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The air-oxidation of high-purity molten aluminum: temperature-dependent rates and energetics

Eric N. Coker, Walt Gill and Burl Donaldson

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NNSA National Nuclear Security Administration

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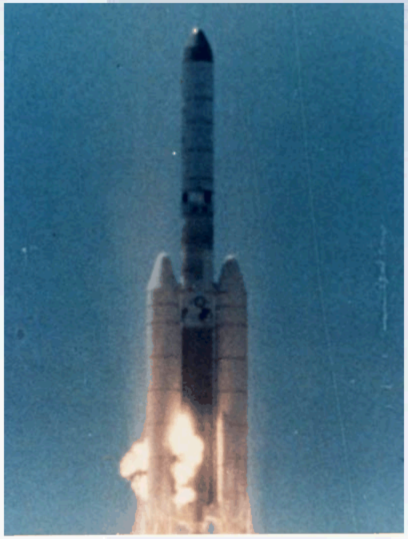
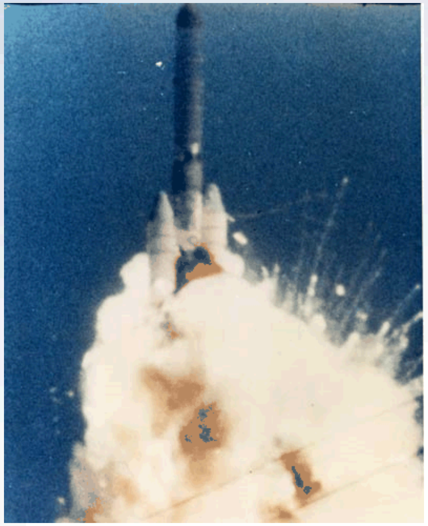
Introduction

Program objectives

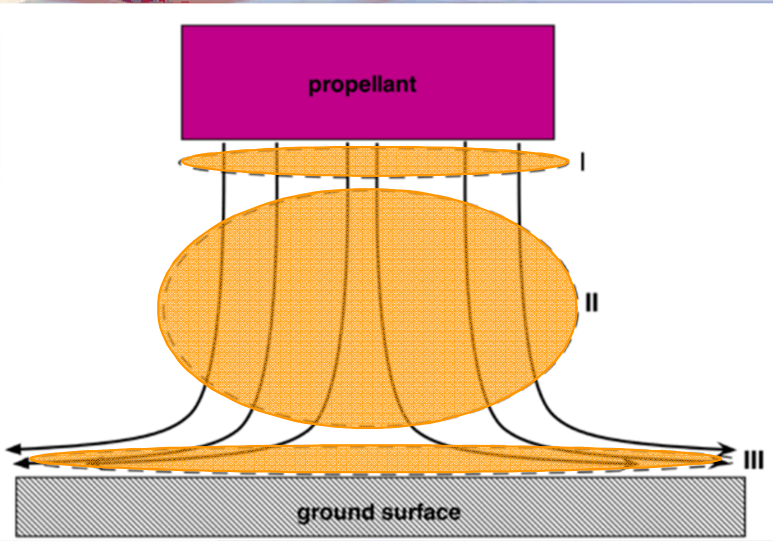
- **Define the thermal-chemical-physical environment in and around a plume formed from a burning fragment of solid rocket fuel at atmospheric pressure.**
- **Use the specification to quantify the risk associated with a launch pad abort involving solid fueled rocket motors and high hazard payloads (e.g., deep space power source).**



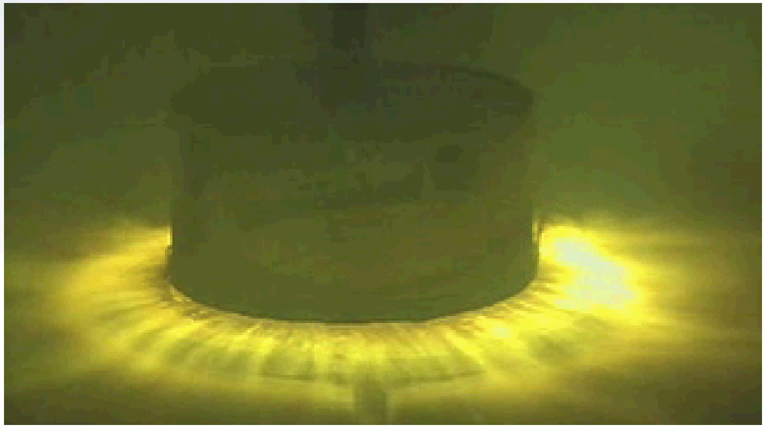


Titan 34D-9 accident sequence, 1986

Regions of Analysis

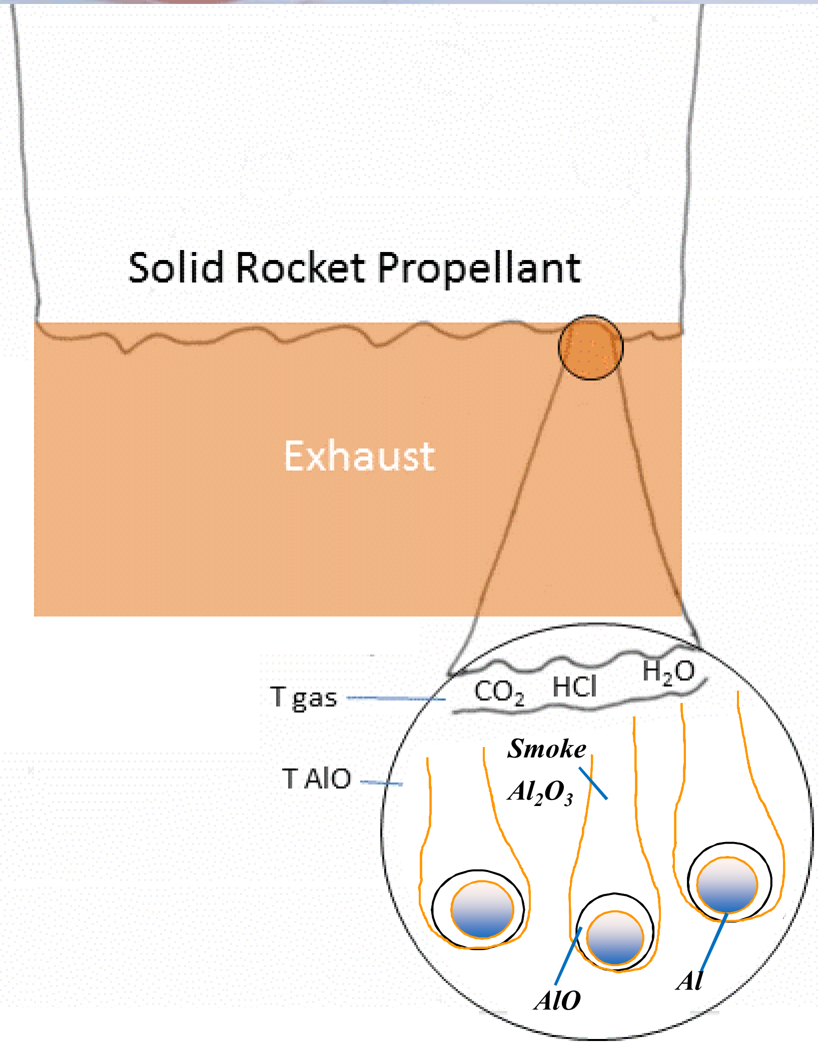


- Region I: combustion of Al + ammonium perchlorate (+ binder, etc.)
- Region II: populated by hot, emitting gases (HCl, CO_2, H_2O) and particles (AlO, Al_2O_3, etc.)
- Region III: interaction with hazardous payload; possible feedback to Region II (e.g., re-radiation)



- *Physico-chemical behavior of burning Al strongly correlated to the environment, i.e., <u>properly functioning</u> motor at high pressure/well defined geometry versus <u>off-normal</u> occurrence.*
- *Major concern: vaporization of hazardous payload material in Region III, condensation on available particulate and transport to the environment.*

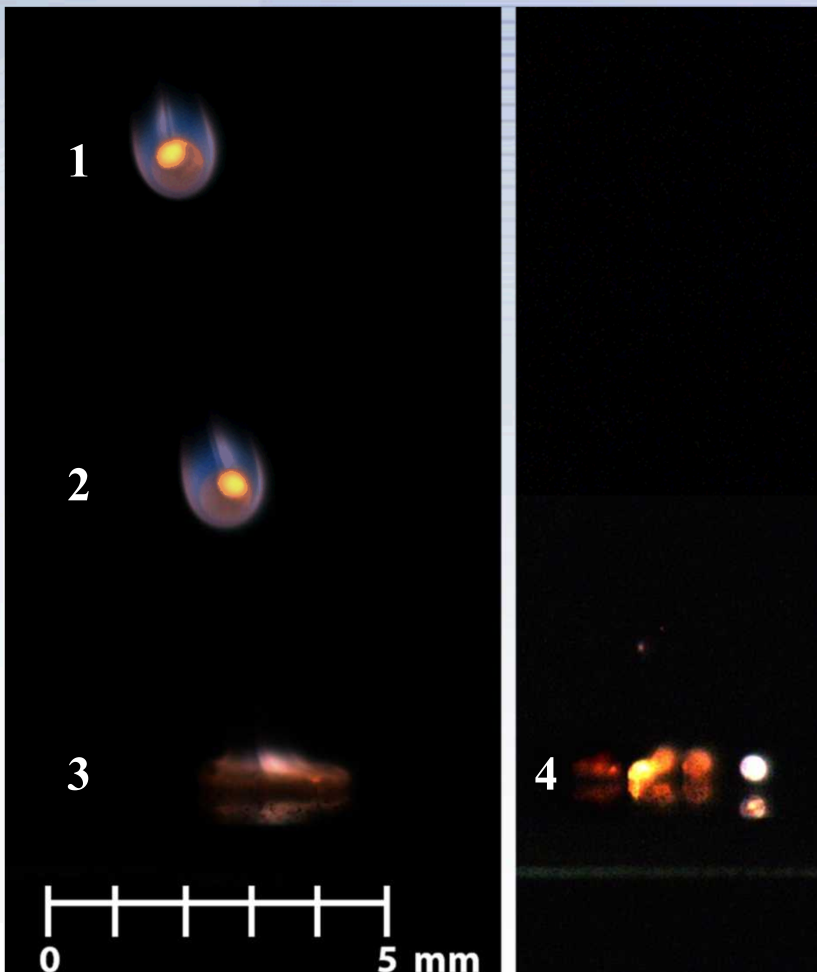
Region I & II Dynamics



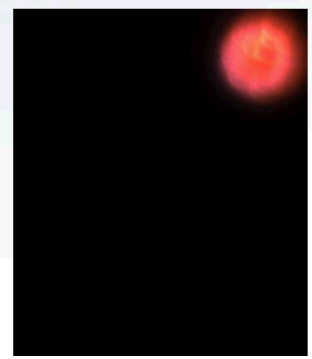
- *Region I shows the burning aluminum and ammonium perchlorate*
- *Multiple temperatures are present in Region II, complicating measurement and models*
- *AlO is believed to have some of the highest temperatures in this region*
- *The smoke and soot (non aluminum related) has the lowest temperature in Region II, probably conforming to a grey body profile*
- *The gases (CO_2, H_2O, and HCl) fall in the middle of temperature ranges*

Complex Aluminum Burning

Model formulation data ***are acquired with small scale bench top experiments. For example, new physics are being introduced into the burning droplet models***



- ***Conventional models view the burning drop as an evaporating molten bare aluminum sphere with an alumina cap.***
- ***The recent literature suggest other modes of burning are possible – a crust covered sphere with burning through cracks.***



Study oxidation behavior of Al under well-controlled conditions

– data to assist model development for launch failure risk assessment

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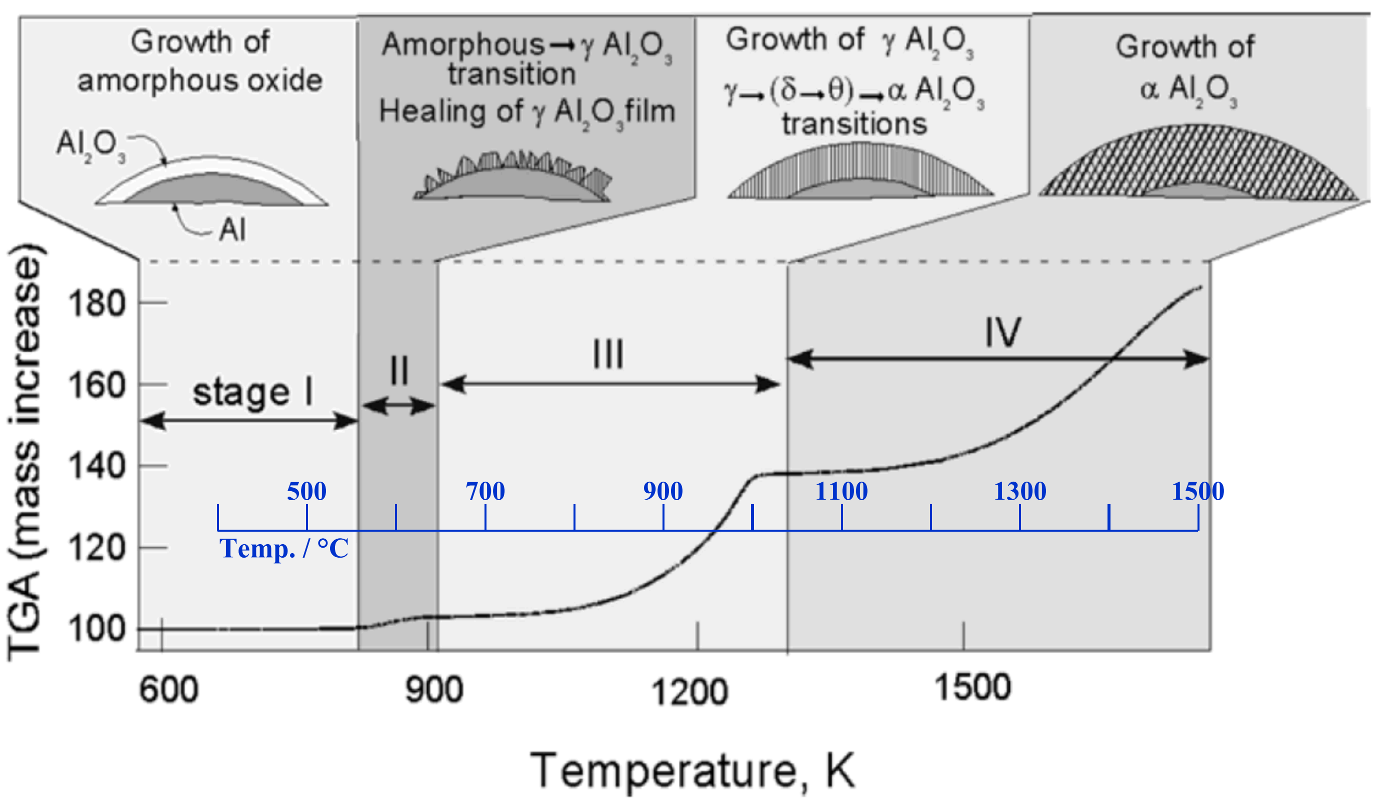
Transitions in Al_2O_3 layer on Al metal



Trunov, M.A., Schoenitz, M. and Dreizin, E.L., "Effect of polymorphic phase transformations in alumina layer on ignition of aluminium particles." *Combustion Theory and Modelling,* **10(4)**, 603-23, (2006).

Thermal Analysis

Materials

- *Al foil; 99.998% (Johnson Matthey)*

Instrumentation

- *Thermogravimetric Analysis (TGA)* — *Δ mass*
- *Differential Scanning Calorimetry (DSC)* — *exo/endotherm*
  - *Simultaneous TGA-DSC: Netzsch STA 449F3*

Procedure

- *~20 mg specimen (6 mm disc) in Al_2O_3 cup (6.6 mm o.d.)*
- *Ramp to desired oxidation temperature under Ar (inert)*
- *Allow system to equilibrate at temperature*
- *Expose to air, hold isothermally*

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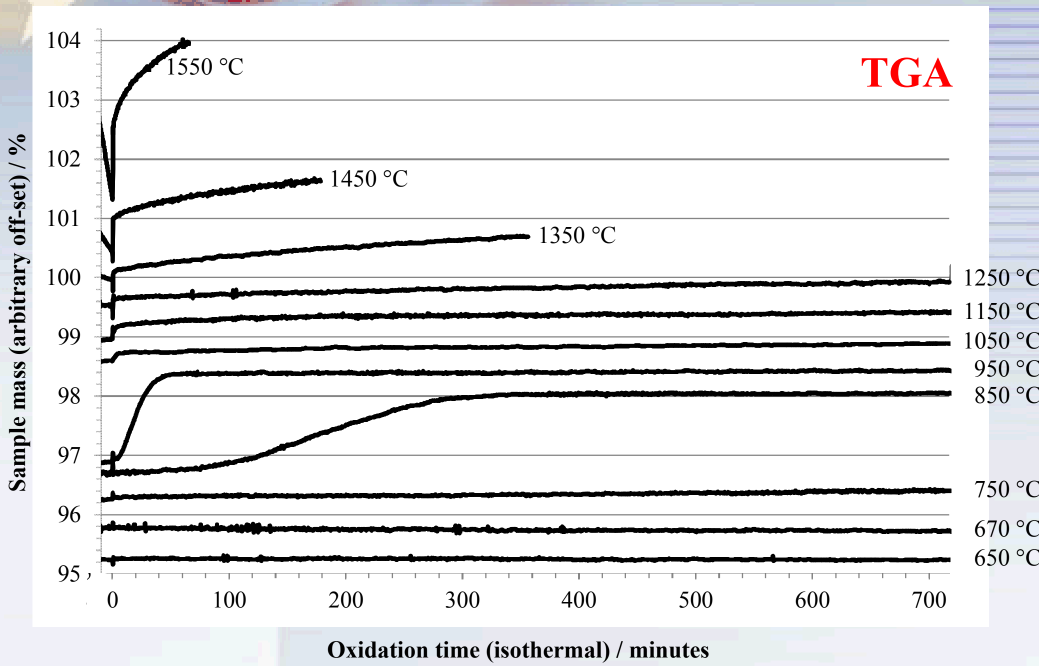
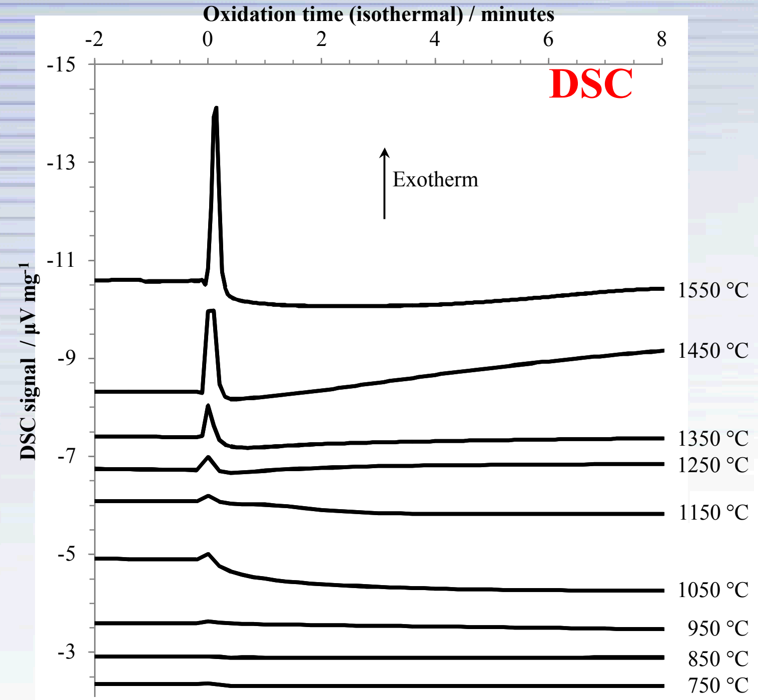
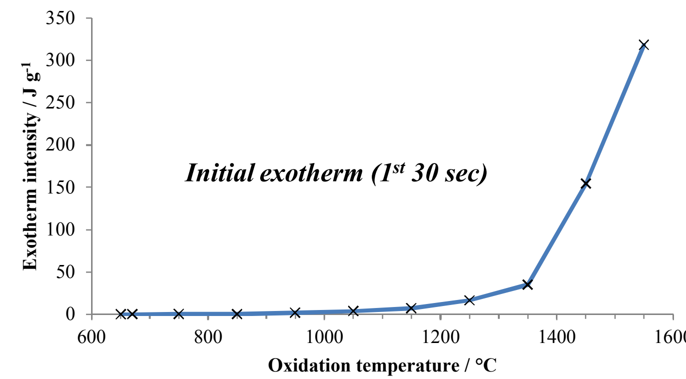
Oxidation of Al foil in air







- **Below 850 °C – little oxidation**
- **850 to 950 °C – oxidation**
- **Above 950 °C – passivation**
- **Above 1250 °C – rapid increase in oxidation rate**

$$2Al + 1.5\,O_2 \rightarrow Al_2O_3 \quad \Delta(mass) \sim +90\ \%$$

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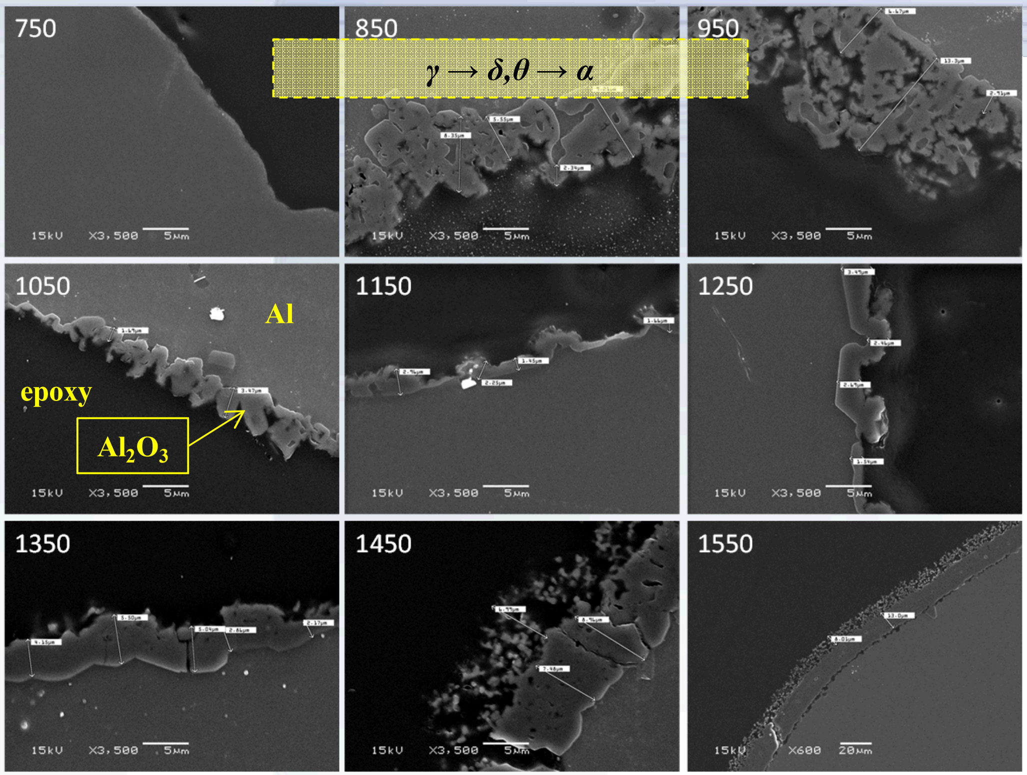
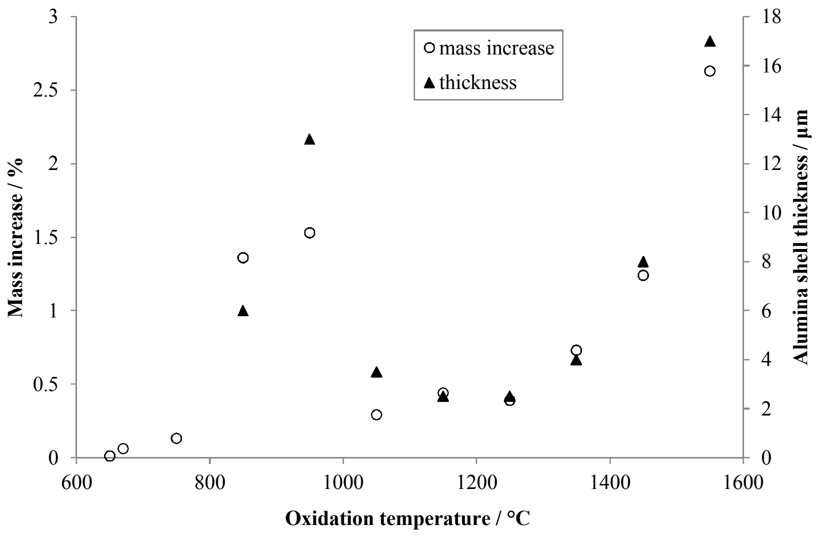
Oxide layer growth

Electron microscopy images of cross-sections of specimen

Oxidation temperature (°C) indicated





Below 1050 °C, foil topology persists.

At 1050 °C and above, spheroidal particle forms.

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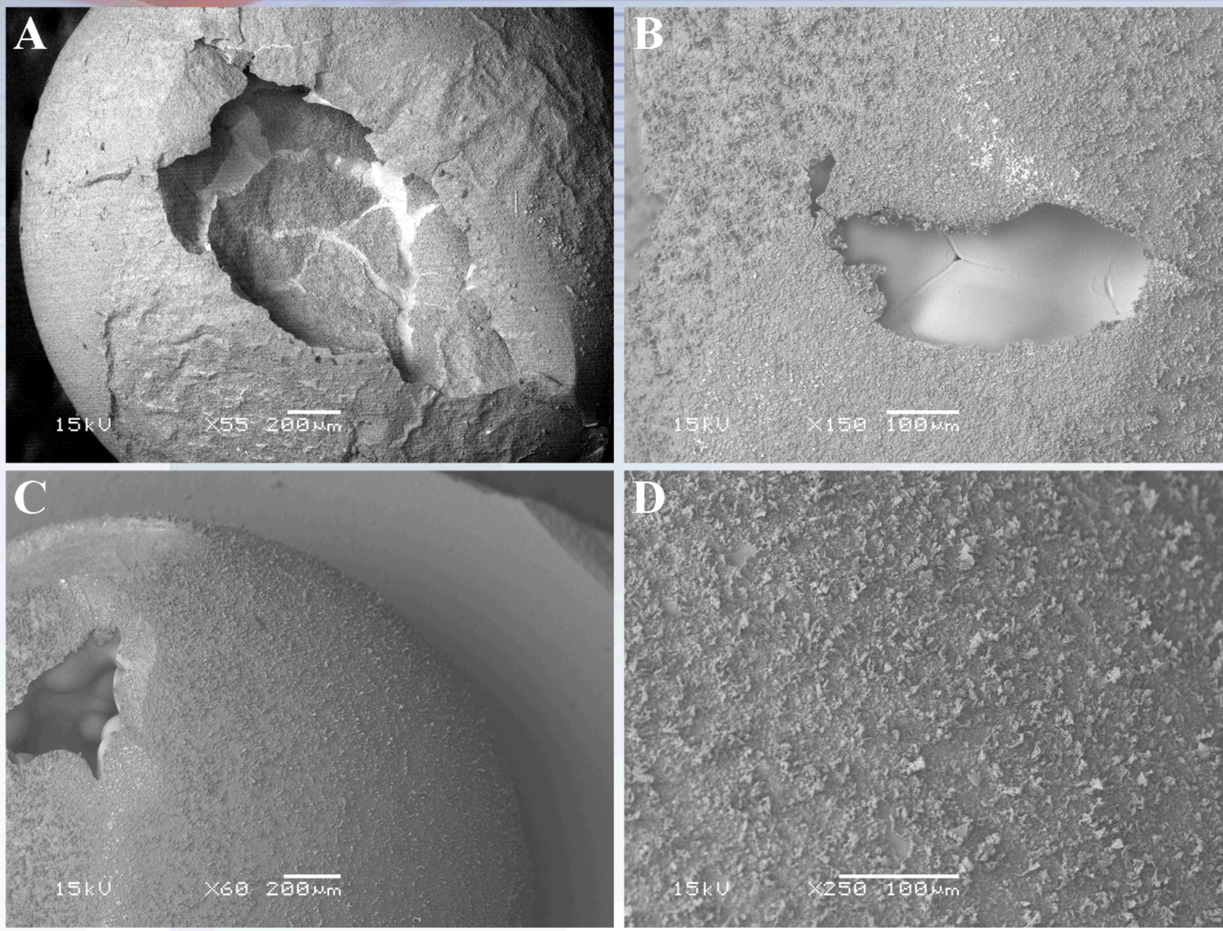
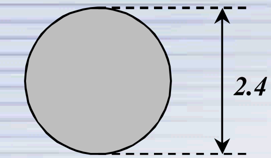
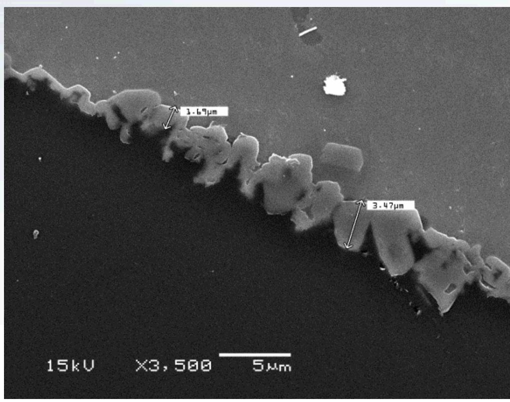
Alumina shell after oxidation at 1050 °C





Cross-section



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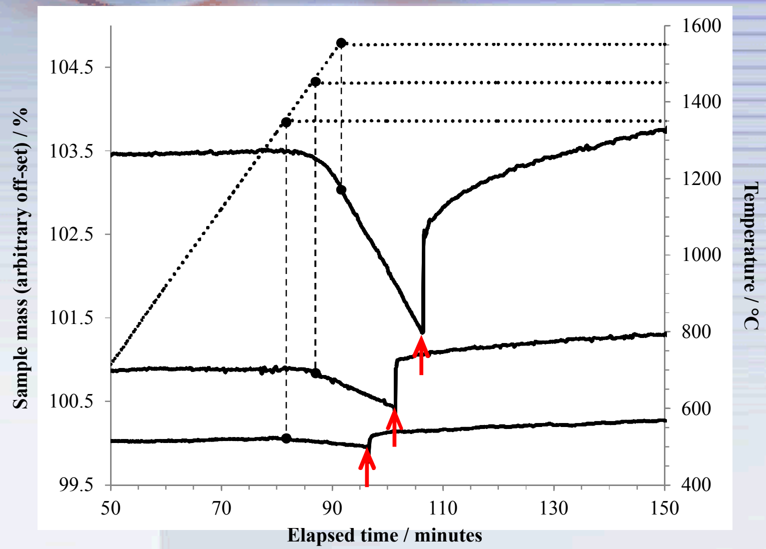
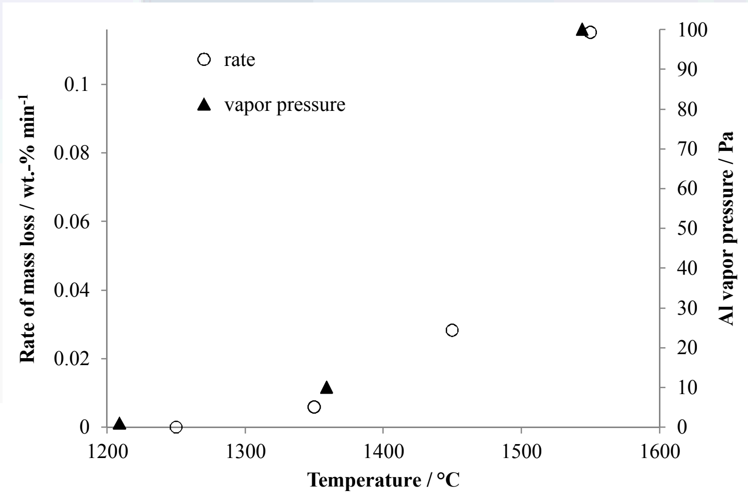
Al volatility at high temperature



Volatility noticeable at 1350 °C and above.

↑ = introduction of air



Measured rate of vaporization proportional to published Al vapor pressure.

Al vapor pressure data taken from:

http://www.knowledgedoor.com/2/elements_handbook/vapor_pressure.html.

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Summary & conclusions

- High-purity aluminum foil heated under inert atmosphere and exposed to air at desired temperature
- Non-linear dependence of oxidation on temperature
  - Below 850 °C minimal oxidation occurred
  - At 850 °C oxidation occurred after "induction period"
  - At 950 °C oxidation occurred with no "induction period"
  - Above 950 °C passivation occurred (oxide barrier)
  - Above 1250 °C oxidation rate increased rapidly
- Degree of oxidation small in all cases (low surface area specimen)
- Oxide layer strength overrides Al liquid surface tension below 1050 °C
- Observed oxide shell thickness (SEM) correlates with mass increase
- Al/Al_2O_3 spheroidal particle shows partial hollowing after 1050 °C oxidation
- Al volatility under inert gas measureable at 1350 °C and above

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Jet Propulsion Laboratory
California Institute of Technology
Pasadena, California

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