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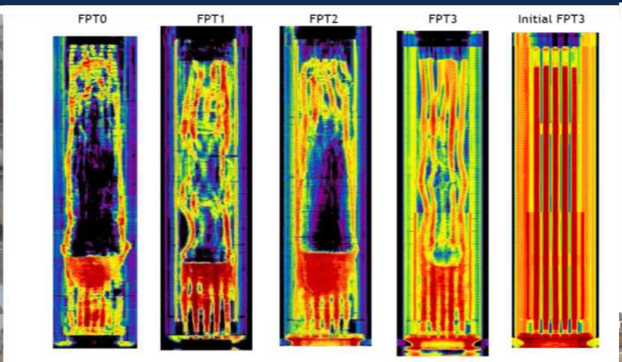

Severe Accident Phenomena

TSG Skill Set

Hydrogen

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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ZR CLADDING REACTION WITH STEAM AND HYDROGEN GENERATION

Cladding Oxidation by Steam

- **Oxidation of the Zircaloy cladding becomes significant when peak fuel temperature reaches about 1000°C**
- **The exothermic reaction is:**

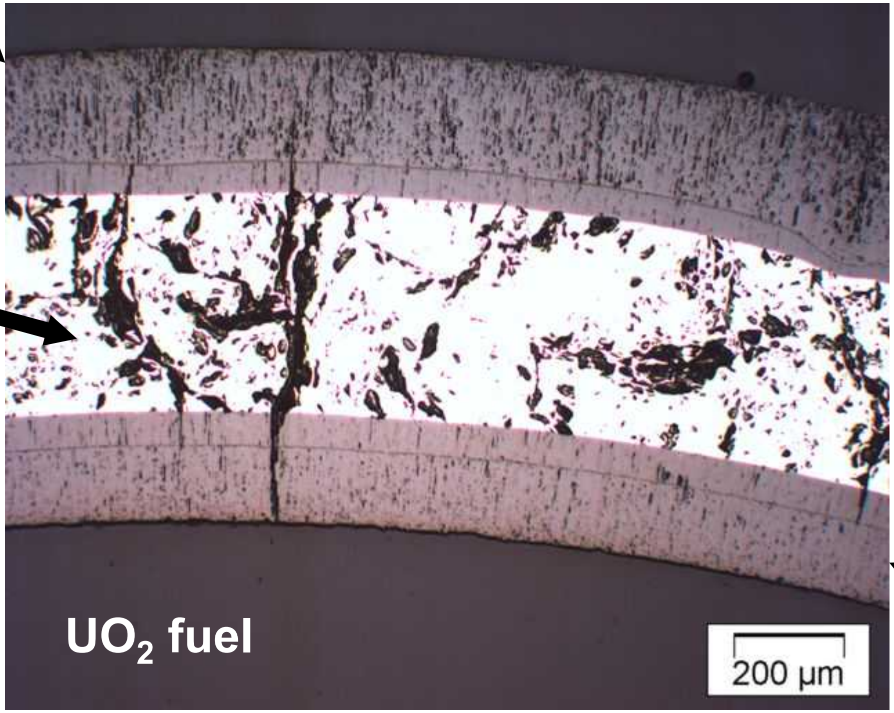
$$Zr + 2H_2O \Rightarrow ZrO_2 + 2H_2 + heat$$

- **ΔH_{rxn} = 6.5 MJ/kg_{Zr} (TNT = 4.6 MJ/kg)**
- **Reaction rate increases rapidly with temperature**
- **Oxidation is a positive feedback reaction**
  - **Exothermic oxidation increases clad temperature**
  - **Increasing clad temperature increases oxidation rate**
- **Oxidation limited by steam availability and by melting and relocation of Zr**
- **Significant quantities of hydrogen produced (650-900 kg from Fukushima Unit 1 uncertainty analysis)**

Steam Oxidation of Zircaloy Cladding

Outer Cladding

Residual oxygen saturated metal



Inner Cladding

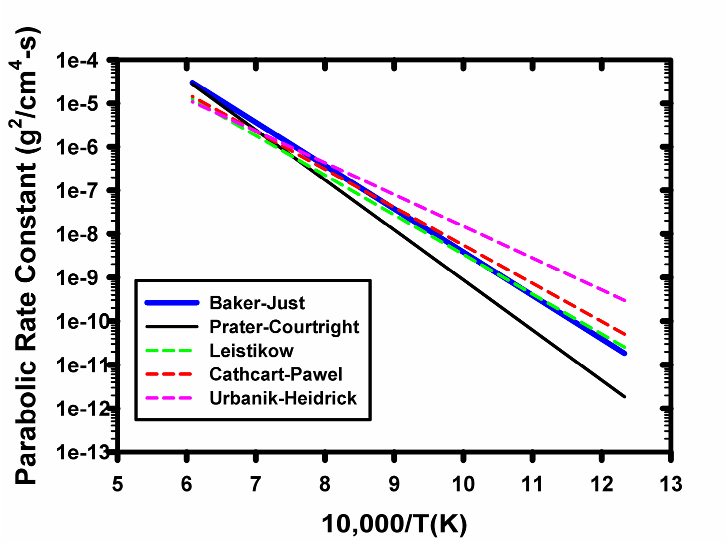
Oxidation Kinetics

- ***Parabolic kinetics***
  - *If limited by oxidant diffusion through ZrO_2,*

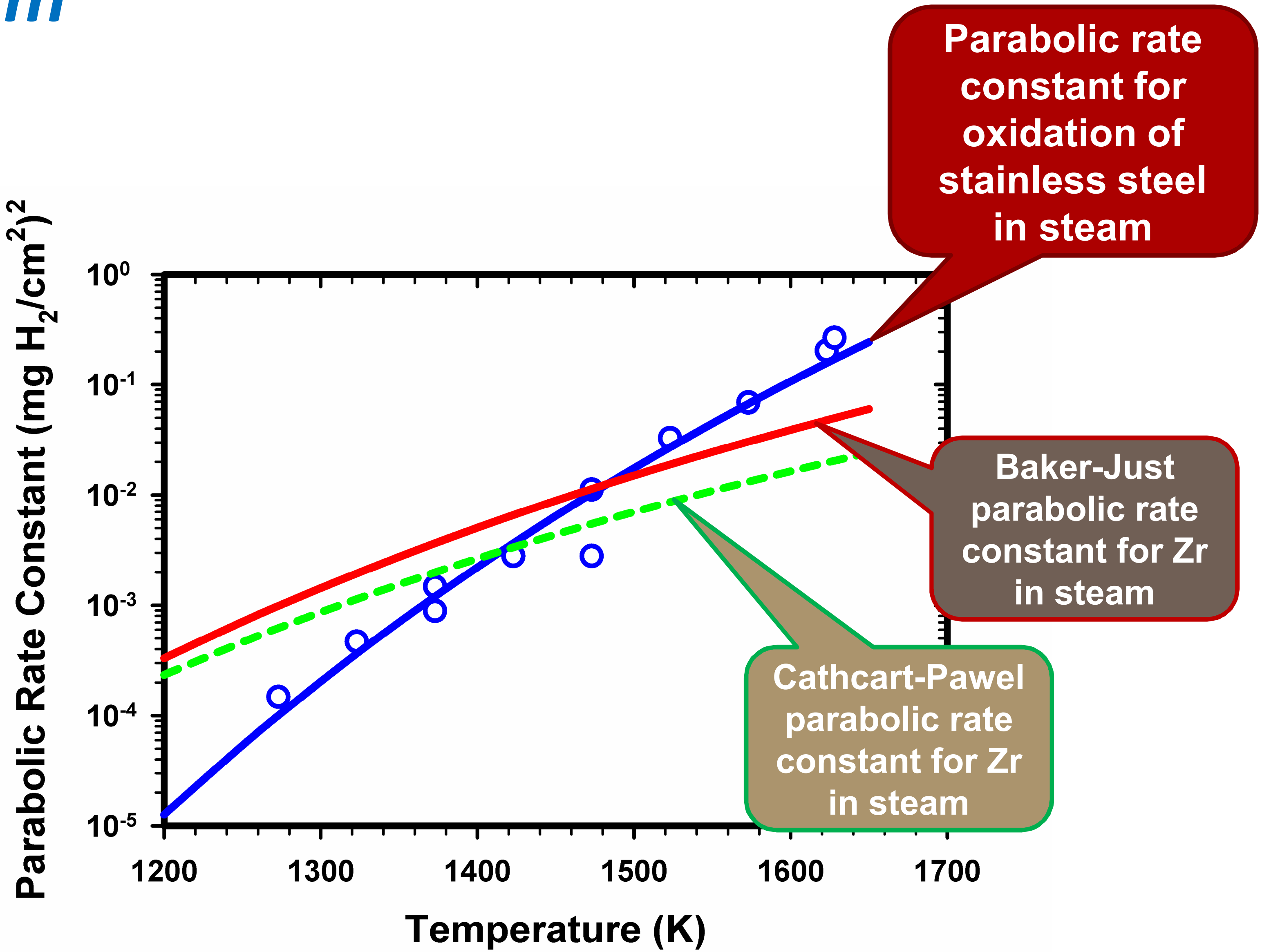
$$W_{Zr} = \sqrt{A e^{-B/RT} t}$$

  - W_{Zr} is mass of Zr oxidized per unit area exposed to steam at absolute temperature T for time t
  - A, B are empirically determined constants
  - R is universal gas constant

Authors	A g^2/cm^4-s	B J/mole
Baker-Just	33.6	190372
Prater Courtright	268	219835
Leistikow	4.26	174288
Cathcart Pawel	2.94	167121
Urbanik Heidrick	0.3	139800



Aside: Stainless Steel also Oxidizes in Steam

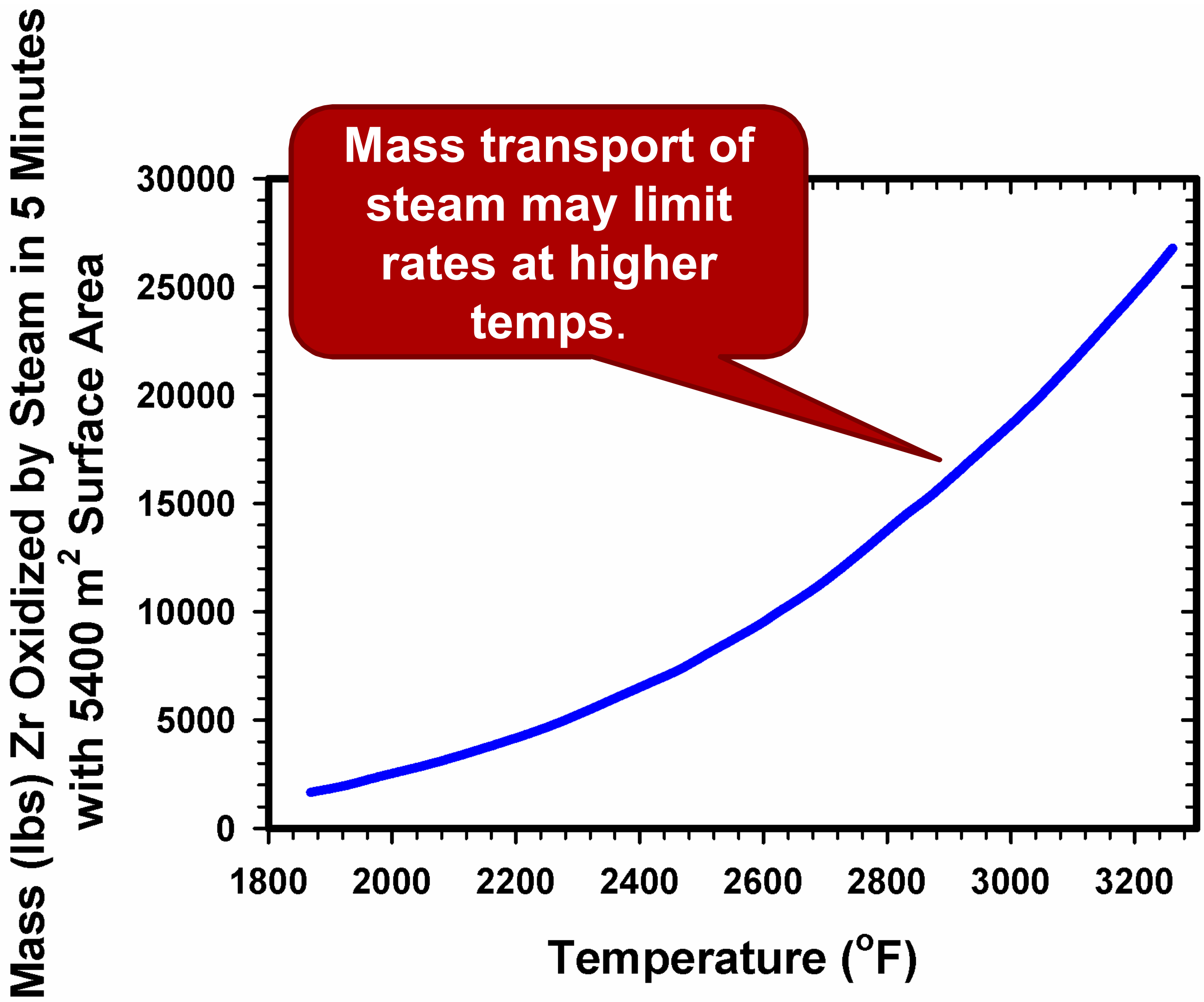


Zr Oxidation Energy

- *2.8x10^3 Btu/lb$_{Zr}$ (-6.5 MJ/kg$_{Zr}$, Exothermic)*
- *6 to 19 times decay power level in covered portion of core when steam limited*
- *Heat transfer from uncovered core to residual water would increase oxidation rate further*
- *Experimental confirmation*

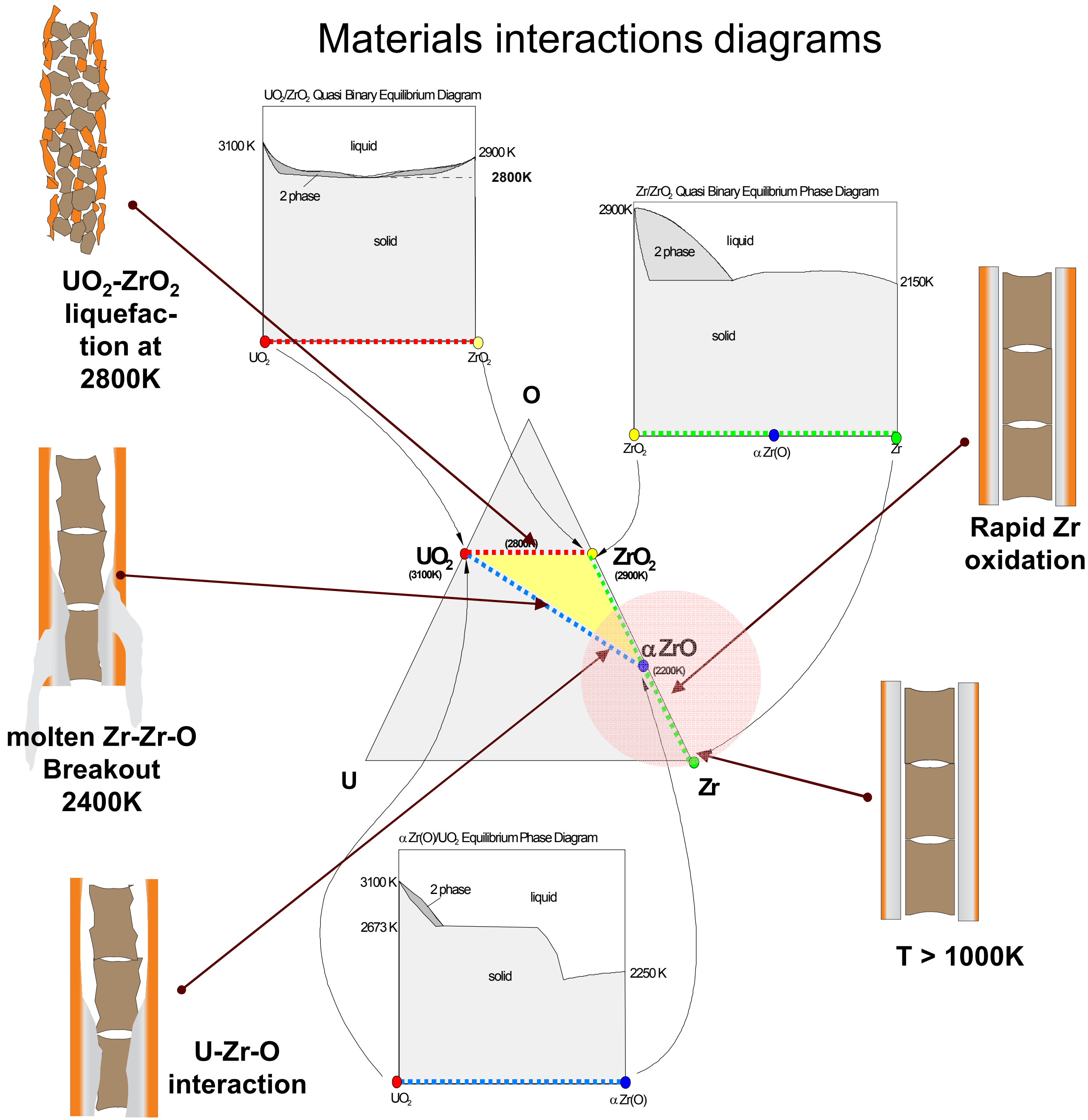
Example

- *Five minute exposure of Zr to 2192°F (1200°C) steam*
- *3,830 lb_m Zr oxidized out of 26,940 lb_m Zr in core (14.2%). Limit is 1% overall and 17% locally!*
- *170 lb (76.9 kg) of hydrogen released*
- *10.7×10^6 Btu (11.3 GJ) of energy released*
- *Idealistic*
  - *Core temperature not uniform*
  - *Energy release would increase Zr temperature*



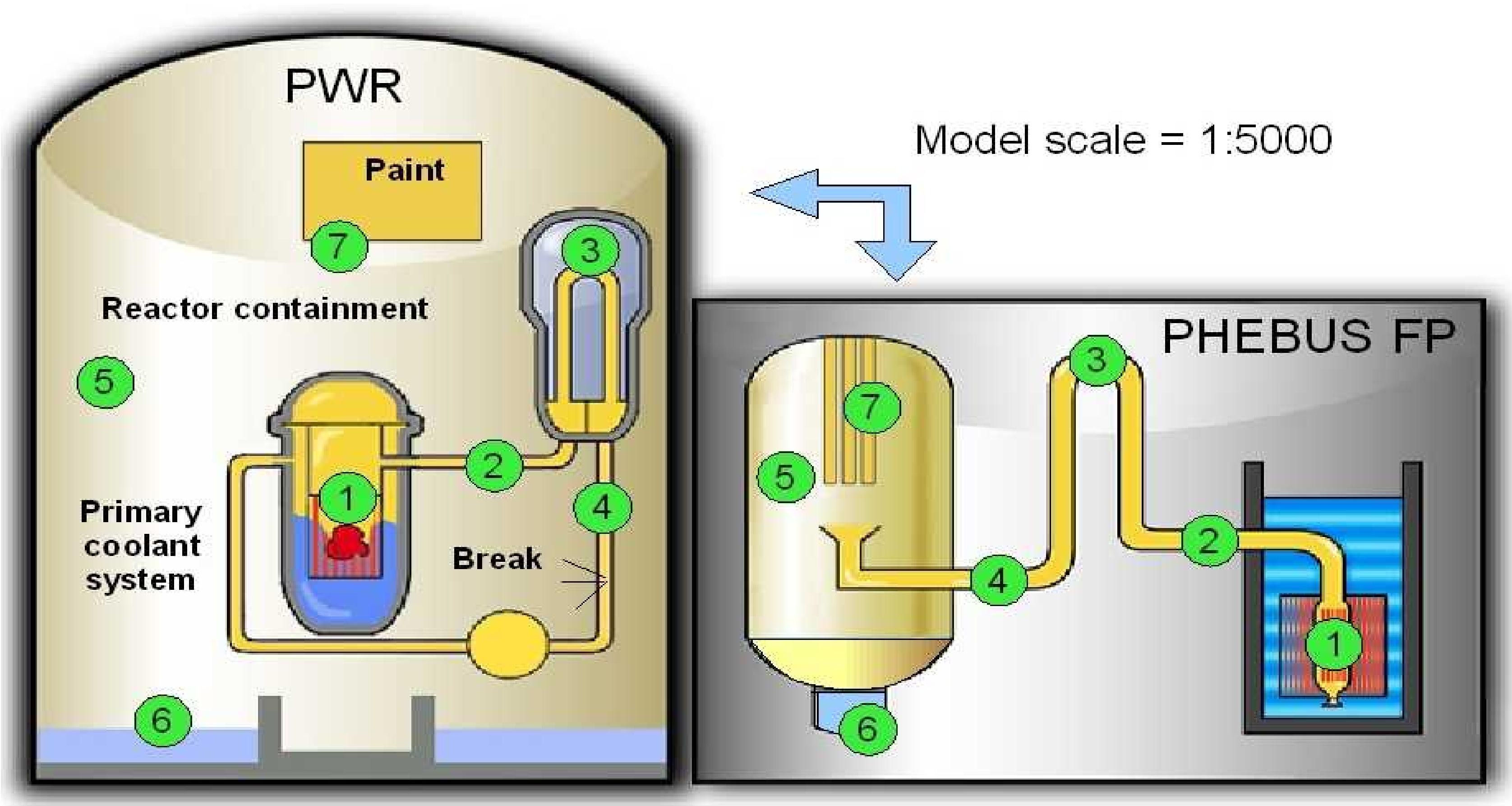
Mass of Zr oxidized in 5 minutes exposure of 5400 square meters Zircaloy

Materials interactions diagrams

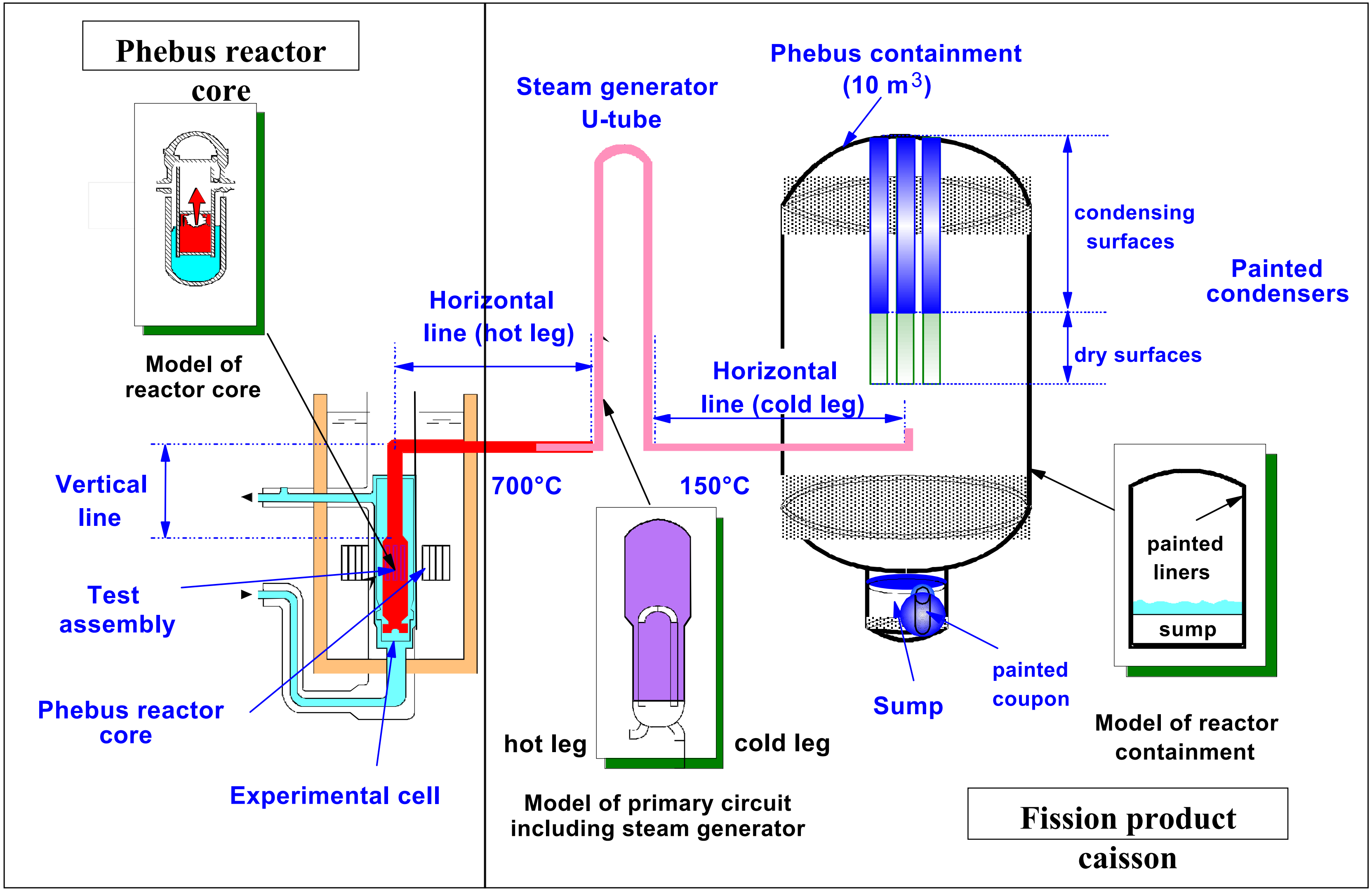


CLAD OXIDATION AND HYDROGEN GENERATION OBSERVED IN PHEBUS EXPERIMENTS

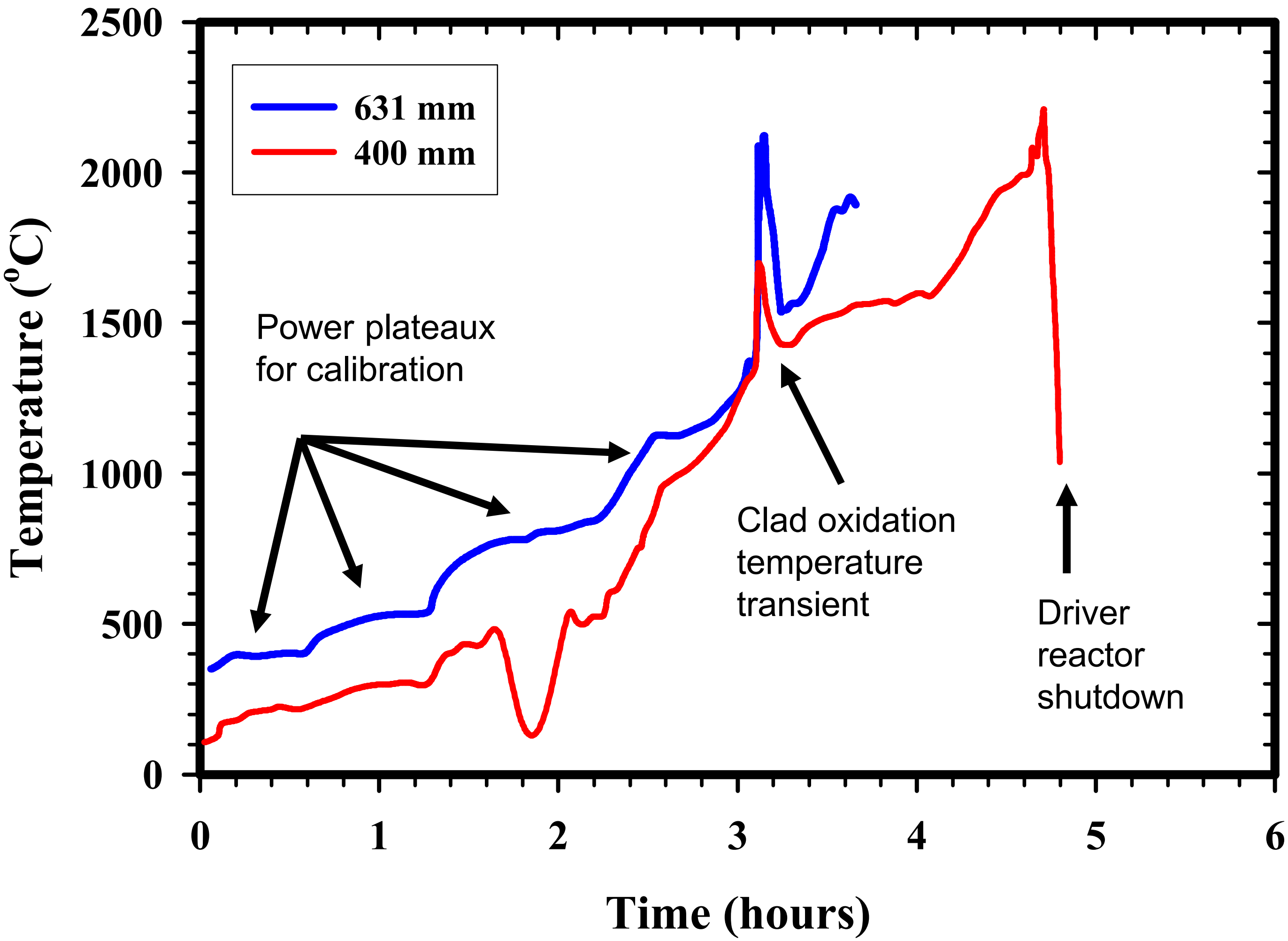
Scaling of PHÉBUS Experimental Facility



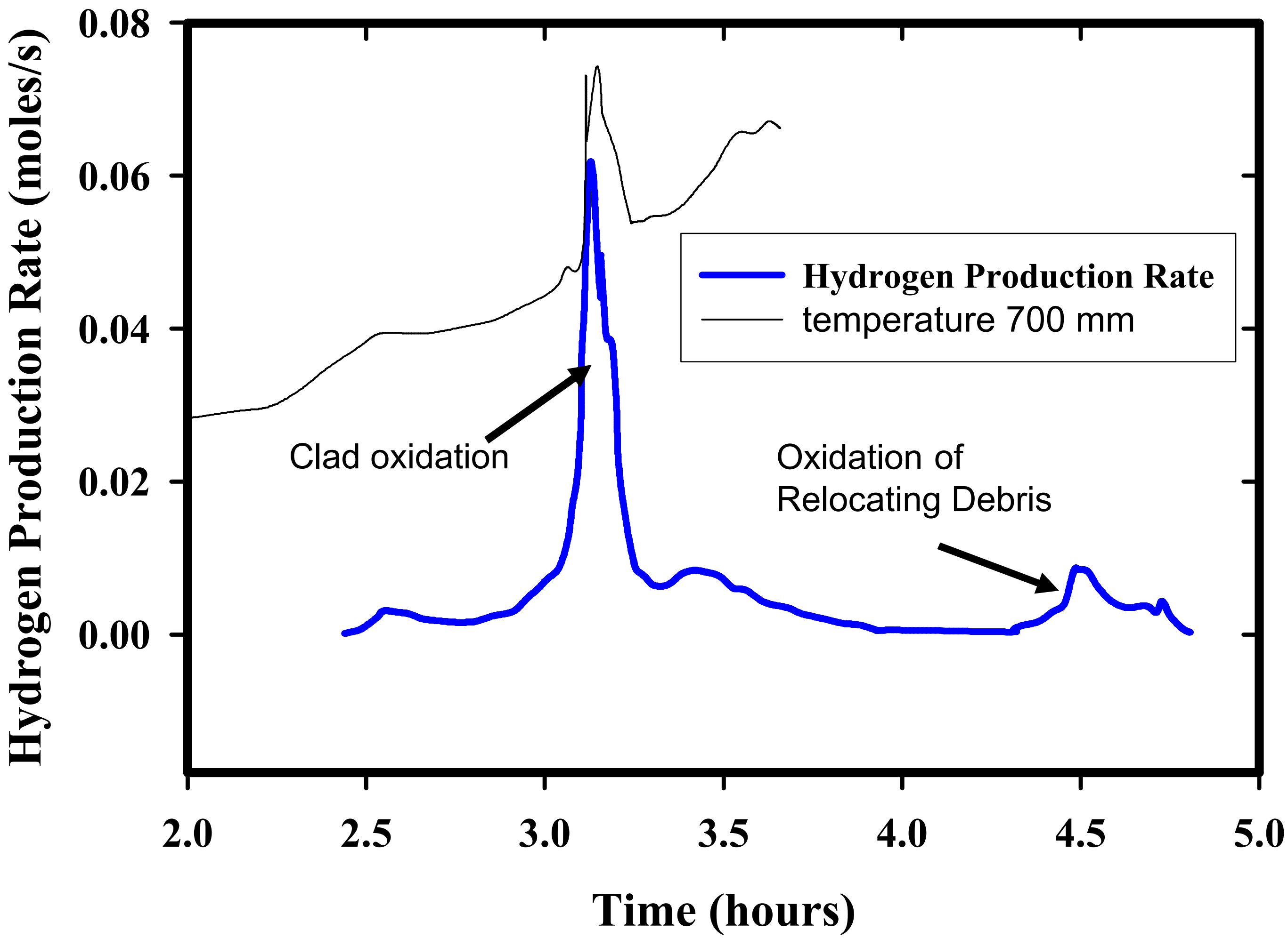
The Experimental Facility



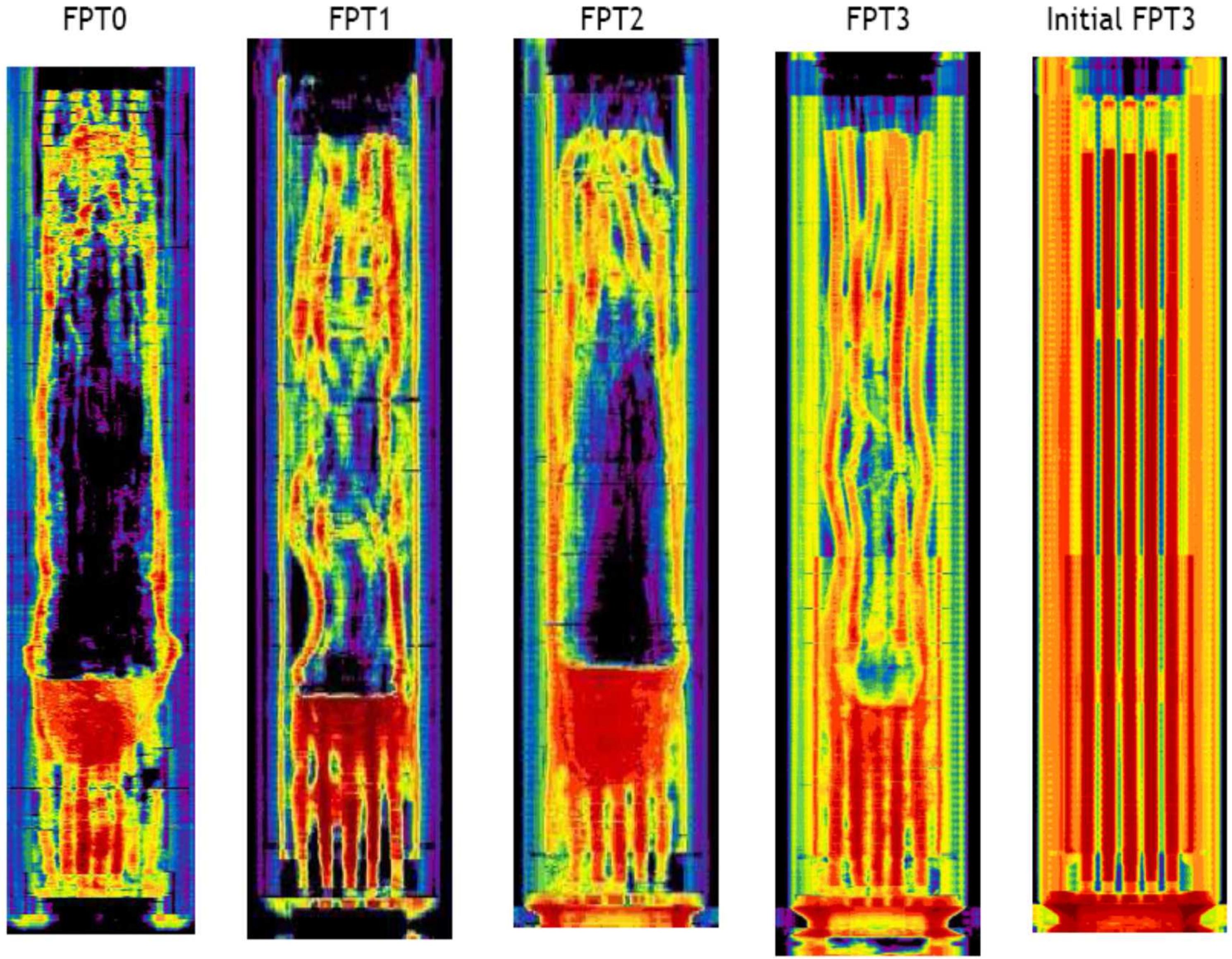
Temperatures at Two Elevations in the Fuel Bundle Used in Test FPT - 1



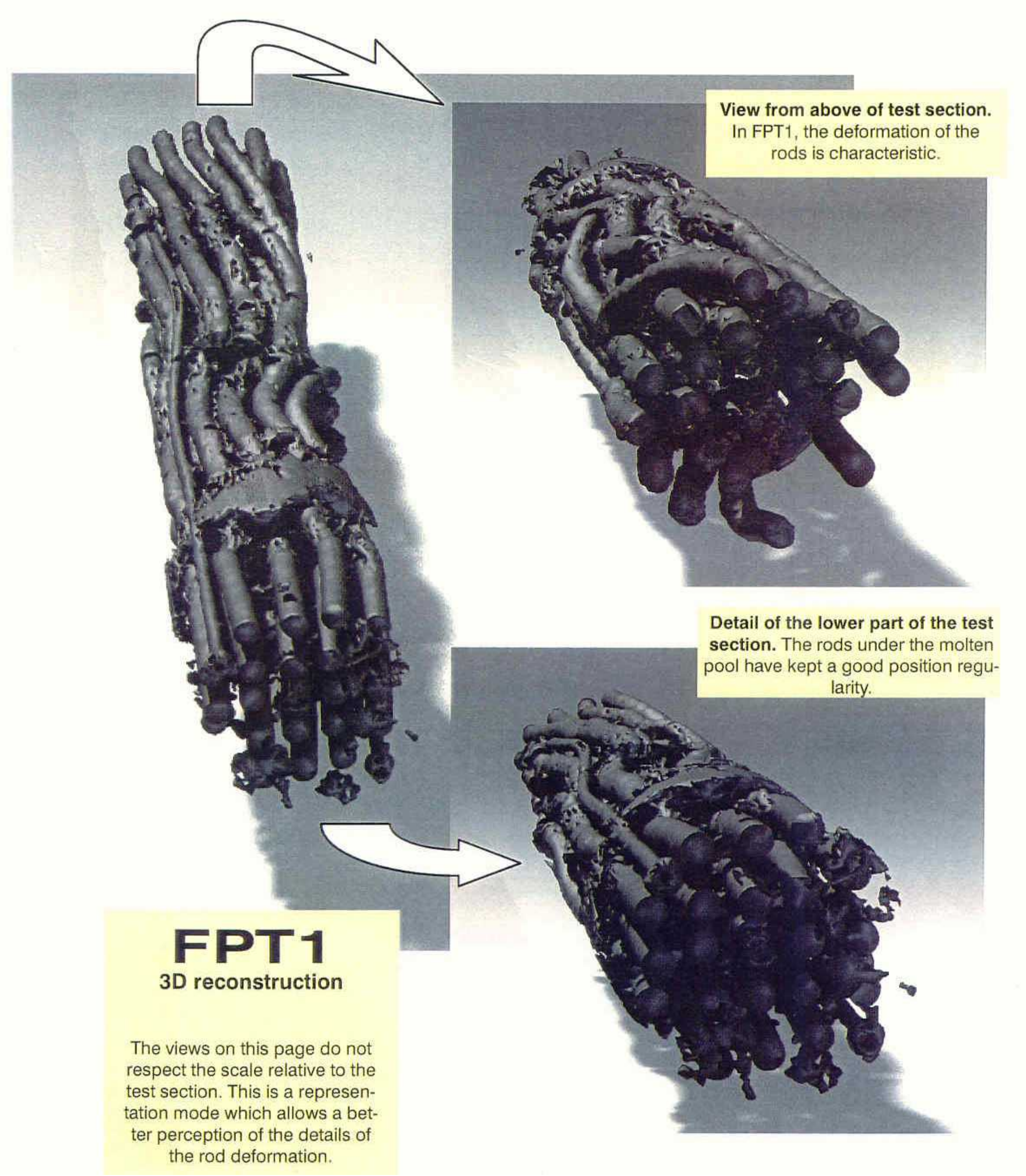
Hydrogen Production Test FPT-1



Examples of Fuel Degradation Observed in PHÉBUS-FP Tests

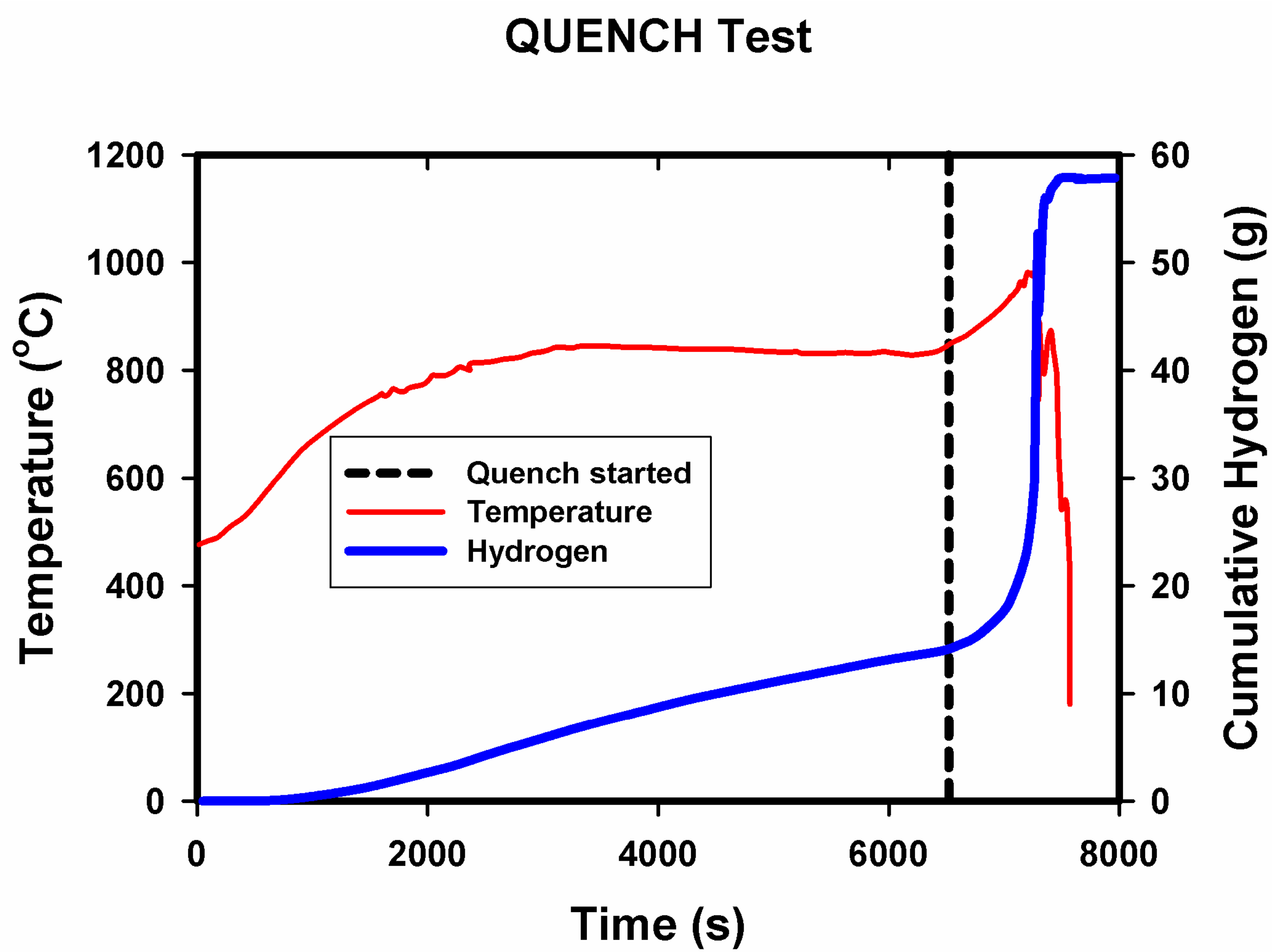


Oxidation Transient Drives Fuel Damage



- Tomography on FPT-1 bundle after fuel damage transient
- Zr oxidation drives severe damage
- Also drives thermal release of fission products

LATE HYDROGEN GENERATION FROM QUENCHING OR EX-VESSEL CONCRETE ATTACK



Hydrogen production becomes an issue when quenching core once degradation becomes extensive.

- MCCI experiment
- Decay heat liberates water from concrete
- Metals (Zr and steel) oxidize and produce H_2 and CO
- Exothermic energy from chemical reactions

RSC: 1/02

Combustible Gas Generation

- Water and carbon dioxide are released from concrete
- Oxidation of core debris yields hydrogen and carbon monoxide (both combustible)

  $Zr + 2\,H_2O \rightarrow ZrO_2 + H_2 + \text{heat}$

  $Zr + 2\,CO_2 \rightarrow ZrO_2 + CO + \text{heat}$

  $Fe + H_2O \rightarrow FeO + H_2$

  $Fe + CO_2 \rightarrow FeO + CO$

  Cr from stainless steel also quite reactive
- Large quantities of combustible gas can be generated
- Combustible gases may burn above the molten pool or may accumulate in containment
- Combustible gases contribute to total pressure in containment
  - Burns or static overpressure can challenge containment

HYDROGEN COMBUSTION

Hydrogen Combustion

$2\ H_2 + O_2 \rightarrow 2\ H_2O$ + energy (heat)

52,000 Btu/lb-mol of H_2 burned
(1 lb H_2 energy equiv. 26 lbs TNT)

Combustion Modes

- Deflagrations - subsonic
- Detonations - supersonic
- Continuous combustion
  - Diffusion flames or jets
  - High temperature recombination

Deflagration versus Detonation

	Deflagration	Detonation
Ignition	milliJoules empirical flammability limits	kiloJoules (or deflagration to detonation transition)
Propagation	Conduction Subsonic 1-1000 m/s	Shock Heating Supersonic 1500-3000 m/s
Loads & Structural Response	Static, Thermodynamic Bound	Dynamic Shock Waves Hard to Model (3D)

Deflagrations

- *Conditions required for ignition are understood*
- *Propagate by conduction from burned gas to unburned gas*
- *Burn front is subsonic relative to unburned gas*
- *Static loading, peak pressure*
  - *Depends on combustion completeness and heat transfer during burn, which are affected by:*
    - *Flame speed*
      - *Initial gas composition and state*
      - *Geometry and location of ignition source*
      - *Turbulence*
    - *Heat sinks*
  - *Bounded by complete, adiabatic, constant-volume combustion pressure*
- *The TMI-2 combustion event was a deflagration*

Containment Vulnerability To Deflagrations

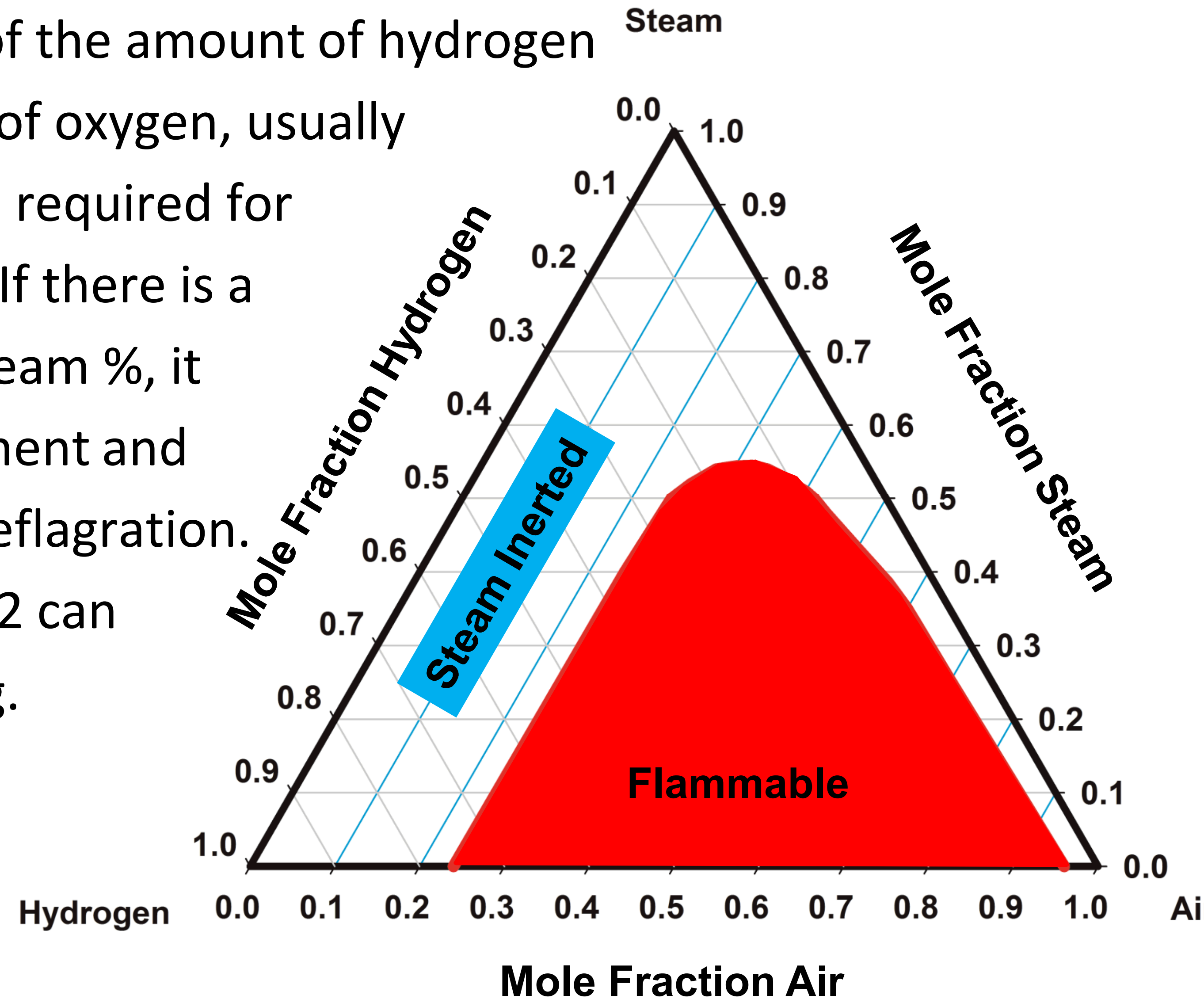
- *BWR Mark I and Mark II*
  - *Inerted containments*
  - *Surrounding reactor building not inerted*
- *BWR Mark III*
  - *High vulnerability of exceeding design pressure without mitigation*
    - *Larger free containment volume but lower design pressure*

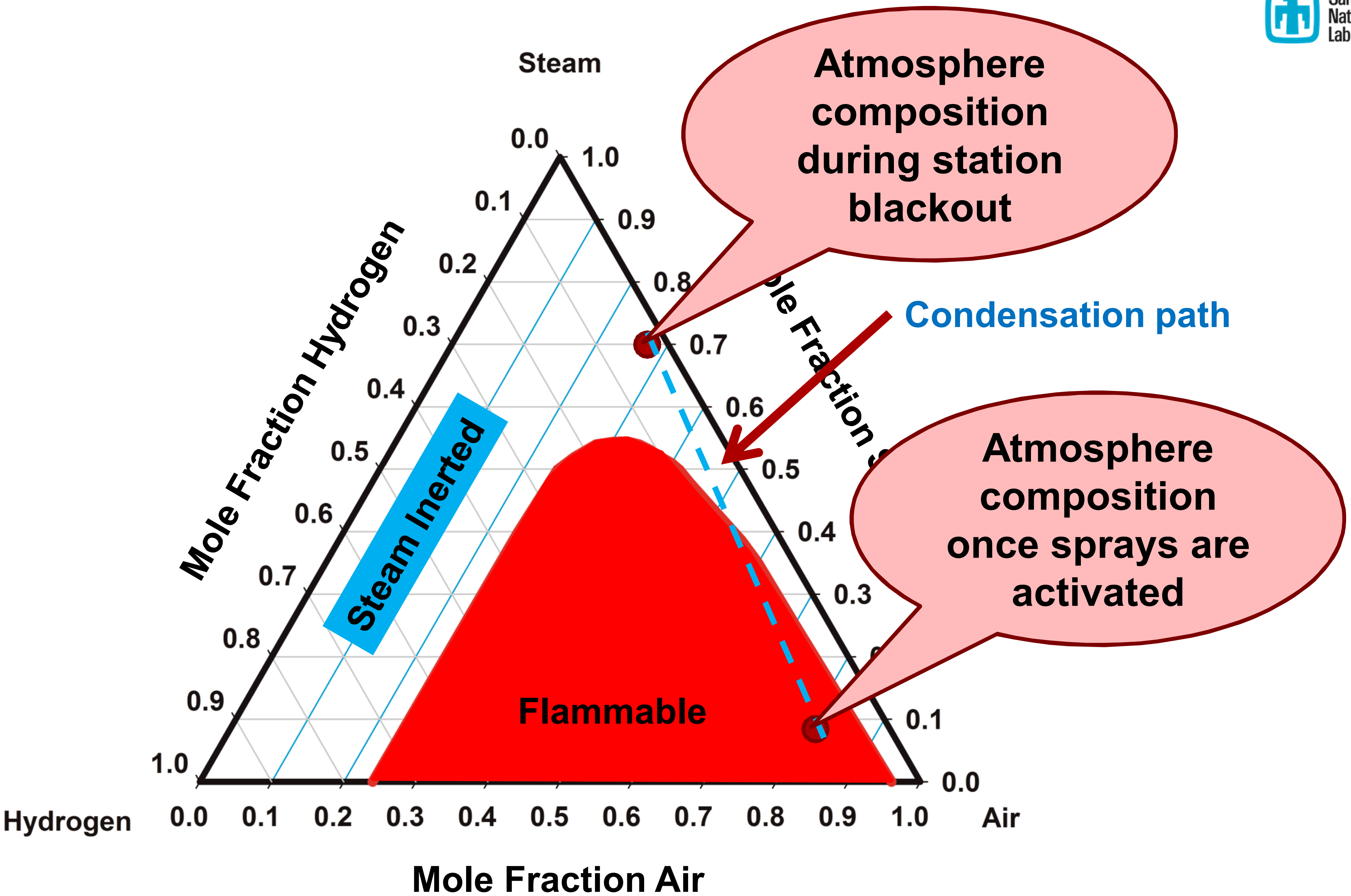
Hydrogen Flammability Limits

Direction of combustion front propagation	Hydrogen Concentration	
	Lower limit	Upper Limit
	%	%
Upward Propagation	4.1	74
Horizontal Propagation	6.0	74
Downward Propagation	9.0	74

Inertion

- Regardless of the amount of hydrogen some amount of oxygen, usually from air, is also required for a deflagration. If there is a high enough steam %, it inerts containment and prevents the deflagration. Nitrogen or CO2 can also be inerting.




Steam
Atmosphere composition during station blackout
Condensation path
Atmosphere composition once sprays are activated
Mole Fraction Hydrogen
Steam Inerted
Flammable
0.0
1.0
0.1
0.9
0.2
0.8
0.3
0.7
0.4
0.6
0.5
0.5
0.6
0.4
0.7
0.3
0.8
0.9
0.1
1.0
0.0
Hydrogen
0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0
Air
Mole Fraction Air

Hydrogen Detonation

- *Supersonic relative to unburned gas*
- *Propagates by shock heating of unburned gas*
- *Dynamic structural loads*
- *Detonations depend on both geometry and gas composition*
  - *No 'detonation limits' akin to deflagration limits*
  - *Limits for detonation quoted in older sources based on experiments with fixed geometry*

Other Detonation Concerns

- *Deflagration to detonation transition (DDT)*
  - *Turbulent flame acceleration*
  - *Contributing factors*
    - *Confinement*
    - *Geometry (obstacles)*
    - *High temperatures*
  - *Difficult to predict*
  - *Compression of gas by deflagration increases pressure when detonation finally occurs*
- *Local Detonations*
- *Missiles*

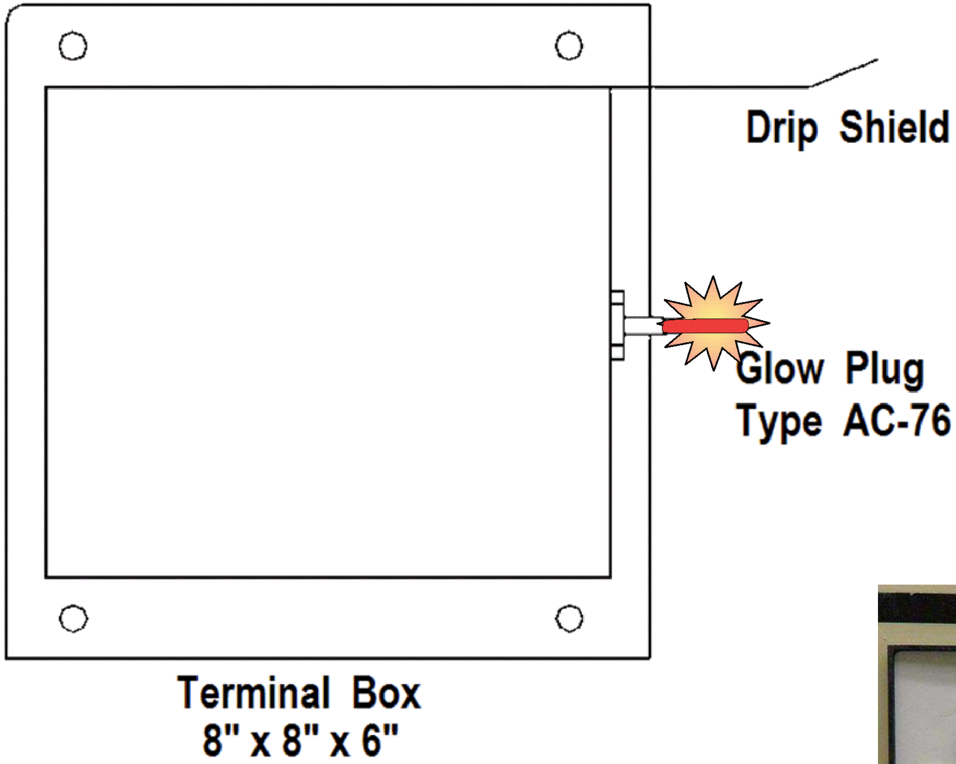
Locations Where High Hydrogen Concentrations May Occur (Assuming It's Not High Everywhere)

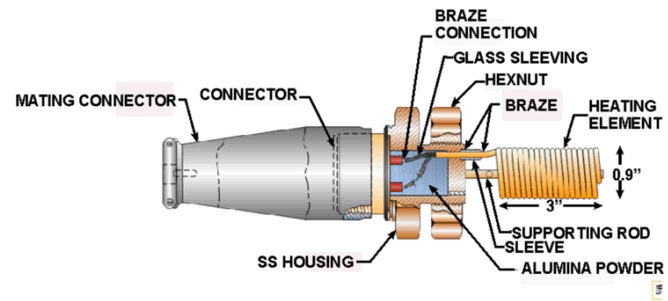
- *Near the hydrogen release point*
- *Under ceilings or dome due to rise and stratification of low density plume*
- *In areas where steam is being removed*
  - *Areas with large heat sinks*
  - *In or above ice condensers*
  - *Above suppression pools*
  - *Downstream of fan coolers*
  - *In containment with functioning sprays*

Hydrogen Rule (10 CFR 50.44)

- *Recombiners or other measures provided for hydrogen control in design-basis accidents would not be effective for severe accidents*
- *In 1981, BWR Mark I and II containments ordered to be inerted*
- *Additional mitigation required for BWRs with Mark III containments*
  - *Deliberate ignition systems (glow plug igniters) installed*
    - *Burn hydrogen when flammable at 5-7% before detonable concentrations are attained*
  - *Dedicated AC power for deliberate ignition systems*
    - *Not cost beneficial for plants with Mark III containment*
  - *Residual concerns: Very rapid H_2 releases, SBO*
- *Recombiners no longer need to be classified as safety-related*
- *Europe - PAHRs (Passive Autocatylitic Hydrogen Recombiners)*
  - *Recombine Hydrogen and Oxygen*

Hydrogen Igniter



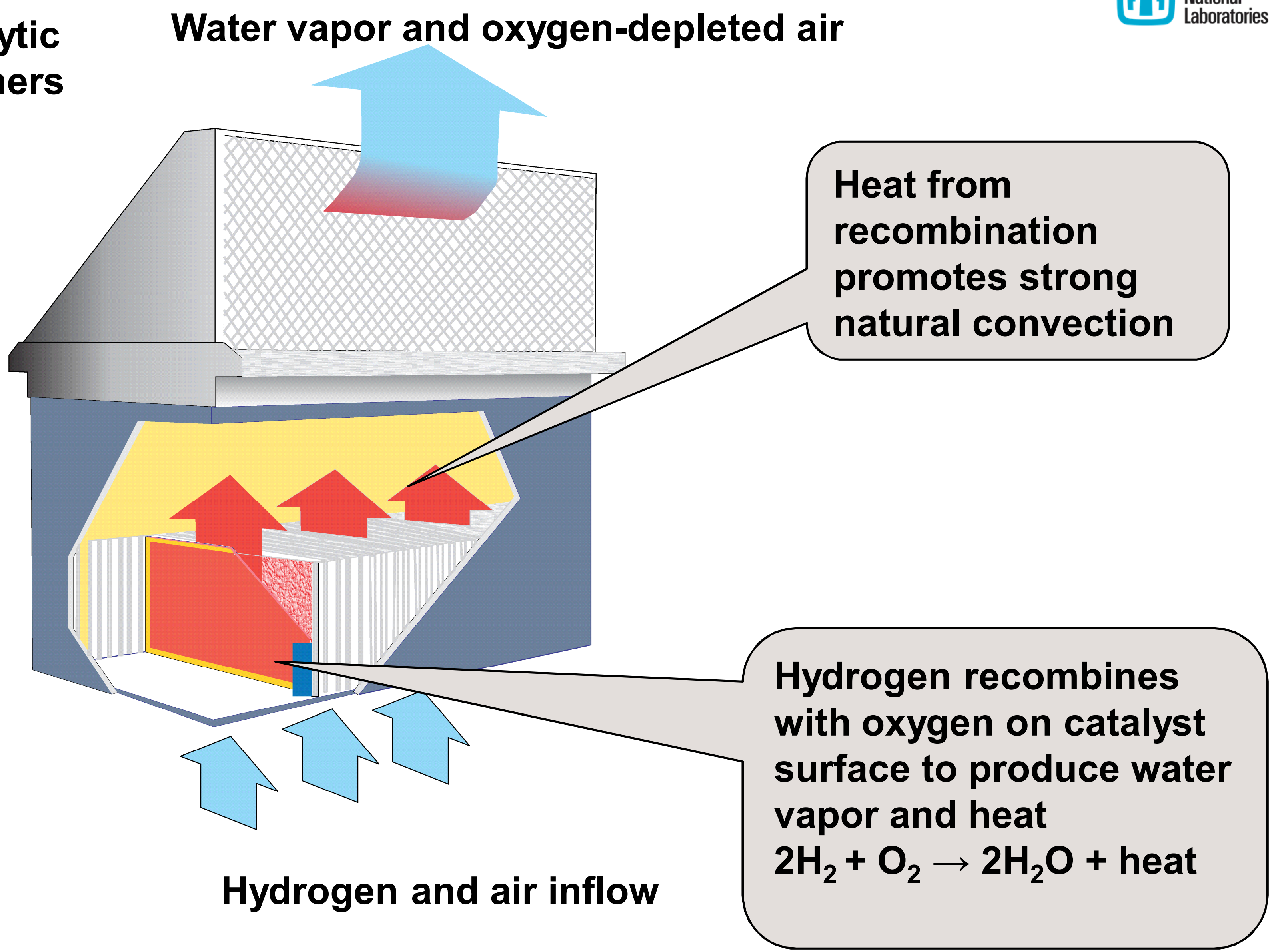


Glow Coil

1700 ^{o}F in 90 seconds

Passive Autocatalytic Recombiners

Sandia National Laboratories



Summary

- *Hydrogen is produced primary by oxidation of Zircaloy cladding, but also by oxidation of stainless steel and MCCI*
- *Oxidation reactions are the driver for fuel damage*
- *Hydrogen combustion one of the few regulated severe accident phenomena*
  - *Temperature threat*
  - *Pressure threat*
- *Different types of combustion are possible*
  - *Deflagration (TMI-2, Fukushima-1)*
  - *Detonations (Chernobyl ?)*
  - *Deflagration to detonation transition (Fukushima-3/4 ?)*
- *Combustion still a concern despite regulatory actions*
  - *Can threaten containment integrity by overpressure*

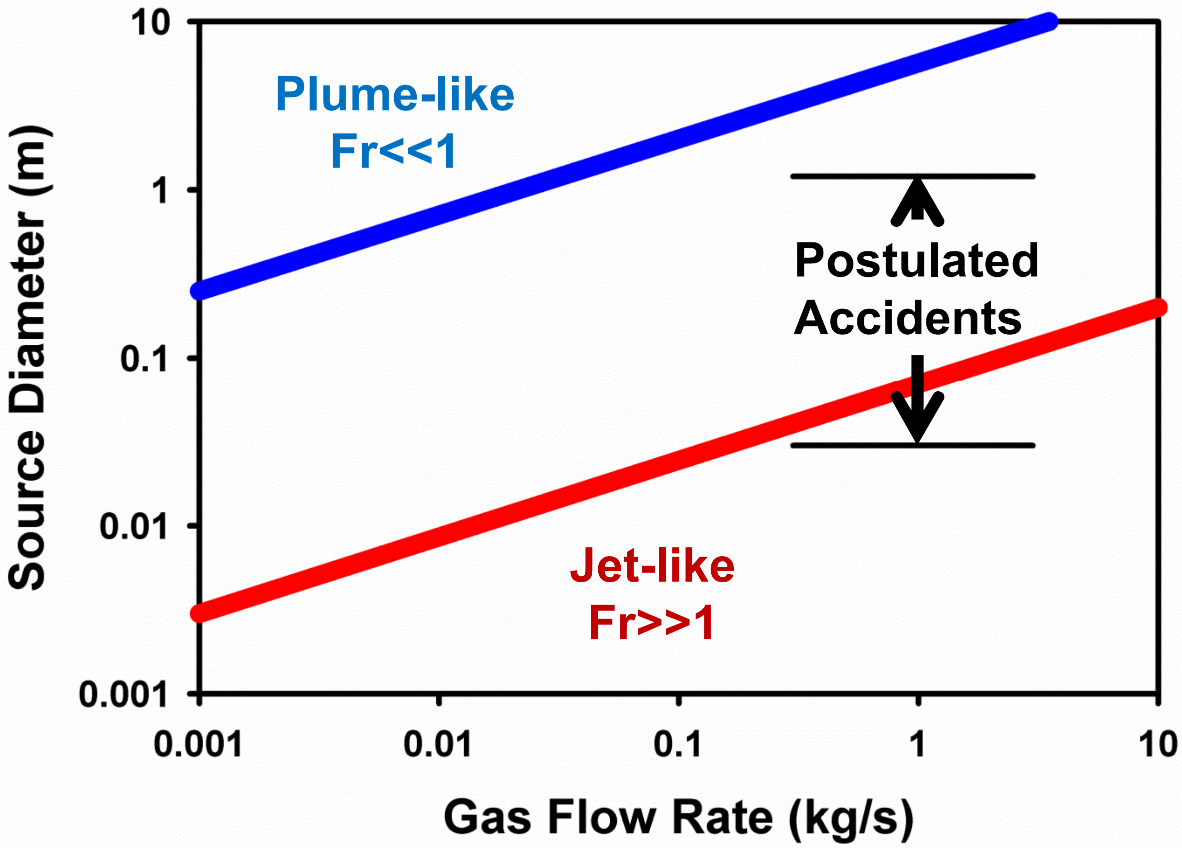
BACK-UP SLIDES

Diffusion Flames

- *Mechanism*
  - *Like a Bunsen-burner (buoyancy driven)*
  - *Like a jet or torch (momentum driven)*

- *Scenarios*
  - *Flame or jet as hydrogen flows from reactor vessel*
  - *Flames as hydrogen emerges from suppression pool*
  - *Low pressure rises but high local temperatures*

- *Key parameters:*
  - *Source gas*
    - *Composition*
    - *Temperature*
    - *Froude number (buoyant versus momentum driven flow)*
  - *Surrounding gas composition*
  - *Ignition source*
  - *Sustainability*


Plume-like
Fr<<1
Postulated
Accidents
Jet-like
Fr>>1
Source Diameter (m)
10
1
0.1
0.01
0.001
0.001
0.01
0.1
1
10
Gas Flow Rate (kg/s)

High Temperature Recombination

- *Mechanism*
  - *Spontaneous ignition temperature exceeded*
    - *See next figure*
  - *Recombination of H_2 with O_2 at hot surfaces*

- *Scenarios*
  - *Hot surfaces near hydrogen release point*
  - *Hot particles dispersed via high-pressure melt ejection (H_2 recombination with DCH)*
  - *Core-concrete interactions in dry reactor cavity if there is air available*

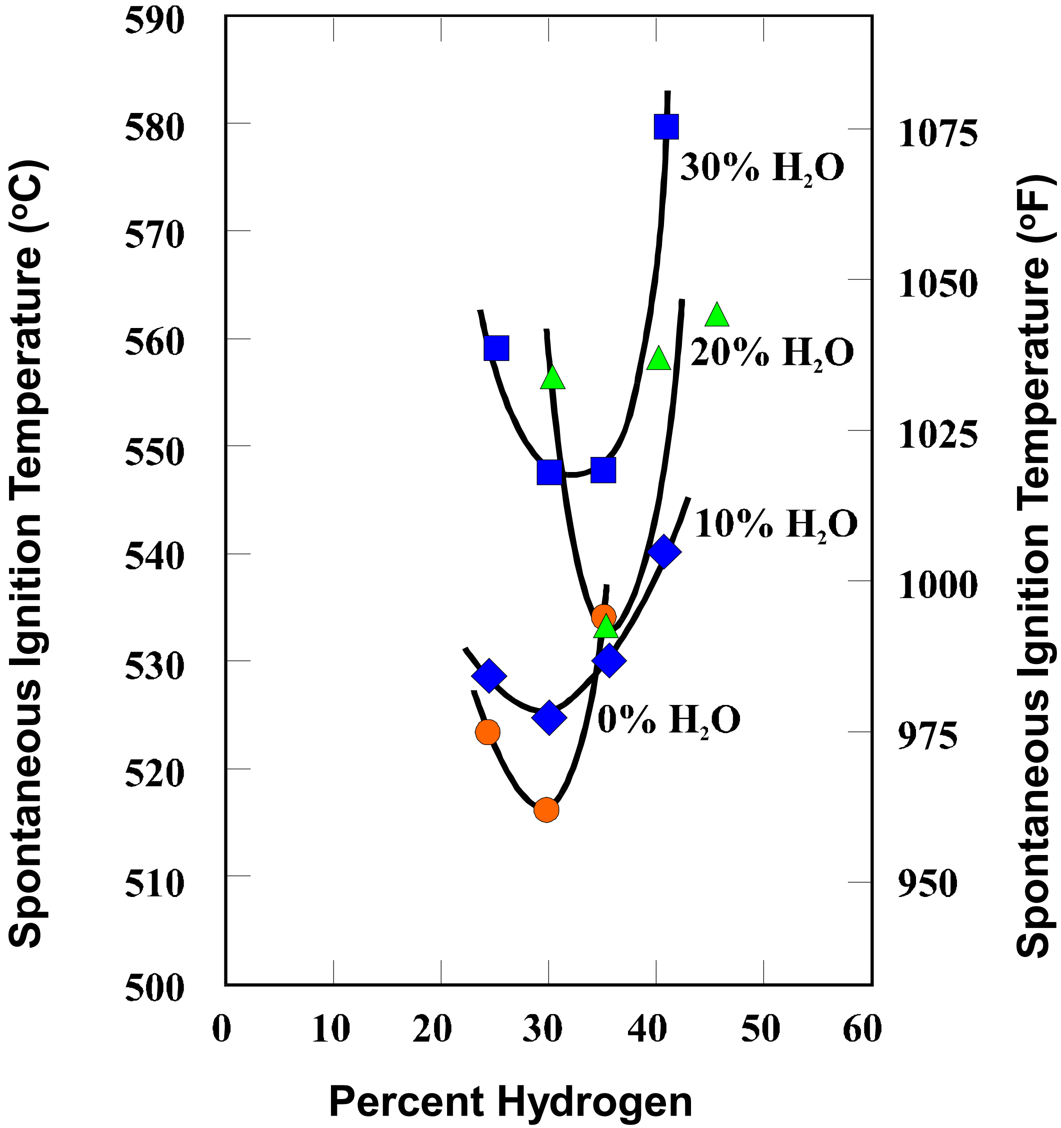
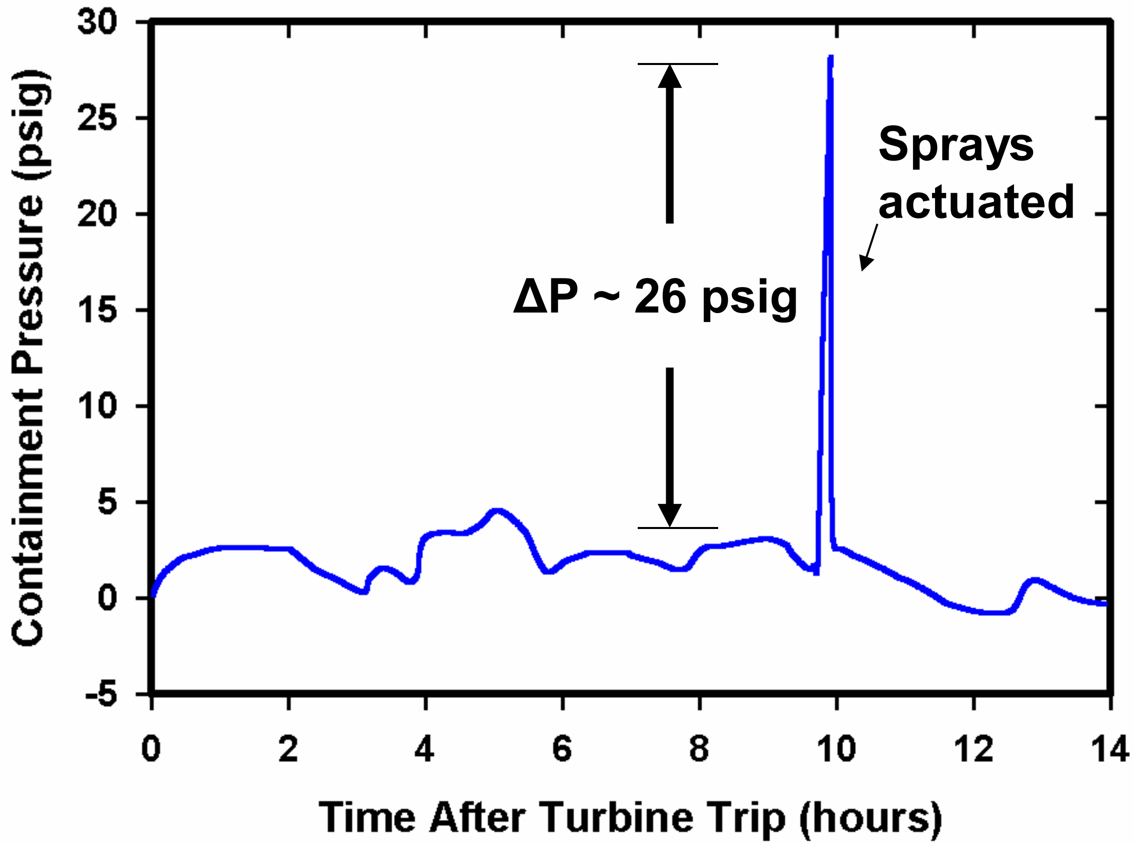


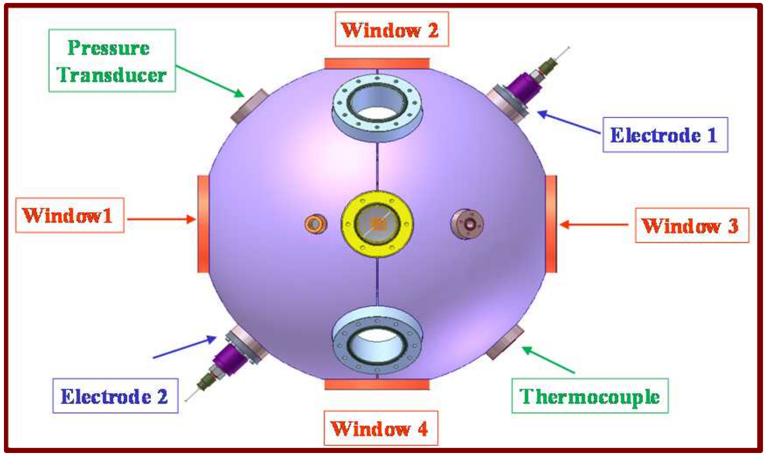
Figure 4.6-16 Minimum spontaneous ignition temperatures

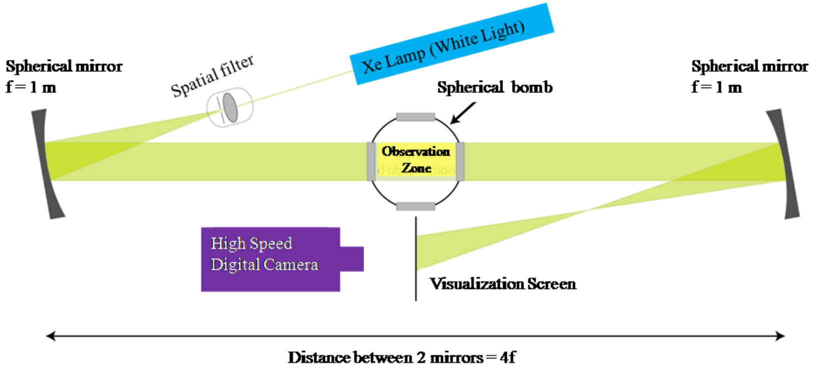
TMI-2 containment pressure versus time And the Hydrogen Burn

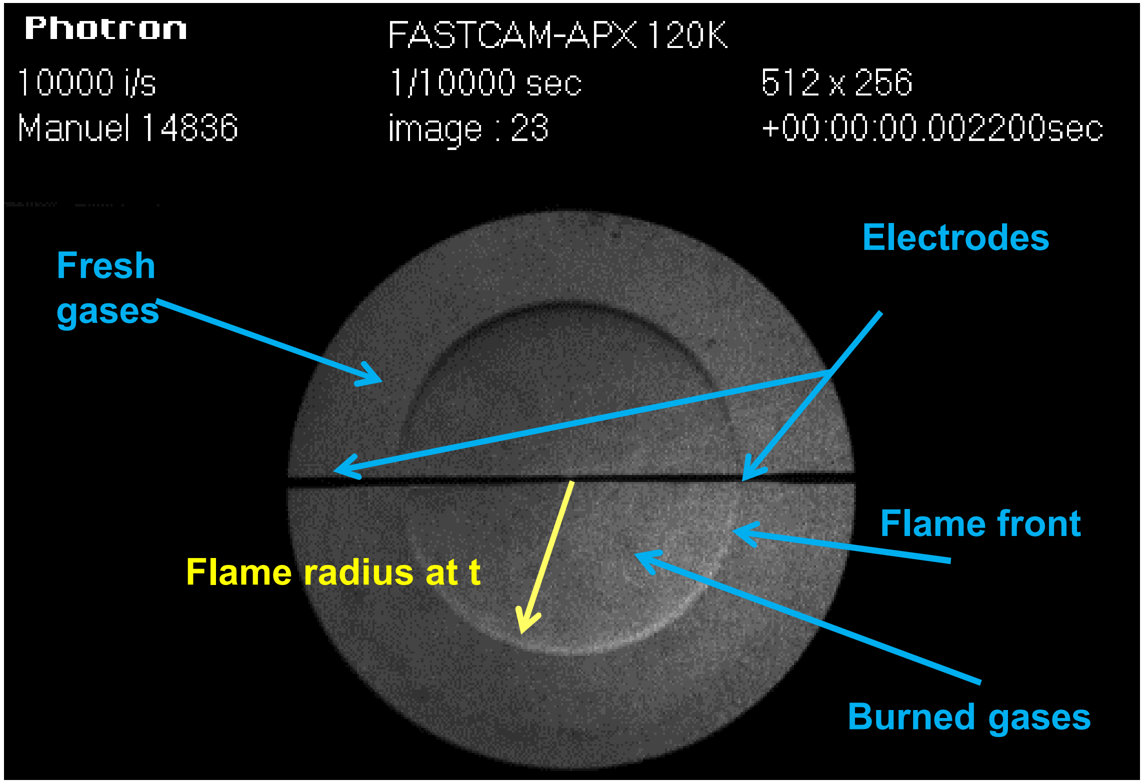


Combustion Limits Tests

Nabiha Chaumeix
Institut de Combustion, Aérothermique, Réactivité et Environnement
Orleans, France






Photron
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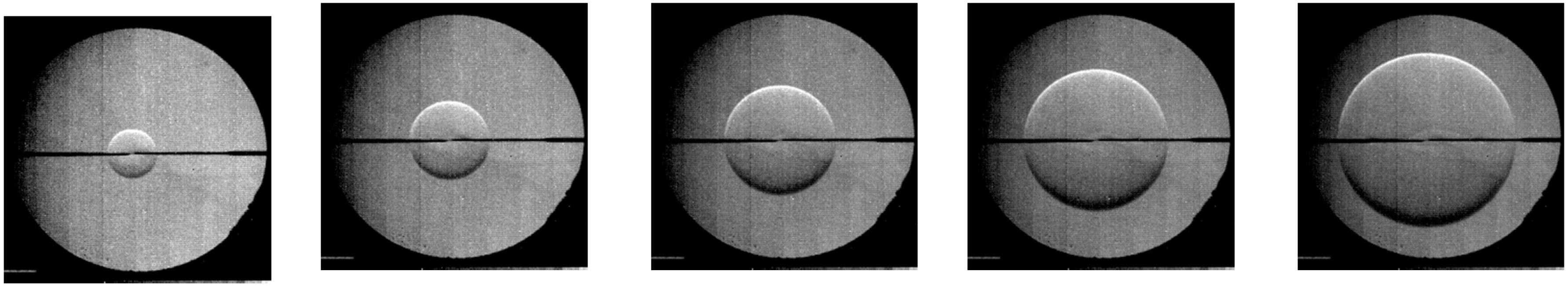
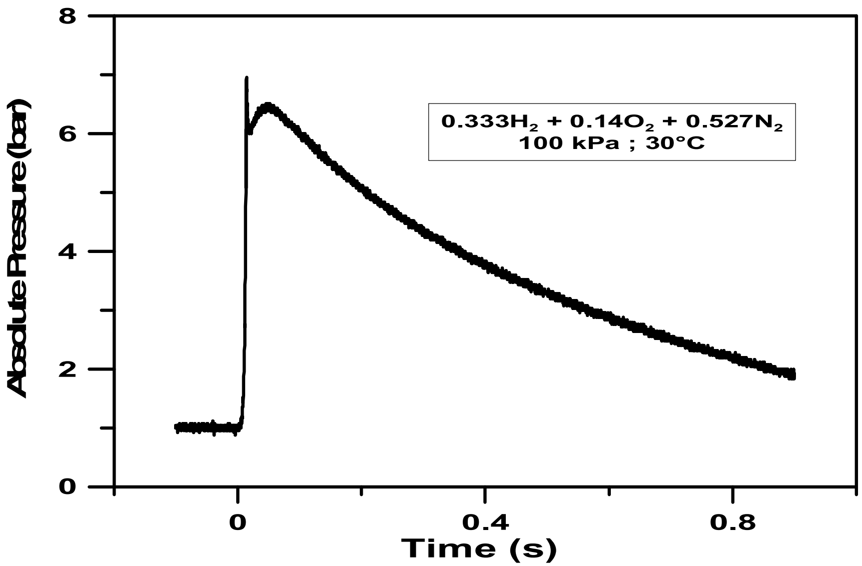
Fresh gases
Electrodes
Flame front
Flame radius at t
Burned gases

Flame Front Evolution with Time



Pressure versus Time



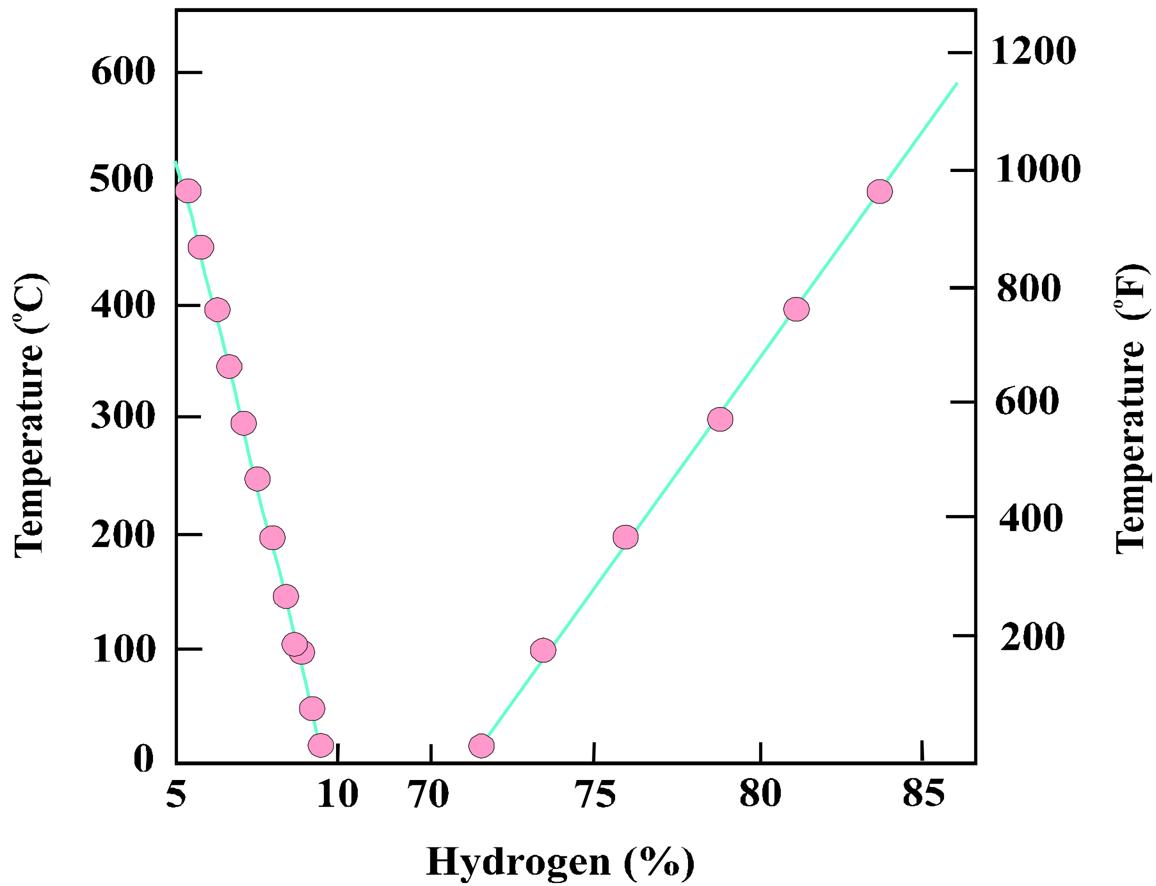


Figure 4.6-3 Effect of initial temperature on downward propagating flammability limits in hydrogen-air mixtures