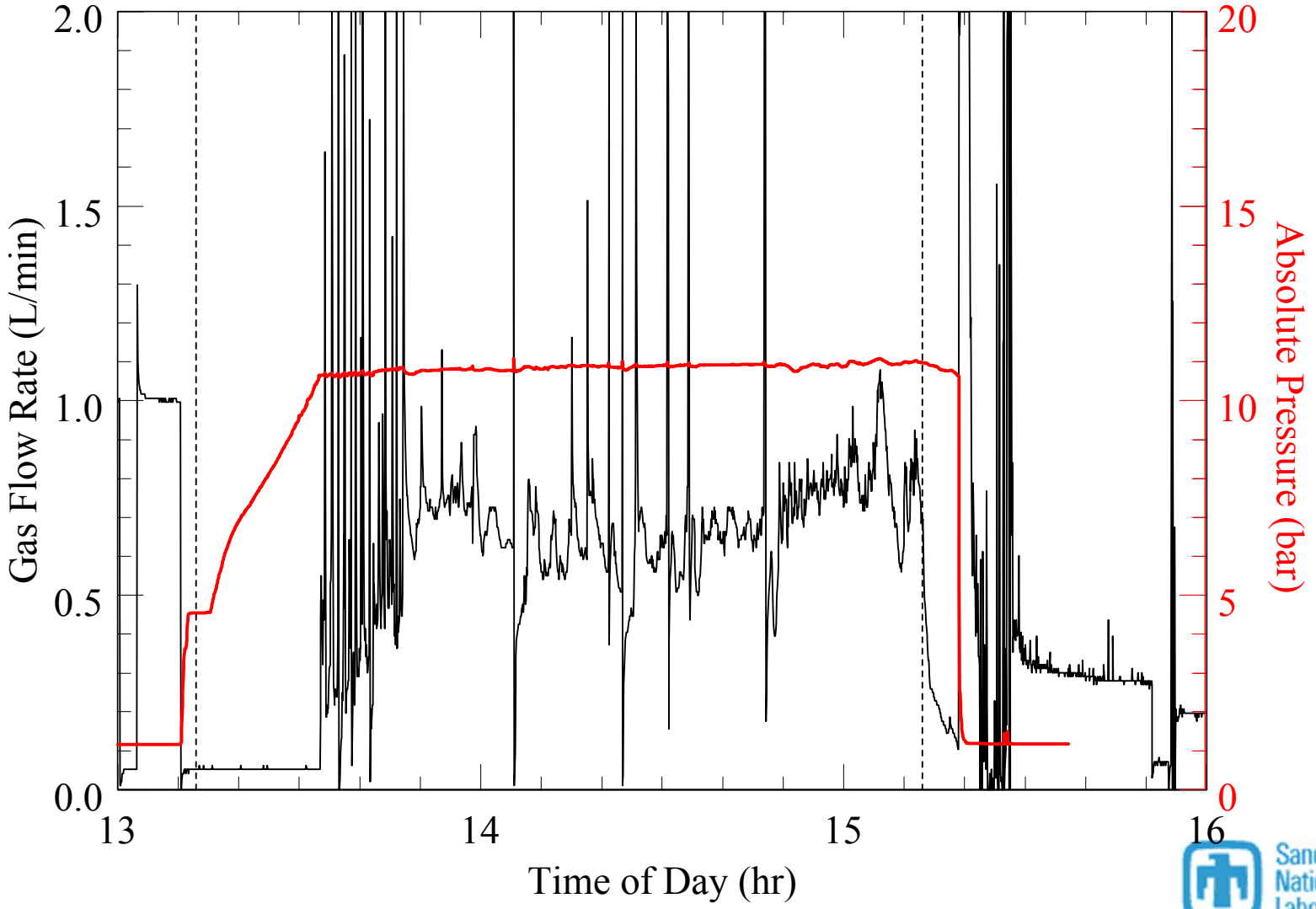
Sandia National Laboratories

Bottom of Decomposer & Top of Condenser Temperature Profiles



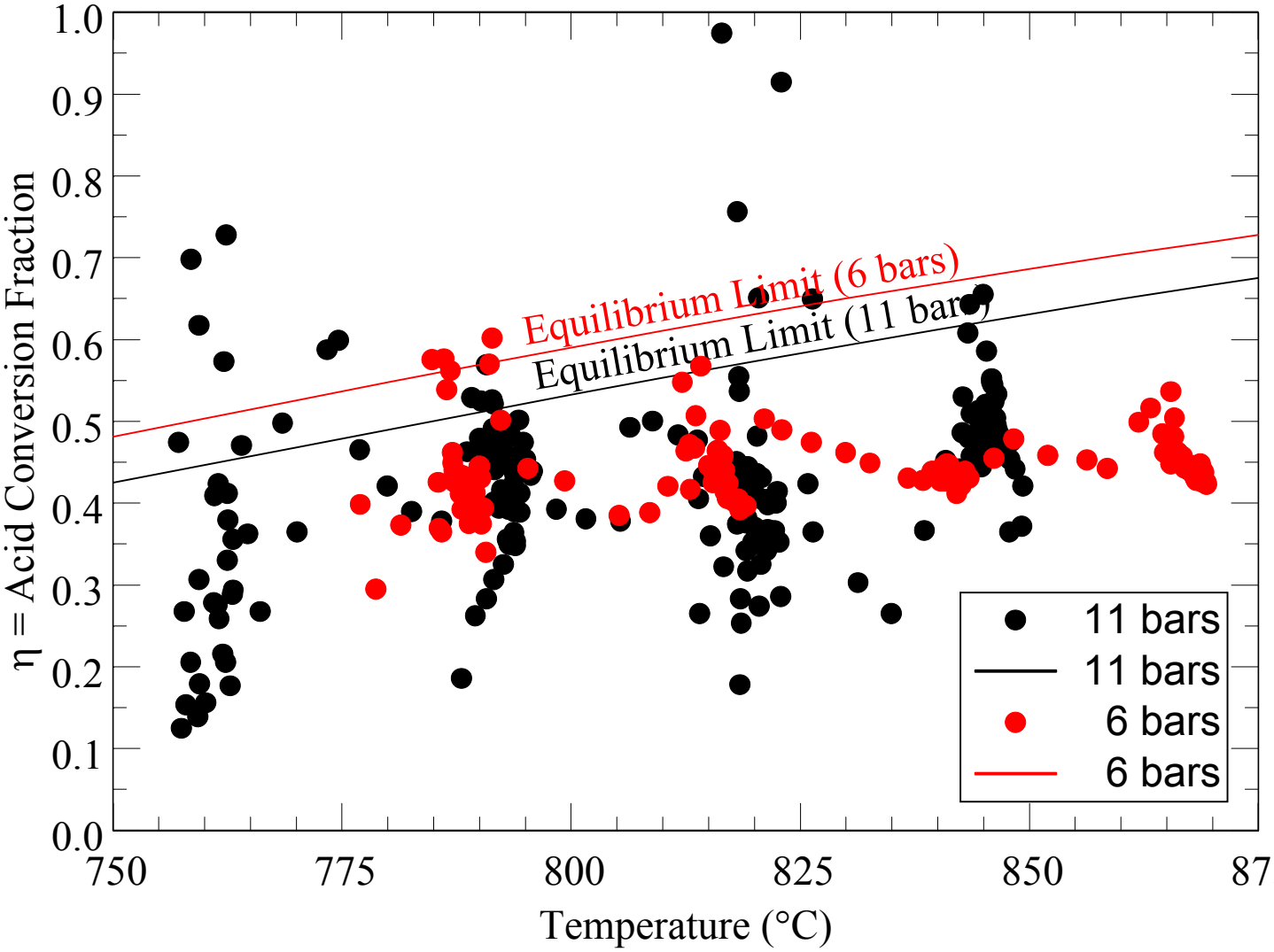
Sandia National Laboratories

Flow Rate & Pressure



Sandia National Laboratories

Acid Conversion Fraction

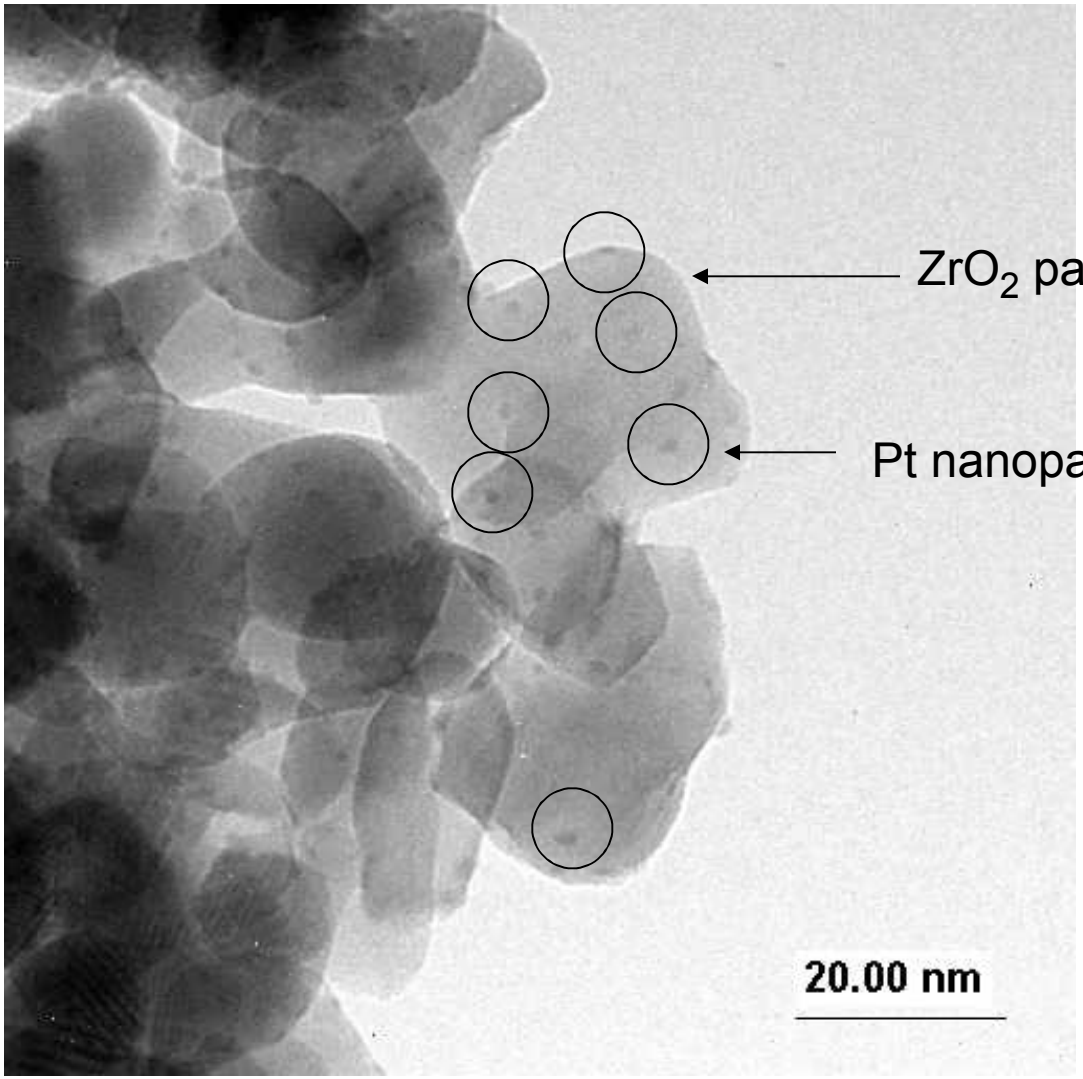


Sandia National Laboratories

Platinum Catalyst on Zirconia

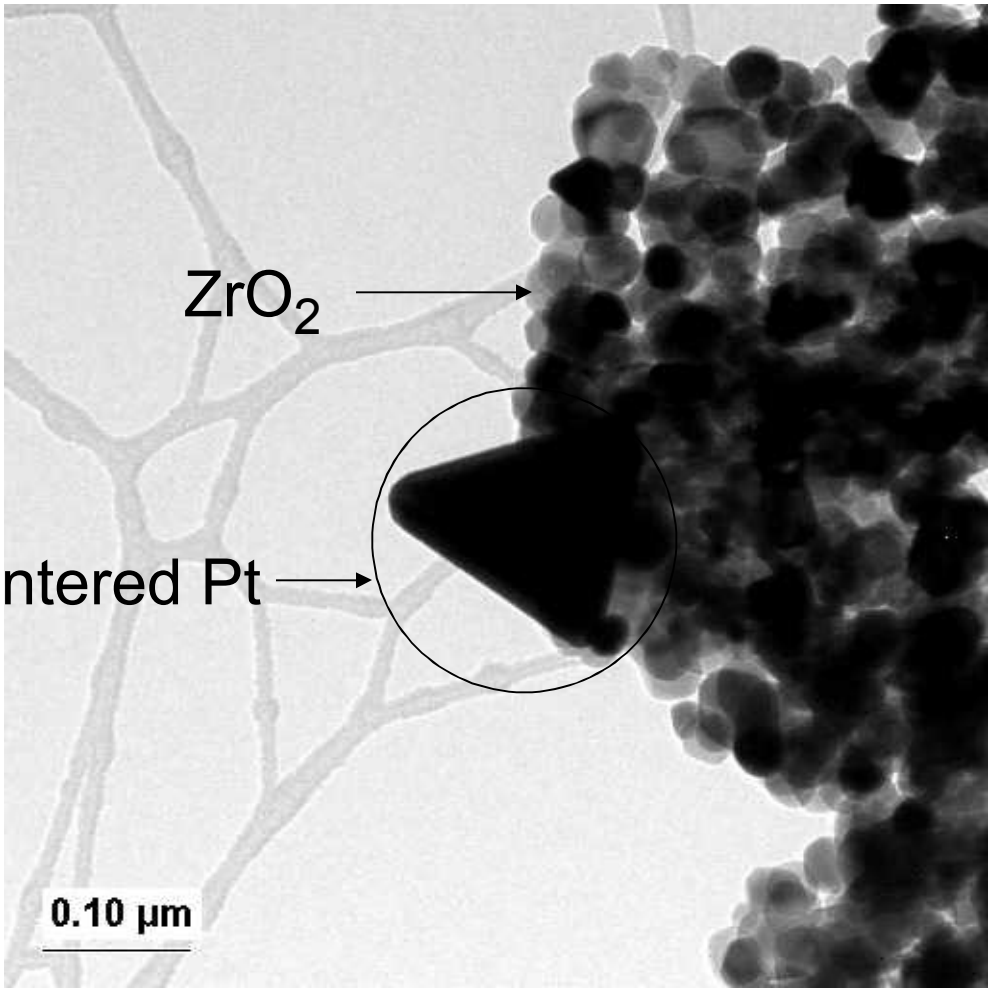
Sandia National Laboratories

Pretest TEM of Catalyst
(Tom Headley, SNL)



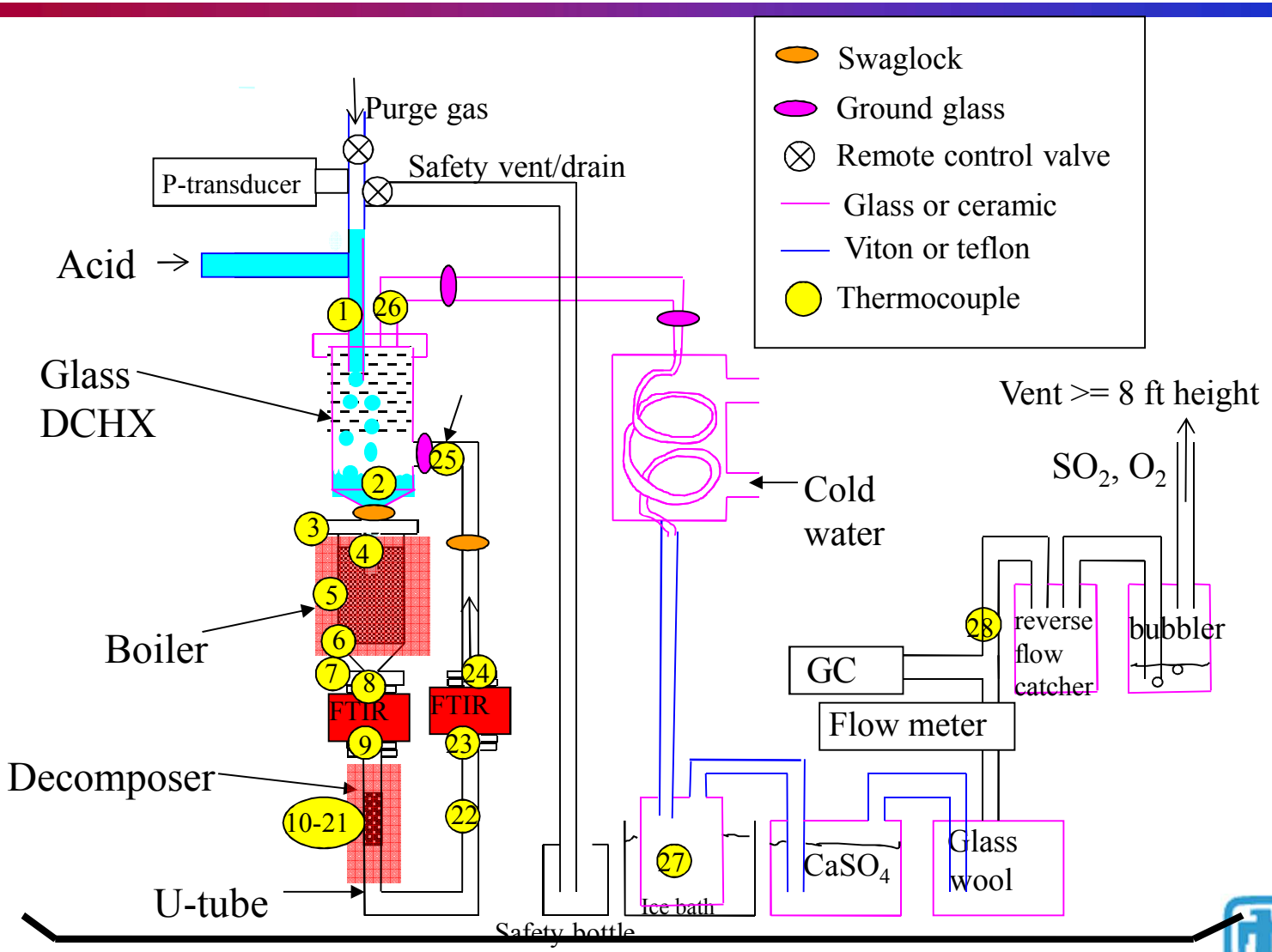
Sandia National Laboratories

Post test TEM of Catalyst (Tom Headley, SNL)



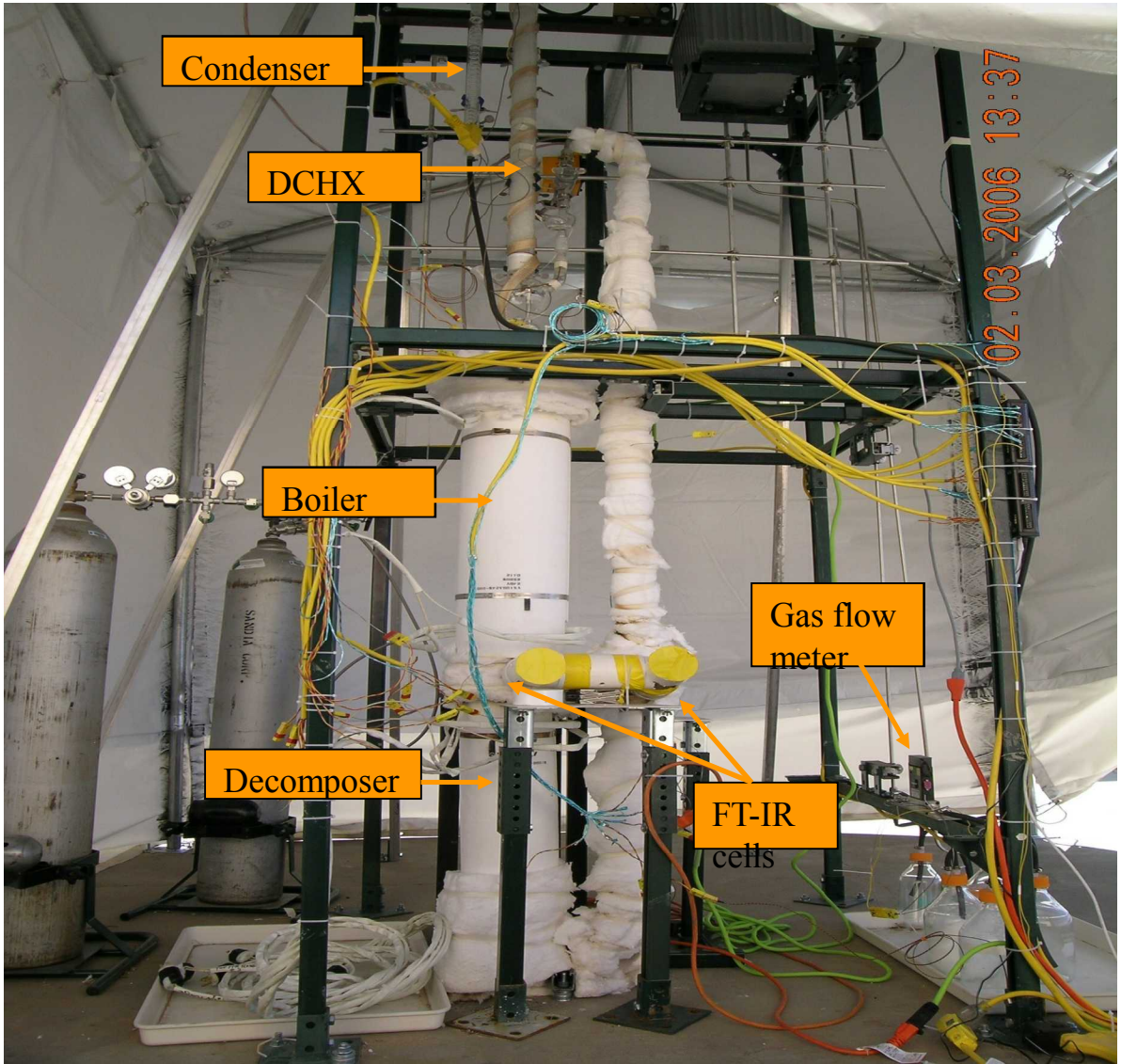
Sandia National Laboratories

DCHX & FT-IR



Sandia National Laboratories

DCHX Apparatus



Sandia National Laboratories

Recycling Undecomposed Acid

- **Direct Contact Heat Exchanger recovers heat and recycles undecomposed SO_3**
- **Corrosion for acid condensation can be greatly minimized by condensing in ceramic/glass tubes (or rapid quenching)**
- **Hardly any corrosion**
  - boiler heated to ~600 C
  - glass insert at top of boiler
  - Condense into glass/ceramic DCHX

Summary

- Pressurized lab-scale apparatus fabricated, assembled, tested, and operational
  - Completed series of pressurized acid tests (2, 6, and 11 bars)
  - Demonstrated real time measurements which are needed for process control
  - Acid conversion fraction 15-20% below theoretical maximum determined by equilibrium
  - Corrosion problems resolved by either rapid quenching or condensing on ceramic/glass materials

Future Plans Leading to Integrated Lab-Scale Experiments

- Metallic components contacting acid will be removed in subsequent tests
  - All ductile metals tested corroded significantly in hot/warm liquid acid
  - Brittle ceramics or glass required for condensing unreacted acid for recycling
- Test incorporating acid concentrator planned for August 2006
- Demonstrate full Section 2 for ILS experiments Dec 2006

Sandia National Laboratories