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Title: PLUTONIUM-238 SCIENCE AND ENGINEERING BENCH-SCALE MOLTEN SALT OXIDATION PROCESSING

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Recovery of Pu-238 by Molten Salt Oxidation Processing of Pu-238 Contaminated Combustibles

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Abstract

Pu-238 heat sources are used to fuel radioisotope thermoelectric generators (RTG) used in space missions. The demand for this fuel is increasing, yet there are currently no domestic sources of this material. Much of the fuel is material reprocessed from other sources. One rich source of Pu-238 residual material is that from contaminated combustible materials, such as cheesecloth, ion exchange resins and plastics. From both waste minimization and production efficiency standpoints, the best solution is to recover this material.

One way to accomplish separation of the organic component from these residues is a flameless oxidation process using molten salt as the matrix for the breakdown of the organic to carbon dioxide and water. The plutonium is retained in the salt, and can be recovered by dissolution of the carbonate salt in an aqueous solution, leaving the insoluble oxide behind. Further aqueous scrap recovery processing is used to purify the plutonium oxide.

Recovery of the plutonium from contaminated combustibles achieves two important goals. First, it increases the inventory of Pu-238 available for heat source fabrication. Second, it is a significant waste minimization process. Because of its thermal activity (0.567 W per gram), combustibles must be packaged for disposition with much lower amounts of Pu-238 per drum than other waste types. Specifically, cheesecloth residues in the form of pyrolyzed ash (for stabilization) are being stored for eventual recovery of the plutonium.

Introduction

MSO is not a new technology; it was developed extensively in the 70's by Rockwell and later at DOE complex sites (West, 1992) as a method of treating hazardous organic wastes, and radioactive wastes (McKenzie et al., 1975). At the time, the recovery of materials retained in the salt was not pursued. MSO was sidelined when incineration became a more attractive alternative. That option is no longer available, so MSO has been revisited as a disposal technique in recent years (Gay, 1992).

Our application has two challenges that have not been addressed by other developers. First, we need to adapt the process unit to work inside a glovebox, with its ergonomic, available space, and safety restrictions. Second, we need to recover the plutonium "contaminant" from the salt.

To address these issues, an MSO system was developed and tested extensively in a facility outside of the radiation controlled area. The system was then modified for operation

^{238}Pu Science and Engineering Bench-Scale Molten Salt Oxidation Process

Presented at
AIChE 2004 Spring
National Conference

Lyndsay Remerowski (NMT-9)

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A Few Facts about Pu-238 Oxide

- "Heat source" material for deep space missions. (Rovers "RUs")
- Currently no domestic production
- Increasingly important material for national security applications

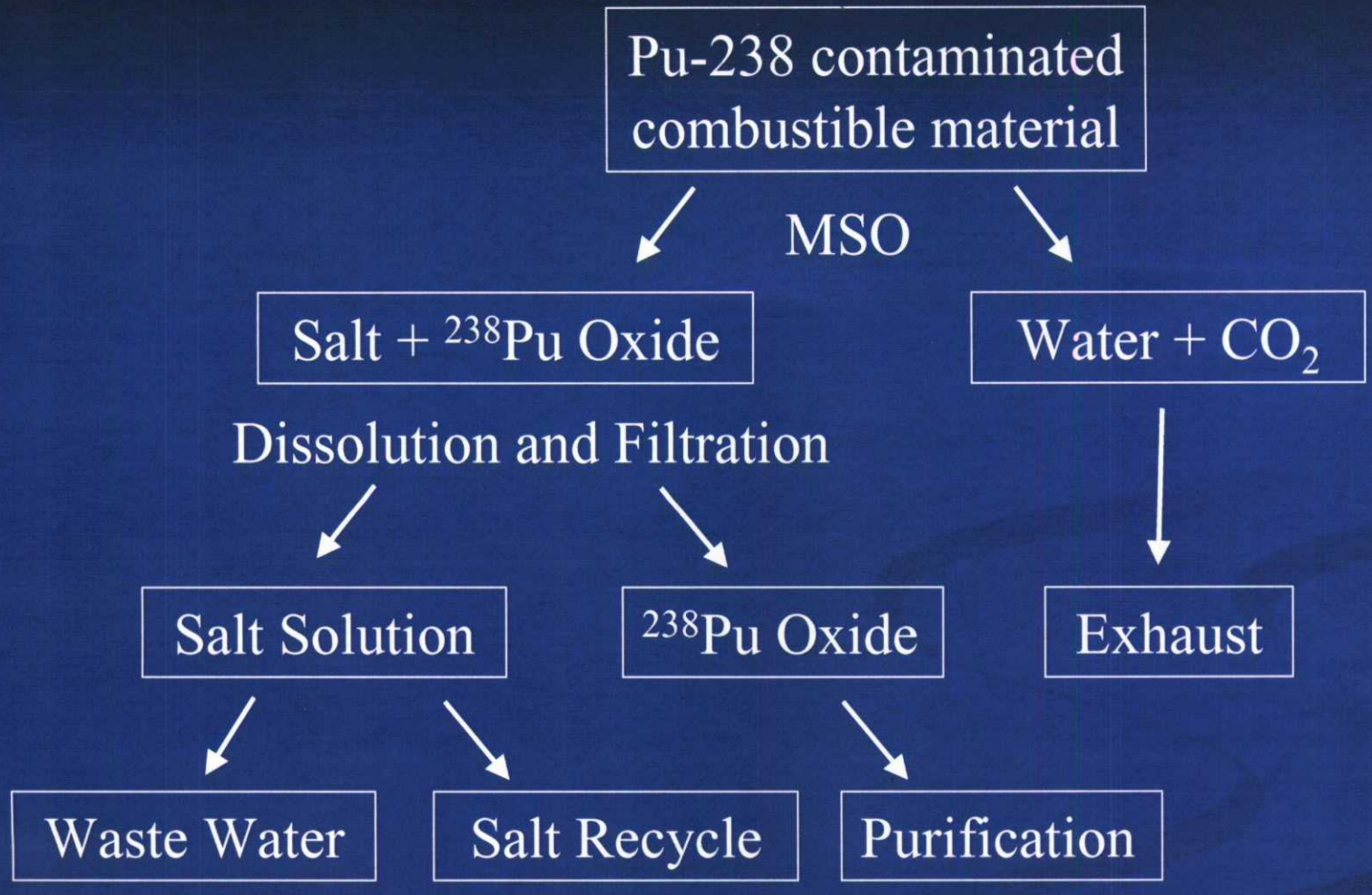
A Few Facts about Pu-238 Waste

- Pu-238 waste has "no-path-forward"
- Limits for Pu-238 combustible waste are a few grams per 55-gal drum
- Significant amount of Pu-238 oxide in the combustible material

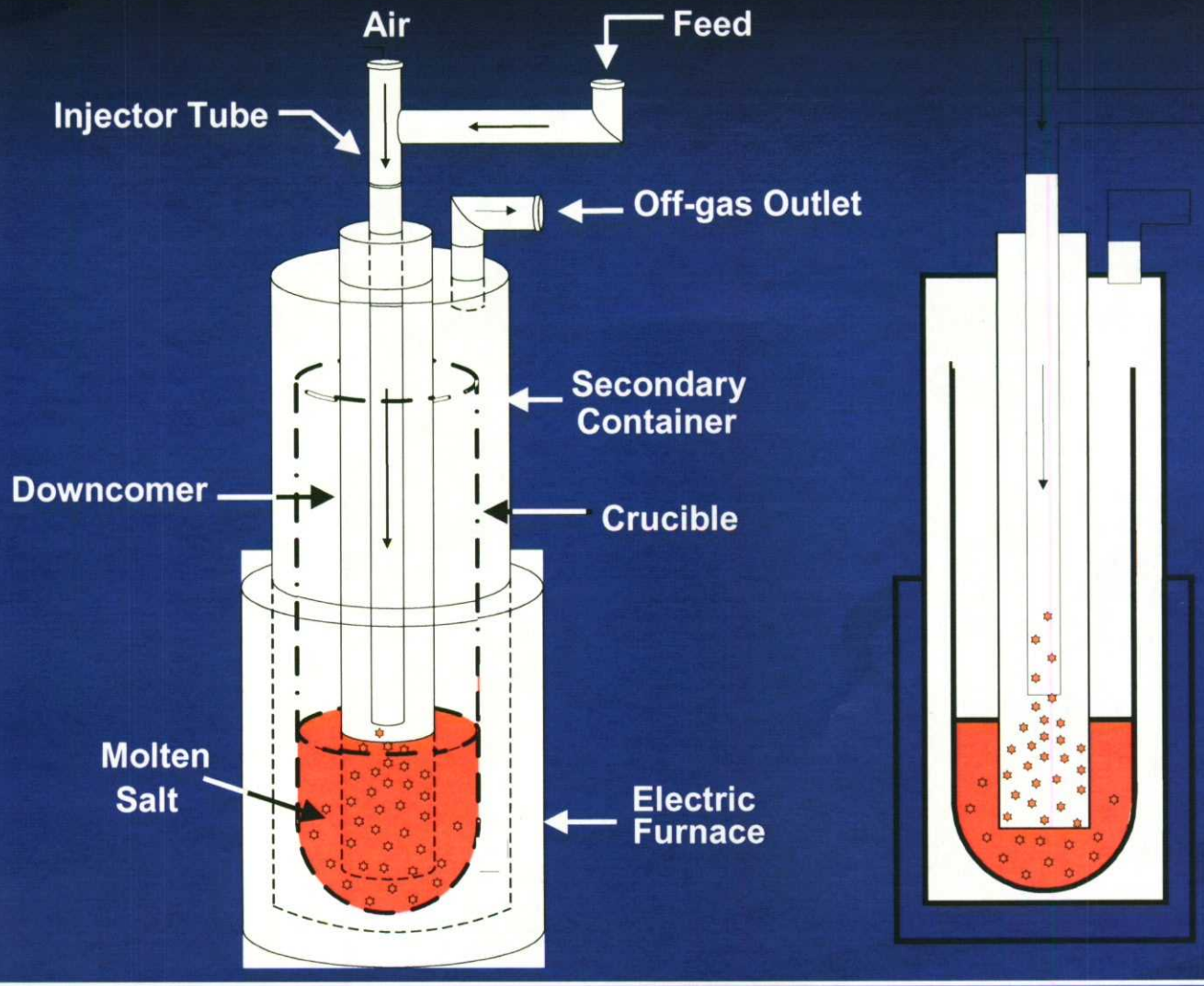
MSO Technology

- Flameless alternative to incineration
- Uses bed of molten salt as matrix for complete oxidation of hydrocarbon material
- Plutonium remains in the salt as insoluble oxide and can be recovered

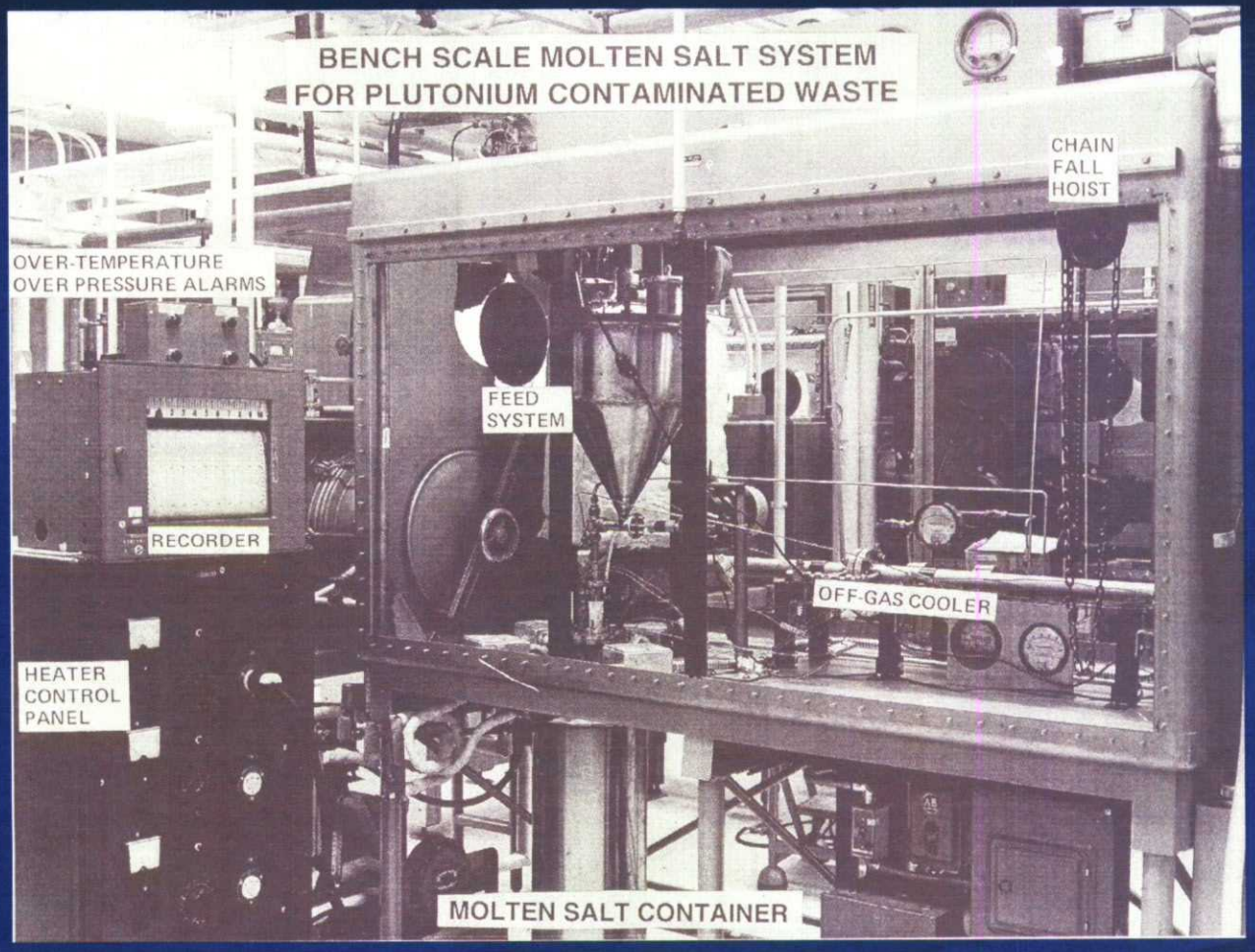
Flow Chart of MSO Process



Schematic of MSO Unit



Bench-scale unit from Rockwell



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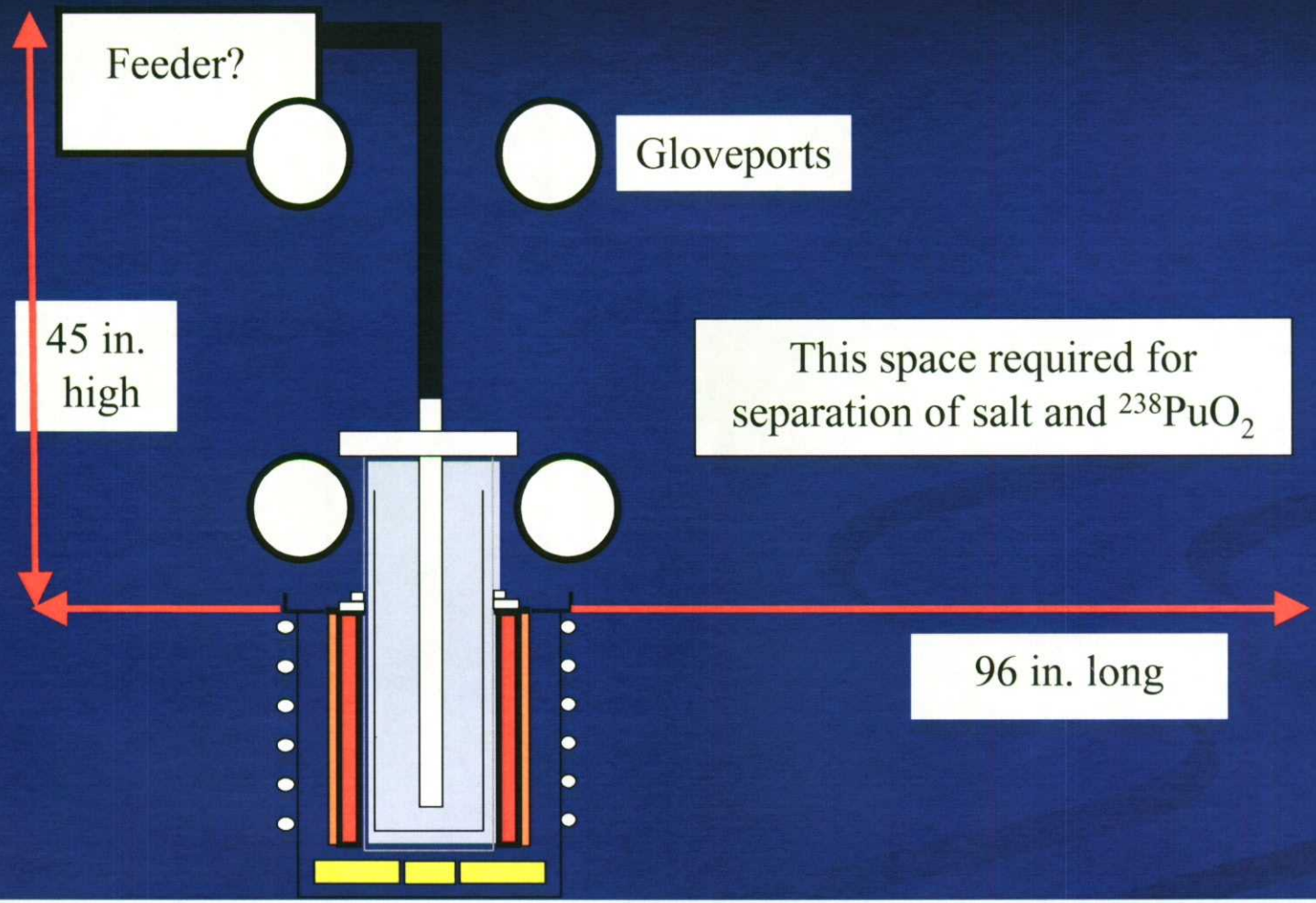
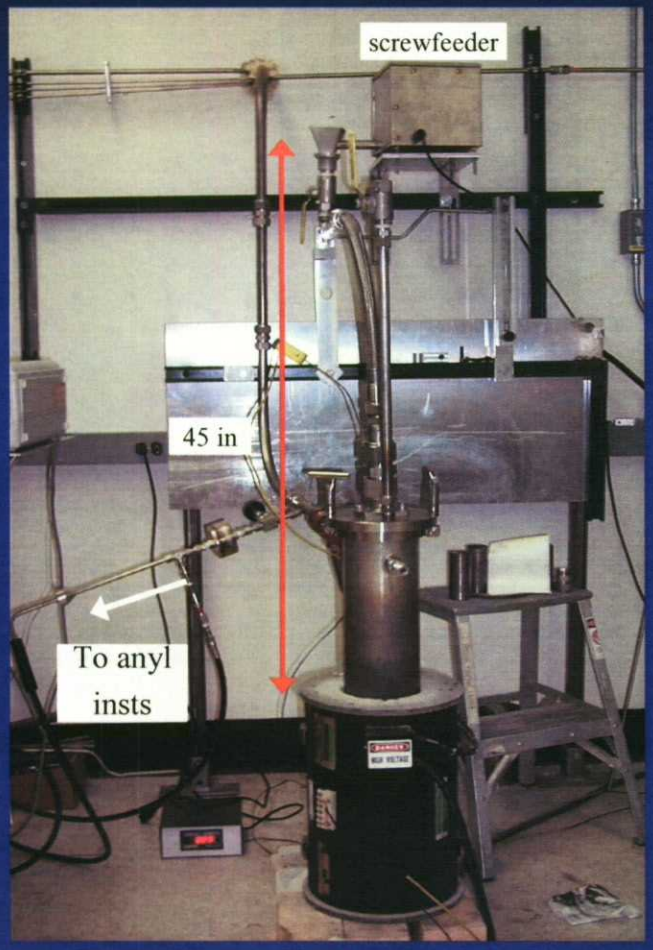
Stringent Space Requirements



Continuous Feed Unit 2002



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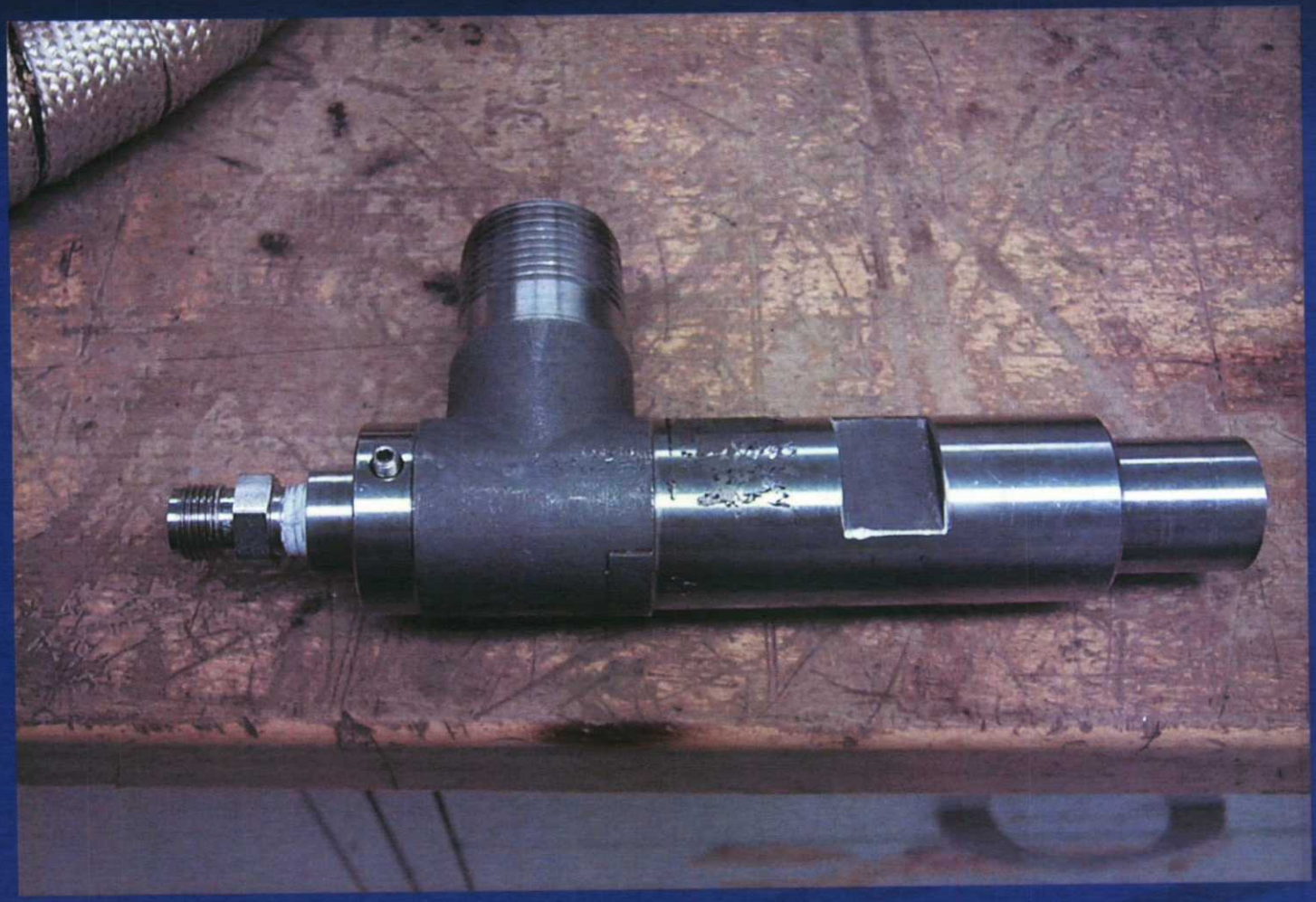
Eductor



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Melt plastic beads and add ash

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Dry to hardness

Jaw crush to right size

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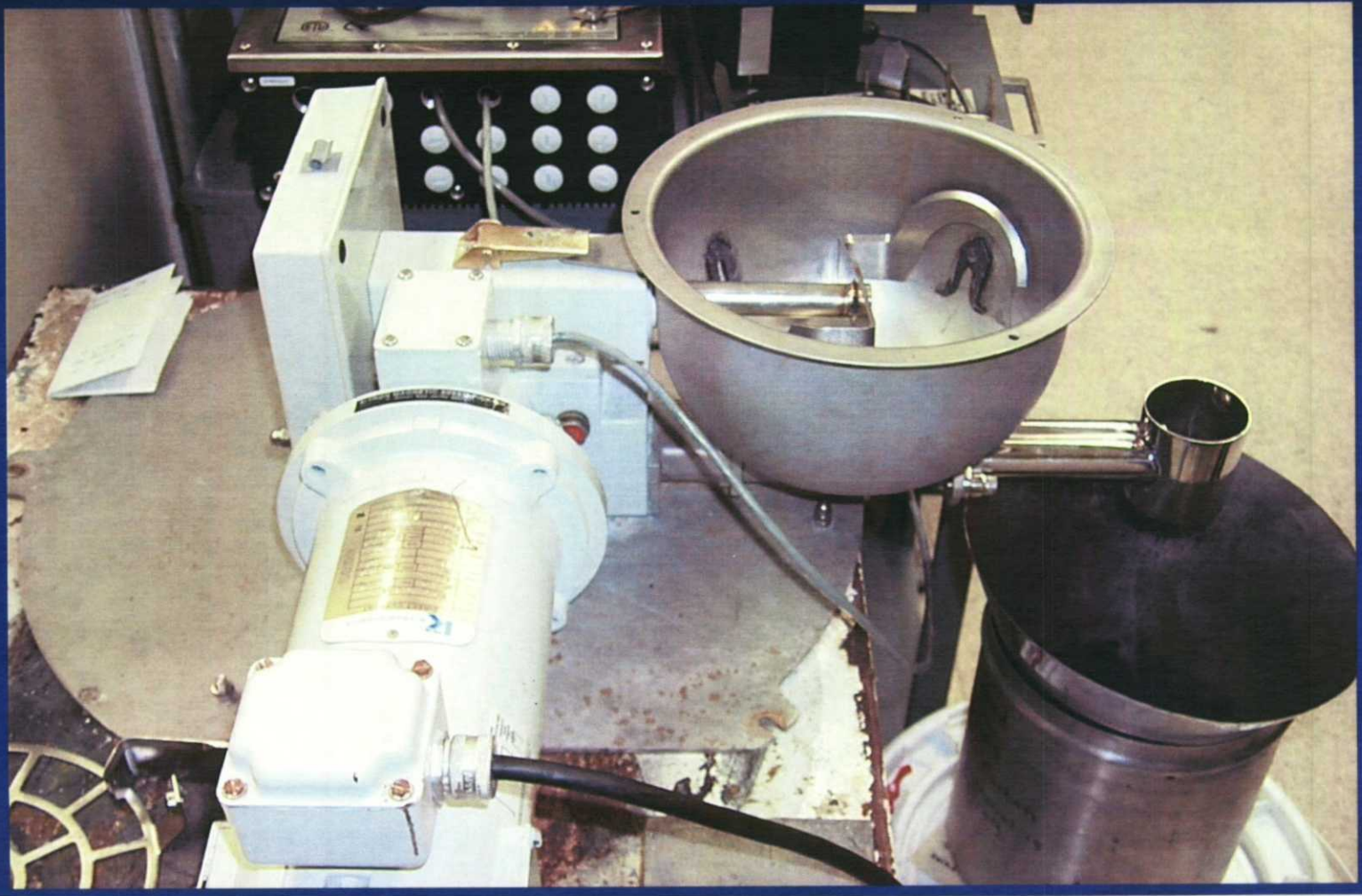
Screwfeeder



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Screwfeeder Agitator and Screws

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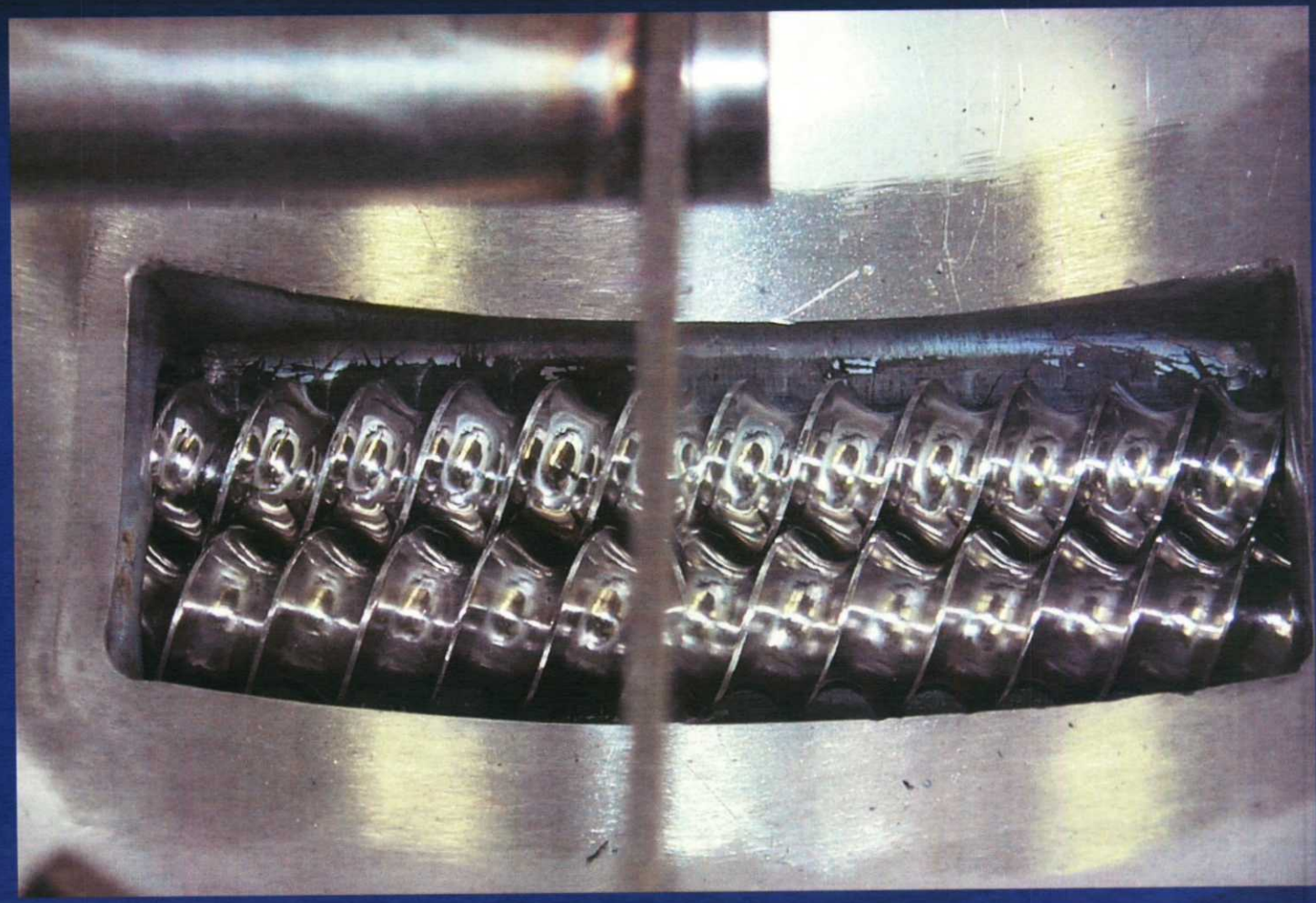
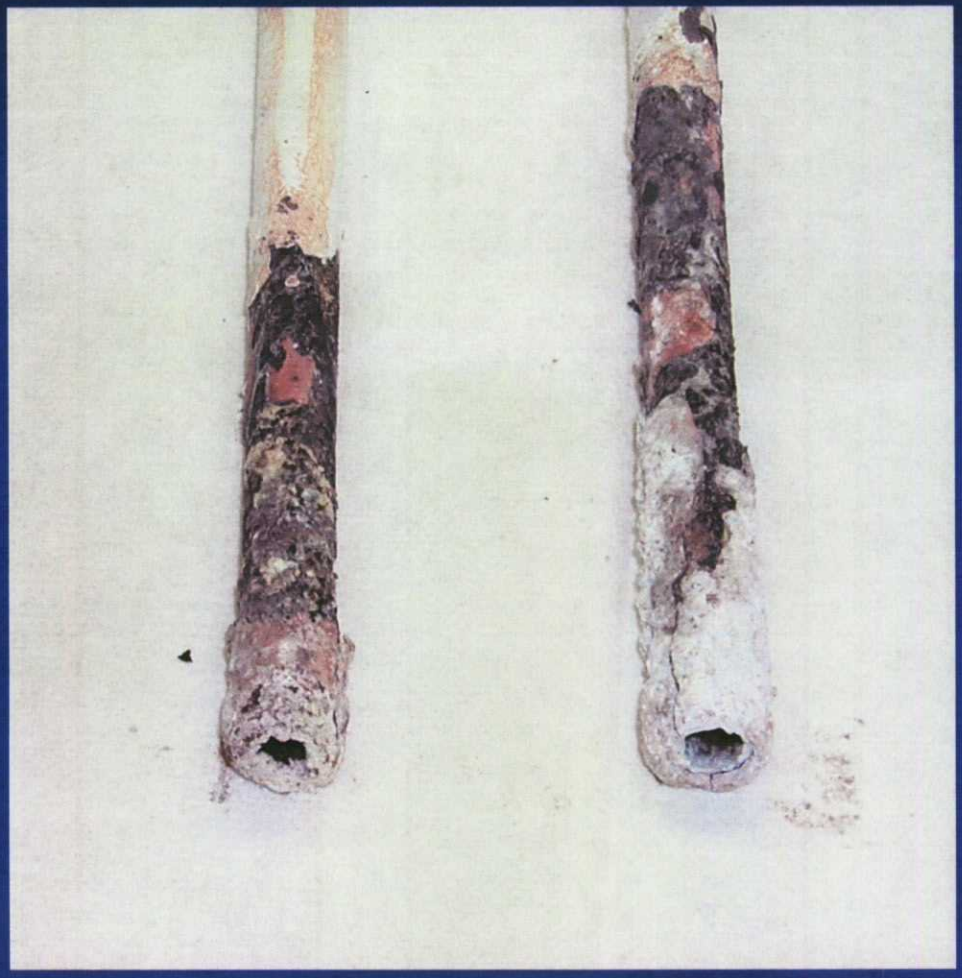
Double Screws



Materials



Inside the Vessel

Alumina crucible and downcomer

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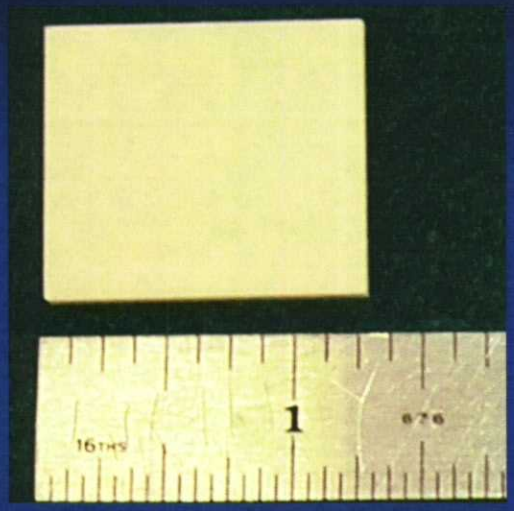
MSO Material Corrosion Tests

- Alumina has the best anti-corrosion characteristics, but has thermal shock sensitivity
- Carry out corrosion tests on thermal shock resistant materials, e.g., SiAlon, titania-alumina composite
- Aluminum nitride has both exceptional thermal shock resistance and corrosion resistance.

Corrosion Test with AlN



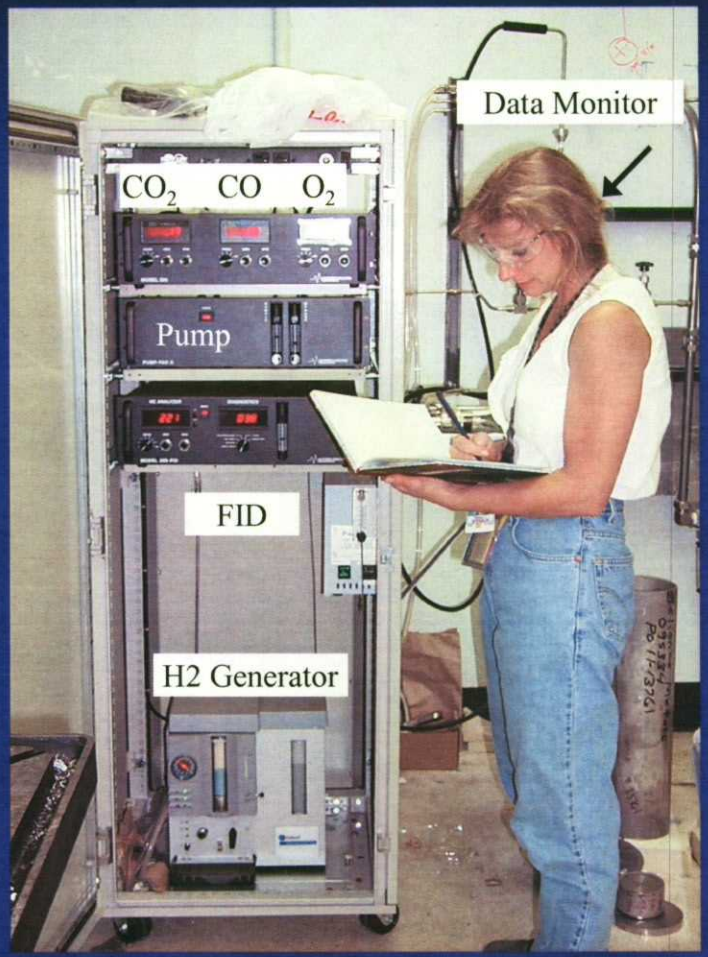
Aluminum Nitride

Aluminum Nitride vs Alumina

Thermal conductivity (high)
175 vs 25

Thermal expansion (low)
3.6 vs 8.1

Analytical Instruments



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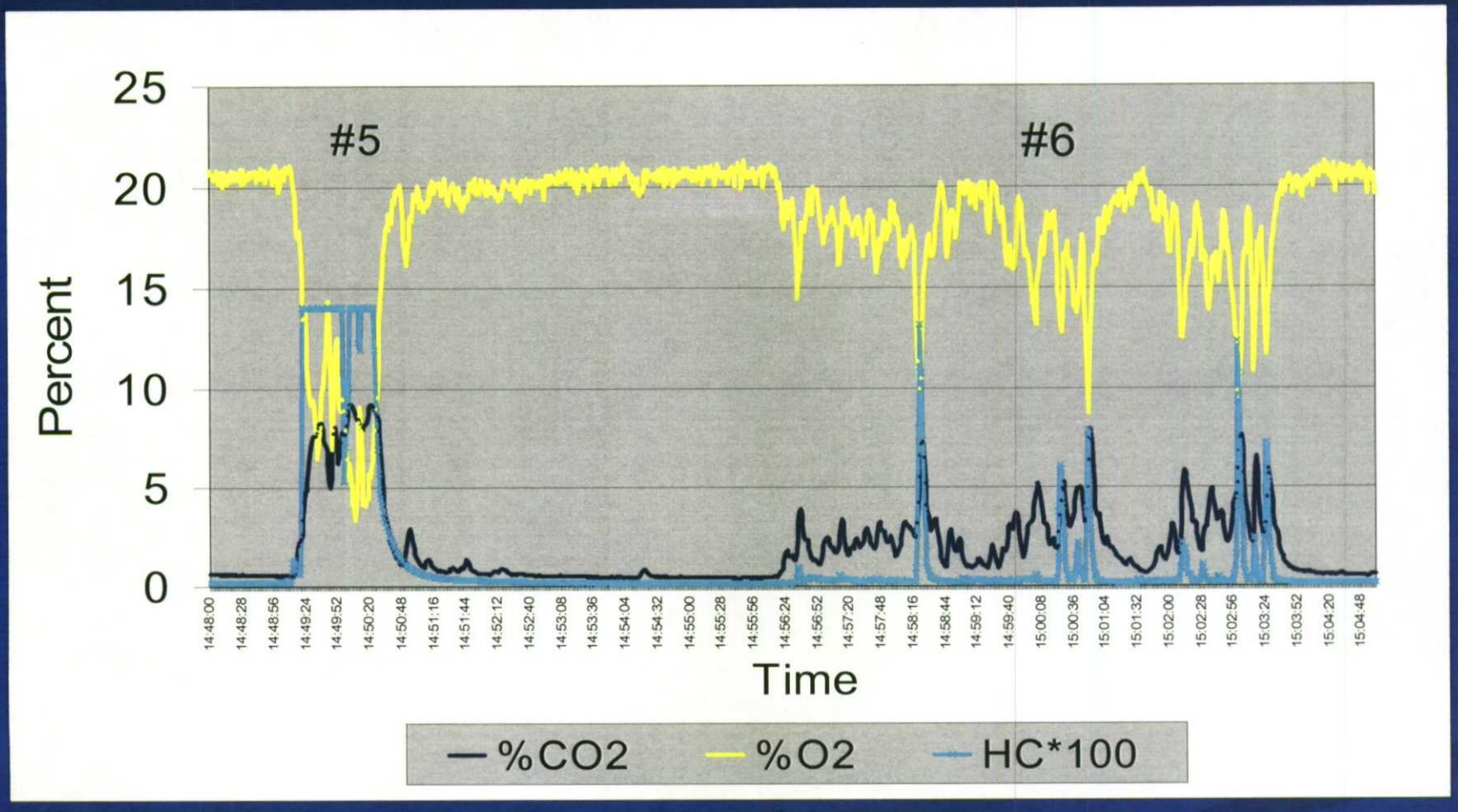
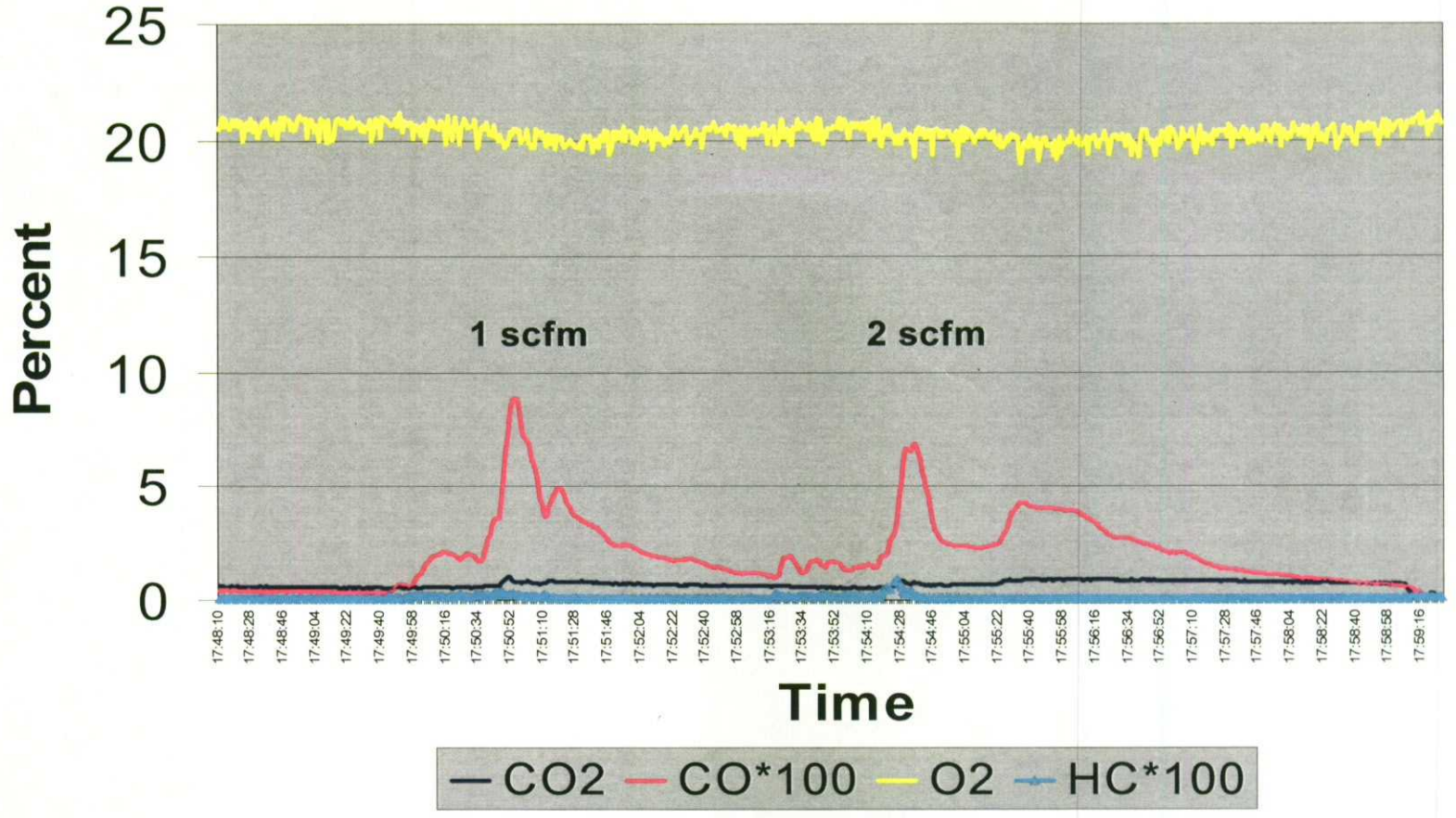
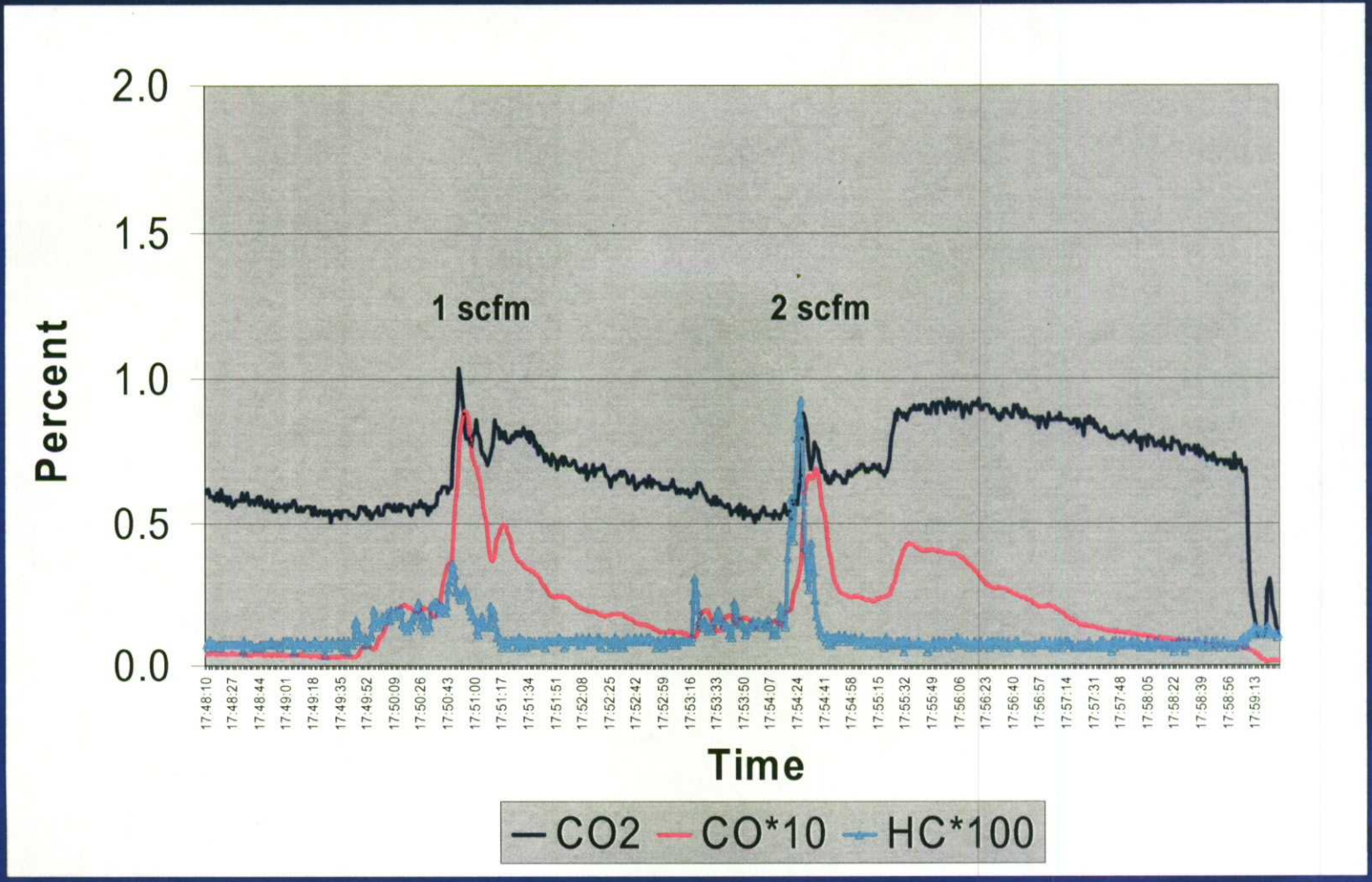
LDPE at 5 g/min (#5) and 1 g/min (#6)



Ash Feedstock



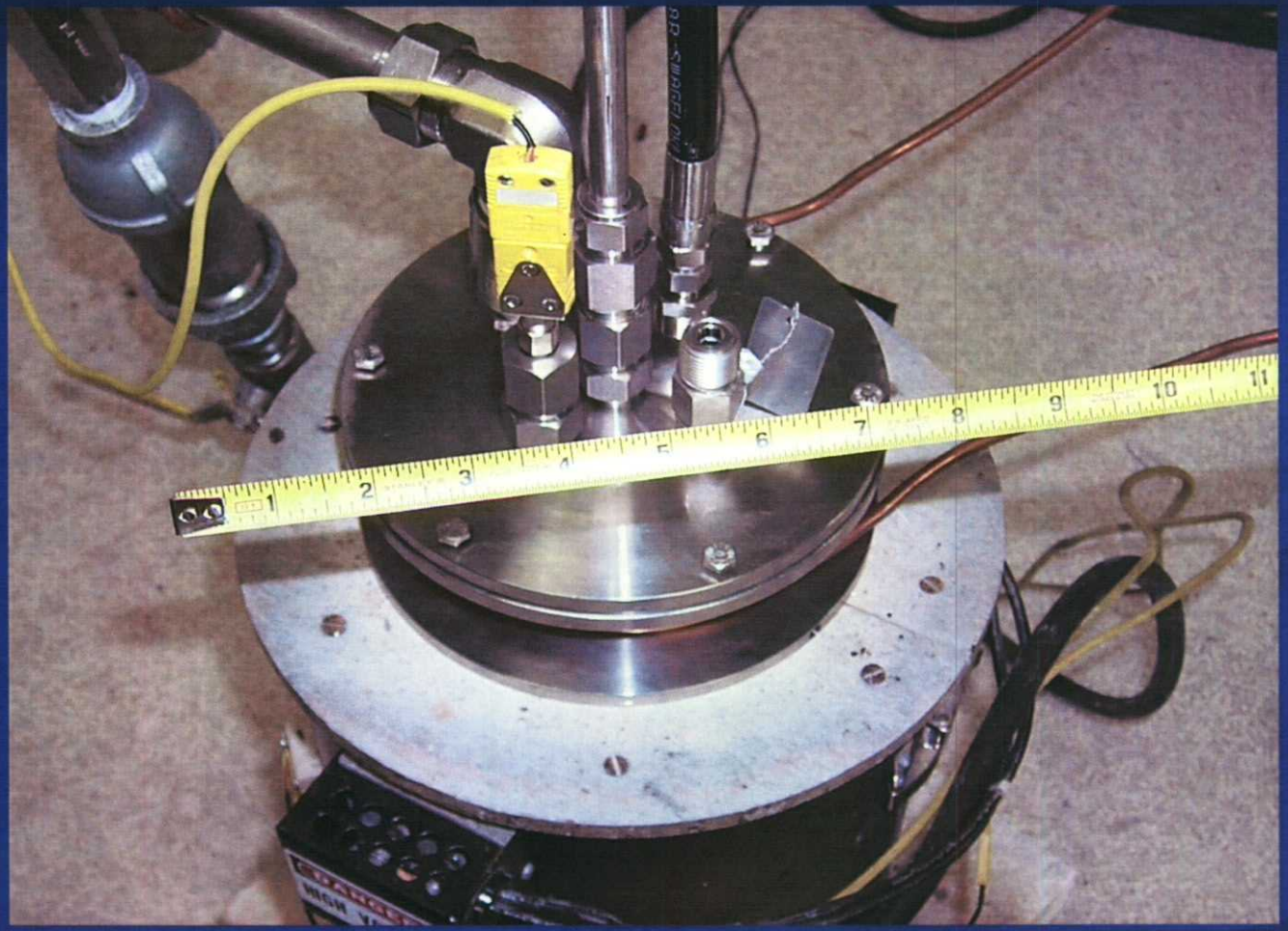
Ash Feedstock



Two-pronged Approach

- Install batch mode unit in Pu Facility
  - ✓ Small unit, authorized for batch mode
  - ✓ Real experience with glovebox and plutonium
- Continue to develop continuous mode unit
  - ✓ Materials testing for crucible
  - ✓ Ash feedstock experimentation

Bird's eye of Batch Unit



MSO Vessel with Inner Liner & Crucible

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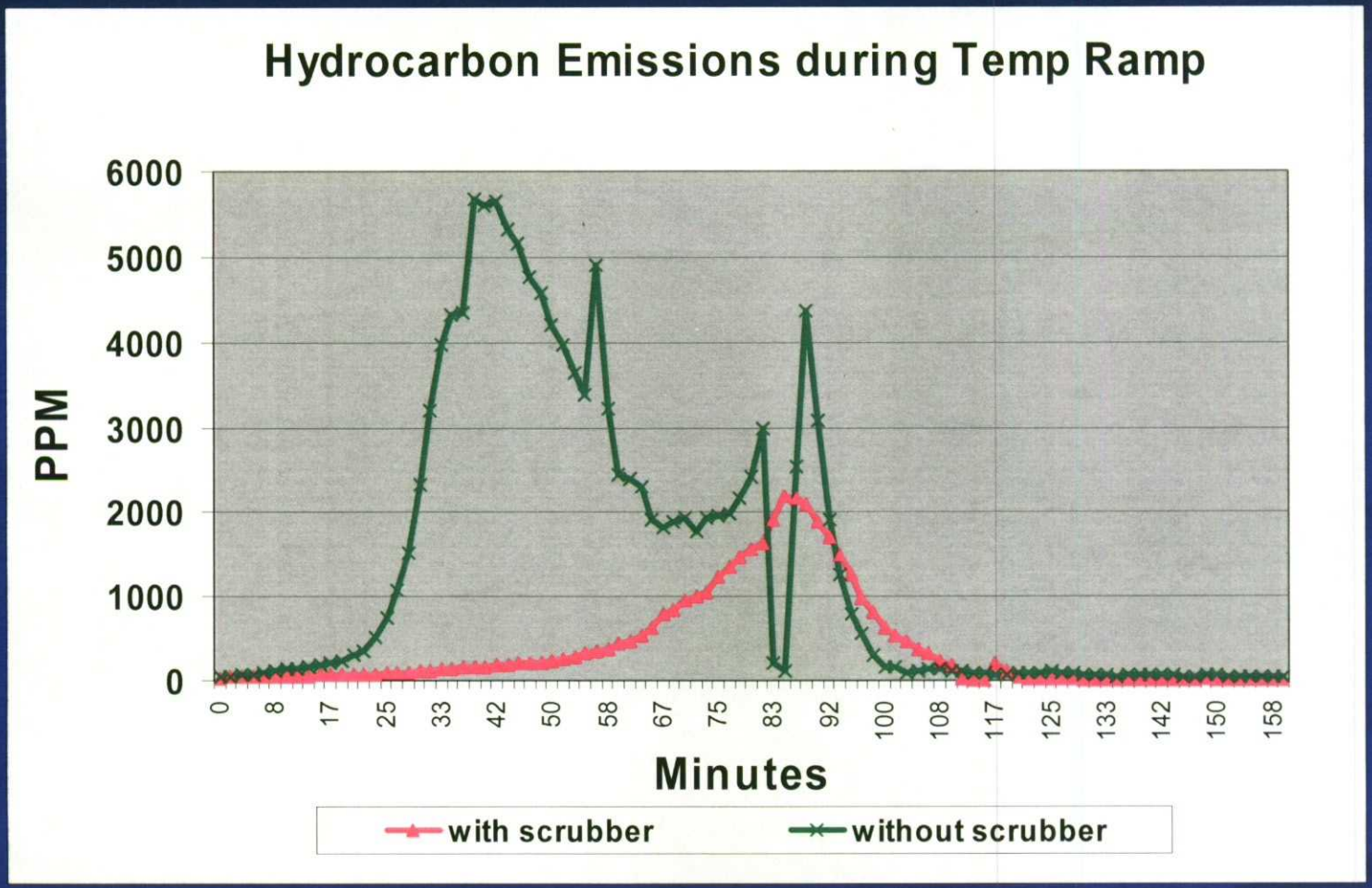
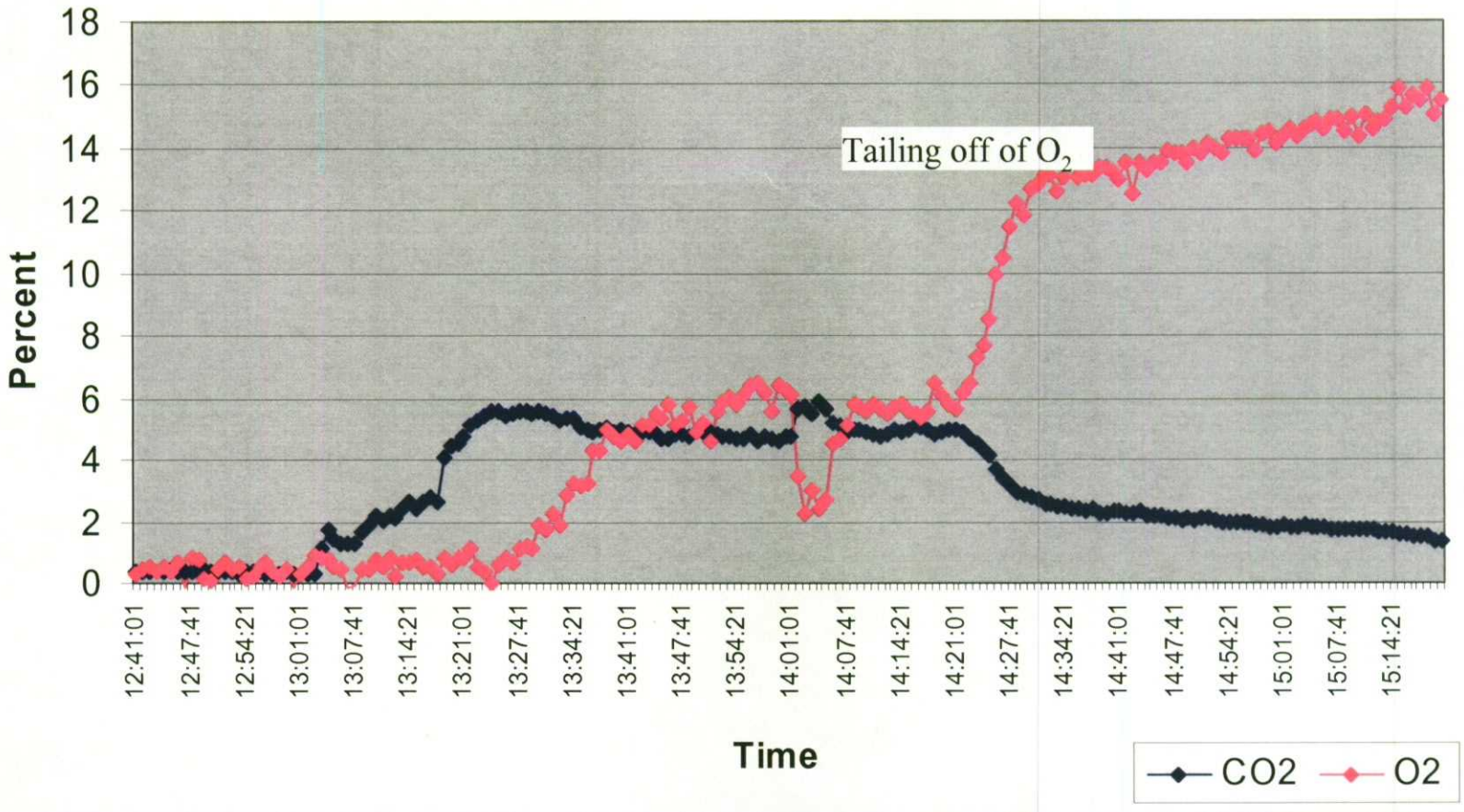
Batch Mode HC Evolution



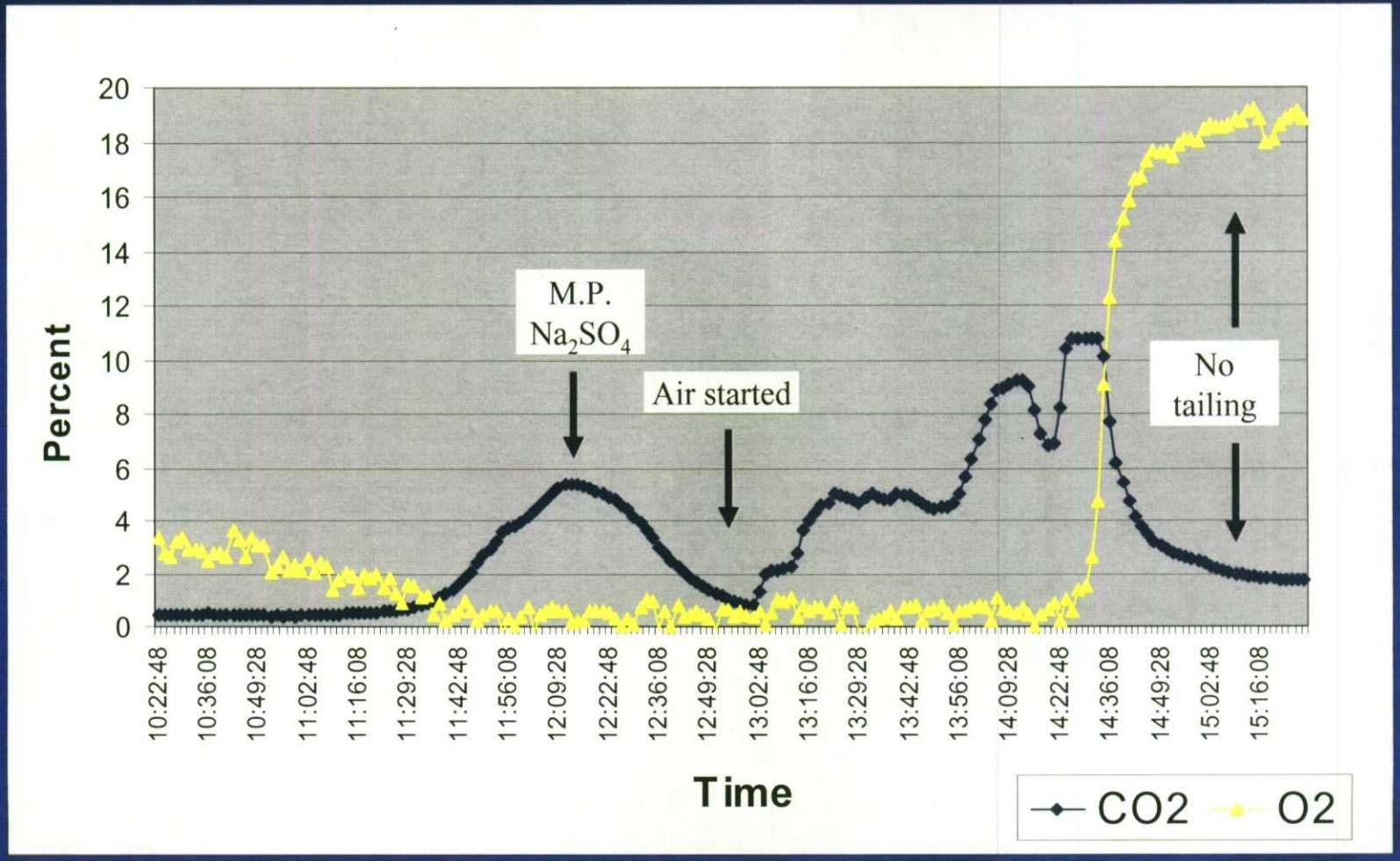
Batch Mode MSO Data



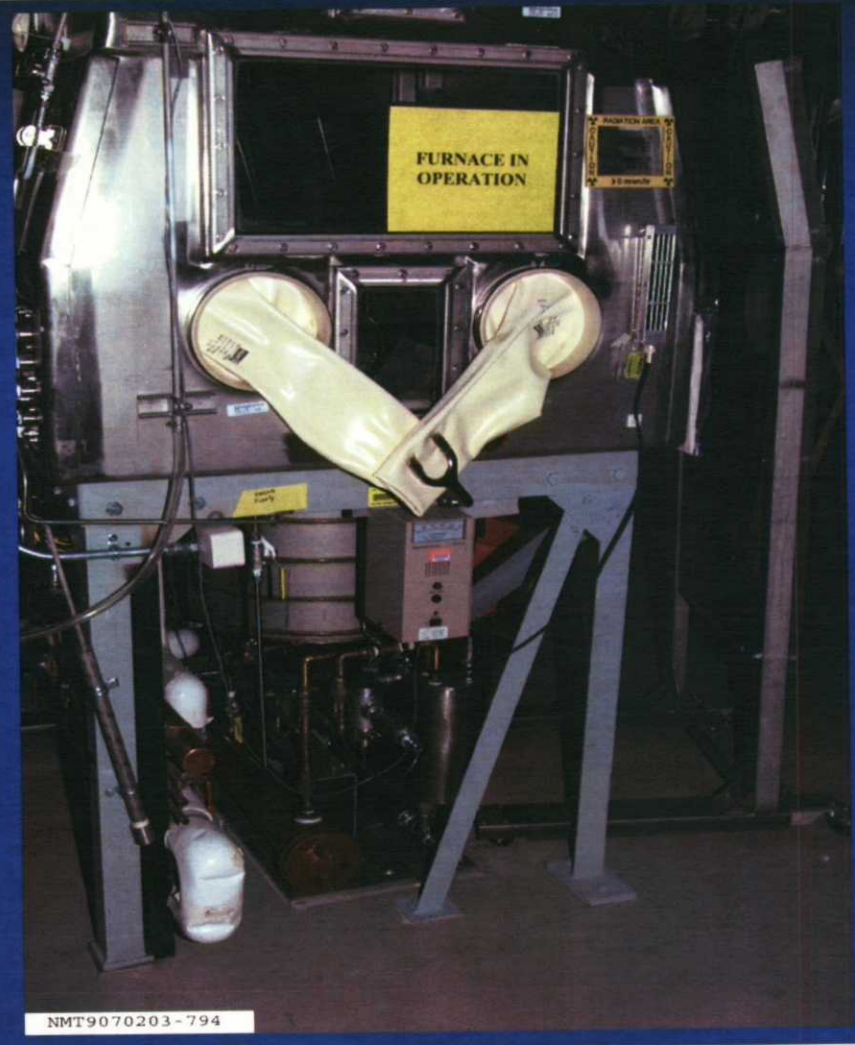
Bad Result

Batch Mode MSO Data

Catalyst added



Glovebox with Furnace Well



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Salt Plug from 1st MSO Run in GB

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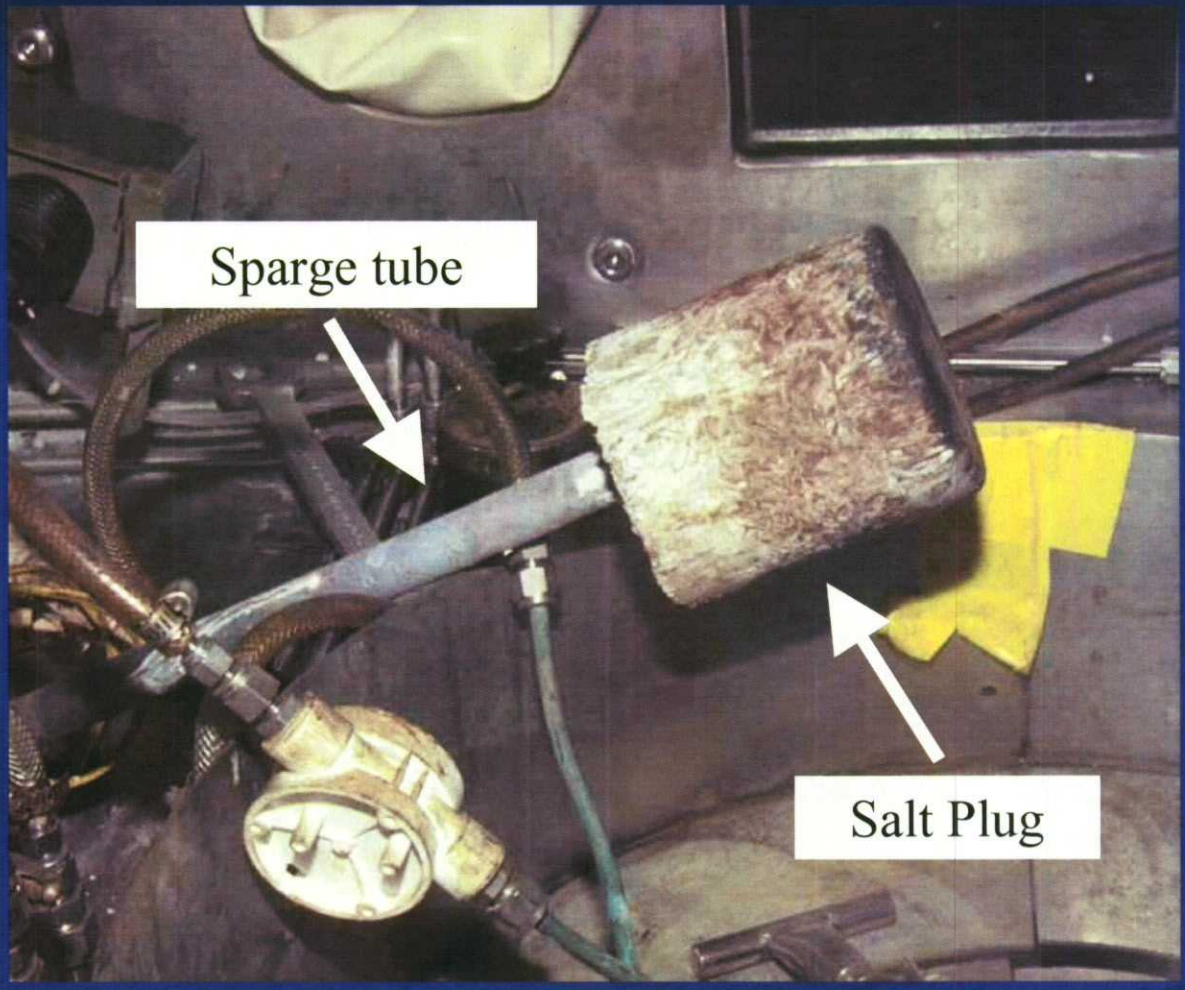
Popsicle Stick



Work, work, work

- Testing materials and crucibles
- Feed system/feed material
- Improve throughput via design changes and parameterization.
- Systems integrated and installed in Facility

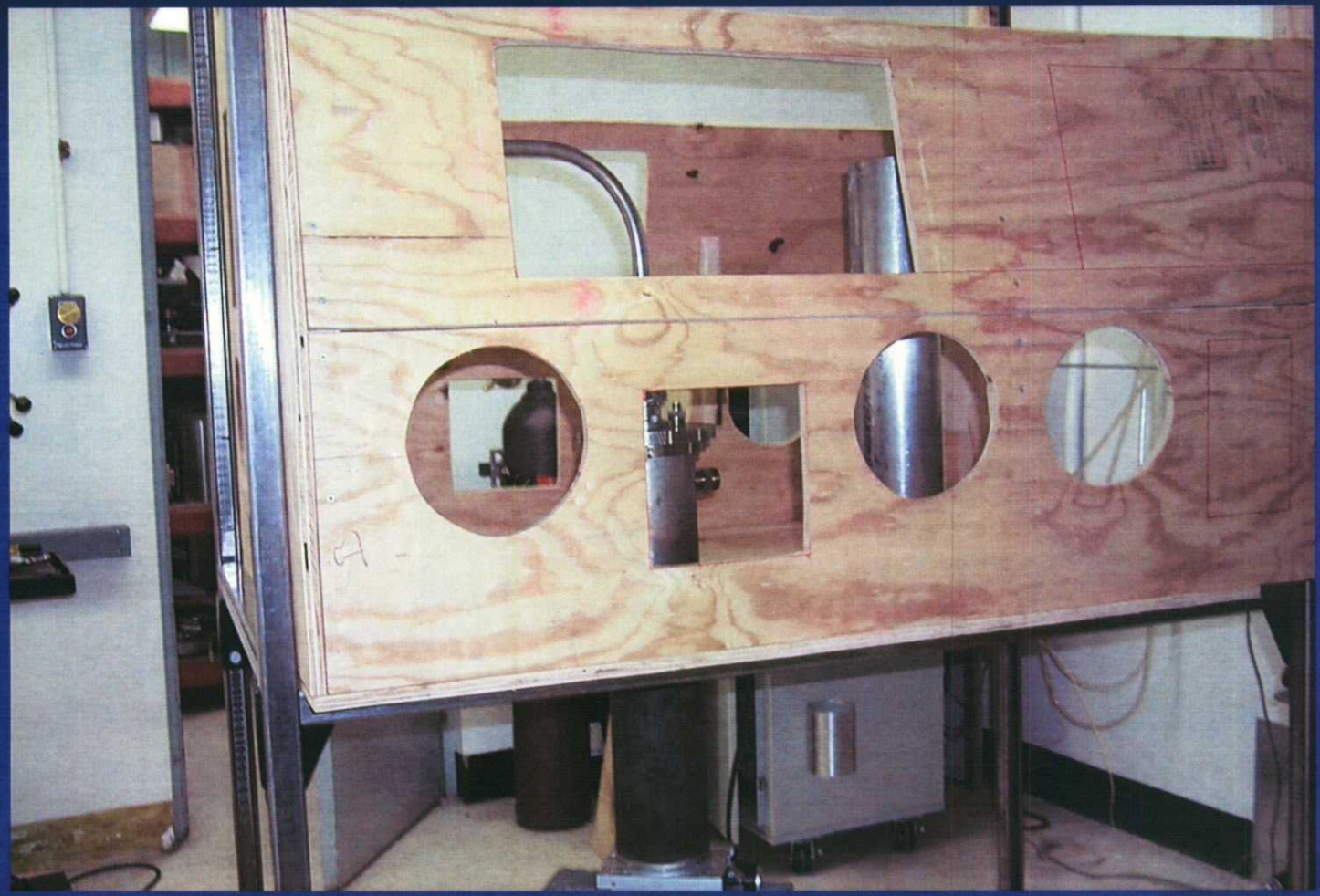
MSO Unit Changes

- Procured inner liners of Inconel 600
- Smaller diameter feed tube
- Shorten vessel by 4 in.
- Inconel 600 downcomer

Ancillary System Changes

- Analytical system
  - ✓ Upgrade of CO and CO_2 ranges
- Furnace & Controller
  - ✓ New controller "fabbed" by our shop
  - ✓ Furnace needs "refurbishing"
- Screwfeeder
  - ✓ Working with vendor on custom order
  - ✓ Improvising hopper agitation system
  - ✓ "Mocking" glovebox space to place feeder

Fits like a glove box



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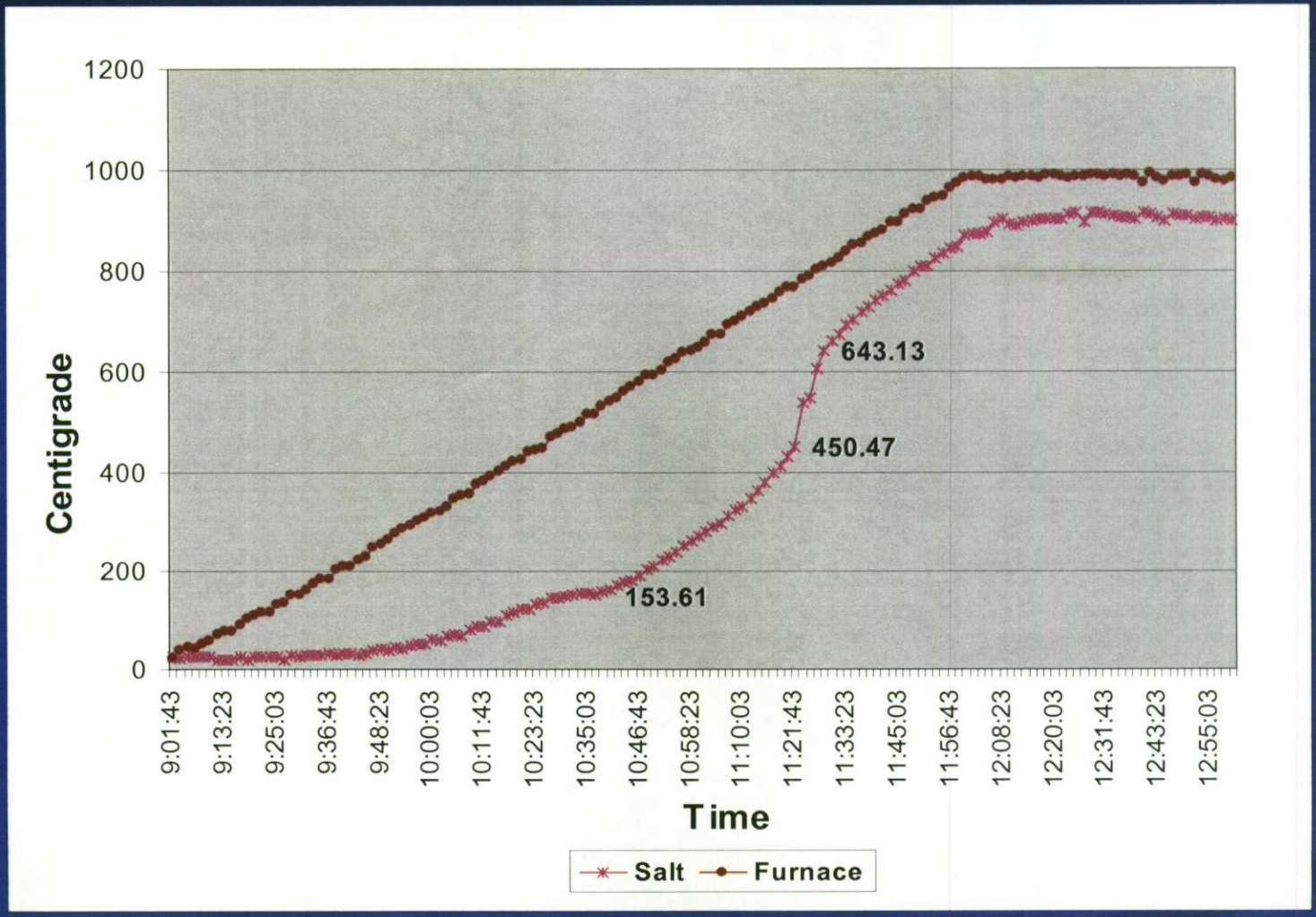
Li/K Eutectic Salt Mix (500 C)



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Ailing Furnace

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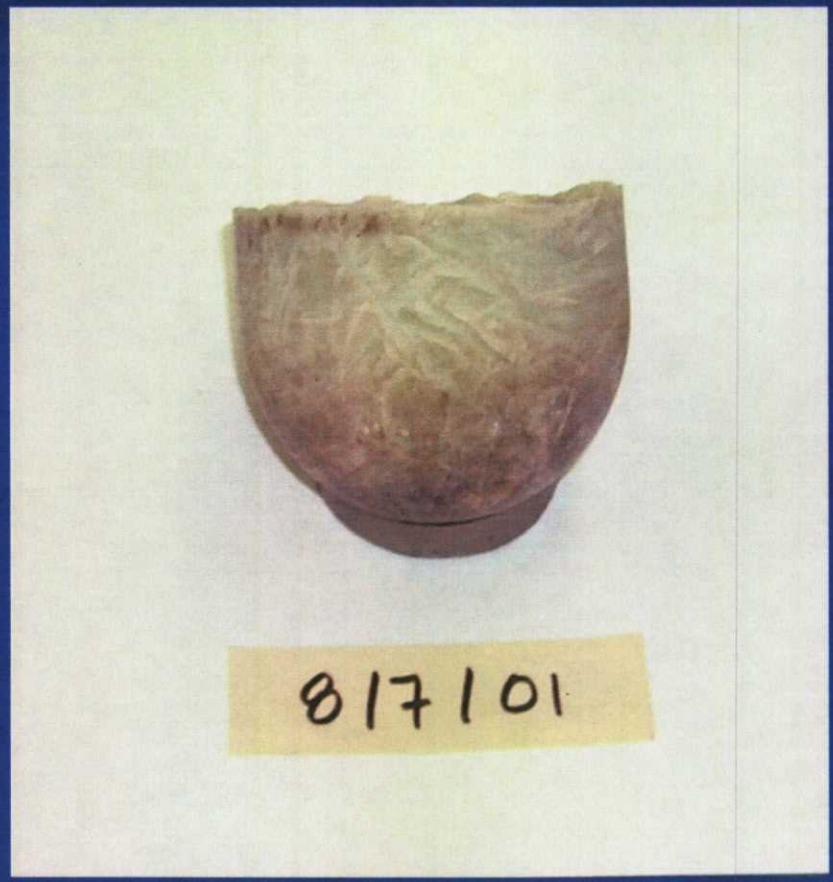
Salt Plug



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Nut Grinder

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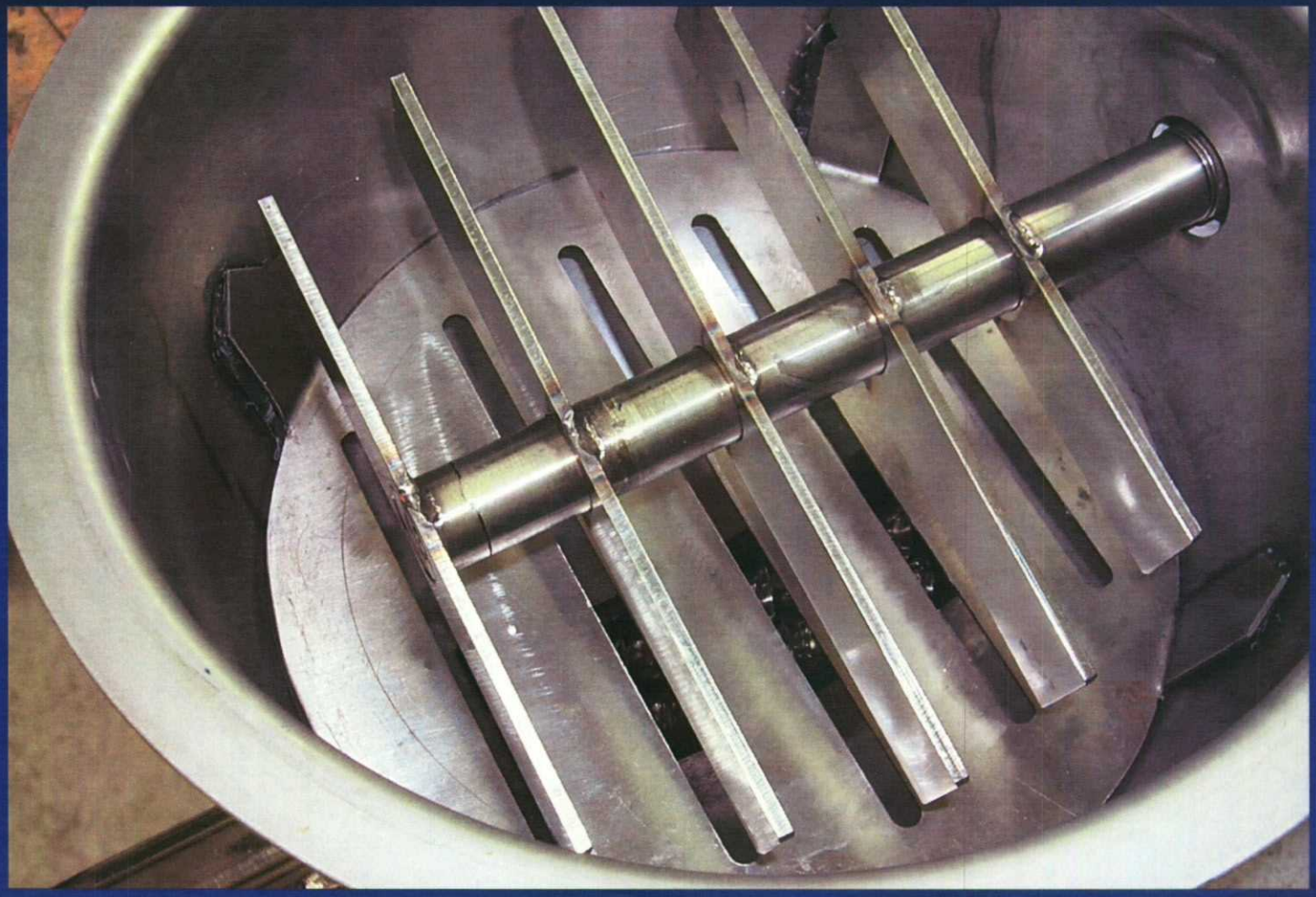
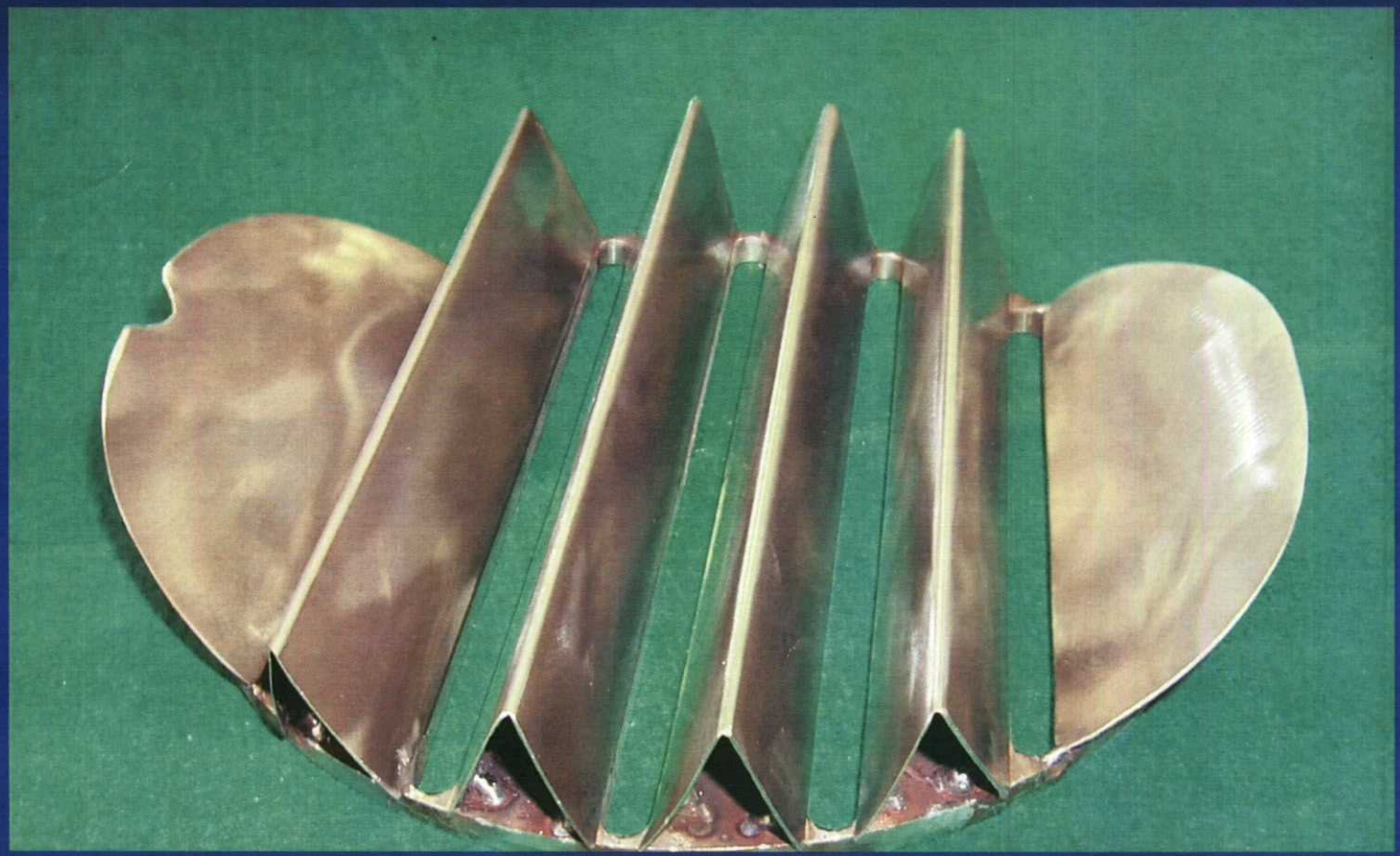
Clump buster



Modified Grill



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